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Urban Environment

Proceedings of the 10th Urban Environment Symposium

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Urban Environment

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Editors

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Proceedings of the 10th Urban Environment Symposium

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Preface

The 10th Urban Environment Symposium (10UES) was held on 9–11 June 2010 in Gothenburg, Sweden. UES aims at providing a forum on the science and practices required to support pathways to a positive and sustainable future in the urban environment. The UES series is run by Chalmers University of Technology within the Alliance for Global Sustainability (The AGS).

UES was initiated by Professor Ron Hamilton at Middlesex Polytechnic (now University) in the early 1980s under the title "Highway Pollution". The initial aim was to measure and assess challenges in highway pollution, with a strong emphasis on urban photochemical smog, ozone formation and particle release. After the first symposium, the emphasis on air pollution issues continued through to Munich in 1989 where diesel particulate issues and the relevance to health through measurements of PM10 emerged. The focus on air quality issues was also strengthened by the co-organisation of the symposium with Professor Roy Hamilton at the University of Birmingham from 1986 to 1998. In parallel, the symposium started to receive an increasing number of scientific contributions from the area of urban run off, indeed to the extent that the title of the symposium was changed to "Highway and Urban Pollution". Also at this time the importance of science in support of policy was emerging as a key aspect of the symposium.

The 8th edition of the symposium was marked by an organizational change with Chalmers University of Technology taking over the organization of the symposium. At this stage, we decided to evolve the name of the symposium to "Highway and Urban Environment" (HUES) to provide a positive view of the challenges in the urban and roadside environment. That said, papers addressing pollution issues remain a central part of the symposium as they help to raise awareness around the issues to be solved. For the first time, the proceedings of the symposium were published as a book in the AGS book series. This continued at 9HUES in Madrid in June 2008.

The 10th edition of the symposium was hosted at our home university, Chalmers University of Technology in June 2010. The 10th symposium was marked by a further evolution of the name, with the term "highway" being dropped. With this change, we hope that the name of the symposium will better reflect its aim.

We would like to take this opportunity to thank all who have contributed to the success of 10UES. We would especially like to acknowledge Alexandra Priatna and Jessica Olausson at Chalmers' AGS office whose organizational skills were essential to the success of this symposium.

The next symposium, 11UES is planned for September 2012 and will be held in Karlsruhe, Germany in collaboration with the Karlsruhe Institute of technology (KIT).

Göteborg, Sweden

Sébastien Rauch
Gregory M. Morrison

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SUSTAINABLE URBAN DEVELOPMENT AND URBAN PLANNING

Transport and environmental regulation – common attitudes and social change

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Abstract

Since the Kyoto protocol in 1997, signatories States attempt by appropriate policies to restrict the traffic in urban space, considered as the first source of greenhouse gas emissions. On the scale of metropolises, the elected representatives borrowed in this policy and the organizing authorities of the transport, elaborate limitative measurements to show their environmental policy. The main stake of environmental policies is the social acceptability. Or, the behaviour change, depends of collective social attitudes, are produced in a comparatively long process. Two approaches are in work in the urban field and especially about urban transportation. The first approach is more technical. It arrests urban policies as being plans of action in a shorter time scale. It leans more on technical choices as main support of urban policy. The second heads rather with the sociological and cultural dimensions of urban phenomena. It takes into account social links, social organization and specific cultural values in every society or a community. Both approaches crystallize academic scientific disciplines dividing up between, the sciences of environment and engineer on the one hand, and human and social sciences of other one. Our objective is to show foundations and borders of both approaches and try to reconcile both acceptations in what we call interdisciplinary approach.

Introduction

The environmental question is in the centre of public policy and public attention. It has become an important paradigm structuring the political, scientific and social spheres in the last three decades. We fear for our common future (natural resources, atmospheric pollution, physical and

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mental health of world inhabitants…). This context has an effect on regulation tools in the urban space. In the field of transportation, of sorting of waste, of production and consumption of goods, of managing natural resources, we attempt to pay more attention to our natural environment in order to respect it. While most solutions have a technical dimension, they are conceived without taking social acceptance into account. We design solutions before looking for a social consensus. It is evident that our entire technical and material environment depends on us joining in the process and adopting it. The success of any public policy means adhesion of social forces. This paper focuses on the analysis of the gap between the two spheres, social logic and technical logic and their fundamentals. The field of research is urban transportation.

Environment and urban mobility

In spite of the close link between economic and social development and the increase of travel, the transportation sector is responsible for nearly a third of greenhouse gasses (GHG) emissions which are increasing rapidly. Emissions of carbon dioxide (CO2), the main greenhouse gas (GHG) produced by the transportation sector, have steadily increased along with the travel, energy use, and oil imports too. Urban transport policy aims at creating sustainable urban mobility through legal tools and technical solutions. The investments increase progressively so as to develop a public service in urban transportation. In France, some of these measures are part of what we call a PDU or urban transportation plan. Cities with more than 100 000 inhabitants must elaborate this PDU in which they commit themselves to reducing their GHG emissions by reducing the use of personal vehicles and encouraging the use of public transportation modes (bus, tramway, train...). These goals reveal a proactive aspect in sustainable development policy. Urban mobility means that transportation must respect the objectives of sustainable development. We give some examples of investments such as adjusting the layout of urban roads (one way/two way roads), developing toll urban parking, social pricing of tickets, creating new tramway lines...

The social challenge

The principal challenge of this policy, via such as tool as the PDU, is to encourage city dwellers to do without the personal car and to use more public transport. Most PDUs were developed and approved in the past decade (2000–2010). Since the “solidarité et renouvellement urbain”

(SRU) law [1], the evaluation of a PDU is an obligation. The authorities in charge of organising urban transportation are obliged to carry out an environmental assessment to see if their policy reached the objectives of reducing the emission of greenhouse gas and if there are more users of public urban transportation than before or not[1]. However, scientific reports on these evaluations, have pointed out the difficulty of demonstrating systematically a relationship between urban transportation policy and environmental goals. At the same time, the car is still the most important mode of transport within the urban space.

Despite the proliferation of restrictive measures and tools for the regulation of car use in cities, the results of these policies remain below the expectations of organizing authorities and experts in urban transportation, in terms of ambition for better protection of the environment and in particular of reducing emissions of greenhouse gases. In several recent studies, it was shown that personal vehicles remain the dominant mode of travel in the city compared to all other modes. This negative assessment puts into question the effectiveness of public policies in urban transport linked to the objectives of "sustainable mobility". Some "politically manufactured" statistics show some evolution in the practices relating to the use of the car but this does not reflect a significant change. The dilemma of urban transportation policy is the gap between aims and results. Our hypothesis in this article is to explain this gap by the fundamental opposition which exists between the technical logic and the social one.

Background of policy makers

The urban transportation policy is designed according to a purely technical logic from a fundamental idea: rational choice theory. The rational choice theory is based on the rule of the rational individual. According to this approach, individuals are rational actors and act as if balancing costs against benefits to arrive at action that maximizes personal advantage. Indeed, social action is explained in terms of the rational calculations made by self-interested individuals. The rational choice theory sees social interaction as social exchange, modelled on

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[1] For example the Eval-PDU project (2009-2012), financed by the National Agency of Research (ANR). The project objective is to develop an optimized methodology to assess the environmental impacts of urban mobility plans (UMP, in French PDU), taking into account their social and economic consequences. With an interdisciplinary approach, the scientific researchers elaborate together indicators to measure the policy and the goals in an environmental dimension.

economic action. People are motivated by the rewards and costs of actions and by the profits that they can make. With this approach, the rational choice theory cannot explain the origins of social norms, especially those of altruism, reciprocity, and trust. This theory did not cease showing its limits vis-a-vis the weight of social determinants such as age, status, social position but also social structure. Any individual is registered in one social organization, having a particular trajectory and carries the characteristics of his social origin but also of their own past experiences. This inscription in the body and the spirit, of the standards, the values and the cultural and these religious precepts, plays an important role in our daily choices. About the collective behaviour, we are faced with diverse kinds of non rational actions. How we can say that social attachment to a car, observed in many cities and different societies, is the sum of rational individual behaviour? Sociologists considered that the individual is modelled by society as well as these manners of view are socially constructed in a long process beginning at birth. Norbert Elias wrote his famous essays: *The society of individuals* and he argued that individual interiorized constraints and differentiation of society do define the behaviour of individuals.

But on another side, we can say that individuals today, in era of more of freedom, of new technologies of communication and information, of less of social control, more choice and chance, have the possibility to do what they like. Some contemporary sociologists note that "the society" does not determine any more the social conduits as straightforwadly as before. Society is more fluid and less rigid in the structures which compose it [2]. Therefore, social acceptability is not guaranteed. All social adhesions are produced within a social process. It's not possible to have social acceptance automatically because it is a collective action. The urban sustainable mobility is a new kind of society here all categories act jointly. It is a collective social consensus. It is not possible to assume that a new tramway line will have the same social sense in different cities or in different kinds of societies. The social uses of techniques are determined by social culture and social history. It depends on social representations of the technical environment. If the car in some societies is just a way of going from one place to the next, in other societies it is a way to show social status. Indeed, we can't have the same consequences for a given public policy. Michel Crozier titled his famous book *We can't change society with a decree* [3].

Rational and non rational actions in sociological theory

The social rational or irrational behavior is at the heart of sociological theory. If we examine Weber's theory, there is a different kind of social action whose reveal different fields of references. Weber's emphasis on the subjective meaning that actors attach to their action implied an individual focus, often described as action theory. He distinguished four ideal types of social action: *traditional action*, justified as a repetition of the past; *affective action*, geared toward the expression of emotion; *value-oriented action*, in which the performance is taken as an end in itself; and *instrumental action* or means–ends rationality, in which actors pursue their economic or other interests. Any specific action might involve one or more of these rationales [4].

In Bourdieu's theory [5], concept of *habitus*[2] means all unconscious and sustainable dispositions accompanying each individual from birth to death. Each individual interiorize these dispositions, in socialization process, in response to objective conditions within which he lived. A large part of the concept of *habitus* is that it brings attention to the fact that there are limitless options for action that a person would never think of, and therefore those options don't really exist as possibilities. In normal social situations, a person relies upon a large store of scripts and a large store of knowledge, which present that person with a certain picture of the world and how she or he thinks to behave within it. A person's habitus cannot be fully known to the person, as it exists largely within the realm of the unconscious and includes things as visceral as body movements and postures, and it also includes the most basic aspects of thought and knowledge about the world, including about the habitus itself.

Then we can act for other reasons which are not rational. The religious actions like example. Some of our daily reactions are linked to love or hatred in fact related to our emotions. If the rationality it means the interest, this interest not systematically economic, it may be symbolic, social or religious. If we applied this approach in the field of transportation policy, we can explain the social resistance who's appears as non rational. But there is others rationalities characterizing the choice of modes of moving. Indeed, the hypothesis that public transportation it is accepted by all social categories is not evident. Many people are more attached at their car than public transportation mode. The personal vehicle

[2] Habitus is defined as a "system of sustainable and replicable rules, structured structures arranged to function as structuring structures, that is to say, as a principle generating and organizing practices and representations that can be objectively adapted to their purpose without assuming conscious aiming for express and control operations necessary to achieve them".

guarantees freedom and comfort. The individuals are very attached at comfort of their lifestyle and conceive the car as private space in which they realizing their personal freedom. Indeed, use car not automatically the alternative of absence of public transport. This social phenomenon reveals the attachment to the car as tool permitting more comfort and flexibility in daily travelling. Therefore, the fundamentals of social action and the goals of urban transportation policy are not coherent. We will see in following lines how the two logics are different.

Technical logic and social logic

The main important objective of sustainable mobility is to encourage urban people at using more public transport. Then the principal stake is to change society, change social behaviour. We hope so that all these investments going to make urban transport conditions more favourable to urban mobility and in fine give up personal vehicles. The investments have mostly a technical nature. Reducing urban space for parking car, modified road direction or created a new tramway is a technical change. We can ask now if it is possible to change society justly with technical innovations.

Anthropologists of techniques such as Robert Creswell [6] demonstrated that is possible after introducing a new technical process to produce social change. On the other hand, the opposite is possible: social change may have effects on the technical system. But the nature of social change is not known before. We can't program the social change. It depends of the effect of technical innovation: the scale of innovation, the social organization system, the social culture, the technical level and social history of inhabitants. Jean-Pierre Digard, anthropologist of techniques too, said that "The technical facts thus are by no means isolated but belong to a coherent technical system, itself not separable from a social and cultural unit broader, whose study requires the contest of several disciplines, in the forefront of which obviously, ethnology and linguistics appear" [7]. Therefore the technical change can trigger a social change as well as the social change can trigger a technical innovation. There is a dialectic relation between social structure and technical structure. We note too that the technical change is on a much shorter scale while the social change occurs on a longer scale.

Societal change, how?

In sociological theory, there are two tendencies to explain social change. Firstly a holistic approach which considers that social change is a global and radical project: the French revolution as example of social movement, or as the labour movement for Marx, Dahrendorf or Touraine. For Alain Touraine [8], the social change is a society project born out of and carried by a labour movement. Changing society is a collective action with common goals. We call this approach, holism. We consider that the characteristics of collective action are different from the characteristics of individuals.

The second approach focuses on individual action. It considers that social change is the effect of the sum of individual actions. The paradigm of this approach is the one of methodological individualism. In the broad sense, one can characterize the methodological individualism by three proposals which postulate that:

- Only the individuals have goals and interests (principle of Popper-Agassi).
- The social system, and its changes, result from action of the individuals.
- All the socio-economic phenomena are explainable in the terms of theories which refer only to the individuals, with their provisions, beliefs, resources and relations.

If we refer to the first theory, today we can't mobilize a large part of population to be more sustainable in their moving practices. In spite of the fact, there are tendencies such as movements against global capitalism or, for example, local associations of producers of organic products having a controlled designation of origin (AOC, appellation d'origine contrôlée).

If we use the second approach, through what means we can change social behaviour? Individuals are the base of society but they are radically different. Society is composed to a diversity of social positions. The working classes do not have the same conditions as middle classes who are different from those of upper social classes. The individual behaviour is closely linked to his social and economic conditions. The response of individuals' vis-à-vis public policy is different because their conditions are specific. In fact, individual response to public policies is never systematic.

Then, the systematic effect of transportation policy on social behaviour isn't evident. This conception applied in the social sphere comes from the sciences of nature. In this sphere of knowledge, the techniques are supreme form of rationality. The social phenomena are of another nature. We can not submit it to a formal rationality as well as we can not be programmed their directions. We can not change society only if work of social mechanisms, and we can not explain what is social only by what is social, as Durkheim said [9].

Ecological culture and contemporary consumerism

In the contemporary world, individuals are socialized according to a model characterized by the following elements:

- Hedonism: research of all kinds of pleasure.
- Comfort: Less effort and more rest.
- Freedom of circulation: moving usually in space and in time like holidays, journey and recreations…
- Consumption freedom: property of goods, eating different kinds of food, clothes according mode.
- Individual interest before the collective interest: exacerbates individualism.
- A global culture of consumerism which more rooted by following means: cinema, publicity, mass media, school, new technology of communication and information…

Jointly to this collective culture of consumption which is very recent in history, we add another fundamental element of modernity: freedom. We compare societies with this criterion of freedom of persons. Democracy is based on freedom. Without freedom, capitalism can't maintain his global domination. I'm free to do what I like and when I like. Therefore, individuals and social groups have become very attached at a standard lifestyle which is characterized by a culture without constraints and with more hedonism. There is a social consensus which legitimates the collective aspiration at having this life mode. Consumerism is the myth that the individual will be gratified and integrated by consuming. It offers the tangible goal of owning a product, and offers only short term ego-gratification for those who can afford the luxury and frustration for those who cannot. For many individuals taking public transportation, they don't own a car because their low income. Usually they belong to popular classes.

We need to introduce a new social culture that we will call: *ecological culture*. The notion of Ecological culture is the sum of collective values which model the social conduits so as to respect the natural cycles and the natural resources. This ecological culture means practicing self-control, a limitation of consumption, less comfort for individuals and less freedom. Therefore, we will be in the age of the global culture of frustration which is the opposite to the global culture of hedonism. This is the political and social challenge today to protect the future generations' interest. As well as the global culture of hedonism resulted from social mechanisms and of a specific socialization, the new ecological culture has needs innovation to socialize models which focus on awareness about our common future and the preservation of natural resources.

Conclusion

It appears from the above that the technocratic logic can only succeed if it is accompanied by a social acceptance or even a total adhesion of all social categories. However, this social consensus is not the work of political institutions or voluntary associations, in spite of their important role in creating greater social awareness of acute environmental problems. It is the result of a long-term socialization of younger generations. Social resistance to fully engage in a more ecological and sustainable culture is a tangible reason of rooting of this lifestyle characterized by hedonism and consumerism socially shared.

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Environmental impact assessment of urban mobility plan: a methodology including socio-economic consequences

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Abstract

The project objective is to develop an optimized methodology to assess the environmental impacts of urban mobility plans (UMP, in French PDU), taking into account their social and economic consequences. The main proposed methodology is based on a systemic approach: multi-factor (air quality, noise, energy consumption, greenhouse gas emission) numerical simulations with a chain of physically-based models, based on alternative and comparative scenarios. The social and economic

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consequences of these alternative simulations are assessed by means of econometric models. Two alternative approaches are explored: (i) the use of composite environmental indicators to correlate the sources to the impacts, especially health impacts, and (ii) the analysis of sample surveys on what makes inhabitants' quality of life, well-being and territorial satisfaction and on citizens' behavioral changes linked to transport offer changes.

Context

Mobility is at the heart of the stakes for urban sustainable development and transportation policies that are set up in cities increasingly incorporate environmental components. Urban mobility plans are, in France as in most European countries [1], an essential tool of urban mobility policies. Their environmental features have received increasing attention during the last decade, so that environmental impacts assessment of the mobility plan actions is now compulsory.

The research program Eval-PDU was launched from the request by Nantes Métropole, the community of communes of the Nantes urban area, for a methodology allowing to assess jointly the various environmental impacts of the actual (2000-2010) urban mobility plan (UMP) and of the future plan (2011-2020), taking into account their social and economic consequences. This situation is a good illustration of the need of local public authorities for rigourously based tools to assess a series of impacts (air quality, noise, ...) effectively associated with various actions (or groups of actions) they lead. Beyond the monitoring of objective indicators, it is a matter of understanding and quantifying a cascade of physical and social causalities and, further, its consequences for the quality of life and its perception by the inhabitants. The need concerns as much *ex post* evaluations of what has already been done, as *ex ante* evaluations of what is being planned. A first one-year research-action grant with Nantes Métropole allowed to imagine the main features of the methodology and to build up consequently the project research team. Since January 2009 the program is funded for 3 years by the French national research agency (ANR) within its "Sustainable cities" program.

A priori, the proposed methodology is based on the assumption that quantitative environmental impact assessment require alternative, comparative simulations with physically-based numerical models of air quality (pollution emissions and dispersion), noise generation, energy consumption and greenhouse gas (GHG) emissions. For a joint assessment of these different environmental compartments, the simulations must be based on the same situations, hence on a common description of the transport traffic – this requires (i) a multi-modal traffic numerical model allowing to represent the actual traffic flows and their counterparts in alternative, hypothetical situations, and (ii) a full description of these alternative situations or scenarios, compatible with each of the involved numerical models. The social and economic consequences of the environmental impacts are to be assessed by econometric model calculations based on the results of the physical process simulations.

This approach raises a large number of methodological questions.

- **About scenarios and data**: What is the pertinence of alternative scenarios to render the changes generated by the UMP? Is-it possible to define different situations "everything else identical"? Can they be translated into coherent data sets? What is the availability of these data, which are necessary to run the various models? Especially in the case where the UMP "starting point" has not been defined in advance?
- **About the physical process simulations**: In the present state of urban transport modeling methods, is a multimodal model able to translate the different scenarios in significant changes of vehicle fluxes and travelled distances? Are multi-factor simulations possible with several mono-factor models? Considering the data and modeling uncertainties, will the simulation results be significant?
- **About the consequence analyses**: Can the simulation results be combined in an integrated assessment? By means of composite indicators? or by "socio-economic" analyses? Are the health impacts identifiable and quantifiable or masked by non-environmental influences? Are the social and economic consequences of environmental impacts of a lower order of magnitude than the direct social and economic impacts? Can they be evaluated in terms of well-being and satisfaction of UMP actions without a full economic computation of the whole UMP?
- **About the methodology itself**: Is-it possible to shorten the long chain of numerical models by using integrated, composite indicators relating the sources (transport fluxes) to the impacts (air quality, nuisances, human health) based on empirical correlations? What is the value of citizen's sample surveys on mobility and behavioral changes in response to mobility offer changes and, since they are more easy to launch by territorial authorities, could they be an efficient alternative?

Project structure

The main approach is based on multi-factor numerical simulations (air quality, noise, energy consumption, GHG emission) representing a set of alternative scenarios (with/without, before/after) rendering the changes in the citizens mobility over the metropolis area generated by UMP actions. These environmental impact simulations require input data sets, among which the emission inventories by the traffic of the different types of transport. Maps of the traffic fluxes are provided by a geographically-based multi-modal traffic model, whose main part is road traffic which is the key of the environmental stacks. The traffic intensities on the rails and road/street segments are the linear sources in the noise calculations. Combined with an engine emission model, they allow to compute energy consumption and emission inventories of regulated air pollutants and GHG. A urban pollutant dispersion model takes into account the meteorology interactions with the urban morphology to evaluate and map the pollutant concentrations and exposures.

The outputs of these numerical model simulations are the inputs of social and economic consequence calculations. The impacts of air quality and noise are computed on two types of socio-economic indicators: (i) well-being and declared satisfaction indicators, extracted from a survey of 1500 ad-hoc questionnaires to a representative population sample on 8 different districts; (ii) property values of housings, spatially correlated with the transport system main features. Econometric methods are further used to isolate the influence of environmental factors from those of other, preponderant factors.

Two alternative approaches are also explored, aiming at skipping all or parts of the numerical model chaining while keeping the capability of analyzing the processes from the socio-economic point of view. The first method, based on the works of COST action 356, consists in designing composite environmental indicators which correlate the sources and the impacts, i.e. the different transport mode intensities and the environmental quality indices; this method will be especially developed for assessing the health impacts, and compared to the results of the previous method. The second alternative approach consists in evaluating the environmental consequences of the changes in citizens' behavior linked to key UMP actions, from another survey among specific inhabitants especially concerned with these actions, by identifying their individual adaptation strategies to the variations in transport offer.

The program includes a large amount of result analysis and experience feedback in view of (1) applying the assessment methodology to the special case of Nantes urban area actual and future plans, (2) revising and optimizing the proposed methodology, (3) taking into account the knowledge obtained from the alternative approaches.

The research program is composed of 11 main tasks: project coordination, construction of alternative scenarios representing UMP actions and methodology optimization, data flux management and storage in a common geographical information system (GIS), multi-modal mobility modeling, simulation of pollutant emissions, GHG and energetic consumption, simulation of air quality, simulation of noise propagation and impacts, socio-economic assessments by econometric and well-being models, assessment by composite indicators, assessment of environmental consequences of the induced changes in citizens' behavior. These tasks are structured into 8 task packages according to their nature and the participation of the eleven research groups.

Coordination, construction of the methodology and scenarios

Coordination

The first task consist in the usual management of the program, including relations with the ANR funding agency, inter-teams and external communications (web, meetings, presentations and publications), and the coordination of the advancement of the different tasks. This last part is especially delicate due to the chaining of the successive modeling tasks and to the relatively large number of young scientists specifically hired for the program. Internal reporting is very important in such a program where several tasks are strongly inter-dependent. The task also includes coordination with Nantes Métropole, which is presently involved in its UMP assessment and revision, and which is the main provider of input data to the research program. Finally it includes coordination with other associated researches as, e.g., a series of student works on the relationships between the UMPs and other local territorial plans for air quality protection, climate, noise protection, ground occupation, and development schemes, and on the "environmentalization" of rules and instruments of urbanism and land law.

Methodology construction and optimization

The construction of the proposed methodology is a continuous task, from the program start since it is necessary to define the research work, until the end since an optimized methodology is the expected result of the works. A preliminary version will be first established, based on the situation of Nantes Métropole. A further analysis of its assumptions, principles, difficulties, drawbacks, … will be pursued during the course of the researches, with the aim of adapting the methodology to other urban areas. An optimized version will finally be drafted based on the results and returns of experience of the other task groups.

The task includes first an in-depth analysis of the previous examples of French and foreign UMP assessments, as well as of the relationships between the principles behind the design of the mobility plan of Nantes Métropole and their translation into actions on the transport network.

These actions cover a very large range: infrastructures, parking, urban toll, traffic restrictions, public transport, eco-driving, multi-modal information … They are categorized as a function of their time-space scales, their mechanisms (actions on behavior, traffic, modal split, prices), and the importance of their expected environmental impact, for selecting the different assessment methods, measurement tools, and pertinence for the urban mobility management.

In the final phase, the experience gained by the different task groups will be integrated to optimize the proposed methodology and to adapt it to either the ex-post assessment of achieved plans or ex-ante assessment of the expected impacts of plans in construction.

Construction of alternative scenarios representing UMP actions

The alternative scenarios are a key to the joint multi-factor impact assessment. For ex-post assessments, they must express the main features of the actual transport system and offers, and those they would have if the UMP actions had not been realized. This implies to select the data sets which define a reference situation "before" the UMP and to imagine the values of these data in situations "without" the UMP but taking into account all the changes which are not related to the UMP actions as, e.g., gas and energy price variations, urban sprawling, local and national economic transformations … Furthermore these data sets must be effectively available for the representative time horizons, e.g. just before and after the period of the plan, or a reconstruction method must be designed.

This task objective is to formulate the methodological principles of construction of alternative scenarios for environmental impact assessment. These principles are further applied to the main actions of Nantes UMP, and their feasibility and effectiveness are analyzed. In a further stage, the additional assumptions which are necessary to define the model input data sets will be explicitly described.

The chain of physical process models and data flux management

Data acquisition, repository and GIS

The data flux is an important key of the proposed methodology since many model inputs are some outputs of the previous calculations. The uniqueness and/or coherency of input data is also a key of the joint multi-factor assessment. All the data obtained from external sources are stored in a common repository geographical information system based on the open source platform OrbisGIS developed by IRSTV for urban researches. Further, all model outputs are stored in this GIS prior to their further use as inputs of other models, ensuring spatial coherency and quality control.

The common GIS construction includes a spatial semantic, definition of common mapping modes, adapted geo-statistical calculation and representation tools. It allows direct comparison, superposition, and fusion of results from the various tasks. It is also a powerful tool for the presentation of the program results.

Multi-modal mobility

The model calculations include two parts: transport offer and transport request.

The multi-modal traffic software VISUMis used to compute the traffic over the road and public transport networks. The calculation domain has an area of 2242 km^2; 300 traffic zones have been defined with a grid density increasing towards the city centre. The simulation includes explicit calculation of inter-zone traffics over 4100 street segments (2300 km, i.e. 25 % of the total) and calculation with an implicit method for intra-zone traffics. It separates heavy duty (HD) and light duty (LD) vehicles, computes transport by trains, trams and buses over the whole network, and includes free and toll parking possibilities, including modal exchange P+R, but not bikes and motorbikes. Modal split procedures involve walking courses.

The model calculations are driven for 4 periods of the average week day: morning peak (7–9 am), evening peak (5–7 pm), night (8 pm – 6 am) and the rest of the day. Long term integrations thus require weighting factors for vacation periods and week-ends. The model outputs are traffic densities, fluxes and speeds over the segments, journey times and costs as a function of transport mode and population type.

The reference situation for transport request is the transport-population sample survey of 2002 over the metropolitan area, completed by regional social and economic data bases estimated for the same year. These data are analyzed with the software VISEM to produce origin-destination matrixes, using transportation times calculated by VISUM. New simulations will also be run for 2008, based on the data from a national survey and local counting, with various options and scenarios.

Noise propagation and impacts

The noise propagation calculations are based on the equivalent point source method where each actual source is modeled by one or several (engine, wheels) point sources characterized by a sound spectrum, a height and a directivity. The calculations are limited to light vehicles and trams in the first stage, then extended to buses and eventually to motorbikes (which are not well documented yet). Street and track segments appear as lines of point sources function of traffic intensity and vehicle types.

The propagation calculations include direct, reflected and diffuse components, towards a regular grid of virtual receptors. They are implemented as a plug-in within OrbisGIS, allowing to handle the sources, the calculations, and the representation of results with a unique software.

The results are maps of integrated indicators as, e.g. the equivalent continuous sound level Leq, for standard periods of time in the days.

Pollutant emissions and energy consumption

The emission inventories are computed with two methods in parallel, COPERT 4 and ARTEMIS, which have been developed by the European Environmental Agency and the European DGTREN, respectively [2]. COPERT 4 is based on a collection of standard emission factors for the different classes of vehicles, propellants, loads, speeds, for a large number of pollutants [3]. Adjusted to the French vehicle fleet, and applied to the traffic cadastres they are used here to compute the emissions of CO, NOx, NMVOCs (incl. Benzene, Toluene, Xylene), formaldehyde, SO_2, CH_4, CO_2, N_2O, NH_3, PM10, PM2,5, PM1, Total particles, PAHs, POP, dioxins, furans, and toxic metals. The energy consumptions are also an output of the calculations.

While COPERT 4 calculations are based on average vehicle speeds, the ARTEMIS method is more dynamic, taking into account the characteristics of each segment (zone, type, slope) and the speed profiles corresponding to instantaneous traffic conditions (fluid, dense, saturated, congested) [4]. This tool includes EURO4 regulation and the further reduction rates. It is being adjusted to the French vehicle fleet and adjustment hypotheses for the Nantes area are being formulated.

The inventory of non-traffic sources (combustion and solvents) is constructed from national and regional consumption data bases using a top-down approach based on population densities (residences and offices density and characteristics) at the scales of communes, districts and blocks.

Air quality

The urban pollutant dispersion model ADMS Urban has been selected from 8 model intercomparison studies [5]. Based on emission inventories, the model is tested and adjusted to the pollutant concentration measurements of the air quality survey network Air Pays de la Loire for the reference years 2002 and 2008.

The UMP impacts on air quality are assessed by comparisons of pollutant concentrations over the Nantes Métropole area and within a selection of streets of the city center during the final year for the scenarios with UMP actions versus "business as usual".

Socio-economic assessments

This task includes a close cooperation of economists, geographers, sociologists and geomaticians to identify and produce pertinent and tractable geographical indicators. The developments are included in OrbisGIS, with the socio-economic data bases. Several population distribution and well-fare data bases of INSEE (National institute of statistics and economical studies), at different dates from 1990 to 2006, are used to characterize the population of Nantes sectors and districts in view of establishing representative samples.

Well-being and territorial satisfaction

The assessment of the air quality and noise impacts on the population well-being is based on an ad-hoc sample survey of citizens' declared environmental satisfaction linked to the UMP actions. A first 30-90 minutes in-depth interview has been applied to a sample of 40 inhabitants of 7 districts during the 2009 spring, bearing on 5 topics: (i) residential choices and territorial satisfaction; (ii) well-being feeling; (iii) environmental experiences and practices; (iv) spatial practices; (v) relationship to public action.

The theoretical and efficiency analyses of this test survey allowed to define a more elaborated questionnaire and a survey protocol which was first tested on a sample of 30 inhabitants, then applied to 1500 inhabitants of 8 districts during the 2010 spring. The districts are different regarding to the socio-economics characteristics of the population (e.g., repartition of the socio-professional categories), environmental amenities (e.g., distance to the city centre, transports offer, access to green spaces), and morphology (e.g., density, age of the building).

Property values of housings

The study requires to identify the environmental variables which may influence the property prices. The PERVAL data base, which includes the housing property transactions and prices in the urban area of Nantes, has been included into the communal GIS, and a preliminary analysis is conducted with OrbisGIS to select the pertinent data and indicators which will be further used to establish geographical correlations between public and private network changes and property values.

Composite indicators

The alternative method purpose is to replace the alternative numerical model simulations by the construction of composite indicators aggregating individual impact indices, based on typologies of the environmental impacts of transport modes. These include a "scientific", process-based typology and a social, perception-based typology based on existing surveys. The indicators must further be related to the impact sources (emission factors and traffic intensities) by empirical correlations. This approach is based on the works of the European cooperation action COST 356 [6].

The method is applied to obtain an atmospheric pollution health impact indicator, computed from the vehicle emissions of the various pollutants. The computation includes weighting factors which are functions of the pollutant toxicity levels and of the population exposure levels, based on the existing health impact studies and life time analyses.

Changes in citizens' behavior

The objective of this task is to identify (i) the individual strategies to adapt to transport offer variations issued of some UMP actions, by taking explicitly into account the determining factors of families' time management strategies, (ii) and the types of journeys and populations which are the most sensitive to these variations, and to deduce the consequences for (iii) the environment, and (iv) the public politics. The study focus is the incidence on individual car traffic of the UMP main actions aiming at promoting a modal transfer, since it is difficult to assess their efficiency at fulfilling their objective, generating longer routes, or just reducing mobility.

The analysis is based on two sample survey of selected populations: (i) reduced mobility persons, and (ii) selected users of the public and private transport systems. The task includes the construction and tests of questionnaires and survey protocols, and their application to about 1000 persons at key sites as, e.g., train and tram nodes, city center parking tolls, modal transfer P+R.

Conclusion

The Eval-PDU program is a deeply interdisciplinary program in which scientists of environmental, engineering, and social sciences cooperate tightly to construct a methodology based on the systemic approach. The construction of a methodology involving the assessment of the physical changes induced by the action of a mobility plan in the traffic of the different transport modes, of their environmental impacts, of their social and economic consequences requires a set of scientists who are not used to work together and this in turn requires a strong effort at constructing a common language, at understanding very alien points of view and at accepting very different working methods. A side benefit is a deep enrichment of each other's thoughts.

At the time of this communication the research program reaches mid-way. Most of the "individual" contributions have already been produced or they will be soon, and are now being proceeded in a continuous process; the time of confrontations, revisions, optimizations is coming; this phase is to be driven in common by all participants, with a constructive perspective, and this may not be the easiest and most smooth task.

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Assessment model of territorial articulation in rural areas. Application to four Spanish "comarcas".

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Abstract

This paper proposes a territorial articulation study model based on a set of indicators. The indicators used in this study were urban distribution, weighted distance of road network and road accessibility. We have applied the indicators to four regions in the Spanish province of Almería: Alpujarra Almeriense, Alto y medio Almanzora, Los Vélez and Poniente Almeriense. The results found show them to be highly heterogeneous, and allow areas with deficient road access or distribution of urban areas to be identified. Indexes were poorest for mountainous regions with high slopes and areas furthest from cities. The conclusions will contribute to improving the development strategies of Settlement and Road transport infrastructures in these rural regions.

Introduction

At the present, territorial cohesion is defined as the possibility that a population in a specific territory has of accessing services of general economic interest [1]. In order to achieve high territorial cohesion it is necessary to have an adequate territorial articulation, which depends on the existing network of urban nuclei and the road network. In other words, there must be an optimal spatial distribution of urban nuclei and high accessibility for inhabitants. This then leads to the promotion of intraterritorial cooperation, the diversification of productive activities and interaction among rural areas and urban areas.

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Although the concept of accessibility plays an important role in the formulation of planning and transport policies, it is frequently misunderstood and poorly defined and structured [2]. Accessibility is generally defined as the ability to access specific activities from one place and with a specific means of transport [3,4]. Geurs and Wee [5] believe that accessibility is related to the role of land use and that of transport systems in society, granting people and groups of people the opportunity to participate in activities in different places.

The concept of accessibility encompasses various elements such as the characteristics of the network and the transport system [6,7], location in a given territory in relation to the desired destination [6], the spatial distribution of possible destinations and the ability to reach them [8], the working population and the economic and industrial activities that these places carry out [9].

If the concept of spatial accessibility is to be examined, two components must be analyzed together: the availability of services and their proximity [10,11,12]. The gravitational model is one of the most commonly utilized methodologies for measuring this. It relates distance and impedance (average travel time) to the attractiveness of a geographic area or service [12]. This model calculates the minimum distance or shortest route between pairs of objects (urban nuclei) [13].

In Spain, rural areas are organized into geographic units called 'comarcas,' which are defined as a group of municipalities with common geographic and historical characteristics. These comarcas cover a surface area of approximately 100,000 ha. and, in general, their small population nuclei are organized around one main municipality.

These main municipalities are urban nuclei that are larger in size and possess the characteristic of being centrally located. The latter implies authority over certain supra-local functions, and the provision of a series of services that cannot be covered by small urban nuclei. Therefore, any initiative carried out in these towns has significant impact potential [15] owing to the fact that they are the organizing centers of their territories and points of dynamism and distribution [16]. Central nuclei are the nexus between rural territory and large urban centers, and they constitute the historical nodes of rural settlement structures [14].

Consequently, this study of accessibility in rural territory must bear in mind two key elements: a) the geographic unit (comarca) and b) the main municipality of each geographic unit. It is important to analyze the connection capacity within the individual comarcas, especially with respect to the main municipalities.

This study proposes a methodology to calculate the territorial articulation in Spanish rural comarcas, which is applied to four comarcas in the Almeria Province. The articulation was calculated according to the spatial distribution of the urban nuclei in the territory and their accessibility, especially to main nuclei.

Method

The four “comarcas” in the province of Almeria, Spain, the object of this study, were: Almeria Alpujarras (Alp), Upper and middle Almanzora (Alm), Los Vélez (Vel) and Almeria Poniente (Pon). The comarcas Alp, Alm and Vel are rural areas in need of revitalization, with low population density, witnessing depopulation and an aging population, little technological development, significant farm land area, etc. As for Pon, it is an area with significant economic development, thanks to farming under plastic, that is in greenhouses, which produce significant production volume on a small surface area. Its high population density stands out, brought about by the demographic growth of the local population and the mass arrival of immigrants to work in the greenhouses. In Alp, Alm and Vel the topography, which will play an important role in accessibility, is rugged. On the contrary, in Pon flat coastal topography, where most of the urban nuclei and population of the comarca are located, coexists with the rugged mountain landscape of the northern and western ranges found in the comarca.

Furthermore, it is important to consider the presence of highways, namely the A91 through the comarca Los Vélez, the A7 through Almeria Poniente, and the A7 and A92 through the Almeria Alpujarras, as they facilitate accessibility between the urban nuclei of these comarcas. The head municipalities, characterized by concentrating public and private services and/or institutions, in each comarca, are: Berja (Alp), Macael (Alm), Vélez Rubio (Vel) and El Ejido (Pon).

In order to construct the articulation index, first the spatial distribution of the urban nuclei was calculated using the Clark-Evans index [18]. This was done to determine the relation that exists between the number of cities in the geographic area and the distance between them [19]:

$$Rn = 2Do * \sqrt{N/A}$$

where, *Do* is the average of the actual road distances between urban nuclei and that which is closest in proximity, in a territory of land area *A* in which there are *N* urban nuclei.

Subsequently, calculations were made of actual and weighted distances between urban nuclei pairs in each comarca. Distance was understood as the actual road distances in km. that separate an urban nucleus from another, independent of the kind of road. The weighted distances for each urban nuclei pair were calculated with the following formula:

$$r_p = \left(Hw * c_{Hw}\right) + (SR * c_{SR}) + (ARR * c_{ARR}) + (BRR * c_{BRR}) + (CRR * c_{CRR}) + (LR * c_{LR}) + (UR * c_{UR})$$

where, *Hw, SR, ARR, BRR, CRR, LR* and *UR* are the km. via highways, state roads, regional roads, local roads and urban roads, respectively. The coefficients (*c)* were fixed between 1 and 2.4 according to the maximum speed limit of each road; these coefficients were lower where the speed limit was higher (expressways and highways). Calculations were made of the average distances ($\overline{r_p}$) in order to make comparisons between comarcas. The actual road distances, and travel time (duration average in hours), of the journeys between urban nuclei were obtained from the 2009 Official Roadmap [17].

Later, road accessibility indicators are calculated for all the municipalities, between each other and to the head municipality. The calculation of road accessibility to main urban nuclei was carried out in function of the weighted distances, the type of road and travel time. To do so, either the gravitational model variant was applied or the Population Potential (POT) developed by Calvo [20] and Pueyo [21]:

$$POT_i = \sum_{j=1}^{n} \left(\frac{P_j}{{d_{ij}}^2} \right) + P_i$$

where, d_{ij} is the minimum distance between *i* and *j*, P_j is the population of the urban nuclei to which people travel *i*, and P_j is the population of the main municipality of the comarca. This formula, which was relativized by the total population in order to be able to make comparisons, considers communication infrastructures (roads) as the elements that link the entire territory [22].

Finally, a territorial articulation index was calculated from these indicators. The index is an additional expression of the distribution of urban areas and overall accessibility. The territorial articulation index (IAT) is defined using the following formula:

$$IAT = \frac{Rn}{\overline{r_p} * t_m} * \frac{POT_i}{P_{total}} * 100$$

where, Rn is the Clark-Evans index, $\overline{r_p}$ is the average of the weighted distances between urban nuclei (km.), t_m is the average travel time between urban nuclei (h), *POT* is the population potential (inhab) and P_{total} is the total population of the comarca (inhab). The indexes that influenced the articulation in a positive way were placed in the numerator, and all others were placed in the denominator. The IAT was multiplied by 100 to obtain manageable values.

The articulation index calculated expresses, in an aggregate and relatized manner, the distribution of urban nuclei, the accessibility among them and to the main municipality, the average travel time between nuclei, and the population potential of the main nucleus of the comarca.

Results and discussion

Spatial distribution of urban nuclei

The spatial distribution of the urban nuclei in each comarca (Table 1), according to the ratio proposed by Clark and Evans [18] (0≤Rn≤2,1491), reveals that Vel and Alp tend to have a random distribution within their land areas, while Pon reveals a regular or uniform distribution of its urban nuclei. Alp has an average value between the two types of distributions.

Table 1. Clark Evans Index.

Comarca	N	A (km^2)	Do (km)	Rn
Alp	31	1 750.39	4.52	1.20
Alm	23	1 521.67	5.78	1.42
Vel	4	1 146.19	9.75	1.15
Pon	9	970.69	9.33	1.80

The values obtained for Vel are a response that is primarily due to the low number of nuclei, despite being a mountainous area. For Pon, the flat topography where most of the urban nuclei are located and the presence of the A7 highway improve the distribution of the urban nuclei in the territory. Alp and Alm have intermediate values.

Average distances, actual and weighted, between urban nuclei, and average travel times

The values of $\overline{r_p}$ obtained were: 55.66, 40.73, 39.89 and 22.62 km, for Alp, Alm, Pon and Vel, respectively. These results show that the inhabitants of Vel have to travel, on average, half the distance of the inhabitants of the other three comarcas. Therefore, its accessibility is higher and this will have a significant effect on its territorial articulation.

In terms of road type (Fig. 1), it was observed that most trips are made via class B regional roads (66.22% in Alm, 48.57% in Vel, 39.07% in Alp, and 25.87% in Pon). Travel via expressways and highways is significant in the comarcas of Vel, Pon and Alp, representing 40.00, 37.17 and 20.44%. Also, travel via class C regional roads and urban roads is significant in Alp, Alm and Pon. These results reveal the heterogeneity of the types of roads between the urban nuclei of each comarca, observing that most road travel between them is made via class B and C regional roads. This influences average travel time (11.33, 19.00, 20.20, 35.68 minutes in Vel, Pon, Alm and Alp, respectively), reducing accessibility. In the case of the comarca of Vel, the average travel time is lower due to the presence of the A91 highway.

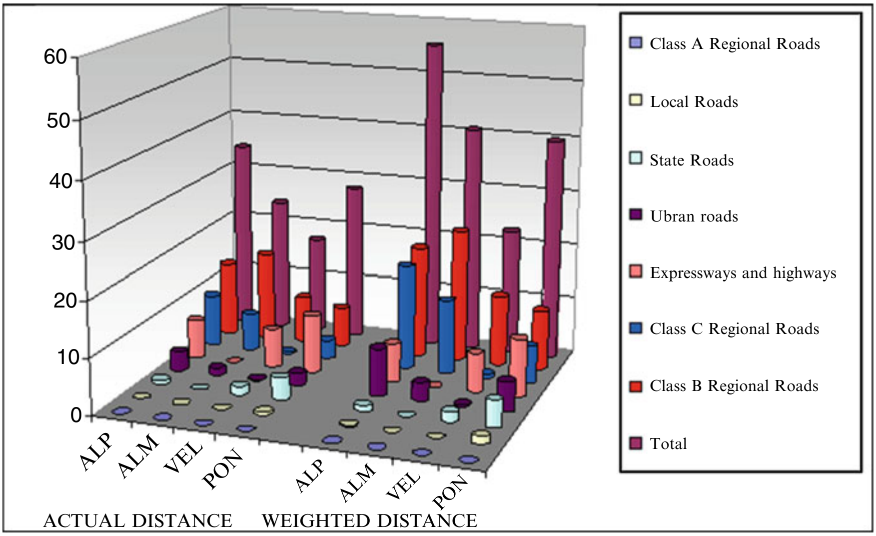

Fig. 1. Average travel distances, actual and weighted, between urban nuclei (km).

Weighting the types of roads by speed reveals that the difference is even greater, with a hypothetical increase of 250% in the average distance between urban nuclei. This increase creates a greater impression of distance among the population, which reduces the perception of accessibility among the urban nuclei. In terms of road type, the average lengths of urban roads and class C regional roads are significantly greater.

Accessibility of urban nuclei in relation to the main nucleus

The POT towards the main nucleus of the comarca demonstrates high heterogeneity among the four comarcas studied (Table 2). The POT value for Pon stands out as it is significantly higher than the rest of the comarcas, corroborating the fact that the greater the population of the main nuclei and shorter the distance from the rest of the nuclei, the greater the reciprocal inferences of potential.

Table 2. Accessibility Indicator.

Com.	Pop. (inhab)	Head Mun.	Pop. (inhab)	POT_1	POT_2	POT_1 P_{total}^{-1}	POT_2 P_{total}^{-1}
Alp	41 929	Berja	15 001	15 074.2	15 025.4	0.360	0.358
Alm	54 918	Macael	6 212	7 110.1	6 494.2	0.129	0.118
Vel	12 693	Vélez Rubio	7 147	7 220.4	7 178.4	0.569	0.566
Pon	232 027	El Ejido	80 987	81 407.0	81 140.9	0.351	0.350

The existing correlation between distance and population, given because the relative position of an urban nucleus originates from possible interaction with other localities in the territory in which it is located [23], determines that POT is less when the weighted travel distances (POT_2) are utilized. The low values of Vel and Alm are mainly due to low population, and in the case of Vel, the high average travel distance, in comparison with other comarcas.

The values of POT relativized by the total population of the comarca reveal the degree of population concentration in main urban nuclei. In the case of Vel, Vélez Rubio constitutes 56.31% of the population in the comarca, and in Pon, El Ejido only 34.90%. In the case of Alm and Alp, the wide scattering of population among the 23 municipalities results in intermediate values with respect to other comarcas.

Territorial articulation of the comarcas

The territorial articuliation index calculated for these four Spanish comarcas (Fig. 2) corroborates the significant differences found for the indicators calculated previously. Vel has an IAT value that is far greater than the other comarcas studied. This is due to better road communication between them (less time and less distance) and the high potential population of their main nucleus (Vélez Rubio) in relation to the total population of the comarca (Table 3), which is favored by the presence of the A91 highway. On the contrary, Alm has a lower value due to the absence of an expressway or highway that alters its structure, and the low potential population of Macael.

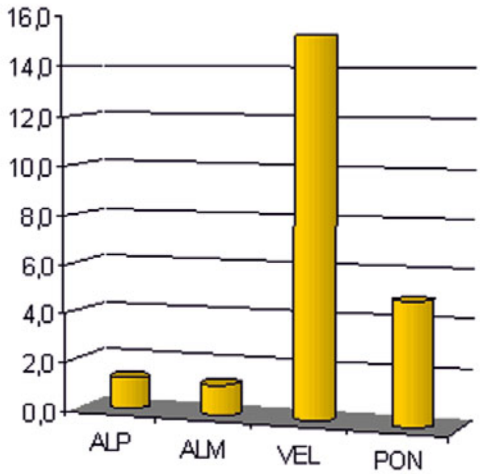

Fig. 2. Territorial Articulation Index.

Table 3. Summary of values of the indicators calculated.

Comarca	r_p	t_m	Rn	POT P_{total}^{-1}	IAT
Alp	55.66	0.59	1.20	0.358	1.30
Alm	40.73	0.33	1.42	0.118	1.24
Vel	22.62	0.19	1.15	0.566	15.25
Pon	39.89	0.32	1.80	0.350	4.98

Alp has a slightly higher value than Alm due to the high potential population of Berja. Also, this is favored by the presence of two highways (A7 and A92), which improve its territorial articulation by facilitating communication between urban nuclei, more dispersed and linked mainly by class B and C regional roads in areas with irregular topography. Pon reveals a territorial articulation that is better than Alp and Alm, because of the A7 highway, its more homogeneous distribution of urban nuclei and the population potential of El Ejido. These results reveal the effects that the type of road network and the distribution of urban nuclei have on territorial articulation.

Conclusions

The results found are highly heterogeneous, and allow areas with deficient road access or distribution of urban areas to be identified. Indexes were poorest for mountainous regions with high slopes and areas furthest from cities. However, the presence of a road network with superior characteristics (highways and expressways) improves its accessibility and favors its territorial articulation. This study represents a preliminary approximation, considering it only took into account the main urban nucleus of each municipal area. A more detailed study of all urban nuclei, which would include less significant locations (groupings of country estates, hamlets, remote rural areas, etc.) would make it possible to discover more about articulation in greater detail. Finally, these conclusions will contribute to improving development strategies of Settlement and Road transport infrastructures in these rural regions.

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Model of territorial distribution of CO_2 emissions reduction target in the transport sector

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Abstract

In this paper CO_2 emissions reduction targets in the Kyoto Protocol and in the energy policy of the EU are transferred to the transport sector. First, we analyze CO_2 emissions from transport in the reference year (1990) and their evolution from 1990 to 2007. Later, we propose a nonlinear methodology for distributing the dynamic CO_2 emissions reduction targets. We have applied the proposed distribution function for 2012 and 2020 to two territorial levels. The weighted distribution is based on per capita and per GDP CO_2 emissions. Finally, we show the weighted targets found for each EU Member State and Spanish Autonomous Communities, compare them to the real achievements to date, and forecast them for the years the Kyoto and EU goals are to be met.

Introduction

The evolution of transport GHG emissions has been uneven in the EU Member States and that there are countries that made a big progress to control emissions and oppositely. The heterogeneity of the transport emissions patterns in different countries underlined the need to establish modulated decisions to be taken at different territorial levels to achieving a common aim. Spanish transport sector and consequent emissions has grown rapidly during last 17 years and will continue growing in the next years unless strong decisions will be taken. Transport sector is the source of GHG with the highest growth rate, especially due to road transport growth. This paper reviews sector impact on GHG emissions and considers effects of different alternative measures on emissions and subsequent energy consumption.

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Evolution of the transport sector and GHG emissions: European comparison

In 1990, transport consumed 39.5% of total primary energy in Spain and 40.7% in 2007 [1]. In 2007, the final energy consumption of the transport sector was slightly more than 38 million toe (tones oil equivalent). Besides being the economic sector with the major final energy consumption, transport is the sector with the major consumption of fossil fuels (55.2%, 2007). In absolute terms, GHG emissions from transport during this period have grown 66% [2]. At an annual growth rate of 3.7%, emissions are expected to double over 20 years. This annual change is quite above the average long-term growth of 1% per year in the OECD countries [3]. Emission growth is due mainly to road passengers and freight transport. Only road transport is responsible of 75% of total sector emissions.

GHG emissions of the transport sector are not explained by either the population growth or the economical growth, because they have lower growth rates. This indicates that production processes in Spain have an increasing growth of transport, contrariety to EU objectives to generate economical growth with smaller increments of passengers and freight transport flows [4]. Current transport demand trends and associated GHG emissions in Spain have higher growth rates than in the rest of EU countries. The mobility of persons and goods grows at a higher rate compare with the mobility of European neighbors. It is observed, in addition, that the growth of transport passengers is greater than the growth of transport freight, when Europe has opposite trend. These data show the greater importance of the problem in our country, and that, in our case, the mobility of persons is still more worrying than the one of goods.

Transport GHG in Europe and in Spain in 2007

The analysis of transport GHG emissions in 2007 shows that the basic indicators are very heterogeneous, varying considerably among EU Member States and Spanish Communities. Of the 27 EU Member States, nineteen presented lower emission rates than the EU-27 mean. Countries with higher population and surface area (Germany, France, the United Kingdom, Italy, Spain and Poland) presented the highest numbers of transport GHG emissions.

Of the 19 Spanish communities, Castile-León had by far the highest Transport GHG emission rate, with 5.21 tones of CO_2 equivalent per inhabitant. This is almost two times higher than the Spanish mean (3.03), while the remaining communities varied between 1.93 and 4.26 tones per inhabitant and year.

The differences between the means of population and absolute emissions for the EU-15 and the EU-27 areas are not great, but they differ significantly in comparison to the average value for the Spanish Communities. The standard deviation and the coefficient of variation are smaller when the number of regions (n) decreases, and when the geographic scope is smaller, as in the Spanish Communities (table 1). It shows more heterogeneity within EU countries than within Spanish Communities.

Table 1. Mean, standard deviation, minimum and maximum values of the CO_2 emission rate (tCO_2eq./inhab.) for NUTS[a] regions in 2007.

Population (million inhabitants)						
Level	n	Avg.	Min.	Max.	SD	CV
EU-27	27	18.34	0.41	82.31	23.20	126.46
EU-15	15	26.13	0.48	82.31	27.51	105.29
CCAA	19	2.38	0.07	8.06	2.41	101.41
CO_2 emissions (million tones CO_2 equivalent)						
Level	n	Avg.	Min.	Max.	SD	CV
EU-27	27	36.39	0.53	153.18	48.16	132.34
EU-15	15	52.24	6.68	153.18	53.97	103.30
CCAA	19	6.21	0.30	17.02	5.17	83.27
Emission rate (t CO_2 equivalent per inhabitant)						
Level	n	Avg.	Min.	Max.	SD	CV
EU-27	27	2.48	0.60	14.04	2.42	97.33
EU-15	15	3.24	1.84	14.04	3.14	96.93
CCAA	19	3.03	1.93	5.21	0.84	27.62

[a] This analyses covers 27 and 15 EU Member States (NUTS0) and 19 Spanish Communities (NUTS2), and is geographically based on the EUROSTAT's NUTS 2003 regional classification [5].

Evolution of transport GHG emissions 1990–2007 in Europe and in Spain

Although the overall evolution of transport GHG emissions in the period 1990–1997 is quite irregular, the amount of emissions in most EU Member States follows a upward trend. By 2007 the EU 27 Member States had achieved an increment of 26.0% in the total transport GHG emissions in comparison with 1990, with an average increment per country of 54.8±57.9%. The EU-15 Member States had been more successful, achieving an increment of 23.7% and a mean of 53.8±53.0%. While only four countries of the EU-27 had made significant progress decreasing emissions (Germany, Bulgaria, Lithuania and Estonia), it should be noted that in most countries transport GHG emissions actually increased over this period.

The results for Spanish communities over the period 1990–2007 present a more standard evolution. Comparison of the transport GHG emission data for 1990 and 2007 reveals also heterogeneous results, ranging from a 129.7% increase in Canarias to a 36.7% increase in Basque Country. The remaining communities have made varying degrees of progress: in 3 communities transport GHG emissions increase by less than 50%, in 6 communities it increases by between 50% and 75%, in 6 communities 75–100%, and in 4 it increases by over 100%.

Desirable threshold and dynamic target values for the reduction of GHG emissions

This study proposes a weighted modulation of the reduction coefficients based on the initial transport emission rates, taking 1990 as the reference year for its application. The desired value of transport emissions would always be fixed by the international commitments of reduction in GHG emissions as EU unilateral target of –20% by 2020 accounting for domestic emission reductions only [6]. Objective progress targets towards this goal must be dynamic and redefined over time, varying according to the area in question. The targets for each area should be obtained as a function of that area's distance from the desired value, in such a way that all areas converge towards it. In this way, the reduction coefficient, expressed in relative terms of improvement per unit, should vary between 1 (i.e. a hypothetical case if reduction is needed as in preindustrial times) and 0 (i.e. 0 carbon emissions from the transport sector). Since the EU aims for a reduction coefficient of 0.8 (emission rate in 2020/emission rate in 1990), on a more local scale the progress would have to be greater if the initial distance from the desired value (0) is greater.

The present work applies this reasoning in two phases. Firstly the EU is considered as a geographic unit (contemplating two scenarios: EU-27 and EU-15), taking each Member State as a subunit. The sum of transport emissions of all the sub-units, once the corresponding modulated coefficients of reduction are applied, will result in 20% reduction of emissions from 1990 in the EU-27 and EU-15 respect to 1990. Secondly, the results obtained for Spain (in the two scenarios: EU-27 and EU-15), are modulated using the distribution formula on a smaller scale in which the Spanish State is the geographic unit. In this case the sub-units are the 19 Autonomous Communities, and modulated reduction coefficients are generated for each one.

Methodology

We propose a methodology to calculate weighed coefficients of reduction of transport GHG emissions at various geographic levels using a weighted distribution, through an inverse logarithmic function of distribution based on the initial emission rate (1990). For each geographic unit (i), the emission rate is the amount of GHG emissions ($GHG_{i,t}$) in a year (t), expressed in tones of CO_2 equivalent, divided by the total Gross Domestic Product at this year ($GDP_{i,t}$), expressed per million Euros, or by the total population at the beginning of the year ($P_{i,t}$), expressed per inhabitant:

$$E(GDP)_{i,t} = \frac{GHG_{i,t}}{GDP_{i,t}} \text{ (tCO}_2\text{ /Million €)} \tag{1}$$

$$E(Inh.)_{i,t} = \frac{GHG_{i,t}}{P_{i,t}} \text{ (tCO}_2\text{ /Inhabitant)} \tag{2}$$

The initial rate of GHG transport emissions was used because it expresses the severity of the environmental problem related to climate change, and as it is a homogenous statistic at international level [7]. The inverse logarithmic function was proposed because it adapts to the objectives of the study as regards obtaining modulated coefficients of reduction.

To achieve the EU target first for 2012 of reducing the GHG transport emissions from 1990 in 8%, Kyoto protocol, (then for 2020: 20% reduction) the following initial premise was considered:

$$GHG_{2012} = c \times GHG_{1990} \tag{3}$$

where c is the total residual coefficient, complementary to the reduction coefficient, and GHG is the absolute value of emissions, i.e. the sum of the emissions in each geographic sub-unit GHG_i:

$$GHG = \sum_{i=1}^{n} GHG_i \tag{4}$$

Therefore, for the EU-27 and EU-15 geographical units:

$$GHG_{EU,2012} = 0.92 \times GHG_{EU,1990} \tag{5}$$

and for each geographical sub-unit, in this case the EU Member States:

$$GHG_{i,2012} = c_i' \times GHG_{i,1990} \tag{6}$$

where c_i' is the residual coefficient of each geographical sub-unit. This coefficient was calculated using an inverse logarithmic distribution function, depending on its initial emission rate E_i:

$$c_i' = f(E_{i,1990}) = a(lnE_{i,1990})^{-1} \tag{7}$$

where a is the factor that modulates the weighting coefficient c_i'. To calculate the value of a, first replace Eq. (7) in Eq. (6):

$$GHG_{i,2012} = a(lnE_{i,1990})^{-1})\ GHG_{i,1990} \tag{8}$$

then, as GHG_{2012} is the sum of the amount of emissions in each geographical sub-unit, $GHG_{i,\,2012}$, we have:

$$GHG_{2012} = \sum_{i=1}^{n} \frac{a \times GHG_{i,1990}}{lnE_{i,1990}} \tag{9}$$

therefore:

$$a = \frac{GHG_{2012}}{\sum_{i=1}^{n} GHG_{i,1990} \big/ lnE_{i,1990}} = \frac{c \times GHG_{1990}}{\sum_{i=1}^{n} GHG_{i,1990} \big/ lnE_{i,1990}} \tag{10}$$

We calculated a weighting factor a for both the EU-27 and EU-15, from which we obtained the residual coefficients c_i' for each Member State in both scenarios. This methodology can also be applied to smaller geographic areas like Spain, where the total geographic unit is the Spanish state and the geographic sub-units are its autonomous communities. In this case, the distribution formula is used, and the total residual coefficient c for the Spanish unit is the one obtained previously from the general targets of the EU for this state. In this way, new results are obtained as residual coefficients c_i', for each autonomous community.

Results and discussion

By applying the proposed methodology, we obtained a set of target values for the reduction of emissions for the EU-27 and EU-15 Member States for 2012 and 2020. The higher the country's initial emission rates, by GDP and population, the greater the reduction coefficients. Two clear examples are Estonia, with a reduction coefficient of 30.2% (2012) and 39.3 (2020) in the case of the EU-27 according to GDP rates and Luxembourg, with 21.9% (2012) and 32.1 (2020) for the EU-15. According to population emission rate, Luxembourg will be the country which need for greater effort to achieve the EU target: 22.7% (2012) and 27.2 (2020) for the EU-27 and 21.7% (2012) and 31.9 (2020) for the EU-15.

The total amount of transport emissions in absolute terms for each country is reduced unevenly, but in such a way that the total sum would achieve the EU's overall target. In addition, the great difference in emission rates, both for GDP and population, between Member States is considerably reduced over time. It is worthy of note that the results obtained for each particular country differ depending on the scenario considered (EU-27 or EU-15) and if the modulated weights are obtained using the GDP or the population. For instance, in the case of Spain the reduction coefficient in 2020 for GDP is higher for the EU-15 than for the EU-27 (21.5% vs. 20.4%). Oppositely, the reduction coefficient in 2020 for population is lower for the EU-15 than for the EU-27 (17.0% vs. 19.0%). However, the average values of reduction, for each of the two hypotheses of aggregation, present only slight differences and both are around 20% in 2020 for GDP and population (23.1% for the EU-27 and 20.7% for the EU-15).

Using the proposed methodology, the target values of reduction for each community was then computed. Also the new emission rates were calculated, using the estimated population in 2020. The amount of emissions, in absolute terms, is reduced unevenly in each community, but in such a way that the total number is reduced by 17.0-19.0% in 2020. Once again it is clear that the reduction coefficients generated are higher when the initial emission rates (by GDP and population) are higher, and that the great differences in emissions rates between communities are reduced considerably. Indeed, in this case the results are even clearer, as the range of emissions rates is greatly reduced, 1225.2–3033.3 vs. 813.4–2179.1 kgCO_2 eq./inhabitant in the case of modulated weights by population.

Comparison of the real evolution of transport emissions in the period 1990–2007 with the proposed weighted values

We compared the percentages of reduction experienced by EU Member States during the period 1990–2007 with those expected according to the modulation carried out for 2012 (fig. 1). It can be seen that overall the EU Member States have increased the transport emissions, not approaching the target proposed by the Kyoto Protocol, with better values for the EU-27 than for the EU-15. However, when the comparisons are made for each country, there are some significant differences. Bulgaria, Estonia and Lithuania are the countries with the best results. On the contrary, the amount of emissions in Cyprus and Ireland, increased over the same period by a percentage much higher (>150%) than the percentage fixed by the two weighted reduction models. According to the proposed models these countries should have reduced the amount of emissions by 5-9%. Only countries like Germany, United Kingdom, Sweden, France and Finland show sufficient progress to achieve the proposed reduction.

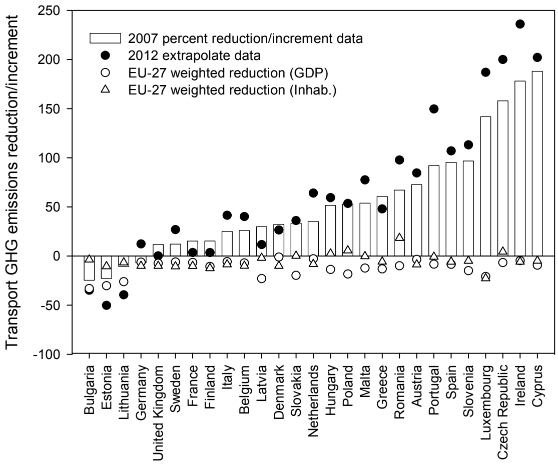

Fig. 1. Percentage of real reduction/increment of the GHG transport emissions, in the period 1990–2007 vs. modulated percentage of reduction/increment for 2012 GDP and Inhabitant modulation procedures. Index = 0 for year 1990, in the order of greatest to smallest real reduction.

The linear extrapolations of data series from the period 1990–2007 to the year 2012 has resulted in negative projections for most of the countries. The increment percentages are higher than those established in this study except for Bulgaria, Estonia and Lithuania, in line with the 8% reduction previously accorded by the European Commission. Once again, Cyprus, Ireland, Czech Republic and Luxembourg present the worst results, and their projections do not invite optimism as regards achieving the EU goal.

In Spain, comparing the results obtained in 2007 with the proposed increment values for 2012, 74% of the Spanish Communities present an increment of between 35 and 85 percentage points of difference. This implies that a lot of work remains to be done in order to increase the amount of emissions in these communities according with the 15% level fixed by the Kyoto Protocol. Catalonia, Basque Country and Navarra are the ones that increased their amount of emissions to 2007 with less than 30% percentage points of difference as proposed in this study (by 43.8%, 43.3% and 36.7% as opposed to the 18.2%, 14.9% and 6.8% proposed respectively), and as such they are examples for other communities to follow. Oppositely, there are communities that need to make a significant progress towards emissions amelioration such as Canarias, Cantabria, Murcia and Region of Valencia, with a difference of more than 85 points between both values. In any case, there is need to review the emissions desirable thresholds and make a new proposal of reduction of the transport emissions rates in Spain for the next years.

Proposal of reduction of the transport emissions rate in the EU for 2020

By 2020, if we extrapolate the values of 1990–2007, overall the EU-27 will achieve a 42.4% increment of GHG emissions. With this result the current EU target would not be reached. We therefore considered that a new commitment should be formulated and agreed for 2020. By this time, those countries with worse rates would be able to redesign and/or improve their transport emission policies and programs.

Considering the dynamism that should prevail in the formulation of quantitative targets, a reduction of 20% in the amount of emissions with respect to 1990 is proposed for the period 1990–2020. We have applied the formula for modulation of the reduction coefficients to the Member States of the EU-27 and of the EU-15 both as function of GDP and population, and to the Spanish Communities.

The new target rates of reduction for 2020 offer different values for each Member State but maintain the target of 20% reduction for the EU as a whole. Once again the states with higher initial transport emission rates are assigned higher coefficients of reduction in the amount of emissions, while in those with lower initial emission rates the progresses would be smaller. The initial dispersion of emission rates between states would be reduced even more over time, changing from a range of 596.0–14035.1 in 2007 to one of 322.9–4227.1 kgCO_2eq. per inhabitant in 2020. The application of the methodology to Spain and its communities for the same period (2007–2020) means that the range of transport emission rate in the different communities is greatly reduced, changing from the range of 5206.1–1939.0 to 2179.1–813.4.

Figure 2 shows the real evolution of emission rates during the period 1990–2007 in the EU-27 Member States and the communities of Spain, and the weighted values proposed for the period 2007–2020 according to population. It can be seen that both the mean and the range of emission rates is reduced with time, particularly due to the greater reduction in those areas with higher initial transport emission rates.

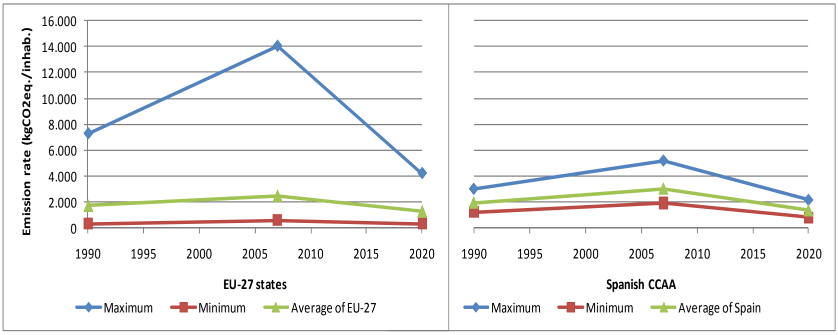

Fig. 2. Real evolution of emission rates in the period 1990–2007 and the modulated proposal for the period 2007–2020 for the EU-27 Member States and the Spanish Autonomous Communities.

Conclusion

The evolution of transport GHG emissions has been uneven and with very heterogeneous values of increment and reduction, both in EU Member States and in the Spanish Autonomous Communities. The countries that have made most progress are those that have implemented transport emission reduction policies and programs in the past (i.e. Germany, United Kingdom, Sweden and France). These countries agree considerably from those countries incorporated into the EU recently (i.e. Bulgaria, Estonia, Lithuania and Latvia) whose overall statistics show decrease on transport emissions. However, due to the economic development of these countries, there will be the need for greater effort and resources to achieve the EU target as they improve their economies as stated in the Transport White Paper in the near future. The Spanish Communities as a whole will also have to make a big effort to converge to the EU target.

To achieve the desirable values, the objective target values of progress should be implemented in a pragmatic way, in accordance with the context and characteristics of each geographic area. The heterogeneity of the statistics underlines the need for "weighted" decentralized decisions to be taken at different territorial levels with a view to achieving a common aim. Under this premise, the proposed methodology allows us to distribute targets for the reduction of transport emissions according to the situation in the starting year for each geographic area. The proposed model has the advantage of being simple to apply. However, its results are orientate and should not be taken as absolute values, but rather as references for the formulation of transport emission amelioration policies.

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Impact of land use and transport policies on carbon emissions in London and Wider South East Region of UK

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Abstract

This study attempts to answer one of the key questions facing academics and practitioners today viz. how far, and by what means, can towns and cities be planned for the future to promote social inclusivity, economic efficiency and environmental sustainability. A range of land-use and transport strategies have been devised and tested using a strategic-level Land-use and Transport Interaction model of the London and Wider South East Region of UK. The four land-use options tested include the trend (reflecting existing policies and committed investment plans), compaction (focussed on the city centre), market-led dispersal and planned urban expansion. The paper presents the results of the carbon emission assessment for four land use options with and without road user charging for year 2031.

Introduction

One of the key questions facing academics and practitioners today is how far, and by what means, can towns and cities be planned for the future to promote social inclusivity, economic efficiency and environmental sustainability. Outer city areas, such as outer suburbs, urban fringes and satellite settlements are of particular interest. This is because these areas are where the majority of people live, most new development is focussed, and where high levels of car dependency, social exclusion and growth exist. Tensions can often arise however when formulating sustainable development policies. For example, promoting a 'compact city' approach with higher densities served by good public transport can conflict with the desires of firms and households to relocate away from urban centres due

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to rising car ownership, and cheaper and better housing. Therefore, much debate currently surrounds the ability of alternative designs of inner and outer town and city areas to achieve economic and environmental sustainability.

As part of the SOLUTIONS project (Sustainability of Land Use and Transport in Outer Neighbourhoods) funded in the UK by the Engineering and Physical Sciences Research Council (EPSRC), a range of land-use and transport strategies have been devised and tested using a strategic-level Land-use and Transport Interaction (LUTI) model LASER of the London and South East Region of UK.

The London and Wider South East (WSE) study encompasses the London, East of England and South East regions covering an area of 4 million hectares and near 20 million people. The study area gravitates around London, a world city of 7 million, which has been under considerable pressure for development as it the most successful economic area of the UK. For more than half a century, there has been a considerable effort to contain the expansion of London with the introduction of a green belt. Most of the growth has taken place beyond the green belt initially in planned New Towns and latter on in the form of urban extensions to existing towns and villages. The outcome has been a mixture of planned and market development leading to severe restrictions in the supply of urban land which resulted in increased property prices. The consequences have been severe in term of reduction of space standards in comparison with other developed countries and housing affordability for lower income groups. There have also been increases in labour cost that impact on the competitiveness of the area. On the other hand there has been protection of green field land around urban areas which is not seen in other developed regions around the world.

Design of transport and land use options

Trend 2001-2031

The Trend option represents a ‘Business as usual’ housing and transport development scenario. The 2001 base year (1997 for transport network) is described using censuses of population and employment, and other observed data, such as that in the Generalised Land Use database. A 2016 base year has also been developed (as many plans are committed to 2016) by updating the land use and transport interaction model LASER using demographic and employment data from the 2001 Census, and from Experian employment projections. The Trend 2031 case is based on the dwellings from the London Plan and Regional Spatial Strategies, and employment, households, and dwellings at the zonal level from DCLG's model TEMPRO v4.3. Densities of new development are based on targets in the Housing Capacity Study for London [1], and in the East and South East Regional Spatial Strategies.

A version of the 2031 Trend is also being tested with road user charging (RUC), in urban areas. The distribution of the land use inputs for the 2031 Trend with RUC is slightly different from the reference case, as it was anticipated that RUC would result in dwellings being allocated slightly closer to the public transport stations and with a slight dispersal of employment from the RUC areas. The net revenue from the RUC is assumed to be invested in additional public transport improvements. All the urban form options (described below) are similarly tested using a RUC.

Compact City

The Compact City option aims to locate people as close as possible to where they work, shop, and carry out personal business, and in areas where there is good accessibility by public transport to reduce travel distances and increase the proportion of trips made by public transport, cycling and walking. This option allocates dwellings within London and several other settlements with strong economies and good public transport. Around 0.9m new dwellings are allocated to London and 0.2m distributed to the other high growth towns. Transport investment is focussed on improving public transport in London, with the emphasis on increasing orbital rail capacity, combined with RUC in urban areas.

Within London, target densities are based on public transport accessibility, proximity to retail facilities and town centres, and employment to dwellings ratio, based on GLA recommendations [1]. The trend in the number of new dwellings on brownfield per borough is estimated from the London Housing Capacity surveys 1999 and 2004. This option would allow development on areas of green belt and metropolitan open land within London that do not have special protection for reasons of nature conservation, amenity value, flood risk or which have steep gradients. It is assumed that the increasing supply of dwellings due to compaction would result in a concentration of employment from the outer metropolitan areas into London.

Market-Led Dispersal

This option aims to reduce the cost of living and labour costs by achieving a better spatial balance between the supply and demand for dwellings. It tests a possible outcome of removing planning constraints except in areas of outstanding natural beauty and sites of special nature, or historic/cultural interest. Transport investment is focussed on highways improvements. Dwellings are dispersed mainly to those parts of the outer metropolitan area where rental values are higher than the WSER average, particularly to Surrey and Berkshire, with additional requirements met from the next most attractive zones. Private sector secondary and tertiary jobs have also been dispersed to areas of economic growth in the outer metropolitan area where the dispersal of dwellings would reduce the cost of living and make the local economy more competitive.

Planned Expansion

This option aims to create planned new settlements and extensions that provide a good quality of life, a vibrant economy, and a high degree of self-sufficiency of local services and jobs, reducing the need for car travel. Planning constraints are applied to achieve densities designed to provide an attractive living environment but which are high enough to support public transport, walking and cycling and have a rail station, and which are large enough to support local services. Areas chosen for development were based on the demand for housing, environmental impacts of the settlement, and the likelihood of attracting sufficient employment. The greatest demand for new dwellings is in the more affluent areas of London and the M3, M4 and M40 corridors where employment growth is outstripping the supply of housing, hence the option includes a local relaxation of the green belt so that the settlements can be located to achieve a better match between housing supply and economic growth in areas close to London to reduce commuting distances, and so offer some the economic advantages of a market-led dispersal but without the sprawl. The option has a similar amount of highway and public transport investment to the Trend, and includes distance based RUC in urban areas.

Sustainability appraisal

As noted above, the purpose of the SOLUTIONS project is to improve understanding of the sustainability performance of the alternative spatial designs for urban areas with a particular focus on areas experiencing high growth rates. The sustainability appraisal at the strategic level uses the LASER model to assess the effect of policy packages with radically different spatial components on the spatial distribution of economic activities, households and travel behaviour. The outputs of the LASER model, summarised are used in this section as the basis for a sustainability appraisal, in which a series of sustainability indicators addressing economic, resource use, environment and social themes are quantified.

The assessment draws on the LASER model transport network outputs (e.g. road location, road type, traffic flow, speeds), or the land use outputs per zone, such as employment by activity type, households by socio-economic group, dwelling numbers and densities. Some indicators, such as CO_2 emissions, are driven by both transport network and land use inputs because both transport and building types affect energy consumption. Where possible, the techniques applied are those widely recognised within the relevant assessment profession (e.g. much of the road network based assessments apply standard Department for Transport methods recommended by the New Approach to Appraisal (NATA) [2] and described in the Design Manual for Roads and Bridges [3]. For others, bespoke methods were developed to address selected sustainability indicators.

The transport sustainability indicators are evaluated using SMARTNET (System for Multi-criteria modelling and Appraisal of Road Transport NETworks) [4], a transport network Decision Support System and a development of an earlier system called TEMMS [5]. TEMMS (Traffic Emissions Modelling and Mapping Suite) addresses atmospheric emissions from road transport, whilst SMARTNET represents an extension to address a wider range of criteria identified as relevant to the sustainability of transport by NATA. Using the information on vehicle flow, speed and road characteristics and additional information on vehicle fleet composition SMARTNET calculates, for each link in the network, a range of evaluation criteria using best practice transport appraisal procedures given in the Design Manual for Roads and Bridges and WebTAG guidance [2]. Criteria addressed include fuel consumption (petrol, diesel), gaseous emissions (seven pollutants, including CO_2), noise emission, water pollution (links requiring diffuse pollution abatement), route severance, route stress (journey ambience), journey delay/reliability and road traffic accidents (fatalities, personal injury accidents, accident costs). Results of the criteria modelling can be summarised for the network, viewed on a link by link basis via database interrogation or GIS network mapping, or summarised on a spatial level (e.g. LASER model zones) to support further appraisal. This study presents and discusses use carbon emissions as an example to illustrate the impacts of the four spatial options.

Emission from transport

CO_2 emission from transport is modelled in the same way as transport energy using the TEMMS-SMARTNET model, and the application of CO_2 emission factors to the vehicle fleet. A range of emission factor databases are now available to European Governments and Local Authorities, including CORINAIR (CORe INventory of AIR pollution), MEET (Methodology for Estimating air pollutant Emissions from Transport), COPERT (COmputer Programme to calculate Emissions from Road Transport), DMRB (Design Manual for Roads and Bridges), the UK EFD (UK Emission Factor Database) and the HBEFA (Handbuch für Emissionsfaktoren des Strassenverkehrs; the 'Swiss/German' handbook of emission factors for road traffic). This study uses UKEFD revised in 2008. This is considered to be the most up to date set of emission factors for use in the UK

Emission from buildings

For commercial and industrial buildings, CO_2 emission is calculated in the same way as for energy use using CO_2 emission per unit floorspace ($kg/m^2/yr$) coefficients determined for each LASER SIC activity group. These coefficients were derived from the national non-domestic building survey in the same way as the energy use coefficients.

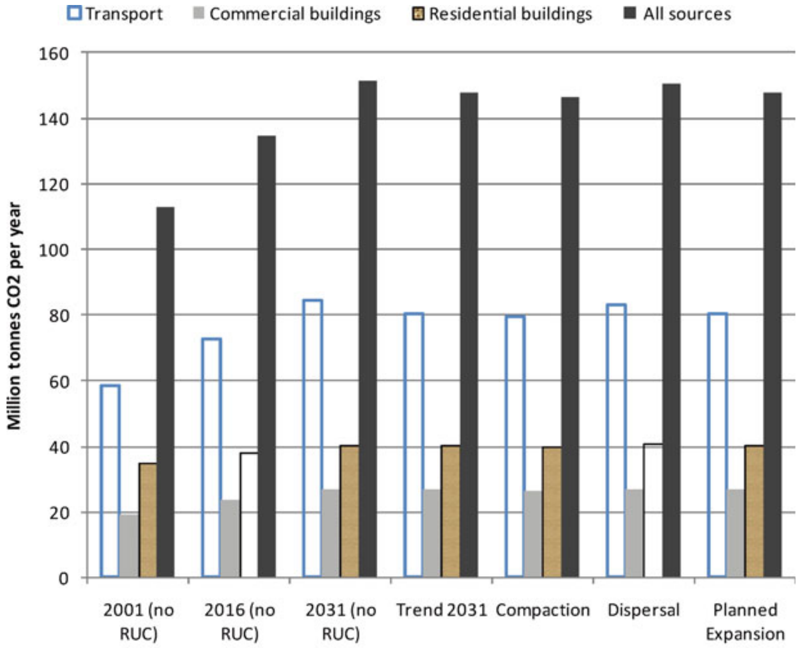
CO_2 emission from the residential building stock is calculated from the building stocks energy use, knowledge of the fuel mix by end use (space heating, water heating, lighting), and emission coefficients per unit fuel by type (g CO_2/Kj). It has been assumed that, whilst different dwelling types have different energy demands, there is no difference between dwellings in terms of the fuel mix used to deliver that energy. This appears a fair assumption, but we note that in future different dwelling types may gain their delivered energy from different sources. Thus dense development, which may include a high proportion of flats, may be fuelled by CHP, whereas low density housing, say mostly detached dwellings, may make relatively more use of passive solar energy.

CO_2 emission factors for delivered energy (kg/Gj for 13 different fuel types) are drawn from the SAP manual [6]. For lighting we apply the factor for delivered electricity and for space and water heating we derive a factor weighted by the share of total delivered residential heating demand, based on EHCS observations. These are that 84.9% of dwellings are heated by gas, 9% by electricity, 3.8% by oil, and 2% by solid fuel (which we assume to be Anthracite, which has an intermediate value for all solid fuels). This process returns a final delivered energy CO_2 emission factor (kg CO_2/Gj) which is applied to the previously modelled energy demand (by dwelling type; and as extant or new build) to return total CO_2 emission from dwellings attributable to space and water heating, and lighting.

Carbon emissions in response to land use and transport options

Total CO_2 emission results

The total CO_2 emission (Mt/yr) for each option is the sum of emission from land transport, commercial and industrial buildings, and residential dwellings. Results are shown below, for the WSER region as a whole, as this is the appropriate spatial scale to report on emission of greenhouses gases.



Note: Transport CO_2 is for 1997 network, 2001 emission factors.

Fig. 1. CO_2 emissions from WSER under spatial options.

Table 1. Total CO_2 under each spatial option.

	Trend			2031 + RUC			
	2001	2016	2031	2031 +RUC	Compaction	Dispersal	Expansion
Transport *	58.7	72.7	84.4	80.5	79.4	82.9	80.5
Commercial	19.3	23.9	26.7	26.7	26.6	26.7	26.7
Residential	34.9	38.1	40.5	40.5	40.2	40.7	40.6
Total	112.9	134.7	151.6	147.7	146.2	150.4	147.8
Change from 2001 (Mt/yr)	0.0	39.4	56.3	52.4	50.9	55.2	52.5
Change (%) from 2001	0.0	19.3	34.2	30.7	29.4	33.2	30.9
Change from 2031 no RUC Trend (Mt/yr)	–38.6	–16.9	0.0	–3.9	–5.4	–1.2	–3.8
Change (%) from 2031 no RUC Trend	–25.5	–11.2	0.0	–2.6	–3.6	–0.8	–2.5

* Base year is 1997 for network, 2001 for emission factors. Mt/yr is million tonnes per year.

These results indicate a substantial increase in total CO_2 emission over the Trend period, for the WSER. Emission from dwellings increases c. 15%, a relatively modest increase given the number of new dwellings, but a reflection of the smaller size and greater energy efficiency of new building. Emissions from commercial and industrial buildings increase to a greater extent but from a lower absolute base (and note that we have not introduced any assumptions re the changing efficiency of commercial buildings, due to a lack of available data). The most substantial increase is observed for the transport sector, with CO_2 emissions increasing by over a third from 1997-2031. This increase is a product of three main factors; a 54% increase in vehicle kms travelled, an increase in the carbon intensity of vehicle kms in the vehicle fleet, and a reduction in trip speeds (emissions are higher at lower speeds). These factors are discussed in the transport energy section.

The spatial options do influence total CO_2 emissions, with relative to the Trend, Market Led Dispersal raising emissions and the Compact City option lowering them. The Planned Expansion option has an emissions profile comparable to the Trend. However, these differences are an order of magnitude less than those observed over the Trend. Furthermore, the differences are achieved through a combination of the spatial option and a road user charge. Table 1 reveals that the road user charge is responsible for a larger proportion of the differences between the 2031 options, than the spatial plan itself. For example, the compaction option experiences a 3.6% reduction in total CO_2 over the 2031 Trend, of which the road user charge alone delivers a 2.6% saving. In other words, for every 10 tonnes of CO_2 saved under the compaction option, 7.2 tonnes are saved by the road user charge and only 2.8 tonnes by the compaction policy.

Conclusions

The analysis presented in this study reveals that the principal drivers behind the predicted growth in transport CO_2 emission are population (number of people taking trips) and lifestyle (distance travelled per person). The impact of spatial organisation has been shown to have a much minor effect on total distance travelled in comparison. Technology has led to an increase in CO_2 emission per vehicle/km, but it is anticipated that it will in future reduce CO_2 emission per vehicle/km. The potential of technology to reduce impact is illustrated by the great success of existing technological measures introduced to reduce emission of CO and NOx.

This illustrates that measures to drive CO_2 down further (or more quickly; spatial plans may be effective over much longer time scales) are likely to revolve around technology measures and measures which affect behaviour (including more severe charging, neighbourhood design, tele-commuting etc). Some advocates of the approach to impact analysis, also warn that treating population as an independent variable, to which policy must adapt, is a mistake. This implies that policies which impact upon the regional population (e.g. national immigration policy) must carefully weigh the economic and social advantage of such policies against the environmental and other costs.

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Carbon sequestration in shrinking cities – potential or a drop in the ocean?

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Abstract

Numerous cities in the industrialized world are loosing population – they are shrinking. Concerning carbon storage, shrinking cities are of particular interest: they have a high potential for urban renewal and related new green space as brownfields are abundant; pressure from development is also low. In this paper, we present a study on the carbon sequestration potential of two urban renewal projects in the cities of Leipzig and Halle in eastern Germany. Results show that both tree planting and biofuel production have a similar carbon mitigation effect and thus are good options for urban renewal. However, compared to overall emissions, the mitigation effect is limited.

Introduction

Humanity has unquestionably entered an urban age, as most people live in cities and the urban population keeps growing on a global scale. The impacts of this ongoing process on the environment are widely studied and increasingly better understood [1]. Concerning the carbon cycle, most CO_2 emissions stem from cities or are related to urban dweller's activities. This is mainly due to burning of fossil fuel, but also due to the destruction and disturbance of natural or agricultural ecosystems [2]. In contrast, urban ecosystems can store and even sequester significant amounts of carbon depending on where urbanization takes place [3]. For example, if cities are located in arid regions, soils and vegetation will develop an increased stock compared to the rural surrounding [4]. While cities in carbon rich ecosystems like, for example, the pacific North-West of the

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US store large amounts in remaining natural green patches, the destruction of forests through urban growth leads to high losses [5]. The general impact of cities on the global carbon cycle is not yet clear [6, 7]. It is clear though, that emissions from burning fossil fuel are far larger than flows in and out of urban ecosystems [8].

Cities are generally expected to keep on growing in the future [9]. While this is true on a global scale, a process that is rarely associated with cities and even less often studied by ecologists is urban shrinkage and decline. However, many cities in the industrialized world have been loosing population and in the future, whole countries are entering a new demographic stage of decline rather than growth. De-industrialization accompanied by emigration in combination with low birth rates has reduced the population numbers of cities in old industrial centres, like Northern England, the North-Eastern USA or Central Germany. In addition, the economic transition after the collapse of the socialist system spread the phenomena to many parts of Eastern Europe, including Eastern Germany [10]. In the last decade, increased effort has been made to actively manage shrinkage, e.g. through state funded demolition of surplus housing units and urban green space developments on brownfields [11].

In this article, we discuss the carbon mitigation potential of two urban reconstruction and urban forestry projects in the shrinking cities of Leipzig and Halle, situated in Eastern Germany. We focus on carbon sequestration by trees and the fossil fuel offset by biofuel plantations. We choose to model the CO_2 balance for 50 years in order to cover a typical lifetime of urban trees. Sequestration by trees increases with age and there is no fixed annual rate. After 50 years, tree mortality will increase and make predictions less accurate.

Study area

Shrinking cities in East Germany

Over the last two decades, Eastern Germany has lost more than 1.2 million inhabitants mainly due to out-migration and suburbanisation. In addition, more than 770,000 new housing units where built. In the late 1990s it became obvious that the development of vacancies was problematic. In 2005, it was estimated that vacant flats added up to 1.3 million or about 1/6 of the total housing stock of Eastern Germany [12]. In 2002 the average vacancy rate was 14.9 %. Some cities had lost a double digit percentage of their population, e.g. Halle saw a reduction of its population from 309,406 in 1990 to 247,736 in 2000 [13]. In the same time, the population of Leipzig reduced from 557,341 to 493,208 [14].

The oversupply of housing and the wide-spread vacancy led to the implementation of the programme “Stadtumbau Ost” (urban reconstruction east) was created – a 2.5 billion EUR fund aiming at stabilizing cities affected by demographic and economic change. Its main tool is demolition, accompanied by the enhancement of neighbourhoods, infrastructure and re-use of vacant land. The programme aimed at demolishing 350,000 units between 2002 and 2009 [11]. The German Federal Environment Agency estimated the total area of brownfields in Germany to be 139,000 ha in 2000 [15]. In 2009, the city of Leipzig estimated the total area of brownfields to be 2.4 % of the city area or 700 ha [16].

Most demolition funded by “Stadtumbau Ost” programme occurred in prefabricated housing estates build between 1950 and 1990. Most enhancements happened in areas of old housing stock, which had been built before 1918 [11]. Therefore, we focus on two urban reconstruction projects; one in prefabricated housing and one in an area built before 1918.

The “Grünes Band” (green belt), Leipzig

The study area is located in a residential area developed in late 19th and early 20th century in the district Volkmarsdorf (Fig. 1).

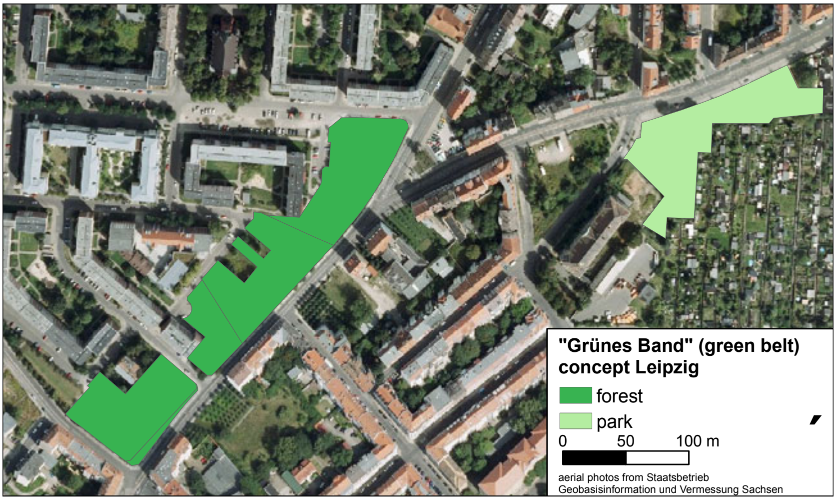

Fig. 1. The urban reconstruction project in Leipzig.

Between 1990 and 2000, this district lost 40 % of its population, ending up with 7911 inhabitants. The housing vacancy rate in 2000 was 44 %. In the course of the "Stadtumbau Ost" programme, the city of Leipzig created a framework plan that aims at preserving building ensembles as well as increasing the size and connectivity of green space [17].

Doing so, a green belt with a length of ca. 600 m has recently been build along a busy road (Fig. 1). It consists of two separate sections, each with a distinct design. The first is 1.46 ha in size and densely planted with trees. The dense, forest-like patch is reminiscent of the houses that once stood there. The second part, 0.7 ha in size, has a park-like design with lawns and fewer trees. For further information and plans, see [17].

The "Waldstadt" (forest city) Silberhöhe, Halle

Silberhöhe was a prefabricated housing estate build between 1979 and 1989, ca. 212 ha in size (Fig. 2). It was erected for the workers of the chemical industry south of Halle and in 1990 ca. 40,000 people lived there. By 2005 the population had dropped by ca. 60 % to 15,608 and according to the prognosis made by the city of Halle for 2015, a population of just 9,600 is expected. The 14,550 units have been reduced to 11,680 by 2005 due to demolition [18]. Still, the vacancy is 27 % and demolition continues.

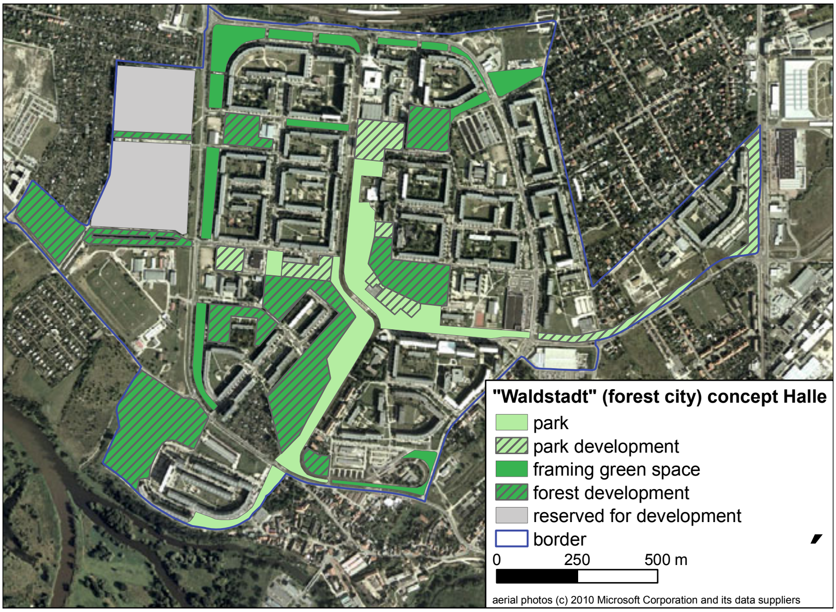

Fig. 2. The urban reconstruction project in Halle.

In order to cope with the development and to improve the situation for the remaining residents, the "Waldstadt" (Forest city) concept was developed. Step by step, preferably from the periphery to the central parts, the surplus of housing stock is replaced by forests. The central part is developed into a landscape park. 25 ha of new forests and 7 ha of new landscape park are planned [18] (Fig. 2). Some land that is kept open for possible future development is used as a short rotation biofuel plantation [19].

Methods: Management options and their carbon footprint

There are several options for urban brownfields. Re-development is often preferred but this is rarely an option in shrinking cities. 85 % of vacant land created by demolition under the "Stadtumbau Ost" programme remained vacant. Roughly two thirds of this land was turned into green-space with most of it becoming lawns [20]. Besides turning the newly created land into parks, other options are sports grounds, gardens, growing biofuel or planting forests. Another option is to do nothing and allowing for natural succession. Each land use option comes with its own carbon footprint. In the following, we look in detail at four management options that are relevant for the studied sites: lawn, forest-like development, park-like development, and poplar plantation for biofuel.

Lawns

One of the easiest and cheapest options for brownfields are lawns. After demolition, soil is spread on the site and grass is sown. The above ground carbon storage of lawns can be neglected [21]. Below ground carbon storage is hard to predict, especially on the very heterogeneous infill like it is encountered in Halle, Silberhöhe [22]. However, as the management involves fossil-fuel lawn mowers, it causes emissions. We have calculated emissions for the case study in Leipzig. Mowing grass once a year by riding mowers causes ca. 2.5 Mg CO_2 emissions per ha in 50 years, mowing 10 times per year causes ca. 6.5 Mg CO_2 per ha in 50 years (see [23] for details).

Forest-like development

While trees grow they sequester carbon through photosynthesis. We conducted a life cycle analysis (LCA) for a case study in Leipzig. We simulated tree growth over 50 years and balanced it against emissions from construction and management. Tree density was 135 trees per ha, consisting of Norway Maple and European Ash in equal quantities. Growth rates were derived from the street tree database of the city of Leipzig. Ivy is used as ground cover, which causes few emissions from management. Overall sequestration after 50 years is 210 Mg CO_2 per ha at average growth rates (154 Mg CO_2 per ha at slow and 282 Mg CO_2 per ha at fast growth, see [23] for details).

Park-like development

The carbon footprint of the park-like design was also calculated in a LCA for the case study in Leipzig. Tree density is 94 trees per ha with Norway maple and European Ash in equal quantities. Grass is used for ground cover, which is mown once a year. Overall sequestration after 50 years is 146 Mg CO_2 per ha at average growth rates (106 Mg CO_2 per ha at slow- and 196 Mg CO_2 per ha at fast growth, see [23] for details).

Biofuel plantation

There is little experience with biofuel plantations in urban settings. The city of Halle has first planted poplars on brownfields, created by demolition in 2007 [19]. There is a similar project in Dessau [24]. On poor soil and without fertilizers, biomass production on short rotation plantations can be expected to be 6 $Mg_{(dry\ weight)}$ ha^{-1} a^{-1}[25]. The calorific value is 18.5 MJ kg^{-1} [26]. This leads to an energy gain of 111 GJ ha^{-1} a^{-1}. Considering the calorific value of fuel oil of 42.61 MJ kg^{-1} and the specific emissions of 267.762 kg CO_2 MWh^{-1} [27] the emissions are 3.17 kg CO_2 kg^{-1}_{oil}. In order to produce above mentioned 111 GJ, one has to burn 2,605 kg fuel oil, which leads to the emission of 8.3 Mg CO_2. One ha of poplar plantation can, therefore. offset fossil fuel emissions of 8.3 Mg CO_2 per year. The true offset is probably slightly smaller as the estimations are not based on a LCA. In a LCA of wood produced on unfertilized short rotation plantations, the emissions where 11.5 kg CO_2 per $Mg_{(dry\ weight)}$ [28]. No LCA for poplar wood produced on urban short rotation plantation exists.

Scenarios

We calculated the CO_2 balance of both study sites for the existing plans described above as well as two scenarios. In the first scenario we assume that the brownfields are simply turned into lawns. This is a very likely scenario as it is the cheapest. The second scenario assumes 50 % lawn and 50 % biofuel production on 2.16 ha in Leipzig and 32 ha in Halle. This takes into account that residents will probably claim some of the newly created space for themselves and that not all areas are suitable.

Results

The reconstruction project in Leipzig can be expected to sequester around 409 Mg CO_2 in the next 50 years. The much larger “Waldstadt” project in Halle will lead to the sequestration of 6,272 Mg CO_2. Just creating lawns on the vacant sites would cause net-emissions of up to 14 and 208 Mg CO_2, respectively. The carbon offset by biofuel production is very similar to the sequestration by trees (443 and 6,565 Mg CO_2 respective). Table 1 summarizes the results for the two case study sites.

Table 1. The carbon balance of the two reconstruction projects for a simulated period of 50 years is shown in line one. Line two gives the carbon emissions if just lawns are created. In line three the carbon offset of biofuel production on 50 % of the reconstruction project area is given.

	Net CO_2 balance [Mg CO_2 •$50a^{-1}$]	
	“Grünes Band” Leipzig	“Waldstadt” Halle
1. plan	409	6,272
2. lawn	–5 – –14	–80 – –208
3. biofuel	443	6,565

Discussion and conclusion

The urban reconstruction plans for both case studies lead to a net carbon sequestration through tree growth. The predicted increase in carbon stock is equivalent to emissions from 2.5 million kilometres travelled by car (passenger car, average traffic mode, year 2000 [27]) in the case of Leipzig and 38.9 million km in the case of Halle. Although this might offset the emissions of a few inhabitants, it is clear that the mitigation potential is small even on a local scale. In this vein, urban forestry projects in shrinking cities are just one of the many small puzzle pieces in the context of CO_2 emission and mitigation. Still, there are several aspects that are worth taking a closer look.

First, we offer an extension to the common environmental impact assessment by estimating the carbon footprint of alternative design options. As it could be shown, the cheapest option of creating lawn comes with net emissions that could be easily offset by planting a few trees.

Second, biofuel plantations are an option worth considering. Even assuming poor soils and using only 50 % of the area, the mitigation potential is comparable to planting trees in parks and forests. For interim use, a biofuel plantation is probably better than park or forest as it is low in construction and maintenance effort and costs and considering that trees only reach high sequestration rates after decades of growth. However, such plantations are more of an option for larger brownfields in the periphery as they can be found in prefabricated housing estates.

Third, the indirect effects of tree planting on the carbon emissions might be larger than the direct ones. The improvement of local climate by well placed trees can significantly reduce energy need for cooling and heating [29]. To our knowledge, there are no studies on this effect of urban reconstruction projects yet.

Fourth, well designed urban green might change people's perception and behaviour. The positive effect of green space on physical and mental health has been identified by several studies [30]. The newly built green space in shrinking cities could encourage people to switch to cycling or walking as a means of transport and encourage more local activities, thus reducing the CO_2 emissions. These effects should be monitored.

Shrinkage, rather than growth are predicted to influence the urban landscape in many parts of Germany in the future [31]. Or as Bückner puts it: "We will have more space for less people" [24]. It should be seen as a opportunity, rather than a burden. While cities should focus on reducing their emissions, well managed shrinkage offers the opportunity for mitigation in combination with increasing the well-being of urban dwellers.

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Urban sprawl, food security and sustainability of Yogyakarta City, Indonesia

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Abstract

The objectives of the study are: (1) to analyze the impact of urban sprawl on farmland conversion by using landsat data, (2) to analyze the impact of land conversion on food supply and food security, (3) to understand the prospect of farming activities in the fringe area of Yogyakarta, and (4) to find implication of urban sprawl towards sustainability of Yogyakarta city. The study shows that urban sprawl causes the loss of food calories, lowering food supply, the decrease of food security and the decline of sustainability of farming activities that result in disharmonious urban and rural relations and threaten the sustainability of the city.

Introduction

The fusion between urban and rural areas not only has been a main issue in urban planning but it has also become a central theme of sustainable city development. Such a "new" paradigm is being promoted based on the fact that high interactions between urban and rural could be the important factors of sustainable urban development. This is in contrast to what people understand as modern city concept in planning system that attempts to separate urban from surrounding rural areas.

The concept of urban-rural fusion by integrating urban and rural areas becomes a promising approach in urban development since making a sharp distinction between urban and rural areas in many cases has failed in controlling urban growth mostly in developing countries. Therefore, any notion to keep away rural sides from urban development is questionable. Even in a broader sense, municipalities could be regarded as cities

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integrating everything from upstream forests to mid-stream farmland and villages to downstream urbanized areas, even down to the coast (Takeuchi *et al*. 2009).

Whatever the concept of urban-rural fusion for sustainable city is used, the role of farmland in supporting the city live is not denied. Hence, effective land use planning and policy in controlling farmland encroachment surrounding the city becomes justified (Conklin and Bryant 1974: Lapping 1975; Kanada and Irham 1998).

The study on the impacts of urban sprawl in the fringe areas has been pioneered by some researchers such as Clawson (1971), Isberg (1975), Furuseth (1982), Lockeretz *et al*. (1987) and Pacino (1990). From our experience, serious impacts of uncontrolled urban sprawl in Asian developing countries could be: (1) the massive loss of fertile farmland causing food insecurity, (2) uncertain prospect of farming activities in the fringe area that lead to continuous haphazard city growth, and (3) sustainability of city lives becomes disturbed as more and more people loss their opportunity to get food supplied by the city and its surrounding.

Other studies also show that urban and sub-urban agriculture often provides a significant contribution to many major cities' food self-reliance (Soni and Salokhe, 2009). Needless to say, that the intensive urban sprawl surrounding Yogyakarta city has lowered the potentiality of sub-urban and urban farming that contribute to cities food supply. Therefore, preservation of farmland becomes a key factor for sustainable city development as urban people highly dependent on the availability of farm production. Without an adequate protection of farmland, the country will find difficulty to meet long term food needs for the growing population in the city regions.

The objectives of the study are: (1) to analyze the impact of urban sprawl on farmland conversion by using landsat data, (2) to analyze the impact of land conversion on food supply and food security, (3) to understand the prospect of farming activities in the fringe area of Yogyakarta city, and (4) to find implication of urban sprawl towards sustainability of Yogyakarta city.

Experimental – Method

The study was conducted in fringe area of Yogyakarta city covering some part of three adjacent districts, namely Sleman district (most favourable region), Bantul district (less favourable region) and Kulonprogo district (least favourable region). Forty seven villages within those three districts are selected as samples, 18 villages in Sleman, 13 villages in Bantul, and 16 villages in Kulonprogo (Figure 1).

Landsat data of 1996 and 2007 are used to analyse the dynamics of land conversion taken place in all villages of the study area. The area of converted farmland is obtained by overlaying those two sequential years of the landsat data. From the selected villages, 201 farmers (50 farmers in Sleman, 48 farmers in Bantul and 103 farmers in Kulonprogo) were interviewed to get information on their views regarding food security and prospect of farming activities.

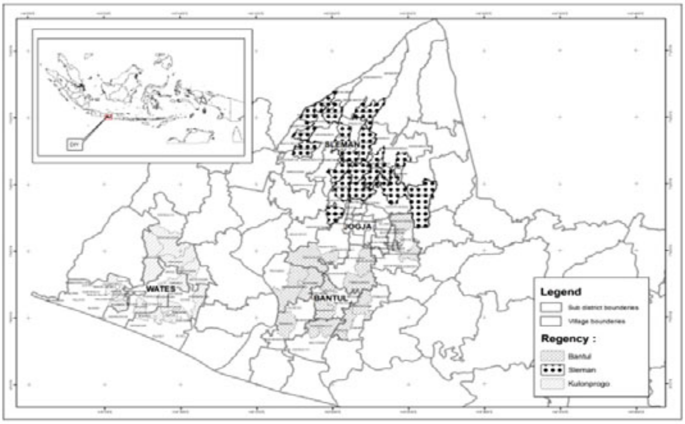

Fig. 1. Location of study**.**

Food loss is calculated from potential rice production of converted farmland based on its planting pattern in each village. The rice production is then converted into calories to get the amount of forgone calories or number of people who lost opportunity to consume the available calories in the study area.

Food security is analyzed by applying Jonsson and Toole (1991) method and its modification. The method is simply calculating the ratio of compartment expenditure on food to percentage of energy sufficiency. The result of the ratio is categorized into four categories: food secure, vulnerable, questionable and food insecure. This method is then modified by using ratio of income to expenditure of farm household instead of farmers' expenditure alone as used in Jonsson and Toole method. The result of the calculation is divided into 8 categories: food secure-stable, food secure-less stable, food secure-unstable, vulnerable, questionable-high saving, questionable-adequate saving, questionable-without saving, food insecure.

Prospect of farming activities is analyzed by applying Lockeretz *et al.* (1987) method by asking land-holding farmers about two aspects (1) the existence of their farmland during the last five years and (2) the farmers plan and expectation about their land for the coming five years. The farmers' view is obtained by giving them questions on those two aspects related to: land size, irrigation, productivity, income, farm inputs, capital and local culture. Another two questions are added to the second aspect related to future plan and inheritance matter of their farmland. Score (1-3) is given to the farmer's response on each question. Score 1 is given to pessimistic response, score 2 is neutral, and score 3 to optimistic response.

Results and discussion

One of the impacts of urban sprawl is conversion of farmland to other uses. Since Yogyakarta is not a typical of industrial city, the most distinctive farmland conversion in the fringe area of the city is to housing. Massive development of housing complexes taking place during the last twenty years becomes the main cause of high conversion rate. There is a strong relation between the rate of conversion and the closeness to the city center as reflected in low, middle and high conversion rate (Figure 2).

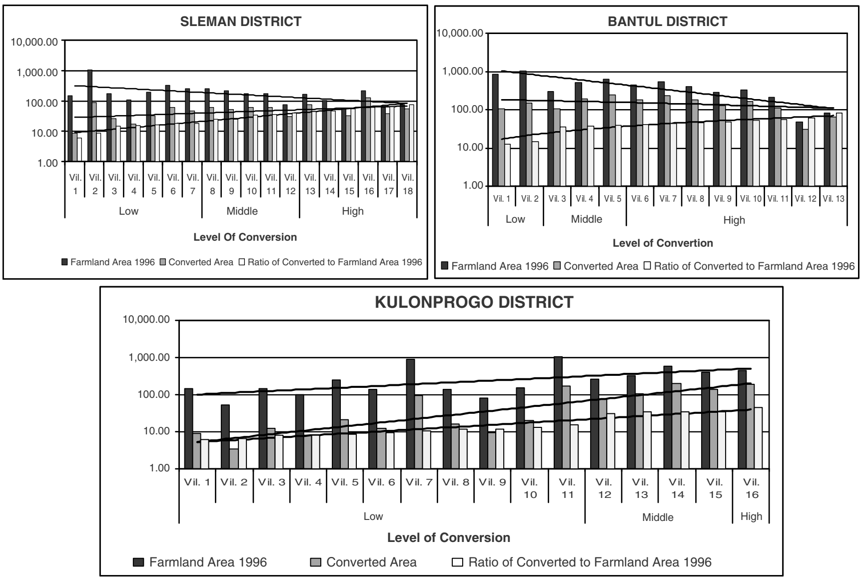

Fig. 2. Convertion rate in fringe area of Sleman, Bantul and Kulonprogo.

In the area of Sleman district (most favorable region), 922 ha of fertile farmland has been converted mostly to housing complex, with the rate of conversion about 33% during the last ten years (3.3% annually). In Bantul district (less favorable region), the converted area is nearly 1900 ha with conversion rate 4.3% annually. Although Sleman is the most favorable region, but the conversion rate is a little bit lower than that of Bantul since the fastest conversion process in Sleman took place during 1990s.

Since Kulonprogo district is the least favorable region, it shows much lower rate compared with that of other two regions. The converted area of farmland is counted to 1,085 ha during the last ten years. The conversion rate is just 1.7 % every year. Similar to Bantul district, the massive conversion has been taking place during the last ten years.

Land conversion and Food Loss

The direct impact of farmland conversion is the loss of food especially rice (as main staple food). The result of analysis shows that almost 20 ton of rice was lost in the study area every year due to conversion (4.6 ton in Sleman, and 9.4 ton and 5.4 ton in Bantul and Kulonprogo, respectively). If the amount of rice is converted into calorie and number of people who consume that calorie, it is found that more than 25 thousand people lost their opportunity to get food in Sleman, 42.3 thousand people in Bantul, and 48.5 thousand people in Kulonprogo (Figure 3).

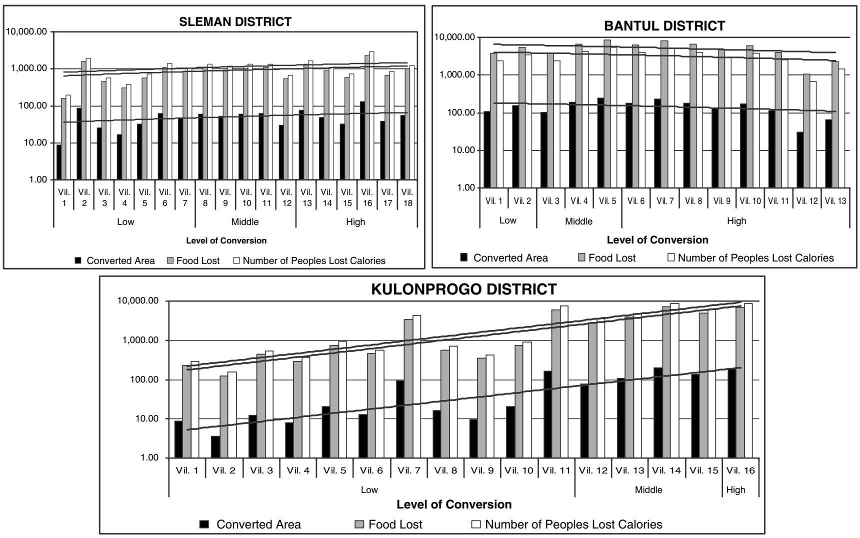

Fig. 3. Land conversion and loss of food calories.

The Figure shows that the loss of calories and the number of people who lost the opportunity to get food due to conversion are increased as the rate of conversion is increased, except in Bantul. The decreasing trend of food loss in Bantul district may be because of the success of local government in improving rice yield.

Food Supply and Food Security

From landsat data 2007 it can be estimated the amount of food supply through potential production of rice in each village sample. It is found that the amount of food supply is estimated to be 51.8 million calories which is equivalent to 64.5 thousand people in Sleman district. The supply of food in Bantul district is estimated to be 55.8 million calories, equivalent to 69.4 thousand people. While in Kulonprogo district, it is estimated to be 42.7 million calories equivalent to 52.2 thousand people (Table 1). Dividing the number of equivalent-people to the total population in each village, the supply of food is enough to provide only 25.3% of the entire population in Sleman, 34.2% in Bantul, and 67.9% in Kulonprogo. Consequently, the rest of the food should be supplied from outside the village.

Table 1. Calorie supply and number of people provided, 2007.

	Calories (million)	Eq. number of people (thousand)	% to total population
Sleman	51.8	64.5	25.3
Bantul	55.8	69.4	34.2
K.progo	42.7	52.2	67.9

The availability of food (food supply) will determine food security in the study area. By using Jonsson and Toole (1991) method and the modified one, it is found that the higher the conversion rate, the lower the food security both in Sleman and Kulonprogo districts (Figure 4). However Bantul shows a decrease trend of food security as the conversion rate increases. This is most probably due to a successful effort of the local government in promoting food diversification.

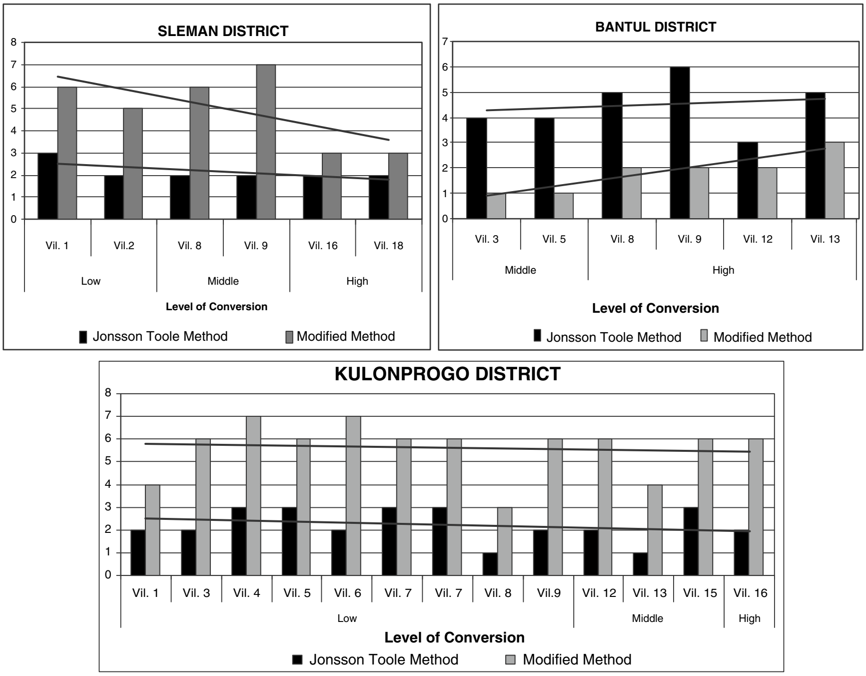

Fig. 4. Land conversion and food security.

Prospect of Farming Activities

The prospect of farming activities to some extent reflect the farming sustainability in the sense that the more prospective of farming activities, the higher the sustainability. The prospect of farming activities can be seen from two combined perspectives of farmers' view on their farmland: the existence of farming during the last five years and the farmers' plan and expectation on their farmland for the coming five years (future plan).

By using LFS (level of sustainability) score, it is found that the higher the rate of conversion, the less prospective (less sustainable) of the farming activities in Sleman distrct (Figure 5). From the farmers' view, such tendency is mainly caused by their future plan rather than by the farming existence during the last five years.

Bantul district shows a similar tendency like that of in Sleman district. The higher the conversion rate, the lower the sustainability of farming. The main cause is due to their future plan on their farmland. This is different from that of Kulonprogo district. Although it has a similar trend in LFS score, but the main reason for the lower sustainability is mainly because of the condition during the last five years, namely when the intensive conversion occurred.

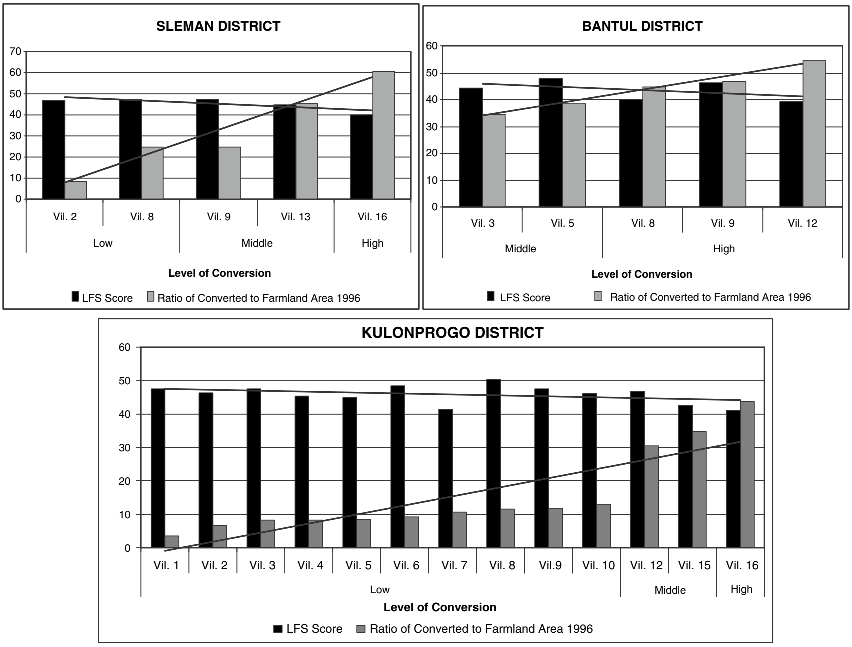

Fig. 5. Land conversion and farming prospect.

Urban Sprawl and Sustainability of City

The impact of urban sprawl towards farmland in the fringe area has a number of implications. Massive land conversion surrounding Yogyakarta city has been resulting in the loss of food production, and hence the decline of food supply. The analysis clearly shows a significant decline of food supply within the fringe area of Yogyakarta city during the last ten years. The most severe impact occurs in Sleman, followed by Bantul and Kulonprogo.This condition eventually threaten the urban-rural relations, hence, urban development becomes less sustainable.

Urban sprawl also becomes the key factor of food insecurity since more people lost their opportunity to get food from the available farmland production in each village. The result of the analysis shows that, the higher the conversion rate the lower the security or the higher the insecurity. Food insecurity not only cause a negative impact on urban development but also towards national food sufficiency program since farmland conversion has also been taking place in all other cities in the country.

Uncertain prospect of farming activities within the fringe area is another threatening factor toward urban-rural relation. The LFS score provides an indication of farming activities to be less sustainable. The higher the conversion rate, the lower the sustainability of farming activities, and therefore, the urban development.

Based on the aforementioned above, therefore, protection of farmland should be part of the urban development planning and policy in order for the city to be sustainable. This is in line with the fusion concept in that urban and rural relation and activities should be harmonious. In the case of Yogyakarta, strong control of housing development is badly needed. Local government needs to implement seriously strict regulation and sanction not only towards housing developers but also district officials who have responsibility towards improper housing development process.

Based on the view of farmers, it is important to keep farming activities attractive for the farmers. Therefore, subsidies for the farmers to keep farming operation feasible can be one of the important solutions. Provision of incentives such as tax relief is another important solution to be applied.

Food sufficiency and food security has become one of the main policies of the Indonesian government. For the sake of food sufficiency and food security program, the government with the support of House of Representatives is now in the process of imposing a fixed farmland area in every district through zoning ordinance. In this case, the role of local government becomes very important in succeeding such a program.

Conclusions and Recommendations

1. The loss of food (calories) and the number of people who lost the opportunity to get food due to the urban sprawl are increased as the rate of conversion is increased, except for Bantul. This is most probably because of the success of local government in improving rice yield.

2. There has been a significant decline of food supply within the fringe area of Yogyakarta city during the last ten years due to conversion of farmland that negatively affects food security in the study area. The study found that the higher the conversion rate, the lower the food security except in Bantul due to the successful effort of local government in promoting food diversification other than rice.
3. Low food security not only cause a negative impact on urban development but also towards national food sufficiency program since farmland conversion has also been taking place in other cities all over the country. Based on the LFS score, the study concludes that the higher the rate of conversion, the less prospective the farming activities and therefore its sustainability.
4. Since urban sprawl has been resulting in production loss, food insecurity and unsustainable farming activities that eventually affect the sustainability of the city of Yogyakarta, the protection of farmland should be part of the urban development planning and policy not only at municipality and district levels but also at central government level. Local government needs to implement seriously strict regulation and sanction not only towards housing developers but also district officials who have responsibility for the improper housing development process.
5. In order to secure food sufficiency and food security in the long run in supporting harmonious urban-rural relations, the government plan to impose a fixed farmland area in every district through zoning ordinance should be fully supported.
6. Provision of incentives for the farmers such as tax relief and subsidies to keep farming operation feasible and attractive should be implemented as one of the conditions for sustainable farming activities in the fringe area.

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The local food system as a strategy for the rural-urban fringe planning – a pathway towards sustainable city regions

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Abstract

Sustainable city region addresses the need for city regions to be self suf lyse the local food system as a strategy for planning the rural-urban fringe, a part of the city region where most of the resources to support the food production are prevalent.

A lesson from the Jakarta Metropolitan Region (JMR), Indonesia shows that the failure in planning the rural-urban fringe undermines the ability of this city region to be self sufficient in relation to the provision of food. To this extent, the local food system serves as distinctive strategy to underpin the rural-urban fringe planning.

Introduction

Background

Agenda 21 addressed two major themes to improve the achievement of sustainable development. First, Chapter 5 calls for actions to enhance the life support system, which addresses the need to provide the growing population with adequate foods for their continuance of life [20, 21, 26, 35,]. Second, Chapter 28 calls for local authorities to undertake local actions towards sustainability, where with their capacity as the closets level of government to the community are capable to set sustainability priorities more effectively [1, 32].

Enhanced life support system refers to the ability of a certain geographical region to be self sufficient in relation to the provision of food, which refers to the use local agricultural resources efficiently [29, 33, 38]. For regions that have been largely urbanised, where most of the resources to support the food production (e.g. agricultural lands and the

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community that is engaged in farming activities) are scarce, the rural-urban fringe would nevertheless be an essential component to endure agricultural activities.

The rural-urban fringe is a set of dispersed landscapes surrounding the urban areas where the agricultural resources comprising significant proportions of prime agricultural lands and the community that is predominantly engaged in the agricultural activities are the attributes of this area [6, 7, 10, 14]. Despite the attributes they have, the rural-urban fringe is a vulnerable part of the region that is facing serious threats from urban sprawl [6, 13, 14, 16, 37, 39].

The urban sprawl itself is often uncontrolled due to failure of planning to address an appropriate strategy to protect the attributes of the rural-urban fringe from the ever expanding urban areas [15, 36]. Compacting the existing built up areas and applying a strict regulatory zoning, which aims to slow down the urban sprawl have been the most common strategies [2, 5, 18, 23, 41]. However, these strategies only focus on spatial matters while disregarding the socioeconomic factors that may underpin the protection of the rural-urban fringe.

Socioeconomic factors are the significant contributor to the urban sprawl. For example, the urban sprawl in the Jakarta Metropolitan Region (JMR), Indonesia had been largely caused by the massive development of new towns and industrial estates, which started in the early 1990s. The urban expansion attracted landowners to take advantage by selling their agricultural lands to private developers in order to gain greater revenues [10, 11]. Meanwhile, the industrialisation itself attracted farmers to abandon their agricultural activities to work in the industries that may offer higher incomes [24]. The effect to agricultural sector for instance, is the declining productivity of rice up to 25.73% between 1996 and 1999 [8]. In this case, strengthening the motivation of landowners and farmers to keep participating in the agricultural activities is considerably important to be addressed in planning.

Research Aim and objectives

The aim of this study is to analyse the local food system as a strategy to underpin the rural-urban fringe planning as an initiative to achieve sustainable city regions. The three objectives of this study are:

1. *To identify the features of local food system as a strategy associated to sustainable city regions in planning the rural-urban fringe;*

2. *To analyse the failure in planning the rural-urban fringe that is affecting the ability of city regions to be self sufficient in relation to the provision of food;*
3. *To recommend a strategic approach to underpin the rural-urban fringe planning to enable city regions to be self sufficient in relation to the provision of food.*

Research methods

This study is a literature-based research that will be investigated through a case study method. The major components of the case study method include the propositions, logic linking data to the propositions and criteria for interpreting the findings [42]. The advantages of the case study method [9] are:

1. It allows the researcher to explore information from particular places in a certain period without a direct observation, which means the effective use of time to collect valuable information
2. Case study method consults the findings from previous studies, therefore, the researcher can learn from the perspectives of different experts who already investigated researches in the related area.

However, the weakness of this method is that the findings are difficult to be generalised and the knowledge resulted from the study may not be applicable elsewhere [17, 19].

Results and discussion

The Rural-Urban Fringe

Rural-urban fringe is a set of dispersed landscape surrounding the urban areas that can contain significant proportions of prime agricultural lands [6, 14]. It is an area where the community is predominantly engaged in the agricultural activities as a source for income and as a media to interact with each other [7, 10].

Ironically, the rural-urban fringe is described as the landscapes that will no longer persist their "rural amenities" and is about to transform to suburbia [6, 13, 14, 16, 37, 39]. There are two main causes for this. First, is the pressure of population growth in the urban areas and second is the invasion of urban infrastructure development.

The rural urban fringe is a part of the city region where the population growth is higher than the urban centre. For example, the population growth in the rural-urban fringe of Jakarta, Indonesia between 2004 and 2007 was above 2.5% in average, which is far above the national average of 1.97%, while the urban centre (Jakarta) only experienced a 0.67% growth per annum. [22]. Apparently, the population growth attracted the growing demands of housing that caused a massive urban infrastructure development in the rural-urban fringe [6, 10, 39].

The invasion of urban development turned the rural-urban fringe to a place where development and conservation compete for spaces. Yet unfortunately, the land is valued from their potential to be developed into urban uses rather than the agriculture or environmental amenities they offer [13, 16, 37, 39]. Moreover, as rural-urban fringe is located only about 10 to 50 miles from the centre of cities, which is considered as "accessible" to the urban centre, more people prefer to commute rather than living in cities that is associated with the high property tax and hustle environments [7, 13].

The Challenge of Planning

Planning is one of the essential tools to achieve sustainable development. The process of planning in achieving sustainable development, which includes the formulation of objectives, methodologies, setting clear strategies, and means of implementation, is a useful guide to addresses the message of "think globally and act locally" [27]. This implies that a planning framework should notice cross boundary issues that will be delivered in local actions. Acting locally means considering the "local people" as the heart of development where planning initiatives must clearly address the human needs and how to shape a better interaction between human and their environment [4].

Literatures on rural-urban fringe identified two major failures of rural-urban fringe planning. First is the scale problem, featuring the difficulties to deal with cross boundary issues involving different levels of authorities [27]. Second, is due to the absence of a distinctive strategy to deliver sustainability outcomes at the rural-urban fringe [36].

Recent studies on rural-urban fringe planning mainly consider the landscapes as "phase" of urban development rather than a "space" with their own rights [36]. Spatial planning had focused more on the management of territorial boundaries and treated the rural-urban fringe as the landscapes that offer opportunities to expand the urban development. Meanwhile, efforts to promote development that will deliver greater benefit to the local people and more likely to protect the rural amenities of the landscape (e.g. urban agriculture) is unprecedented [3]. Planning often failed to address a distinctive strategy to manage the lands and improve the mutual interactions between human and the environment, because they consider that changing the landscape into urban uses can be more economically beneficent [36]. As long as rural-urban fringe is valued as a "phase" of the urban development, the rural amenities (e.g. agricultural lands and the community that is engaged in farming) within the rural-urban fringe will disappear alongside the urban expansion.

The Significance of Local Food System

The mainstream of sustainable city region as to the context of this study is to develop an enhanced life support system, which implies to the ability of city regions to be self sufficient in relation to the provision of food.

The local food is contextualized as a "strategy" that incorporates strong community participation and the efficient use of local resources to produce the food for the fulfilment of local consumptions [29, 33, 40]. There are two main reasons why a local food system represents a high level of sustainability. First, besides promoting the efficient use of resources (from being self sufficient in food), it encourages the city region to allocate adequate amounts of green open spaces to accommodate agricultural activities. Second, as it requires strong community participation, a local food system empowers people to take advantage of income generation from a sustainable job (e.g. participating in farming activities) while also improving social cohesion.

For densely populated city regions, where the built up areas are intensive, there is a question regarding the available spaces for the agricultural activities. In this circumstance, considering the rural-urban fringe is reasonable. The rural-urban fringe within city regions can contain prime agricultural lands [6, 14]. These lands can also be the homes of people who are mostly engaged in the agricultural activities, providing cultural identity as well as an important source of income [7, 10]. The agricultural lands and the community that is engaged in the agricultural activities are the attributes of the rural-urban fringe, which are potentially capable to support the local food system.

Ironically, rural-urban fringes are vulnerable landscapes threatened by urban sprawl. They are often valued as a landscape in transition, where the agricultural lands are about to transform into urban uses and the community's source of income will shift from primary activities (agriculture) to secondary and tertiary based activities [6, 13, 14, 16, 37, 39]. Therefore, the loss of rural-urban fringe attributes implies to the loss of opportunities to develop a local food system as a strategy to enhance the life support systems of city regions. In other words, to plan a local food system successfully means preventing the attributes of the rural-urban fringe from disappearing due to urban sprawl.

Failures in Planning the Rural-Urban Fringe – Lesson from the JMR

The experience of JMR illustrates the tangible problems relating to the research question. Urban sprawl in JMR resulted in a decline in the productivity of the agricultural sector due the loss of agricultural lands and the socioeconomic transition experienced in the rural-urban fringe. This transformation made it difficult for this city region to be self sufficient in relation to the provision of food.

Overview of the Region

The JMR includes five regional territories namely Jakarta, Bogor, Depok, Tangerang and Bekasi, covering an area of 618,919.00 Ha and located at the most densely populated areas in Indonesia. The total population of this city region in 2007 was more than 24 million people with an average annual growth rate of 2.02% [22].

The centre of this City Region is Greater Jakarta, the capital city of Indonesia. The rural-urban fringe surrounding Jakarta (Bogor, Depok, Tangerang and Bekasi) is a vast and rapidly growing region and is affected by urban sprawl and the impacts of economic spillovers from Jakarta [12]. The population at the urban centre (Jakarta) has shown a slow and steady growth of 0.67% per annum from 2004 to 2007, while the surrounding regions experienced a significant growth of higher than 2.5% per annum [22]. This is caused by a high rate of migration from Jakarta to the rural-urban fringe as a response to the urban sprawl phenomenon.

The JMR Sprawl

Urban sprawl started during 1980 to 1990 where the population growth was at the rate of 3.5%, which was above the national average growth of 1.97% per annum. However, the urban sprawl of Jakarta was largely influenced by the private sector that began developing new towns and industrial estates, turning the JMR into a large polycentric region. Further to this phenomenon, on average, 2.71% of the agricultural lands were converted into urban uses every year during 1994-2001.

Up to 2005, the development of new towns and industrial estates in the JMR consumed 52,001 ha of lands covering 8.4% of the total area of JMR. The new towns are mainly low-density settlements occupied by residents in the middle to higher-income groups [11]. Some of the houses are unoccupied because the owners purchased the property only for speculative investment purposes, while they remain settled in Jakarta. Meanwhile, other residents preferred to move to the new towns due to rising property taxes and cost of living in Jakarta [10].

The urban expansion (new towns and industrial estates) is a result of the 'leapfrogging' of development. The polycentric development transformed this region into a powerful growth pole that stimulates the growth of its own surrounding areas. Bekasi was once Jakarta's rural-urban fringe, which was dominated by traditional agricultural activities, and now had become a densely populated urban area.

The Socioeconomic Transition

Apart from the leapfrog development, the urban sprawl and economic spillovers of Jakarta also resulted in a transition in relation to economic and social structures. As the economic activities have shifted from the primary sector (agriculture) to the secondary and tertiary sector (industry and services), there has been a considerable decline in the number of people engaged in the agricultural sectors [24].

There are two main reasons for this shift in the labour force from the primary agricultural sectors to the industrial sectors. First, the ex-farmers with higher educational qualifications were able to meet the required skills in the industrial sector and moved to this sector to improve their income. Second, as most of Jakarta's rural-urban fringe has been urbanised, fewer farmers were willing to maintain their activities due to the increased land rents [10].

Besides the declining percentage of workers in the agricultural sector, the influx of people into the rural-urban fringe contributed to a change in the lifestyle of the community. The rural-urban fringe community is adopting an urban lifestyle, moving away from their rural attributes (cohesive community with their traditional farming activities) [24]. Furthermore, the urban sprawl also attracted landowners to take advantage by selling their agricultural lands to private developers, which offered them greater revenue than could be achieved by occupying the land and engaging in agricultural activities [10, 11].

The Impacts toward JMR's Food System

The decrease in the number of people working in agricultural sectors, coupled with the conversion of agricultural lands to urban uses has resulted in the declining productivity of the agricultural sector. The Department of Agriculture recorded a 27.53% deficit of the paddy field productivity in the JMR during 1996 to 1999. Despite the slight increase of the total harvested area, the paddy fields in the JMR, especially in the surrounding regions declined at a rate above 5% per annum in every region.

These phenomena raised a question regarding the JMR's food supply. The absence of adequate agricultural lands to accommodate food production in the JMR is an obstacle to the optimisation of local resources for food production. For example, the national average rate of rice consumption per capita in 1999 was 0.12 tonne per annum [8]. Assuming that the population of JMR consumed rice at the same amount as the national rate, it implies that there was approximately 2.96 million tons of rice consumed within the JMR. By looking at the annual productivity of the JMR's paddy field, the local production of rice can only cover about 13.18% of the total demand in the JMR.

Recommendations

Approach to Problem Solving

To make resilient the agricultural sector in regions like JMR needs more than just a "spatial" approach (e.g. green belt, urban compaction). It needs a strategy that 'motivates' farmers and landowners to participate in the agricultural activities and understand the value of it rather than a strategy that focuses only on preventing the lands from disappearing. From this point, solving the typical problems experienced by the JMR would be best by considering the "Urban Agriculture" approach. Urban agriculture refers to involvement of the community's participation in the food production by giving them the opportunity to grow crops in their neighbourhoods, where larger portion of the food produced will be consumed at the household level [30]. Yet, not many of the urban and regional planning considers integrating this strategy as a larger part of the initiatives.

Implementing the Local Food System – a Lesson from Havana, Cuba

The reason to learn from the experience of Havana is that the local food system was practiced as a part of their cultural identity, which has successfully improve the community's income and considerable improvement to the environment. Unlike other regions (e.g. Tanzania, Uganda, Zambia, Zimbabwe and other African Nations) where the community was forced to grow their own foods because of their lack of purchasing power (poverty), the practice of urban agriculture in Havana was a prestigious program that is supported by the government [28, 31].

The practice of urban agriculture emerged following the collapse of the Soviet Union together with the termination of trade and agricultural aid from the Council for Mutual Economic Assistance (COMECON) in 1989. During this period, the Cuban agriculture sector reached its nadir causing the decrease in the average daily per capita caloric and protein consumption by 20% and 27% respectively between 1989 and 1992 [28].

In response to the situation, the government through the Ministry of Agriculture initiated the urban agriculture program in 1991. This program encouraged the community to produce foods in their own gardens for the most of their daily consumption. For farmers who were unable to obtain spaces in their nearby neighbourhoods, the government encouraged them to build a club and provide them the state owned gardens to be cultivated at no cost. The government organised this program by addressing a strong and centralised policy directions, combined with decentralised actions to monitor and provide technical assistance. As a result, more than 26,000 gardens located at the inner urban and the rural-urban fringe of Havana were occupied ranging from high yield organic gardens to household sized site for household self-provisioning where involving more than 22,000 producing units [28, 34].

Through Urban Agriculture, Cuba showed a remarkable resilience after the gloomy episode of the agricultural sector during 1989 to 1992. The urban agriculture program managed to increase the production of foods dramatically. For example, the production of vegetables increased by 78% during 1994 and 2005 and reached an average of 1 kg per capita per day, which is far above the FAO's recommendation of 0.3 kg [25].

Further to the dramatic increase of food production, the urban agriculture was successful to improve the economy at the community level. Besides the profit they gained from selling the produce, the local food production covered approximately 60% of the household food consumption, which enables the farmers to save up to 40% of their income that they usually spend on foods [28]. Moreover, this program managed to generate employment, where there were 350,000 well-paid workers engaged in the urban agriculture in 2006 [25].

Besides the income opportunities offered by the urban agriculture, another factor that supported the willingness of the community to participate in this program was their cultural values. For farmers who cultivated their own gardens, which is mostly an adjacent space to their house, the gardens served as an extended part of their home. The way they design and display their gardens represents the family's identity and their pride of being a part of the urban agriculture. Meanwhile, for farmers who joined a club, their agricultural activities served as a media to improve the social cohesion [34].

By looking at the gap between the experience of the JMR and the great achievement of Havana, there three key concepts that the strategy to implement the local food system in the JMR should consider:

1. The government should resolve the scale problem and build on collaborative planning to develop the local food system strategy.
2. To the evidence that that the greater cause of urban sprawl was influenced by the private developers, the need to manage the urban sprawl should consider to cooperate with the private developers in order to control the urban development.
3. Regarding the loss of the rural-urban fringe attributes (agricultural lands and the community that are engaged in farming activities); there should be a mechanism to encourage the community, especially farmers and landowners, to maintain the agricultural activities. The mechanism should ensure that agricultural based jobs are capable of generating better income opportunities together with the improvement of the social cohesion.

Conclusion

The local food system represents a high level of sustainability because it promotes the efficient use of resources (from being self sufficient in food) and empowers people to take advantage of income generation from a sustainable job (e.g. participating in farming activities) while also improving social cohesion.

Based on the findings, the JMR is considerably unsustainable. The development of the JMR, which triggers the urban sprawl, caused the loss of agricultural lands in the rural-urban fringe at a rate of 2.71% per annum during 1994-2001. Furthermore, the number of the community who were engaged in the agricultural sector declined by 13.52% during 1980-1990 which was followed simultaneously by an increase in employment in the industrial sector.

The declining agricultural lands and number of community working in the agricultural sector contributed to the decline of agricultural productivity. For instance, the paddy fields experienced a sharp decline by 27.53% during 1996 to 1999 making the local resources can only cover 13.18% of the JMR's demand on rice. The findings indicate that the JMR is far from being able to be self sufficient in relation to the provision of food.

To make resilient the agricultural sector within the rural-urban fringe of the JMR needs more than just a "spatial" approach. It needs a strategy that 'motivates' the willingness of farmers and landowners to participate in the agricultural activities. To this extent, the local food system serves as the distinctive strategy that is expected to prevent and improve the productivity of the available agricultural lands and ensure the community to have better income from farming, while simultaneously enable the city region to be self sufficient in relation to the provision of food.

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AIR QUALITY AND HUMAN HEALTH

A case study of chemical weather forecasting in the area of Vienna, Austria

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Abstract

AQA, the Air Quality Model for Austria, uses ALADIN-Austria meteorological forecasts in combination with the chemical transport model CAMx to conduct forecasts of gaseous and particulate air pollutants over Austria. For a short episode in January 2007 with enhanced PM_{10} concentrations caused by a pronounced temperature inversion in Kittsee in NE Austria, model predictions of vertical profiles of wind and temperature are compared to data of a Sodar-RASS operating at this site from 2006 to 2007; predictions of PM_{10} are compared to data of the Kittsee air quality station. The results indicate that the model is able to reproduce this complex situation only after an adjustment to the low-level wind field based on a detailed analysis of the situation.

Introduction

The area around Vienna in Eastern Austria is often affected by high ozone levels in spring and summer as well as high PM_{10} pollution in winter. The regional weather forecast model ALADIN-Austria of the Central Institute for Meteorology and Geodynamics (ZAMG) is used in combination with the chemical transport model CAMx (www.camx.com) to conduct forecasts of gaseous and particulate air pollutants over Austria. The Air Quality model for Austria (AQA) uses ALADINs horizontal resolution of 9.7 km. Since 2008 the higher resolved ALARO with a horizontal resolution of 4.9 km and more vertical layers than ALADIN is also available.

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A SODAR-RASS system of ZAMG was operated from February 2006 to May 2007 at the Austrian site Kittsee near Bratislava, Slovakia. Based on the profile measurements of this system and simultaneous PM_{10} measurements, an episode in January 2007 with a developing inversion which led to elevated PM_{10} concentrations is chosen to investigate if the models can reproduce such complex situations. The meteorological forecasts of the two models ALADIN and ALARO are compared for this selected episode to the measurements. Air quality simulations with input fields obtained from both models for CAMx are conducted and the calculated PM_{10} concentrations are compared to the measurements at Kittsee.

Model components and experimental setup

The air quality model system consists mainly of three parts that are linked together: the meteorological model ALADIN/ALARO, the emission model that converts different emission inventories to the modelling grid and the dispersion model CAMx (Fig.1).

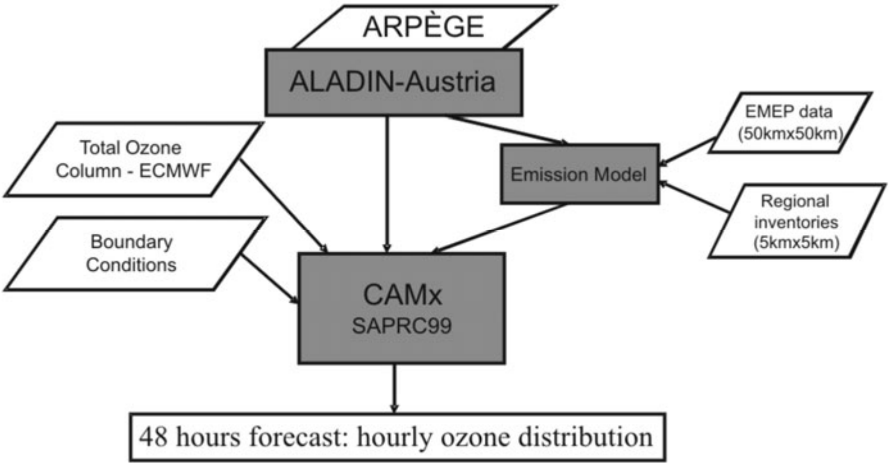

Fig. 1. ALADIN-CAMx – air quality modelling system.

CAMx (v5.00, "Comprehensive Air quality Model with Extensions") simulates the emission, dispersion, chemical reaction, and removal of pollutants in the troposphere by solving the pollutant continuity equation for each chemical species on a system of nested 3D grids. A two-grid nesting is used with a coarse grid over Europe and a finer grid for the core area covering Austria. The meteorological fields are supplied by the limited area models ALADIN-Austria (http://www.cnrm.meteo.fr/aladin/) and ALARO-Austria. The meteorological fields have a temporal resolution of one hour.

The model system generally uses EMEP emissions. For the countries Austria, Czech Republic, Slovakia and Hungary, the original 50 km x 50 km data are downscaled to 5 km x 5 km based on an inventory from 1995 [1]. In addition, a new highly resolved emission inventory for the City of Vienna [2] is used for this area. The emission model SMOKE (Sparse Matrix Kernel Emissions Modeling System, [3]) is used to calculate the biogenic emissions.

The measuring principle of the Sodar/RASS system is remote sensing by electromagnetic and sound waves. It is a combination of an acoustic radar (Sodar) and two electromagnetic grid antennas. The antenna of the Sodar consists of 64 horns and serves as a transmitting and receiving system for sound waves (2 kHz). The frequency shift of the signal due to the Doppler effect is used to determine the 3-D wind. The scattering of radio waves (1290 MHz) to sound waves (2 kHz) is used to determine the temperature, by applying the temperature dependency of the sound propagation velocity. Inversion heights, mixing heights and temperature gradients can be determined from the temperature profiles.

The system was operated in Kittsee in the East of Austria near the Slovakian border from February 2006 to May 2007. The location of the measurement instruments is characterized by very flat agricultural terrain. The dominant anthropogenic emission sources are in the vicinity of Vienna (NW, 40 km) and Bratislava (NE, 5 km). Fig. 2 shows the alignment of the instruments. The air quality station (green container) and a

Fig. 2. Measurement instruments in Kittsee.

ground based ultrasonic anemometer (10 m mast) can be seen in the background. The SODAR-RASS system measures meteorological profiles from 50 m above ground in 25 m intervals up to about 600 m.

Results and discussion

For the evaluation, a short period of an evolving inversion in mid-January 2007 (Fig. 3) detected by the RASS and leading to enhanced PM_{10} concentrations in Kittsee was chosen.

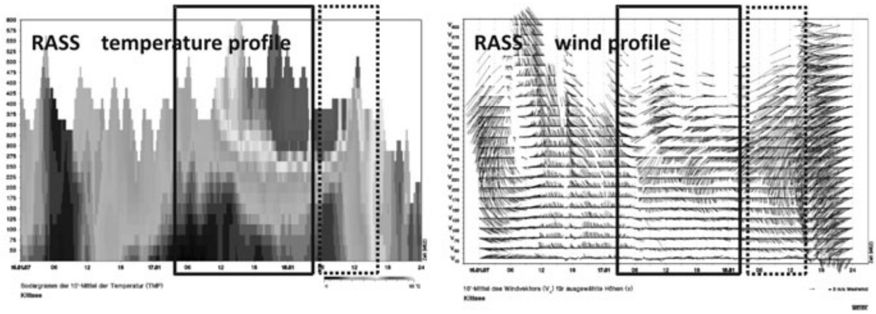

Fig. 3. RASS temperature (left) and wind (right) profiles up to 600 m from 16 to 18 January 2007.

On the 17th the air is characterized by stable conditions. Within the left rectangle in Fig. 3, the inversion strengthens until the temperature increase reaches values of up to 10K/400m at midnight to 18th January; the base of the sharp temperature increase descends from about 400 to about 200 m. The wind turns from south-east to north-east. The PM_{10} concentrations increase with the wind turn and the evolving inversion and reach maximum values of 100 µg/m³ at midnight (Fig. 4). Both models cannot predict these high concentrations during this episode. However, in [4], a good performance of especially the ALARO model in predicting PM_{10} concentrations during the whole month of January 2007 is demonstrated.

In the following, the vertical temperature and wind profiles of the RASS at 13:00 UTC on 17th January 2007 at the onset of the pronounced inversion are compared to the ALADIN and ALARO predictions. The RASS shows the inversion base in around 200 m (Fig. 5). Both models fail to predict the strength of the inversion, but ALARO shows slightly stronger temperature gradients than ALADIN.

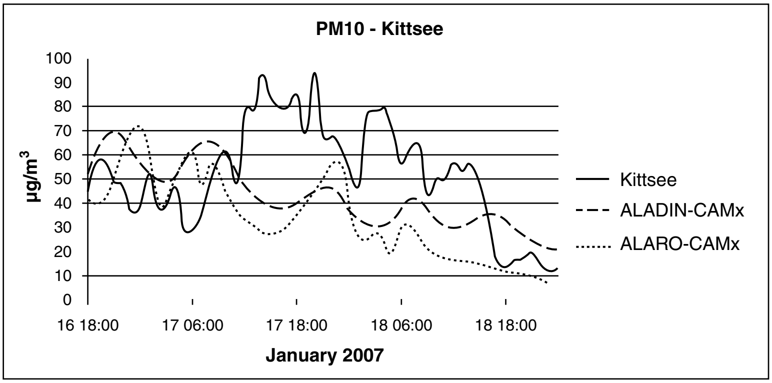

Fig. 4. Time course of PM_{10} concentrations measured at Kittsee and predicted with ALADIN-CAMx and ALARO-CAMx; period is from 16th to 18th January 2007.

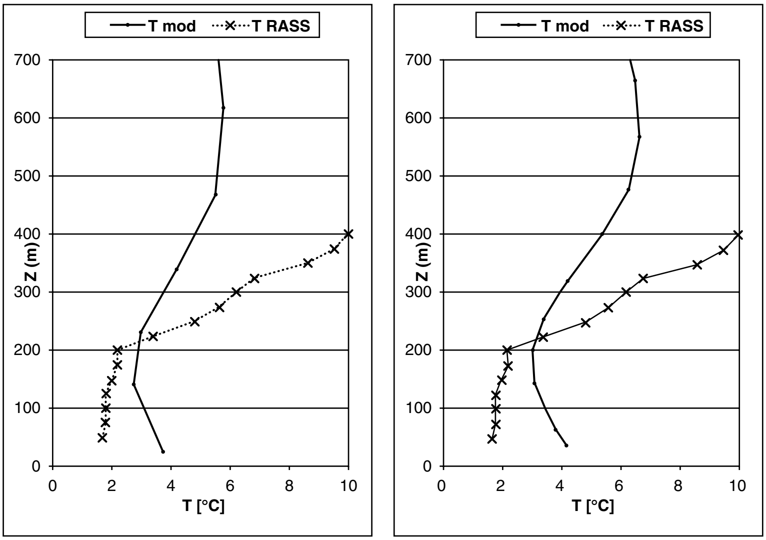

Fig. 5. Temperature profiles of ALADIN (left) and ALARO (right) compared to the RASS temperature profile (symbols) on 17th January 2007, 13:00 UTC.

At the same time, ALARO-CAMx calculates lower PM_{10} concentrations than ALADIN-CAMx. The modelled concentrations of both models lie below the measurements (Fig. 6).

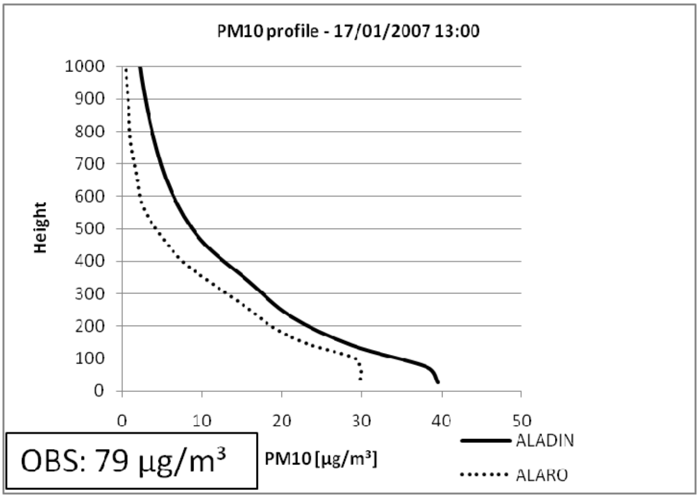

Fig. 6. PM_{10} profiles of ALADIN (solid) and ALARO (dotted) on 17^{th} January 2007, 13:00 UTC.

The reason for the discrepancy between observed and predicted PM_{10} concentrations displayed in Fig. 6 can be attributed to deviations between observed and modeled near-surface winds (Fig. 7). The absolute wind speeds are by a factor of approximately 2 weaker in the models than observed, whereby ALARO performs slightly better than ALADIN. The observed winds from NE transport the high PM_{10} concentrations of anthropogenic origin from Bratislava to the rural site of Kittsee. Both models predict, however, winds from SE. Therefore, the models show areas of elevated PM_{10} concentrations north of Kittsee, also downwind of Bratislava according to the modeled wind directions (Fig. 8). When the modeled concentrations in the main wind direction are compared to the observations in Kittsee, the agreement between observations and predictions is increased considerably (compare Fig. 9 and Fig. 4). Both models show two peaks which occur slightly before and after the period of enhanced PM_{10} observations.

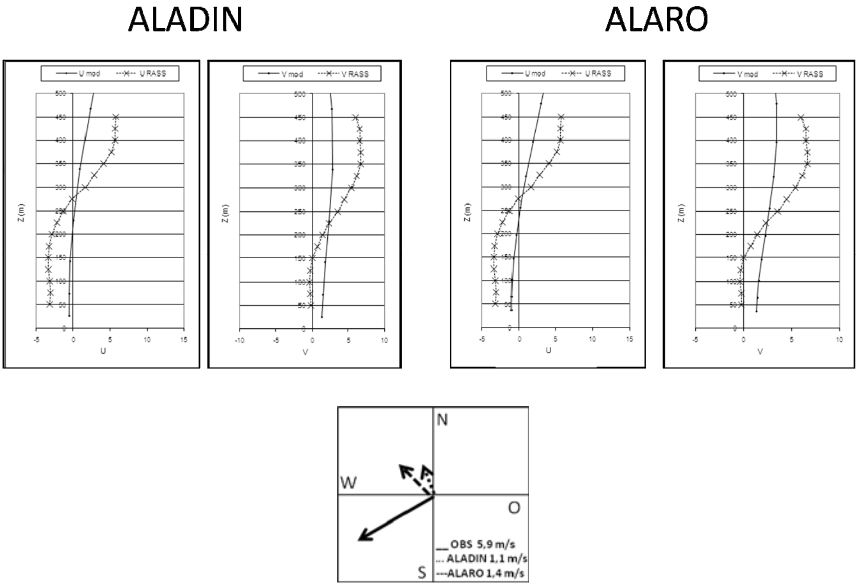

Fig. 7. Vertical wind profiles (u- and v-components) of ALADIN (left) and ALARO (right) compared to the RASS wind profile (symbols) on 17th January 2007, 13:00 UTC; bottom centre: wind vectors at lowermost level.

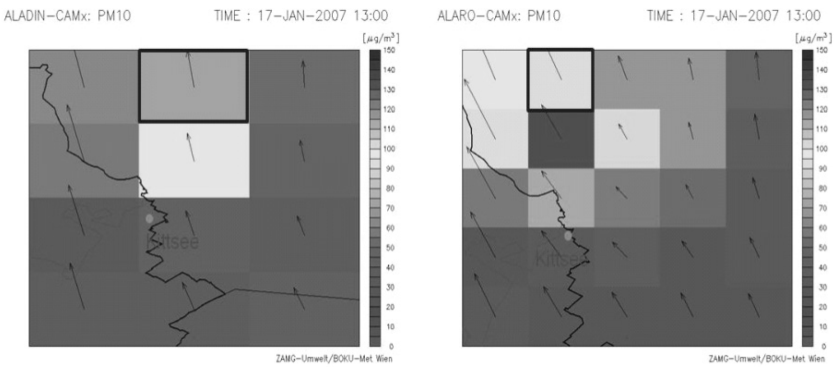

Fig. 8. Spatial distribution of PM_{10} predicted with ALADIN-CAMx (left) and ALARO-CAMx (right) on 17th January 2007, 13:00 UTC.

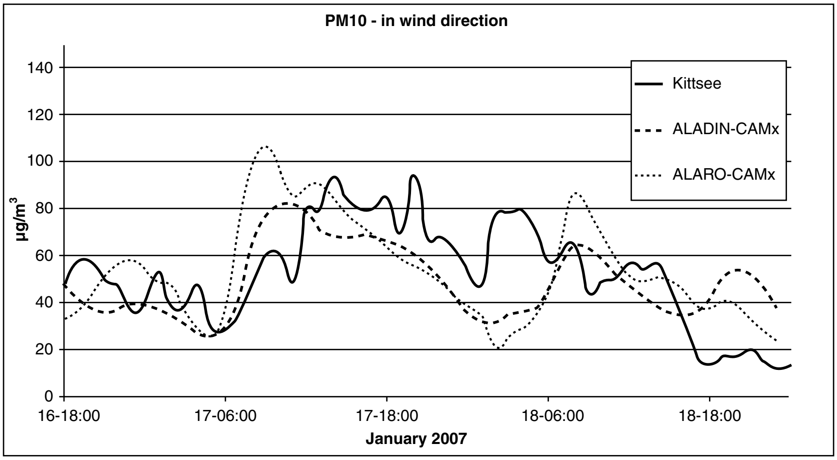

Fig. 9. Time course of PM_{10} concentrations measured at Kittsee (black line) and predicted with ALADIN-CAMx (dashed) and ALARO-CAMx (dotted) in wind direction; period is from 16[th] to 18[th] January 2007.

Conclusions

AQA, the Air Quality Model for Austria, uses ALADIN-Austria meteorological forecasts in combination with the chemical transport model CAMx to conduct forecasts of gaseous and particulate air pollutants over Austria. AQA uses ALADINs horizontal resolution of 9.7 km. Since 2008 the higher resolved ALARO with a horizontal resolution of 4.9 km and more vertical layers than ALADIN is also available and used here for a comparison to the coarser ALADIN forecasts. A short episode in January 2007 with enhanced PM_{10} concentrations caused by a pronounced temperature inversion in Kittsee in NE Austria is chosen to investigate if the models can reproduce such complex situations. During this time a Sodar/RASS operated in Kittsee delivering vertical profiles of temperature and wind up to about 600 m. Model predictions are compared to the measured profiles and to observed PM_{10} concentrations.

The results show that the models are not able to reproduce the enhanced elevated inversion measured by the Sodar/RASS (Fig. 5). The main reason for the under-estimation of observed PM_{10} concentrations (Fig. 4 and Fig. 6) is however deviations in the near-ground wind directions (Fig. 7). Observations in Kittsee show winds from NE which transport high PM_{10} values caused by the agglomeration of Bratislava, which is only 5 km away, to this rural site. The models predict winds from SE and thus show the area of enhanced pollution north of Kittsee

(Fig. 8). When the modeled concentrations in the main wind direction are compared to the observations in Kittsee, the agreement between observations and predictions is increased considerably (compare Fig. 9 and Fig. 4). Both models use EMEP emissions which, in the investigated area, are downscaled to 5 × 5 km. This is most probably the reason why the observed PM_{10} levels are predicted with good agreement, but in this special case not at the correct location.

The reason therefore is, as mentioned, the deviation between observed and predicted near-surface winds. In Eastern Austria, winds from SE are very common in connection with anticyclones or ahead of approaching fronts from the West. The NE wind in Kittsee in these situations is caused by the surrounding topography: in stable conditions, air flows around instead of across the end of the Carpathian mountain chain North of Bratislava, causing a NE wind in Kittsee which turns to SE only further west towards Vienna, then deflected by the Alps. These local complex flow conditions will only be resolved by the models when, in the future, model resolution will further decrease to resolve detailed topographic structures more accurately.

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Micrometeorological effects on urban sound propagation: A numerical and experimental study

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Abstract

The present work is in line with the EM2PAU project whose aim is to quantify the influence of micrometeorological effects on sound propagation in a street "canyon". This study is carried out both numerically and experimentally. Experimental data are issued from a long-term *in situ* experimental campaign. A ½ street scale model is installed in a quiet outdoor environment in order to control noise emission. Different values of street width (W_S) will be considered in this project. The first studies are conducted with the value W_S/H_S = 0.7, H_S being the street height, which corresponds to a typical aspect ratio in most European cities. The turbulent flows and temperature fields in the street are recorded by using 9 sonic anemometers and 48 thermocouples. Moreover, reference meteorological parameters are recorded from two 10-m meteorological towers, located around the site. Simultaneously, acoustic measurements are carried out along the main propagation axis of the street, across the central cross-section and outside the street. A few acoustic results from preliminary measurements are presented and discussed.

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Introduction

Among all urban annoyances, noise is the most mentioned pollution by French people: according to INSEE [1], 54% of french households claim to be disturbed by noise at home. In this context, legislation in terms of noise control lays down, through the European Directive on Assessment and Management of Environmental Noise 2002/49/CE, to set up strategic noisemaps notably. Thus accurate acoustic propagation models are needed for forecasting noise levels in urban areas. Yet acoustic modelling softwares do not take into account a few influential phenomena, as micrometeorological effects that can have a major impact on sound propagation.

Very few works have been published concerning micrometeorological effects on urban acoustic propagation, both from an experimental or a numerical point of view. Except an old study published by Wiener *et al.* [2], these effects are analyzed through numerical modelling [3, 4, 5]. However, micrometeorological fields are quite difficult to predict in urban area and inaccurate micrometeorological input data in propagation models can result in strong discrepancies in term of sound levels. Thus the aim of the EM2PAU project consists in contributing to knowledge and to assessment concerning the interactions between micrometeorological effects and acoustic propagation at a street scale, and then, in integrating these effects in computational codes for sound propagation modelling in urban area. This project is based mainly on experimental campaigns that are carried out both in laboratory and in outdoor site.

This paper focusses on the outdoor site campaign which is performed with a ½ street scale. First, general topics concerning micrometeorology elements and scale models experiments are given. The meteorological and acoustic experimental devices are then described. Lastly, a few preliminary acoustic experimental results are presented and discussed.

Preliminary topics

Atmospheric boundary layer

The micrometeorology stands for microscale meteorology and consists in studying the atmospheric boundary layer (ABL) structure, *i.e.* the first kilometer of the atmosphere. Figure 1 presents the radiative balance variations impact during a typical daily cycle within the ABL. During the day (S1), the friction effect near the surface induces mechanical and thermal turbulence inside the surface layer due to the thermal convection. At the end of the day, the surface cooling results in a transition regime from unstable to stable within the surface layer.

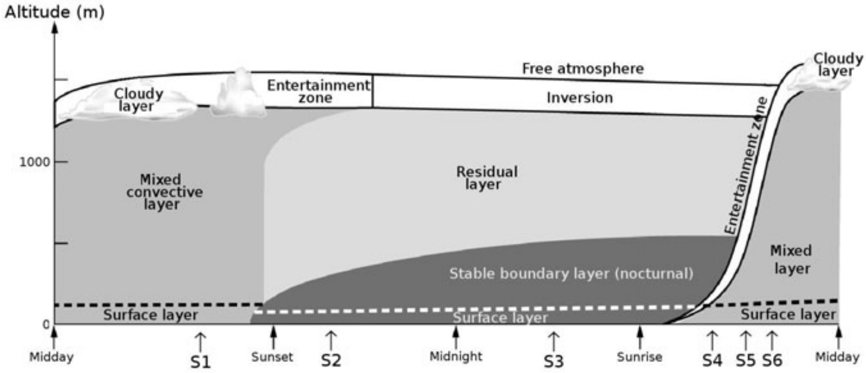

Fig. 1. Daily cycle of the atmospheric boundary layer [6].

Over cities, the atmospheric layer spreading to the mixed layer top is currently named urban boundary layer. Inside the urban canopy, which corresponds to the lower zone of the roughness sublayer included between the soil and the rooftop mean height, the transfers of energy and momentum are very important and multiple interactions between turbulent wakes and canalized flow effects make this layer highly heterogeneous. Within a street, micrometeorological fields are so generally highly turbulent and *in situ* measurements allow a better understanding and knowledge of the phenomena.

Acoustic scale models: fundamental notions

Practical difficulties encountered in setting-up *in situ* measurements lead to consider scale models experimentations. This technique has have been widely used for understanding physical phenomena in various scientific fields, especially in architectural acoustics. Indeed, this technique allows to study acoustic mechanisms inside an *in situ*-like environment. It implies to emit a sound wave of wavelength λ_m in the model for simulating a sound wave propagation of wavelength λ at real scale though keeping the model/real site dimensions scale ratio, that is:

$$\frac{\lambda_m}{\lambda} = \frac{l_m}{l} = S, \quad S < 1, \tag{1}$$

where l_m and l are the scale model and the full-scale model dimensions respectively, and S corresponds to the scale parameter that is defined from the scale factor N such as $S = 1/N$. Consequently, as the same gas constitutes the propagation medium both inside the scale model and in the full-scale one (*i.e.* the air), the sound celerity in the scale model c_m is equal to the sound velocity in the full-scale environment c. The sound wave frequencies at the model and at the real scales must then verify:

$$f_m = \frac{1}{S} \times f. \tag{2}$$

The time component is also affected by the scale modification and is governed by the following similitude law:

$$t_m = S \times t, \tag{3}$$

with t_m and t the time components in the scale model and in the full-scale one respectively.

From this theoretical background, it is clear that the experimental set-up and process must be chosen adequately in order to give rise to micrometeorological effects on the full-scale acoustic observables. On the one hand, these micrometeorological effects do not depend on frequency, but intensify the ground effects that are frequency-dependent. On the other hand, these effects are time-fluctuant. In the present study, for which the scale model is placed in outdoor environment, it is important to keep in mind that acoustic time fluctuations vary according to Eq. (3), whereas micrometeorological conditions deviation are full-scale ones.

Experimental campaign

The acoustic field behavior in a street can be fully described by measuring the impulse responses (IR) at many locations in the street [7]. Indeed this measurement enables the post-computation of the sound level (SL) and the reverberation time (RT) giving informations on the sound attenuation and the sound decay in the street respectively. Difficulties in setting up long-term experimental campaigns in a real city street lead to consider street scale models experiments in laboratory for studying urban sound propagation [8, 9]. It can be also preferable to carry out this kind of study in a controlled and quiet environment to avoid any unwanted intruding effect like background noise or else time-fluctuations of the propagation conditions (humidity, temperature, air flow). However, it is difficult to recreate realistic meteorological effects in laboratory. The originality of the present work is that the scale model is set up outdoor, *i.e.* in an environment where micrometeorological conditions are not controlled.

Street scale

The scale model used in this project is made up of eight marine containers set in order to re-create a ½ scale street with the following dimensions (*see* Fig. 2):

- street length: $L_s = 24.45$ m;
- block width (*i.e.* container width): $W_b = W_c = 2.44$ m;
- street height: $H_s = 5.18$ m;
- street width: $W_s = 3.63$ m.

Thus the street scale ratio of building height to street width is $H_s/W_s = 0.7$ which corresponds to a typical aspect ratio in most European cities. In addition, the street axis forms an angle of 45° with north, corresponding to the prevailing wind direction.

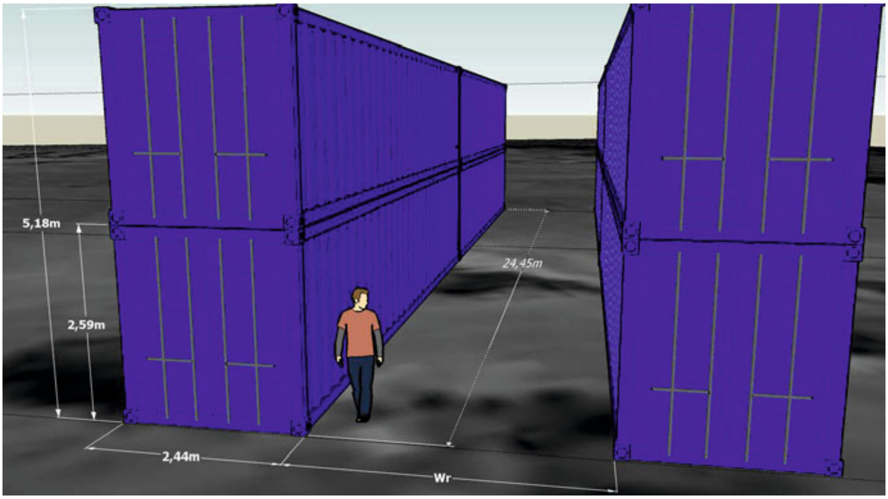

Fig. 2. 2 ½ street scale dimensions.

Experimental system

The experimental site is set up with a lot of micrometeorological and acoustic sensors (*see* Fig. 3). The acoustic device is set up on the experimental site punctually in order to avoid any sensors spoiling, whereas meteorological sensors ensure data acquisition permanently.

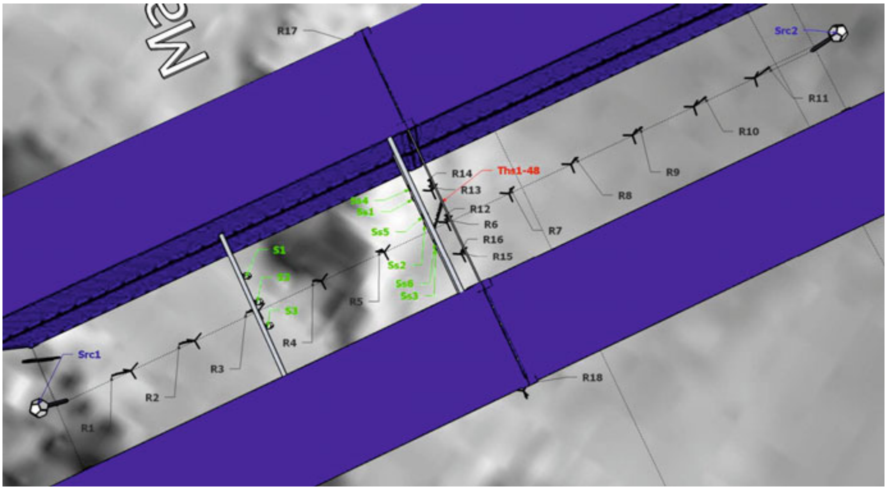

Fig. 3. Overall view of the ½ street scale acoustic and micrometeorological instrumentation.

Acoustic device

A high power omnidirectional sound source emits a random noise at one street extremity (Src1 or Src2 at Fig. 4(a)) and at 1.3 m high above ground. Fifteen ½-inch free-field microphones are spread inside the street as depicted at Fig. 4(a): eleven microphones (R1-R11) are located every two meters from the source along the street axis and at 1.2 m high, and an array of six microphones is positioned at the central street cross-section at 1.2 m (R6, R13 and R15) and at 3.5 m (R12, R14 and R16) high above ground. Two microphones are also located outside and on both sides of the street to estimate the acoustic energy propagating above the roof tops. Data acquisition is ensured by a B&K Pulse™ system.

Micrometeorological data acquisition

The experimental site is also instrumented with micrometeorological sensors recording wind velocity, sound speed, temperature and relative humidity at many locations permanently.

Two 10-m meteorological masts located around the site record the wind and temperature fields profiles non-disrupted by the containers. One of them, located at northeast of the site (Mast 1 at Fig. 4(b)), is equipped with two tridimensional ultrasonic anemometers Young 81000 at 3 m high, with a ventilated temperature and relative humidity probe, and at 10 m high, which are connected up with an acquisition computer. The second mast (Mast 2 at Fig. 4(b)) is fitted up with an ultrasonic anemometer Metek USA-1 overhead and is placed southwest of the street. Another temperature and relative humidity probe is positioned in front of one of the containers at the northeast street extremity.

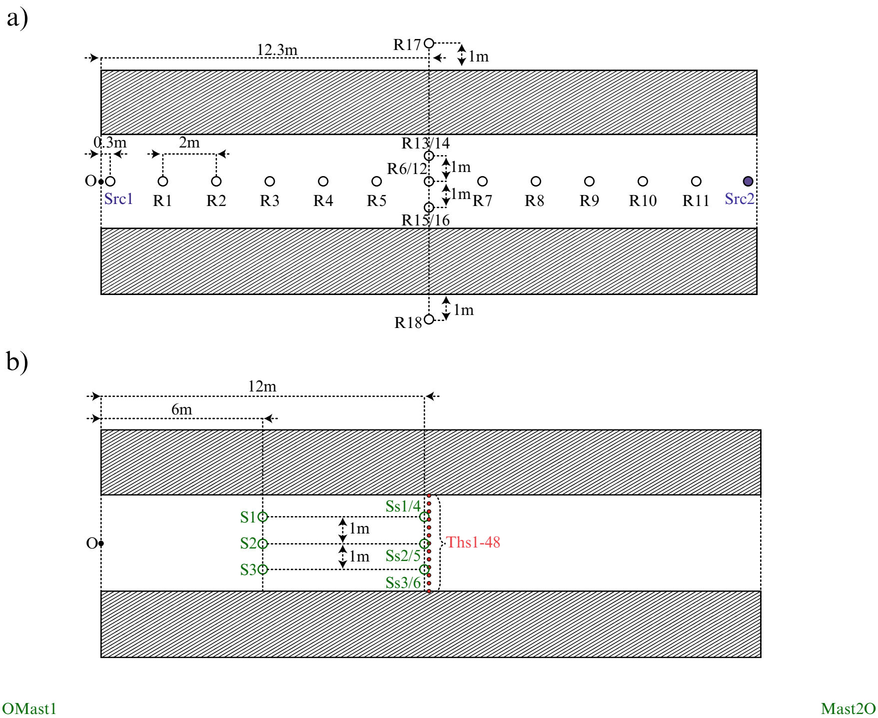

Fig. 4. Locations of (a) the source and the microphones and of (b) the meteorological sensors on the experimental site.

Three tridimensional ultrasonic anemometers Young 81000 are fixed on a transversal rod, at 3 m high, on a street section located at 6 m from the northeast street extremity (S1-3 at Fig. 4(b)). Six ultrasonic anemometers Gill Windmaster are hanged up on two others transversal rods at the middle of the street, one at 2 m (Ss1-3 at Fig. 4(b)) and the second one at 4 m (Ss4-6 at Fig. 4(b)) high above ground. Thermocouples rakes (Ths1-48 at Fig. 4(b)) are also fixed at the center street section: seven on each frontage, three on the ground, twelve on the central vertical axis and nineteen horizontally at 4 m high.

Preliminary results and discussion

This experimental campaign has just begun. At this stage, only preliminary acoustic results are then treated and presented. Micrometeorological results will be subjected to intended publications.

Acoustic data processing

As meteorological conditions should have an influence on the sound attenuation and on the reverberation time in the street, the Schroeder's integrating method [10] is used to compute the steady-state SL and the sound decay simultaneously from the IR for each 1/3 octave band. The excess of atmospheric attenuation in the scale model compared with the full-scale street is not compensated.

Sound distribution in the street

Figures 5 and 6 present the SL measured consecutively along the street axis and across the central cross-section respectively. The sound source is placed at Src1 position (*see* Fig. 4(a)). Each measurement consists in computing the IR on a 320 ms duration and in applying 20 linear spectrum averages, what results finally in a 6.4 s acquisition duration. This averaging enables to free from background noise fluctuations and to smooth the turbulent flow effect. However, only wind varies between each measurement and, consequently, the SL discrepancies are only due to the meteorological conditions fluctuations. The sound attenuation deviations along the street (Fig. 5) appear downstream of the street centre mainly, *i.e.* behind the widely instrumented section that constitutes cluttering from a meteorological point of view, and reach until around 5 dB at the street end. This effect is also observed on the SL for the street cross-section (Fig. 6). Only a detailed study can enable to determine if these discrepancies are the result of micrometeorological fluctuations effects on sound propagation, or of the turbulence induced by the sensors.

Conclusion

The EM2PAU project should provide valuable informations on the interactions between micrometeorological conditions and sound propagation at a street scale. This project is widely based on an experimental campaign in a ½ street scale model set up in outdoor environment. Permanent micrometeorological sensors allow long-term observations with short-time data acquisitions. At the same time, punctual acoustic measurements are performed to give rise to the wind and temperature conditions influence on the sound distribution along the street and on the central cross-section. The preliminary measurements show that, either the flow fluctuations, or the turbulent flow induced by the sensors in the street, have an impact on the sound distribution. Future works will consist in analyzing the meteorological data rigorously in order to give rise to the impact of the flow on the sound propagation in term of sound levels. This experimental campaign will be useful for integrating micrometeorological effects into acoustic propagation models based on energetic approaches (Sound Particle Propagation Simulation (SPPS) method, diffusion model) and undulatory models (modal approach, Parabolic Equation (PE) method, Transmission Line Matrix (TLM) method).

a)

b)

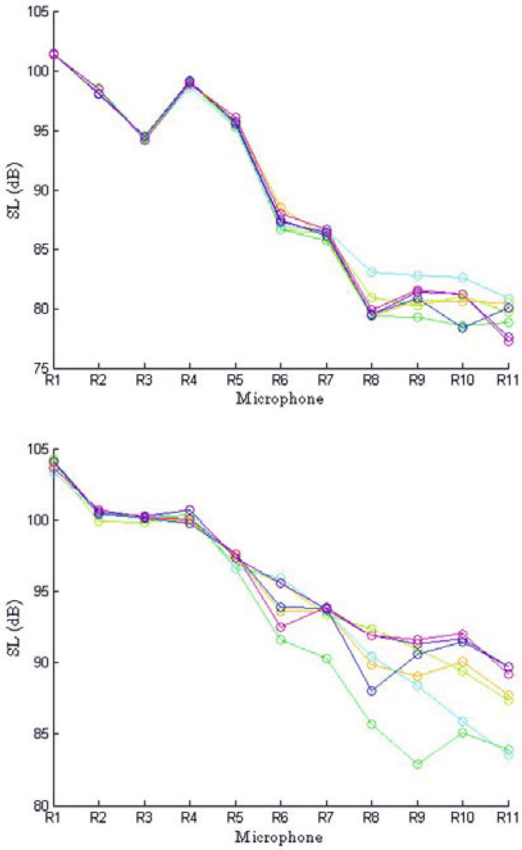

Fig. 5. Third-octave band SL along the street axis with centre frequencies (a) 500Hz and (b) 1000Hz for consecutive measurements.

a)

b)

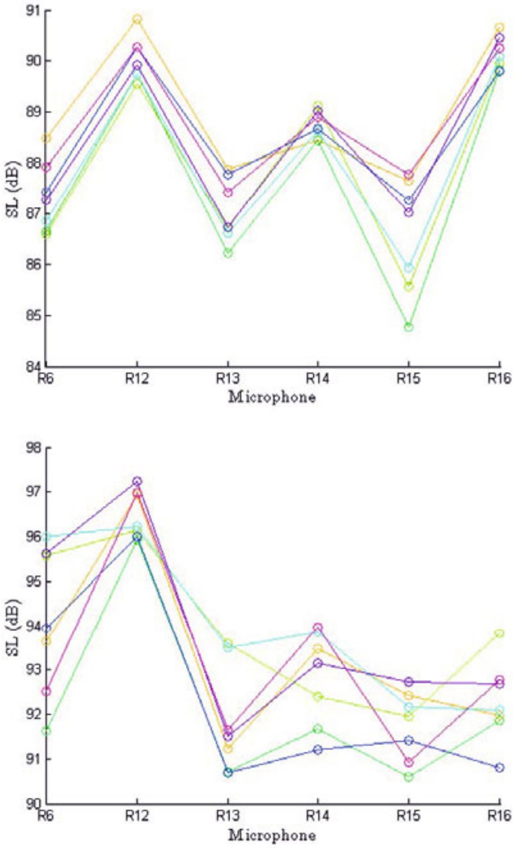

Fig. 6. Third-octave band SL across the central street cross-section with centre frequencies (a) 500Hz and (b) 1000Hz for consecutive measurements.

Acknowledgements

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Performance evaluation of air quality dispersion models in Delhi, India

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Abstract

Several attempts have been made worldwide to understand the gravity of the air pollution problem. Researchers and policy-makers have been trying to understand it through scientific, regulatory and non-regulatory options. This study reports on the applicability and performance of some well-known air quality dispersion models like AERMOD and ISCST3 models for Indian conditions. The evaluation of these models has been carried out in Delhi, India using the historical air quality, meteorological and traffic data for the year 2007. The models have been evaluated using standard performance indicators like fractional bias, mean bias, normalized root mean square error, index of agreement and geometric mean bias.

Introduction

It is well documented that exhaust derived pollution can have adverse effects on human health [1] and [2]. Such impacts on human health are becoming ever more frequent in developing countries, particularly in China and India, where rapid economic growth, urbanisation and improved road infrastructure have led to the severe contamination of air from road vehicles [3]. India's capital city, Delhi is a prime example of this and is in fact ranked amongst the most highly polluted cities in the world [2]. Over the years Delhi has seen a gradual shift in passenger and freight movement from rail to road based transport, which has led to a marked increase in fuel consumption by the road sector [4]. Source apportionment revealed that India's transport sector now accounts for up to 90%, 74%, 12% and 22% of CO, NO_x, SO_2 and PM emissions respectively of which road transport is the dominant share [5].

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It is therefore necessary for local, regional and national authorities in India to accurately assess the level of air contamination from the road network to allow them to develop new policies and strategies. The ability to identify those areas within a city which have poor air quality is paramount if such policies are to be successful. In reality monitoring everywhere in a city is neither physically or financially feasible. Instead policy makers must rely on air quality models to predict the spatial distribution of pollutants over a given area. Over the years there have been many different types of dispersion models developed, but despite such development their application in India has been limited. It was therefore the aim of this investigation to assess the ability of AERMOD and ISCST3 to predict the pollutant dispersion from a heavily trafficked intersection in Delhi, India. The models have been described in detail elsewhere [6].

Study Methodology

AERMOD and ISCST 3 were used to predict concentrations of $PM_{2.5}$, CO and NO_2 for a winter period (23rd to the 29th January) in 2007. Model performance was evaluated through comparison against monitored data and via the use of statistical descriptors. The Bahadur Shah Zafar (BSZ) Marg commonly known as Income Tax Office (ITO) intersection in Delhi was chosen as the study area for this investigation (Fig. 1). The intersection comprises of 4 links. A 2 × 2km grid comprising the intersection was defined as the simulation domain. Within the domain a pollution monitoring site governed by CPCB is situated 12m from BSZ Marg outside the premises of Indian National Science Academy. Data from this station was used to evaluate model performance. The four roads of the intersection are numbered as Road 1 to 4, for convenience.

Traffic Characteristics

ITO is one of the busiest intersections in Delhi. Fig. 2 shows the diurnal variation in traffic at the ITO intersection. Roads 3 and 4 carry more traffic than Roads 1 and 2. 87% of traffic at the ITO intersection occurs between 7 am and 10 pm within which there are two daily peaks. The morning peak occurs between 9 am and 11 am and the afternoon peak between 5 pm and 8 pm. The average fleet composition for Delhi was use to describe the traffic at the site [7]. The fleet composition at ITO is dominated by two wheelers (64%). The second largest share is held by cars and jeeps (30%) and goods vehicles, three wheelers and buses account for a small fraction of the fleet (3%, 2% and 1%, respectively).

Fig. 1. Schematic of the ITO intersection, Delhi; 1 = Road 1. 2 = Road 2, 3 = Road 3, 4 = Road 4 and R = monitoring site.

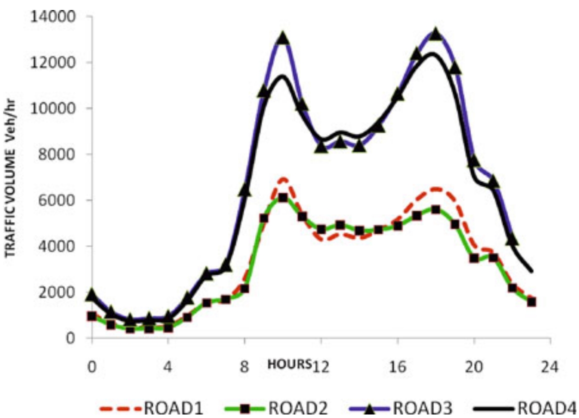

Fig. 2. Diurnal flow pattern of traffic at the ITO intersection.

Emissions

The methodology followed by [8] was adopted to calculate link emissions: an emissions rate (g/s) was assigned to each road based on volume of traffic (veh/h), vehicular emissions factors (g/km) and road length (km). The 2007 emissions factors established by the Automotive Research Association of India [9] were used and the proportion of each vehicle type was derived from the average fleet composition in Delhi.

Meteorological Dataset

The compulsory meteorological data required for AERMOD and ISCST3 is document by [6] and will not be described here. Sequential hourly meteorological surface data for the winter period (23rd to 29th of January, 2007) were obtained from the IMD Delhi weather station at an hourly resolution, which is broadly representative of the weather conditions within the simulation domain. The surface meteorology for the week is characterized by mixed patterns of wind (Fig. 3). The week had 43% of calm (wind speed <1m/s) conditions. Data characterising upper air conditions were acquired from Wyooming University website [10].

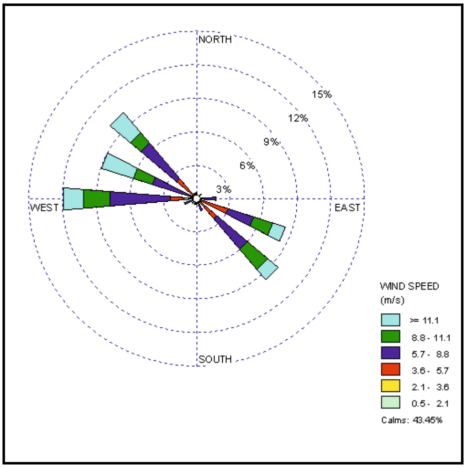

Fig. 3. Wind Rose diagram for January (23rd to 29th)

Surface (and upper air meteorology in AERMOD) data were input into the models. In addition the base elevation of the meteorological monitoring site above mean sea level (MSL) was specified as 216m and the base elevation of ITO above MSL input as 208m. Albedo, Bowen ratio and roughness length were specified as 0.18, 2 and 1m respectively. An AutoCad map comprising topographic information was input into AERMAP in order to pre-process terrain data for AERMOD. For ISCST3 the terrain type was specified as urban.

Statistical Analysis

Air Quality Monitoring

CO, NO_2 and $PM_{2.5}$ data from the monitoring site for the week period in January are presented in Fig. 4. The week's average pollutant levels were

2392 μg/m^3, 164 μg/m^3 and 256 μg/m^3 for CO, NO_2 and $PM_{2.5}$ respectively. High pollutant concentrations experienced on the 23rd of January are more than likely due to the persistence of calm conditions (wind speed < 1m/s). Similarly high pollutant values were observed towards the end of the week (between the 26th and 29th of January) and this can be attributed to the wind direction (100^0 to 150^0) blowing directly towards the monitoring site. During high wind speed conditions low pollutant levels were observed.

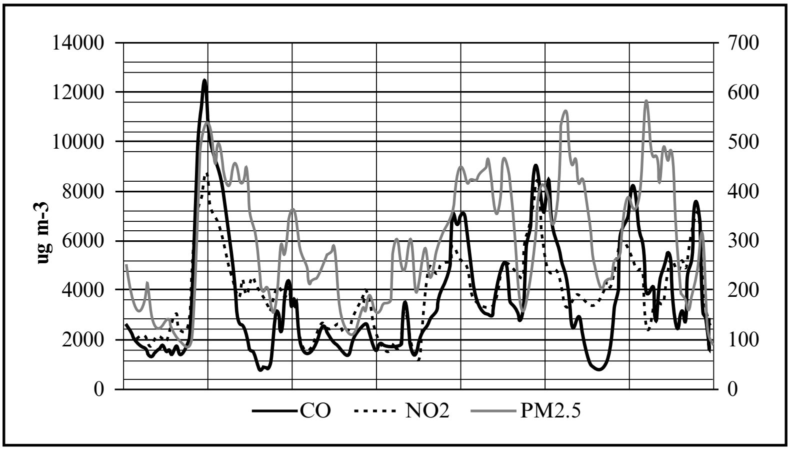

Fig. 4. Variation in CO, NO_2 and $PM_{2.5}$ concentrations at the ITO intersection for the 23rd to the 29th of January 2007.

Background Concentrations

The methodology adopted by [11] was used to estimate the background concentration for CO and was determined to be 1400ug/m^3. The background concentration used for NO_2 and $PM_{2.5}$ were derived from the monitored data and were 61 μg/m^3 and 83 μg/m^3 respectively.

Statistical Analysis Method

Evaluation of model performance was carried out through comparison of model predictions with monitored data. The data containing calm conditions was removed prior to statistical analysis in accordance with the [17]. The following statistical descriptors were used to analyse model performance; index of agreement (IA), fractional bias (FB), normalised root mean square error (NRMSE), geometric mean bias (MG) and the geometric mean variance (VG). They are defined as shown below:

$$IA = 1 - \frac{\sum_{i=1}^{n}(P_i - O_i)^2}{\sum_{i=1}^{n}(|P_i - \bar{O}| + |O_i - \bar{O}|)^2} \tag{3.1}$$

$$\mathrm{FB} = 2\frac{(\bar{\mathrm{P}} - \bar{\mathrm{O}})}{\bar{\mathrm{P}} + \bar{\mathrm{O}}} \tag{3.2}$$

$$NRMSE = \frac{RMSE}{O_{max} - O_{MIN}} \tag{3.3a}$$

$$RMSE = \frac{\sqrt{\Sigma_{\mathrm{i}=1}^{n}(\mathrm{P_i} - \mathrm{O_i})^2}}{n} \tag{3.3b}$$

$$MG = exp(lnO_i - lnP_i) \tag{3.4}$$

$$VG = exp(lnO_i - lnP_i)^2 \tag{3.5}$$

In above equations, P_i is the predicted value, O_i is the observed value, $\bar{O}$ is the mean of the observed data and $\bar{P}$ is the mean of the predicted data. O_{max} is the maximum value in the observed data set and O_{min} is the minimum value. IA and NRMSE indicate the degree of correlation between observed and predicted time series data, while FB is a measure of agreement with the mean concentration [8]. NRMSE is an estimator of the overall deviation between the observed and predicted values. MG test the predicted mean on a log normal basis, and gives the bias of predicted mean from observed mean so that the extreme values or the outliers of the sample do not affect the comparison. An MG value <1 indicates model overprediction, whereas MG 1> indicates underprediction. Similarly, VG checks for the variance (or scatter) of predicted mean from observed mean on a log normal. If model performance is perfect (i.e. predicted match observed values) then FB and NRMSE should be zero and IA, MG and VG should be one. In reality this is never the case, however according to [12] the performance of a model can be deemed acceptable if: NRMSE ≤ 0.5; $-0.5 \leq$ FB ≤ 0.5; $0.75 \leq$ MG ≤ 1.25; and $1 \leq$ VG ≤ 1.25.

Results

Carbon Monoxide (CO)

The statistical descriptors of the CO predictions are shown in Table 1. ISCST3 shows reasonable agreement between predicted and observed values and performed better than AERMOD. ISCST3 predicts the maximum concentrations well and meets the criteria for model acceptability [12]. AERMOD however does not. ISCST3 had a tendency to underpredict by 20% and AERMOD underpredicted by 43%. Model underpredictions are further highlighted by the average values which are both significantly lower than those of the observed. Both models have NRMSE values that indicate reasonable correlation between the observed and predicted data.

Table 1. Statistical descriptors for observed and predicted CO.

CO	IA	MG	VG	FB	NRMSE	Average ($\mu g/m^3$)	Maximum ($\mu g/m^3$)
AERMOD	0.25	1.43	1.14	–0.36	0.02	1670.59	3939.86
ISCST3	0.66	1.20	1.04	–0.19	0.02	1985.70	7585.92
Observed	1.00	1.00	1.00	0.00	0.00	2392.47	7580.00

Nitrogen Dioxide (NO_2)

The statistical descriptors of NO_2 are given in Table 2. The results for ISCST3 show relatively poor agreement between predicted and observed values. In comparison AERMOD outputs show good agreement, having a mean bias (in favour of underprediction) of just 6%. In contrast ISCST3 had a tendency to overpredict by around 30%. This is further highlighted by the positive FB score. AERMOD overpredicts the maximum concentration by a factor of 3 and ISCST3 overpredicts by a factor of 4. ISCST3 has high scatter and fails to meet criteria for model acceptability [12]. AERMOD has minimal scatter and meets the criteria defined in [12].

Table 2. Statistical descriptors for observed and predicted NO_2.

NO_2	IA	MG	VG	FB	NRMSE	Average ($\mu g/m^3$)	Maximum ($\mu g/m^3$)
AERMOD	0.14	1.06	1.00	–0.06	0.06	154.35	1068.88
ISCST3	0.32	0.73	1.10	0.31	0.08	224.67	1557.61
Observed	1.00	1.00	1.00	0 00	0.00	163.99	340.00

Particulate Matter ($PM_{2.5}$)

The statistical descriptors of $PM_{2.5}$ are shown in Table 3. Both of the models show poor agreement between predicted and observed values, as is illustrated by the IA scores. ISCST3 and AERMOD had a tendency to underpredict significantly as can be seen from the high geometric mean values (MG). In addition AERMOD underpredicts the maximum $PM_{2.5}$ concentration value by a factor of 4 and ISCST3 by a factor of 2.

Table 3. Statistical descriptors for observed and predicted $PM_{2.5}$

$PM_{2.5}$	IA	MG	VG	FB	NRMSE	Average ($\mu g/m^3$)	Maximum ($\mu g/m^3$)
AERMOD	0.02	2.90	3.10	–0.97	0.05	88.44	121.52
ISCST3	0.06	2.74	2.76	–0.93	0.05	93.54	184.03
Observed	1.00	1.00	1.00	0.00	0.00	256.16	484.00

Discussion

NO_2 predictions by AERMOD were deemed adequate according to the criteria of [12], ISCST3 however showed poor performance. AERMOD's satisfactory performance can be attributed to the chemistry scheme which is implicit in the model algorithms. ISCST3 has no such scheme. In contrast ISCST3 predicted CO concentrations well but AERMOD significantly underpredicted. Both models failed to predict $PM_{2.5}$ concentration values accurately. The poor performance observed by AERMOD (for CO and $PM_{2.5}$) and ISCST3 (for NO_2 and $PM_{2.5}$) could be attributed to the uncertainty of emissions from non-road source releases, model sensitivity to wind conditions and link emissions and meteorological input data. These factors are discussed below.

The emissions factors used to calculate total link emissions were constructed by the Automotive Research Association of India [9] and were developed from laboratory dynamometer tests under Indian drive cycle urban conditions. These emissions factors do not take into account real world vehicle speed, evaporative emissions, cold start emissions (possibly not an issue in India due to high ambient temperatures) the influence of brake and tyre ware, vehicle maintenance and the use of auxiliaries (air conditioning, radio etc.). In addition it is highly likely that the Indian driving cycles used to construct the factors were not representative of the heavy congestion that is evident at the ITO junction. The accuracy of model predictions is reliant on input data that is representative of the domain. Model outputs indicate that the best available emissions factors used here are not representative and may have significantly influenced model performance. On a similar note anomalous meteorological data could also have contributed to poor performance. Significant over and underprediticons indicate that the meteorological data used was not representative of the domain. The upper air data used, although the best available, may not have reflected the conditions within the domain.

The uncertainty of emissions sources other than those from vehicle exhausts appears to have been a significant cause of error in predicting pollutant concentrations. The model underpredictions observed indicate the influence of non-road source releases. The ITO intersection is situated close to the heart of the city and a heavy layer of road dust is evident at the site. Resuspension of particulate matter is therefore inevitable and unavoidable. The influence of resuspened particulate matter (RSPM) was not accounted for in the investigation due to lack of data. In addition the background concentration used may not have accounted for RSPM, as the single value was derived from night time conditions when traffic and ultimately traffic induced resuspenison is low.

It is well documented in the literature that Gaussian models perform poorly under low wind speed or calm conditions [13] [8] [14] [15] and [16]. Although all the calm conditions were removed from the datasets (in accordance with [17] prior to statistical analysis there remained some wind speeds that were close to 1m/s. Such wind conditions could have been responsible for significant model underpredictions but most likely not to the extent observed. It is also documented in the literature that Gaussian models perform poorly when air-flow (wind direction) is away from the receptor [8] [14] and [15]. For the latter part of the week the wind direction was indeed away from the receptor.

Conclusions

ISCST3 and AERMOD were used to predict the spatial distribution of exhaust pollutants emitted from on-road sources at a heavily trafficked intersection in Delhi, India. Model performance was in generally adequate for NO_2 and CO, but not for $PM_{2.5}$. It is advised that air quality modellers in India acquire data that is representative of the study domain before informing policy makers of their findings.

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Characterization of atmospheric deposition in a small suburban catchment

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Abstract

The objective here is to characterize atmospheric deposition in a small urban catchment. Our results show that micro pollutant concentrations are very low. Polycyclic aromatic hydrocarbons are only detected during the winter, while pesticides are detected in the summer. Due to the low traffic density and to the absence of industrial activities on the Pin Sec catchment, anthropogenic particles are rarely identified during scanning electron microscopy observations. On the contrary, particles of biological and detritic origins are ubiquitous. This characterization will be completed by a new approach aiming at a better knowledge of dry deposition.

Introduction

More than 50% of the world population is now living in urban areas; this percentage reaches 70% in developed countries.

Atmospheric pollution is of major concern all over the world especially because it has multiple effects on human health and the environment [1-4]. Atmospheric particles also interact directly or indirectly with earth energy balance and can, therefore, affect the global climate.

Atmospheric pollutants are either of natural or of anthropogenic origins. Among anthropogenic sources are road traffic, rail and air traffics, heating, industries, working sites, agriculture, incineration plants. Natural sources are mainly volcanoes, forest fires, soil erosion (especially in dry regions), oceans (marine aerosols). Once in the atmosphere, pollutants can be transported over long distances and are deposited on surfaces through wet or dry atmospheric depositions. Dispersion of the pollutants

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depends both on the characteristics of the wind and of the chemical properties of the particles [5].

Many studies concern micro pollutant concentrations and fluxes in total atmospheric deposition [6-8]. Heavy metals (such as Cd, Cr, Cu, Ni, Pb and Zn), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dioxins, furans and pesticides are the most commonly measured in environmental studies.

Our objective is to characterize air quality and atmospheric deposition; heavy metals (Cd, Cr, Cu, Ni, Pb and Zn), PAHs and pesticides were measured. This chemical characterization is completed by scanning electron microscopy (SEM) observations. These first results will be completed by a new approach aiming at a better knowledge of dry deposition; the original new experiment is presented.

Experimental – Method

The study site

This study was conducted in the Pin Sec catchment located in the East of Nantes city (France), between the Loire and Erdre rivers. The Pin Sec catchment has a surface of 31 ha with 2505 residents. The habitat is mostly constituted by personal households or collective buildings. The impervious surface represents 49% mainly roofs, streets, pavements and parking lots. Roof surface represents 18% of the total surface of the catchment. The mean slope of the catchment is about 1.1%.

Sampling and equipment

Samples were collected from May to December 2008. During this period the total rainfall was 532 mm and the total rainfall collected was 358 mm (67% of total rainfall). The collected rainfall represents 40 % of the total 2008 rainfall (889 mm). The total rain collected was 85 mm in May - June, 24 mm in June – July, 91 mm in August – September, 108 mm in October – November and 50 mm in December. The duration of each period was 28, 30, 43, 33 and 20 days respectively.

Bulk atmospheric deposition (dry and wet depositions) was collected by “Air Pays de la Loire”. A funnel connected to a 10-liter bottle was used for the sampling. Glass material was used to collect samples aimed at organic pollutant analyses. Polyethylene material was used for samples aimed at heavy metal analyses. The funnel areas were 0.034 m^2 and 0.066 m^2, respectively. The samples were collected weekly. One monthly sample was constituted and analysed. Rainfall was measured continuously by three bucket rain gauges installed in this zone. Data validation is made by comparison between the 3 rain gauges.

Atmospheric dusts were collected with Filter Dynamics Measurement System, Model 8500 with a filter of 13 mm effective diameter. The flow rate of air filtered was 2 L min^{-1}. The characterization of the morphology and composition of atmospheric particles was made using scanning electron microscopy (SEM) HITACHI S 570 with an energy dispersive X-ray spectroscopy (EDX BRUKER AXS). All the samples were dried at room temperature and coated with gold.

Chemical analysis

Fifteen out of the 16 PAHs recommended by the Environmental Protection Agency (EPA) were analyzed by IDAC laboratory according to [9]. The quantification limits for PAHs vary from 2.0 to 10 ng L^{-1}. Heavy metals (Zn, Ni, Cd, Cr, Cu, Pb) were analyzed too. The analyses were made at the LCPC environmental and chemical laboratory by atomic absorption spectrometry according to [10]. The quantification limits were 0.10 μg L^{-1} for Cd, 1.0 μg L^{-1} for Pb, 2.0 μg L^{-1} for Cu, 0.5 μg L^{-1} for Cr, 8.0 μg L^{-1} for Zn and 1.0 μg L^{-1} for Ni. Pesticide analyses were performed by the IANESCO-CHIMIE Laboratory in Poitiers. Glyphosate and AMPA were evaluated using HPLC with a fluorimetric detection. Prior to analysis, the homogenised sample was derived with 9-fluorenyl methyl chloroformate (FMO-Cl) at pH 9. For diuron, 250 mL of the sample were extracted (liquid/solid extraction). The extract was then analysed using HPLC coupled with a double mass spectrometer (LC/MS/MS). Quantification limits were 0.05 μg·L^{-1} for glyphosate and AMPA, and 0.1 μg·L^{-1} for diuron.

Results and discussion

Bulk parameters and metal concentrations

In this study, the pollutant concentration is presented in normalised concentration (NC). Normal concentration was calculated according to equation 1 [8]. Where MMC is the mean monthly concentration, H_c is the total rainfall collected for each period and H_m is the total rainfall calculated from mean between tree bucket rain gauges.

$$NC = MMC \times \frac{H_c}{H_m} \tag{1}$$

During the survey period pH values were low ranging from 5.3 to 6.8. The highest values were obtained between July and September 2007, which can be explained by the lower NO_X levels measured during this period (NO_X concentrations are twice as high in 2008 than for the same period in 2007). It is well known that the highest the NO_X and SO_2 concentrations, the lowest the rain pH.

Conductivity varies strongly between the years with values around 100 µs cm^{-1} in 2007 and 20 µs cm^{-1} in 2008. This can be due to higher ion concentrations (NO_3^-, SO_4^{2-}, Cl^-) measured in the samples collected in 2007. These ions originate from anthropogenic sources (NO_3^-, SO_4^{2-}) as well as from marine inputs (Cl^-, SO_4^{2-}).

Regarding suspended solids (SS) and total organic carbon (TOC) differences are observed according to the period of the year. Suspended solid concentrations range from 2 to 35 mg L^{-1}, whereas TOC concentrations vary from 0.5 to 12.3 mg L^{-1}. For both parameters, the highest values were measured in June and July 2008 and can be attributed to an increase in biological activities. This is corroborated by SEM observations showing mosquitoes, pieces of leaves and pollens.

Automobile traffic, incineration plants, domestic heating and airports are the main metal sources in urban environment [5, 11, 12]. Due to its characteristics (residential area), traffic is the main source of metals in the Pin Sec catchment. However, other sources located near the catchment (such as the Valorena incineration plant and a nearby highway) could also play a role.

Figure 1 shows metal concentrations in total atmospheric deposition. Apart from cadmium (detected in 60 % of the samples only) the 6 surveyed metals were systematically detected. As can be seen on the figure metal concentrations are variable.

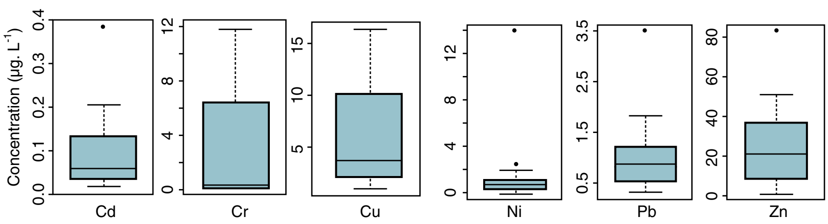


Fig. 1. Trace metal concentrations in bulk atmospheric deposition.

Zinc constitutes 72 % of metals, which is in agreement with the literature. The relative abundance of other metals is as follows : Cu > Cr ≈ Ni > Pb > Cd. According to the literature [5, 13, 14] this order was different before 2000 (Zn > Cu > Pb > Cr ≈ Ni > Cd). This difference can be explained by the decrease in the use of leaded gasoline since the year 2000.

Organic micropollutants

Polycyclic aromatic hydrocarbon and pesticide concentrations were generally low and in some cases close to the quantification limits. For PAHs, concentrations above the quantification limits were measured in 50 % of the samples, mostly at the end of the summer and during the winter. For the campaigns carried out in 2007 and in May-June 2008 no PAH was detected. Acenaphtylene and phenanthene were the only one detected in the June-July campaign at a concentration of 13 ng L^{-1}. For the October-November 2008 campaign, fluoranthene, pyrene and chrysene concentrations range from 2 to 14.5 ng L^{-1}. Finally, for the 2008 winter campaign, the sum of the 15 PAHs is 1090 ng L^{-1}. The higher winter concentrations are due to domestic heating (which is not working during the summer) and to an increase in fuel consumption when vehicles start in cold periods. Similar observations were made in 2008 by [15] for a survey campaign carried out on an urban site in Nantes. During cold periods, the sum of the 7 studied PAHs (B(a)An, B(j)Fl, B(b)Fl, B(k)Fl, B(a)Py, Db[a,h]An, In[1,2,3-cd]Py) was 11 µg m^{-3}, whereas in the warm period the total concentration ranges from 0.09 to 0.8 µg m^{-3}. Individual PAHs concentrations are regularly below the quantification limit (0.014 µg m^{-3} for a sample of 720 m^3). Figure 2 shows the annual variations of the 7 PAH concentrations.

Other similar studies also show variations in PAH concentrations and fluxes between cold and warm periods [5, 15]. This difference is mainly due to heating, which is responsible for 85 % of winter PAH emissions in urban environment. In their study on atmospheric deposition of 2 French cities –one industrial (Le Havre), the other urban (Rouen)- [16] measured max winter concentrations of 1659 ng L^{-1} (Rouen) and 1012 ng L^{-1} (Le Havre).

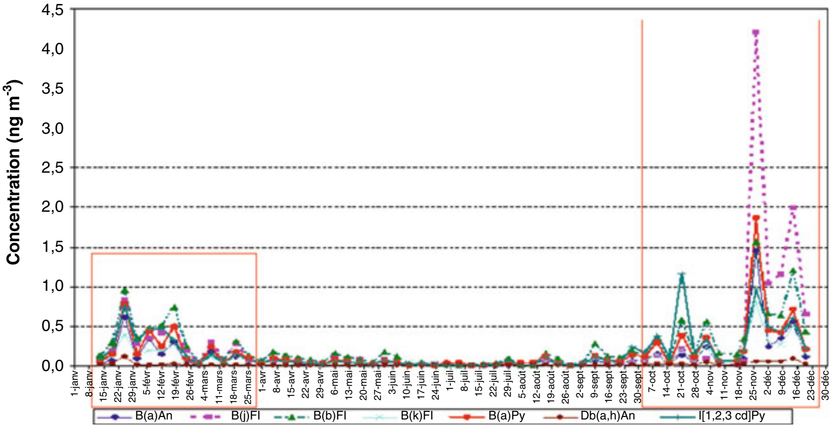


Fig. 2. Air concentration of PAHs in Nantes [15].

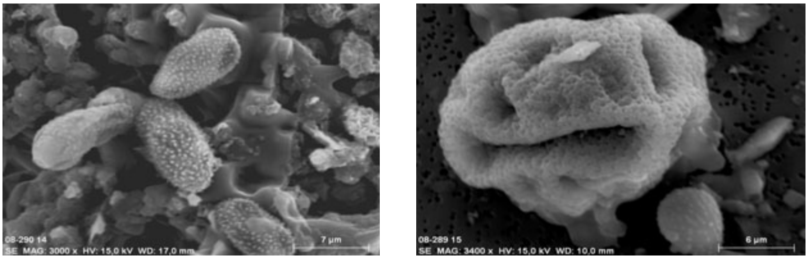
Regarding pesticides, none of the surveyed molecule was detected in the winter, whereas glyphosate and AMPA summer concentrations range from 60 to 470 ng L^{-1} for glyphosate and from 50 to 770 for AMPA. To our knowledge, glyphosate and AMPA have rarely been identified in atmospheric deposition Though their volatility is low, these molecules are found in the atmosphere due to their transport by vaporisation when applied on the catchment and in the neighbourhood. For a typical wind speed of 3 to 5 m s^{-1} pesticides can cover a distance of 250 to 500 km per day. The detection of glyphosate and AMPA in about 10 % of the samples is reported by [16]. On the Pin Sec catchment the possible sources of glyphosate to the atmosphere are chemical weed killing by homeowners, the treatment of impervious surfaces by municipalities and external input from cultures around Nantes such as vineyards, orchards, market gardening.

Identification and analysis of organic micropollutants

A classification of atmospheric particles based on their morphology, size and chemical composition was made. Four groups were defined: 2 groups for organic and mineral particles of natural origin and 2 groups including organic and mineral particles of anthropogenic origin.

70 % of the particles belong to the first group: organic particles of natural origin among which pollens are dominant (figure 3) though there is a large diversity of particles.

In the second group are particles of mineral origin, either isolated or agglomerated. The most commonly observed are irregular particles with a smooth surface and with a size ranging from 5 to 10 µm. The size of the agglomerates varies from 30 to 230 µm. Due to the protocol used to collect the particles; it is unlikely that the agglomerates have been sampled. They were probably formed during the sampling period (15 to 34 days).



(a) (b)

Fig. 3. Atmospheric particles of biologic origin. (a) bacteria, (b) pollen**.**

Organic particles of anthropogenic origin are in the third group. These particles are mainly flying ash formed during fuel combustion. Figure 4a shows a spherical particle with a porous surface originating from the combustion of oil [7] Figure 4b, shows shining fibres which are probably soot particles generated during the combustion of fuels.

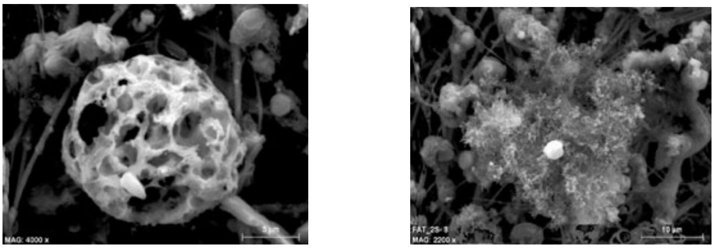


Fig. 4. Atmospheric particles of anthropogenic origin: flying ash formed during the combustion of fuels.

In the last group are spherical particles of anthropogenic origin with a size ranging from 2 to 15 µm. Elemental analysis of these particles as well as a cartography of the elements identified (Si, Al, Fe, Ba) were performed. Si and Al-rich particles probably originate from the combustion of fuels. Microspheres enriched in Fe are probably iron oxides from industrial source. The cartography also shows two small particles containing barium. The presence of Ba mainly associated with S ($BaSO_4$) was reported by [17] who consider that $BaSO_4$ is generated by the abrasion of brakes or road paints. However, S was not detected in our samples, this hypothesis has then to be rejected.

Towards a new approach to measure atmospheric deposition

In order to improve our knowledge of atmospheric deposition on the Pin Sec catchment an original approach is being implemented, comprising particle collectors, pluviometers and a frame on which test samples of different urban materials are fixed (figure 5). The objective here is to get a better estimation of dry deposition taking into account the nature of urban surfaces and air turbulence.

To quantify dry deposition, a transfer coefficient called speed of dry deposition Vd (m s^{-1}), is defined as the ratio of the surface flow of airborne deposition (kg m^{-2} s^{-1}) on the atmospheric concentration of the aerosol near the surface (in kg m^{-3}). To study the transfers of atmospheric pollutants on the surface, Vd must be quantified for different types of urban surfaces typical of the Pin Sec catchment (tiles, glass, bitumen etc) and for various weather conditions (speeds and wind directions, atmospheric stability, air temperature, sunshine period, rate of humidity). The temperature of the substrata will also be taken into account. Besides, the washout of dry deposition by the rain will be studied for the various types of surfaces and according to the meteorological data (rainfall, speeds and wind directions).

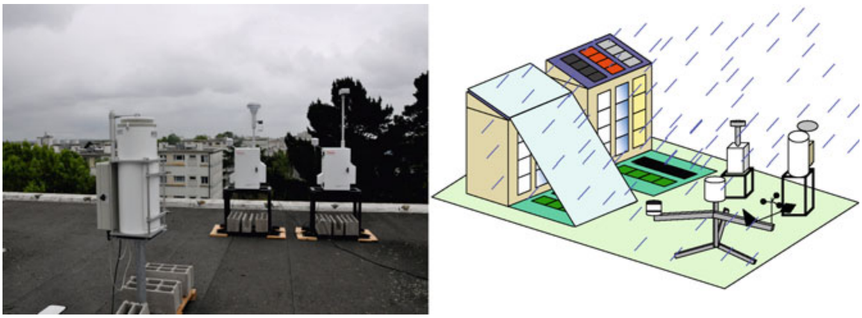


Fig. 5. Atmospheric deposition instrumentation (Particle collectors, pluviometers and frames)

Two types of *in situ* complementary experiments, long and short durations will be carried out. In both cases, the protocols for the study of aerosol deposition will be similar: a tracer of the aerosol and its deposition (Beryllium 7 (^{7}Be) or fluorescein) is used. Test samples are exposed to the tracer during a definite time. Sampling on filter is made near test samples, during their exposure. The measure of the quantities of tracers deposited and collected gives the surface flux of deposition and the aerosol concentration in the air, thus the speed deposit. A meteorological station and an ultrasound anemometer will supply meteorological and micrometeorological data on the conditions met during the experimentation.

The long-term experiments will take place on the “Pin Sec”, during approximately 18 months. They will allow the study of the processes of dry deposition in their entirety and the leaching of the deposition by the rain. Dry deposition will be studied thanks to test samples protected from the rain, and leaching by comparison of the deposition on these test samples protected with test samples exposed to the rain. Beryllium 7 will be used as tracer of aerosols vectors of pollutants as PAHs, metals or pesticides. This radioelement is naturally present in the atmosphere and measured by gamma spectrometry.

Finally, in order to validate the use of ^{7}Be as tracer of atmospheric deposition a Electrical Low Pressure Impactor (ELPI) will be used. This device will allow the measurement of aerosol particle size.

Conclusion

Our study on atmospheric deposition on the Pin Sec catchment (Nantes, France) shows that micro pollutant concentrations are very low. Metal, PAH and pesticide concentrations in atmospheric deposition vary from one campaign to the other. PAHs are only detected during the winter, due to heating, which is the main source of PAHs in urban environments during cold periods.

Most pesticides concentrations are lower than the quantification limits, glyphosate and AMPA, however, are detected in the summer. These molecules probably originate from the dispersion of herbicides containing glyphosate sprayed over vineyards and other cultures in the Nantes region. They may also come from the aspersion of herbicides on impervious surfaces.

Due to the low traffic density and to the absence of industrial activities on the Pin Sec catchment, anthropogenic particles were rarely identified during SEM observations. On the contrary, particles of biological and detritic origins are ubiquitous in our samples.

New research have been launched to complete our knowledge of atmospheric contribution to the global pollution of urban watersheds. This future work will enable us to get a better estimation of dry deposition.

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Atmospheric elements deposition and evaluation of the anthropogenic part; the AAEF concept

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Abstract

The atmospheric deposition on tree trunks is commonly used for evaluating air contamination on a large time scale. However, the deposits are mostly composed of organic matter (generally more than 80 %) and of minerals of geogenic origin. From the elemental composition of the whole deposit, measured by ICP-MS, a calculation was conceived which allows to separate the amounts of elements corresponding to organic matter, to geogenic compounds and, finally, to anthropogenic minerals. For this purpose, the weight of organic matter was obtained through incineration at 550 °C. The elements composition of organic matter was deduced from plant composition. The weight of geogenic compounds was evaluated from the Si, Al content. The formula of geogenic elemental composition took into account the composition of the local soil. This calculation was carried out on four different situations showing the contribution of the main anthropic atmospheric contaminants (Sb, Cd, Sn, Pb, Cu, V, Zn, W, Cr, Ni, Co, As).

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Introduction

The analysis of atmospheric samples is currently the main process for controlling air quality. However, the analysis of the atmospheric deposit on the earth surface is also a very valuable information source for long term studies on atmospheric changes. The atmospheric deposition occurs on soil and on seas but sampling it in its pure state is extremely difficult. Consequently, in a quest for better conditions, it was shown that tree trunks were much better matrices, less contaminated by soil components, remaining alive for a long time, often over one century and being very common under our climatic conditions. From the seventies, numerous studies were carried out by several scientific teams all over the world in order to establish the chemical and especially elemental atmospheric deposition on tree trunks [1-4]. These studies were a main contribution to the understanding of several atmospheric contaminations for instance by heavy metals, Cd, Cr or Cu [5, 6]. They allowed to compare non-polluted reference stations to heavily contaminated ones and to demonstrate whether the concentration of the contaminants increased or decreased over time. It was, for instance, the case of Pb which was seen to decrease after it was forbidden in 1994 as additive for gasoline [7].

More accurate studies concerning elements speciation and origins seemed then to be feasible on the trunk deposits but they required a much better understanding of the deposition process and of the evolution of the deposits on the bark, over time. For this purpose, several points were established:

1. A deposit, mostly of atmospheric origin exists on the tree bark surface. It can be isolated through gentle washing with water [8].
2. It is mainly composed of organic matter either originating from the bark tissues, from plant or animal fragments transported by wind or from beings living on the bark (algae, fungi, bacteria…) [8, 9].
3. As it is, the so-called “deposit” represents a reviviscent ecosystem in which biological activities and growth depend on water supply [8].
4. The superficial bark tissue in dicotyledonae and gymnosperms is suber. It is constituted of cells which die rapidly, the walls of which are impregnated with a lipophilic polymer named suberin [10]. This tissue can be isolated easily and is able to trap a large amount of atmospheric components [11].
5. The elemental compositions of the free deposit on the bark and of the suber tissue are not exactly the same and change with a relative independence overtime.

6. In the two types of matrices, the constitutive elements may have three different origins: a) they may be the elements contributing to the formation and activity of the living matter, mainly plant cell constituents. This is the case of large amounts of Ca, K, N, S, P, Mg and oligo-elements such as Fe, Cu, Zn… b) they may be the elements contributing to the geogenic structures, such as $CaCO_3$ or primary or secondary minerals (feldspars, clays, quartz…). Small particles of such minerals are transported from the soil by the wind and represent one part of the atmospheric deposits. c) they may be the elements having an anthropic origin, for instance from industrial activities or car traffic.

The purpose of the present report was to try to separate, through calculation, the amounts of the different elements coming either from a biological origin or from a geogenic origin or from an anthropic origin.

Materials and methods

Sampling sites

Three places were chosen for a comparative study. The three sampling sites were located in the Rhone-Alpes region (France) near the city of Grenoble. The sites: 1- the Grenoble campus (Joseph Fourier University, Saint Martin d'Hères, Isère, France, altitude 200 m)**;** 2- the "Seiglières" peat bog which is located in a wild, protected area on the North West side of the Belledonne range (altitude 1100 m)**;** 3- the A_{43} highway l'Epine tunnel with a length of 3200 m and with a traffic of 60,000 vehicles per day (Figure 1).

Sampling methods

In sites 1 and 2, the deposits on tree barks were collected from measured bark areas (2 to 10 dm^2) by repeatedly rinsing the bark surface with 10 ml distilled water per dm^2 with the help of a soft plastic brush as described in [8]. The obtained suspension was centrifuged at 8000 rpm (corresponding to 9500 g). The pellet was dried and the supernatant was evaporated. The resulting dry matter weight (DW) was measured. Following this procedure, the supernatant contained the elements issued from the soluble and colloidal fractions of the superficial deposit. The pellet was composed of insoluble particles. All dried samples were kept in the dark in closed Teflon flasks at room temperature.

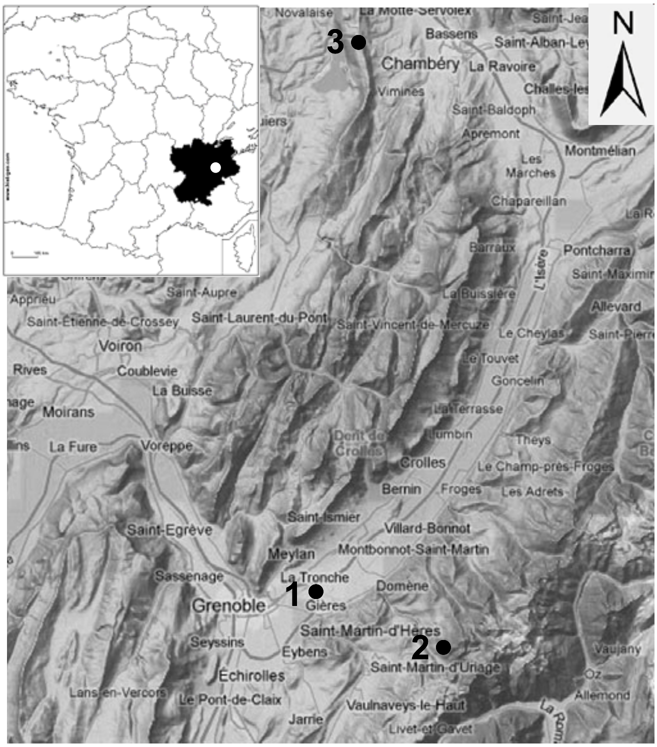

Fig. 1. Scheme illustrating the location of the 3 studied sites. Site 1: Grenoble campus, site 2: Seiglières peat-bog, site 3: l'Epine tunnel.

On site 1, an alveolated wall protected from rain was shown to accumulate atmospheric dust. Dust was collected using a vacuum cleaner and elemental composition was measured by ICP-MS.

On site 3, inside the tunnel, the walls have been washed and the deposit collected. The elemental composition has been analyzed by ICP-MS.

Total mineral content and organic matter content determination (Loss on ignition)

An aliquot of each sample was heated at 550° C overnight under controlled conditions to minimise mineral volatilization [12, 13]. The ashes were weighed and the ashes/DW ratios calculated. The organic matter content corresponded to DW minus ashes weight.

ICP-MS: determination of elemental contents

The digestion step was conducted in PTFE closed flasks, on a digestion plate (Digiprep, SCP Science) as previously described [11]. The acid mixture (10 M HNO_3, HF, $HClO_4$) was chosen to dissolve all materials, whether of organic or mineral origins. Two mL of nitric acid were added to the samples in the flasks, which were then heated for 24 h at 120 °C. After cooling, 2 mL of hydrofluoric acid were added and the flasks were heated for 3 h at 120 °C. After cooling again, 1 mL of $HClO_4$ was added and the flasks were heated for 3 h. Three successive additions of 10 M HNO_3 were performed. The solutions were then heated to near dryness in opened flasks. The residual solutions after complete digestion were brought to the appropriate volume with ultra-pure water. The elemental content was analyzed by inductively coupled plasma–mass spectrometry ICP–MS (Xseries, Thermo Electron), according to the procedure described by [14], modified by the use of a collision cell technique (CCT) to reduce the isobaric interferences of the Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As. The data quality was controlled with reference materials (lichen-336 and sediment-SL1, both from the International Atomic Energy Agency, Vienna). The values obtained with this procedure agreed well with the certified values as verified for each experiment.

SEM–EDX: determination of particles content

Samples were studied by scanning electron microscopy. Samples characterisation was performed using a ZEISS SUPRA 55 VP Scanning Electron Microscope (3rd generation of GEMINI field emission column), allowing spatial resolution down to 1.0 nm, coupled to an energy dispersive X-ray microanalysis system (SAHARA Silicon Drift Detector with Spirit Software of PGT) allowing high counting rate. Samples were studied using electronic imaging (secondary electrons [SE] and back scattered electrons [BSE]), X-ray qualitative analysis and X-ray elemental mapping. Analytical conditions were as below:

- Accelerating voltage: 15 kV.
- Working distance is about 7 mm (optimal distance for EDXanalysis).
- Samples were carbon coated to reduce charging effects.

Results

Composition of the studied deposits

Figure 2 illustrates the composition of the superficial deposit on the bark of 1 to 40-year-old ash-tree stems of the Grenoble region (France). 81.3 % of the weight of dry matter is composed of organic matter (OM) as shown by weight loss at 550°C. The estimation of the total weight of the atmospheric geogenic fraction (AGF) is based on the mineral composition of the surrounding soil, sampled at a 50 cm depth, under the ploughing depth. It is supposed that the Si content of the organic matter can be neglected and that the main source of Si was geological. In country, urban or suburban atmospheres without specific industries (ceramics, steel industry, mining), the Si content of the anthropic fraction was also considered as negligible. In this context, the Si concentration in the sample allowed to deduce the amount of soil minerals to which it corresponded. In figure 1, this estimation gave 9.4% for the atmospheric geogenic fraction (AGF). In places where anthropic emission of Si occurs, the ratios between the main elements (Si, Ca, K, Al, Mg) in the bark samples change, as compared to the soil samples. The total weight of the atomspheric

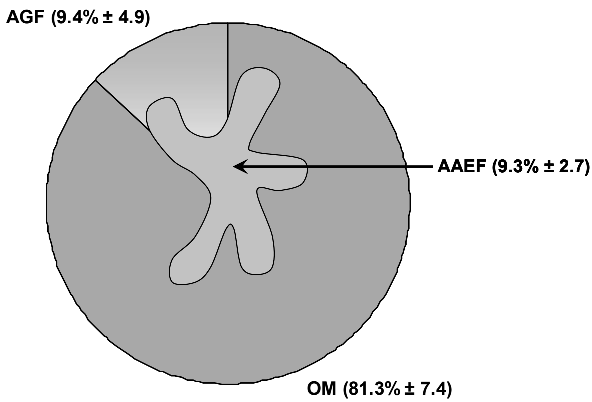

Fig. 2. Diagram showing the composition of the superficial deposit (mean ± SD, n=11) on the bark of ash-trees (*Fraxinus excelsior*) of 1 to 40-year-old sampled in the campus station. OM, organic matter; AGF, atmospheric geogenic fraction; AAEF, anthropic atmospheric elements fraction.

anthropic elements fraction (AAEF) expressed in % of the sample dry weight is obtained as follows: AAEF% = 100 – (OM+AGF). It reaches 9.3% ± 2.7 in figure 2.

Estimation of the elements content of the organic matter fraction (OMF) and of the atmospheric geogenic fraction (AGF)

The organic matter to be found in trunk deposits may have several different origins: through microscopic observation and diverse chemical analyses, one can deduce that the main components are: 1. plant material, 2. humic substances. The other origins (microbes or invertebrates) seem to remain very low. Previous studies [15, 16] established a plant material global composition for elements, on a large number of species. Our own control on ash tree grown under an undisturbed atmosphere agreed with such a formula with few small changes affecting mainly Ca, K concentrations, probably depending on the transpiration rate (not shown). The use of Bargagli formula was chosen for OM in the 4 stations presented in this report.

As shown in table 1, the mineral composition of the soil in the different sites was not the same and it was impossible to use a general formula of the superficial earth crust composition as previously published [17-19].

The SEM-EDX studies of the deposits show that a main part of the AGF is composed of solid particles which can be readily identified as clays, feldspars, quartz, $CaCO_3$… (Figure 3).

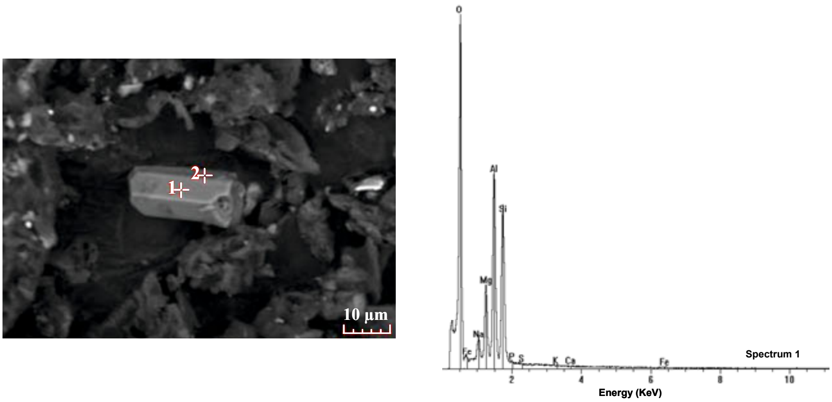

Fig. 3. SEM-EDX picture (80 × 55 µm) of a superficial deposit on a washed bark of a 6-year-old ash tree stem sampled in the Seiglières station. Two spots were chosen for chemical composition spectrum analysis as shown for the particle. The spectrum corresponds to the spot n°1 on the picture.

Table 1. Composition formula for OM and AGF in 2 different studied sites.

Element	OM	AGF (campus)	AGF (tunnel)
K	8000	19600	9500
Ca	6000	78500	2500
Mg	1500	15700	3600
Si	500	607200	369000
Mn	80	1100	510
Na	155	13000	4500
Fe	125	86000	16700
Al	50	165800	26000
Ti	12	10900	2700
Zn	57	200	44
Sr	30	290	45
Rb	30	79	79
Ba	30	670	180
Cu	12	67	4
Ni	1.8	73	22
Cr	1.8	127	62
Pb	0.5	65	16
V	0.55	65	48
Mo	0.8	1	1
Ce	0.55	65	65
W	0.22	5	5
Sn	0.14	7	7
La	0.24	40	40
Cs	0.22	7	7
Co	0.22	23	8
Zr	0.063	238	238
Sb	0.1	3	1
Ga	0.1	20	20
As	0.1	20	12
Hf	0.06	7	7
Cd	0.04	0.5	0.2
Sm	0.045	7	7
U	0.01	4	4

The corresponding spectra issued from the X-ray emission contribute to complete the soil elemental formula established through ICP-MS (Figure 4).

The choice of the surrounding sieved soil as reference formula for AGF is sustained by the fact that the main part of AGF is constituted of small particles coming from The neighbouring area. This use is appropriate in geologically homogeneous sites such as the Isère valley (Site 1) with a high rate of atmospheric emission probably due to agricultural practices

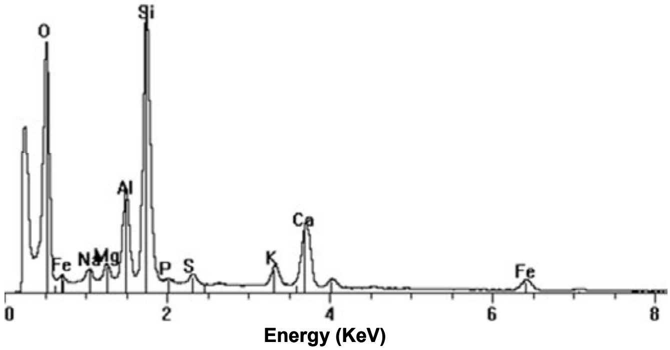

Fig. 4. Global X-ray emission spectrums of the campus soil analysed by SEM-EDX.

(naked soils in early spring and ploughing). It might require supplementary care in places having a complex geology. In site 3, the AGF is mostly influenced by the composition of Hauterivian clay level.

An example of AAEF calculation: anthropogenic copper in a deposit

On 1 to 40-years-old ash tree stems, the copper content measured by ICP-MS is 102 $mg.kg^{-1}$ dry weight. Biological Cu reaches 12 $mg.kg^{-1}$ in OM which represents 81.3 % of the deposit. The biological Cu in the deposit is therefore 12 × 0.813 = 9.8 mg. Cu in the AGF reaches 67 $mg.kg^{-1}$ as shown by the soil formula. In the deposit, the AGF reaches 9.4 % of the weight. The geological Cu in the sample is equal to 67 × 0.094 = 6.3 mg. The “anthropic” Cu in the deposit is: 102 – (9.8 + 6.3) = 85.9 mg. The Cu concentration in the AAEF is (85.9 × 100) / 9.4 = 914 $mg.kg^{-1}$. The ratio between AAEF Cu content and sample whole Cu content is (85.9 / 102) × 100 = 84 %.

The results of the AAEF study

Table 2 shows the values of the ratio between the amount of each element of anthropic origin and the whole amount of this element in the sample (in %). The elements which reach a high concentration either in OM or in the AGF or in the two undoubtedly show a low value for this ratio. A null value of this ratio, seen for K, Rb or Ti is probably not true. For an element such as Fe (AAEF = 27, 68, 14 and 4 % respectively in the four samples), the changes in the ratio value depends on the content of the AGF, which is submitted to important changes. Table 2 compares the values of the AAEF ratio for some elements in the 4 studied sites.

Table 2. A comparison of the AAEF ratio for the different studied sites, N.A: not analysed. Site 1a: campus ash trees, site 1b: campus alveolated wall, site 2: Seiglières peat-bog ash trees, site 3: highway tunnel walls.

Elements	Site 1a	Site 1b	Site 2	Site 3
Sb	93	97	80	96
Cd	92	99	89	95
Sn	89	N.A	N.A	91
Pb	87	96	84	97
Cu	84	92	67	96
V	83	N.A	71	33
Zn	81	96	26	92
W	72	N.A	N.D	58
Cr	72	78	51	80
Ni	72	84	61	49
Mo	68	N.A	N.A	94
Co	59	98	53	N.A
As	57	90	N.A	12
Ba	44	80	37	63
Mn	31	57	48	36
Na	30	N.A	N.A	37
Fe	27	68	14	4
Mg	25	45	10	2
Sr	12	76	N.A	84
Ca	8	79	27	66
K	0	22	0	3

The lowest anthropic contribution on ash-tree was observed in the Seiglières station (alt 1100 m) in the mountain close to Grenoble. On the Grenoble campus, site 2 shows a heavy contamination for the 9 elements considered (Sb, Cd, Pb, Cu, Zn, Cr, Ni, Co, As) with some specific features (high Co and As concentrations). The fact that site 2 was protected from rain was to be taken into account. However, SEM-EDX study (not shown) demonstrated that this site was submitted to a specific industrial contamination. In the A_{43} tunnel, the level of anthropic contamination was high with a noticeable typology: anthropic V, W, Ni, As were specifically at a low level.

Discussion and conclusion

As seen in table 2, the AAEF calculation allows to see several interesting differences between sites, which seem to depend clearly on atmospheric activities. We can expect that a more extensive use of this method will increase its accuracy and reliability. Furthermore, it may induce some diversification in the reference formula for OM, possibly depending on the composition of the local flora.

If crossed with observation methods such as SEM-EDX, this method may contribute to give a much better insight of the type of contamination of a sample.

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Sulfur dioxide and sulfate in particulate matter scavenging processes modeling in different localities of metropolitan region of São Paulo with different cloud heights

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Abstract

This article deals with the scavenging processes modeling of the particulate sulfate and the gas sulfur dioxide in the Metropolitan Area of São Paulo [MASP). Three sampling sites were chosen from MASP center to surroundings. The results show that the in-cloud processes were dominant (80%) for sulfate/sulfur dioxide scavenging processes, with below-cloud process indicating around 20% of the total. Clearly convective events (higher than 20 mm), are better modeled than the stratiform events, with *r* of 0.92. Additionally, the suburb sampling site as expected due to the pollution source distance, presents in general smaller amount of rainwater sulfate [modeled and observed) than the center sampling site, Mackenzie, where the characterization event explains partially the rainfall concentration differences. Cloud height seems also to clarify some results.

Introduction

Air pollution and its impacts on ecosystems are considered one of the problems in the current world topics. Air pollution can either be of anthropogenic origin but also from natural emissions due to decomposition made by the microbiota, and volcanic eruptions, among other sources. There are several atmospheric processes that promote air pollution scavenging, which the rainfall amounts are essential. The rain pollutant compounds in the rainfall and their effects on ecosystems have long recognized the importance of classical antiquity where sulfates play

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an important role. In the *Acid Reign* '2005 Conference ", held in the Czech Republic, there were several works in Eastern Europe demonstrates that significant improvement in air quality in the region, particularly SO_2, and consequent reduction of acid rain, and its sulfate concentration [1,2;3].

Our study area, the metropolitan area of São Paulo [MASP), in addition to several other problems, being a large urban center, has serious environmental problems, air pollution being one of its most relevant. Taking in account this problem, the role of the scavenging processes by raindrops and cloud is important in order to remove air pollutants. Preliminary results of numerical modeling indicated that the air pollutant scavenging, during the rainy season, is dominated by "below-cloud" for the city of São Paulo and adjacent areas [4,5].

Studies on the chemical composition of rainwater from the city of Sao Paulo began in the eighties and nineties [6,7]. Despite having been performed by different groups, which adopted different methodologies, it was observed to decrease the acidity of rainfall over recent years. Results of average values of pH around 4.5, average weighted by volume, (MPV) was observed in studies relating to the period until 1995 [8]. It is important to remember that average pH values are obtained from the mean concentration of H + ions (pH =-log [H +]), while results for samples tested between 2002 and 2004 had values of pH 5.2 (MPV), i.e., five times less acidic than in the '80s and early 1990s, according to [[9]). This variability in acidity was associated predominantly reducing atmospheric concentrations of SO_2 (gas) and, consequently, SO_4^{2-} in rain water [[10]). Another important characteristic of the chemical composition of rainwater in São Paulo has been the remarkable presence of ammonium compound, whose presence in the atmosphere is associated with ammonia gas [NH_3), whose main removal process is the wet deposition due to its high solubility. Other species found in rainwater in São Paulo have been the nitrate ions (NO_3^-), sodium (Na^+) Chloride (Cl^-), calcium (Ca^{2+}), potassium(K^+), magnesium (Mg^{2+}), and also the anions of carboxylic acids [acetic, formic and oxalic). These latter species have shown significant potential contribution to the acidity of these samples [9,10].

Consequently, numerical modeling wet deposition, or scavenging of pollutants, is a topic of great importance, particularly regarding the sulfate. This theme has been approached in the aforesaid work of Gonçalves *et al.* [4,5,11] and Nakeama [12], for the MASP and the Amazon (Silva *et al.*, 2009) with continuous improvement of modeling, including intra-cloud effects, more detailed spectrum of droplets and particles, and collections in more than one site for comparisons within and below the cloud, among others. It should be noted that the work of Gonçalves [5], was studied only one event in two locations, for this reason, this paper focuses on a significant increase of events and locations.

As a result of air pollution, there are numerous works in MASP showing its impact on human health both in cardio-vascular [13,14] and respiratory [15,16]. This air pollution is associated with the presence of sulfate and sulfur dioxide.

Therefore, this work aims to continue this improvement, checking the removal processes in the case of particulate sulfate and SO_2 gas in several different events in two different locations of the MASP to verify the dominance of processes within and below the cloud and efficiency of numerical modeling and the difference between locations, including 2 cloud heights.

Methodology

Three sampling sites were chosen: GV (Granja Viana) at MASP surroundings, IAG-USP and Mackenzie [MASP center). The methodology is divided in following sections: 1- Synoptical characterization of the chosen events; 2- Modeling of the scavenging processes through BV2 model. BV2 is one-dimensional model which model the scavenging processes in-cloud and below-cloud, described in Gonçalves *et al.* [4,5,11].

Ionic chromatography analyzed the observed sulfate in rainwater. The used chromatograph is Metrohm model 761, with condutometric detection and analytical conditons: anionic column aniônica Metrosep A-Supp5 (250mm x 4mm), eluent solution of Na_2CO_3 4.0 mmol L^{-1} / $NaHCO_3$ 1.0 mmol L^{-1}; ratio of 0.7 mL min^{-1}; supressing column Metrohm and regenering solution of H_2SO_4 50 mmol L^{-1} -deionized water with flow of 0.8 mL min^{-1}. Basing on synoptic conditions, it was chosen a group of events where the numerical modeling the scavenging model BV2 was used. These synoptic conditions were usually convective cloud storms, which are usual at MASP, normal around 40% of the total event and 60% of the total rainfall volume. Two cloud heights were used, 8 km and 12 km, being the usual convective cloud tops at MASP.

Results

The results show that the in-cloud processes were dominant (80%) for sulfate/sulfur dioxide scavenging processes, though below-cloud process indicates around 20% of the total (see Tables 1 and 2). Generally, the

Table 1. Modeling results for each event with *below cloud* (B) and *in-cloud* (C) outputs, comparing to the observed sulfate in rain water (A). The model output (with 12 km) is the concentration of SO_4^{2-} in rainwater in µmol.L^{-1}. The % column shows the ratio between the modeled *below cloud* (B) observed by (A) and the percentage ratio between the *in cloud +below cloud* (C) and observed (A). GV is Granja Viana and CM's Consolation / Mackenzie. Observe that the sulfate in rainwater is in µmol.L^{-1}.

EVENTS	LOCAL	SO_4^{2-} OBS.(A) (µmol.L^{-1})	SO_4^{2-} MOD.(B) (µmol.L^{-1})	B/A %	SO_4^{2-} MOD.(C) (µmol.L^{-1})	C/A %
12/Feb/04	GV	28.40	0.29	1.00	1.5	8.63
	CM	28.10	0.27	0.95	1.1	6.40
19/Feb/04	CM	30.10	0.72	2.40	2.9	16.23
	CM	22.60	0.58	2.60	2.3	17.23
	GV	26 50	0.58	2.80	2.3	14.75
22/Feb/04	GV	1.57	0.04	2.50	1.2	118.58
	CM	6.54	0.35	5.40	1.4	42.70
20/Jan/05	IAG-USP	4.65	0.64	13.00	3	104.85
	CM	12.00	1.6	13.00	7.6	103.30
	GV	4.55	0.74	16.00	3.4	122.24
04-05/Apr /2005	GV	5.80	0.56	9.70	2.3	65.92
	CM	8.32	0.93	11.00	3.7	75.72
21/Apr/05	GV	14.83	0.08	0.56	0.33	25.86
	CM	11.40	0.51	4.50	2.0	73.20
21-22/May /2005	GV	1.96				18.89
	GV	1.84				41.90
	GV	2.02	0.26	13.00	1.0	0.00
	GV	2.03				51.59
20/Jun/05	GV	7.53	0.29	3.80	1.2	122.24
	CM	6.30	0.66	11.00	2.8	3.75
10/Mar/06	GV	10.73	0.30	2.80	1.2	25.86
	IAG - USP	14.00	0.86	6.10	3.5	73.20

Table 2. Average concentrations of sulfate observed and modeled total and by location and two different cloud heights.

	average (µmol.L^{-1})		
	all events	GV	CM
Observed	13.0	11.0	16.0
Modeled (total) with 8 km	2.9	2.0	3.7
Modeled (total) with 12 km	4.1	2.8	5.2
Modeled below cloud	0.54	0.4	0.7
Modeled in cloud with 8 km	2.4	1.6	3.0
Modeled in cloud with 12 km	3.6	2.4	4.5

modeling sub-estimates the SO_4^{2-} rainwater concentrations below-cloud, as expected by previous works [4,5,11]. However, the in-cloud processes are far more efficient by a factor of 4.6 higher, even so, smaller the observed sulfate rainwater concentrations using both cloud heights. In general, the below-cloud modeled events were 6.7 ± 5.2% of the observed ones, with a maximum of 16.0% at event of 20/01/05, in GV. In cloud modeling shows values around 31.0 ± 25.2% with a maximum of 76.0% at event of 22/02/04 with 8 km (122% with 12 km), also in GV. The maximum values, summing *in* + *below* cloud processes, reach high values, over 40% of the observed at 4 events with 8 km and 60% with 12 km, all convective events. The explanations for these results are due to many factors such as no mass advection and cloud height differences when it is fixed in 8,000 m, mainly. Therefore, when it changes up to 12,000 m high, the concentrations are closer than the observed data, sometimes even higher [the event of 22/02/04, for example, see Table 1), indicating that this height is a best guess.

These results show a different result comparing to previous work [5,12], where only two events were analyzed. In those events, the below-cloud processes were dominant. Therefore, this article illustrates an improvement, emphasizing the in-cloud scavenging processes, with strong impact of the variable cloud height. The general mean concentrations in the surrounding sampling site of MASP, GV (Granja Viana), show concentrations smaller in air [consequently in the modeled rainwater concentrations) and observed rainwater. Nevertheless, there were events where this ratio was not obeyed, GV presents some events with higher sulfate values in rainwater than the center region (Mackenzie), generally due to events presenting the lowest rainfall volumes (30 and 40 ml, comparing to the mean of rainfall volume of 168 ml, presenting rainfall amount of 1.5mm and 12.3 mm, respectively) and, therefore, they are related to the stratiform events. However, the differences and similarities between both sampling sites suggest dominance of the in-cloud process in both situations, linked to convective cloud events, with higher cloud top. It must be notify that those events correspond to 40% of total number of events at MASP and 61% of total rainfall volume [17]. It must be observed that when all events present the correlation coefficients between the ratio modeled/observed and precipitation of 0.65 (Figure 1) Additionally, clearly convective events, with total rainfall higher than 20 mm, are better modeled than the weaker ones, with correlation coefficient of 0.92 (see Figure 2).

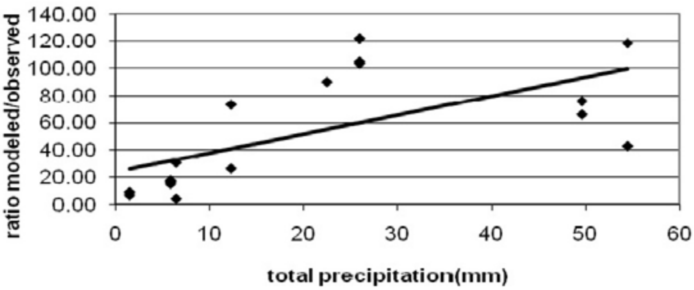

Fig. 1. Correlation between rainfall and the ratio between the concentrations of sulfate in water modeled and observed rainfall for each event. Correlation coefficient of 0.65.

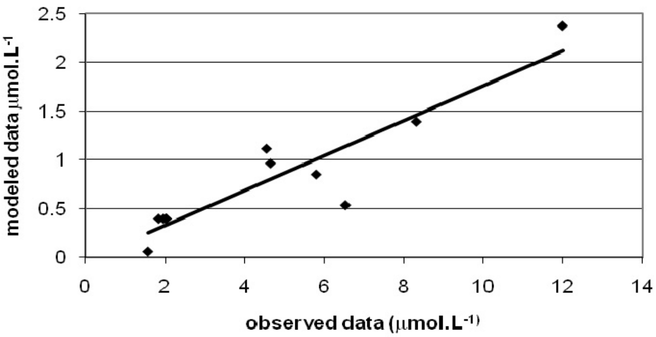

Fig. 2. Comparison between modeled and observed data with total precipitation higher than 20 mm with correlation coefficient of 0.92.

Conclusion

The overall results show that the in-cloud processes were dominant (80%) for sulfate/sulfur dioxide scavenging processes, with below-cloud process indicating around 20% of the total. Clearly convective events, with total rainfall higher than 20 mm, are better modeled than the stratiform events. There is also a clear association with events presenting higher rainfall amount and the ratio between modeled and observed data set. Additionally, the suburb sampling site, GV, as expected due to the pollution source distance, presents in general smaller amount of rainwater sulfate [modeled and observed) than the center sampling site, Mackenzie, where the characterization event explains partially the rainfall concentration differences. Cloud height seems also to clarify some results

As perspectives, it should be pointed to clarify the differences among the events. This improvement can be done through numerical modeling using RAMS (*Regional Atmospheric Modeling System*), basing on Gonçalves *et al*. [5] work, in order to calculate the more details about cloud size and mass advection and satellite images with more data of the cloud top.

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Shop opening hours and population exposure to NO_2 assessed with an activity-based transportation model

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Abstract

In this work we assess for the first time the impact of a policy measure on population exposure to NO_2 by using the activity-based model ALBATROSS, the emission model MIMOSA and the dispersion model AURORA. We found that widening shop opening hours changes the activity pattern of the adult population in the Netherlands. It increases kilometres driven and, as a consequence, emissions. When matching the concentration maps with the dynamic population, we observe an increase in population exposure to NO_2.

Introduction

The most straightforward way of determining population exposure is by making use of concentrations measured at fixed monitoring stations in combination with data on population density. Although this is a simple and commonly used methodology, it does not represent actual population exposure because it ignores movement of people and concentration variations on a local scale. The second issue is dealt with in numerous studies by applying interpolation maps to introduce spatial variation. The first issue is much harder to take into account.

A decade ago Shiftan [1] already explored the advantages of activity-based modelling for air-quality prediction purposes. An activity-based model basically predicts a diary for every individual: which activities will be performed where and for how long and if a trip is involved which

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transport mode will be used [2, 3]. By using an activity-based transportation model we can improve on both of the issues stated above: we can use dispersion modelling based on more accurate emission estimates of modelled trips and we can model the location of every individual for every hour of the day. This way a truly dynamic exposure analysis can be made by geographically matching hourly concentrations and hourly population densities. Moreover, we are able to differentiate between different subpopulations and different activities allowing a more detailed exposure analysis.

This methodology has the potential to provide valuable information for air pollution epidemiology [4] and policy purposes [5]. As an example this paper will look at a scenario (widening of shop opening hours) and evaluate the effects on population exposure.

Methodology

The modeling framework will be summarized in a nutshell since this is not the main focus of this paper. An extensive description of the framework can be found in Beckx *et al*. [6-9].

Activity-based transportation model

The activity-based transportation model ALBATROSS, an acronym for A Learning Based Transportation Oriented Simulation System, was used to predict activity-travel patterns for the Dutch population [10, 11]. The model starts by developing a synthetic population using demographic and socio-economic geographical data from the Dutch population and attribute data of a sample of households originating from a national survey including approximately 67,000 households. Every adult inhabitant of the Netherlands (or more precisely, household head), is incorporated in the synthetic population. The 4-digit postal code area (4PCA) was chosen as the spatial unit for the ALBATROSS model.

Activity-travel schedules are simulated for all the individuals by using decision trees representing each choice (e.g. a stochastic choice of activity type, duration of activity, choice of location, transportation mode involved, etc.). While making these decisions several constraints, e.g. institutional constraints and household constraints, are taken into account to make resulting activity diaries more realistic. We used the output of one run of the model representing one-day diaries across all days of the week of all people in the study area. Thus, the activity patterns should be representative for an entire week. Presently, possible seasonal differences in weekly activity patterns are not captured. To predict traffic flows, we extract from the activity patterns generated O/D-trip matrices. Those predicted trips for the entire population are assigned to a road network ('Basisnetwerk') by using an all-or-nothing assignment (shortest path in distance).

Emission model

Traffic flows are converted into vehicle emissions by applying the emission factor approach from the MIMOSA emission model [12].

Dispersion model

In a next phase, the activity-based approach is further extended by converting the emissions into pollutant concentrations. For this purpose, the AURORA model is applied to simulate the dispersion and conversion of the emissions into concentrations [13].

Integration of the models

The goal of the modelling framework is to assess population exposure both in the base case, as well as in a scenario. The predicted hourly concentration fields from the ALBATROSS-MIMOSA-AURORA modelling chain are combined with hourly information on people's location to calculate the exposure. By using the population information from the activity-based simulation, hourly population maps are simulated and dynamic exposure values can be estimated (Figure 1).

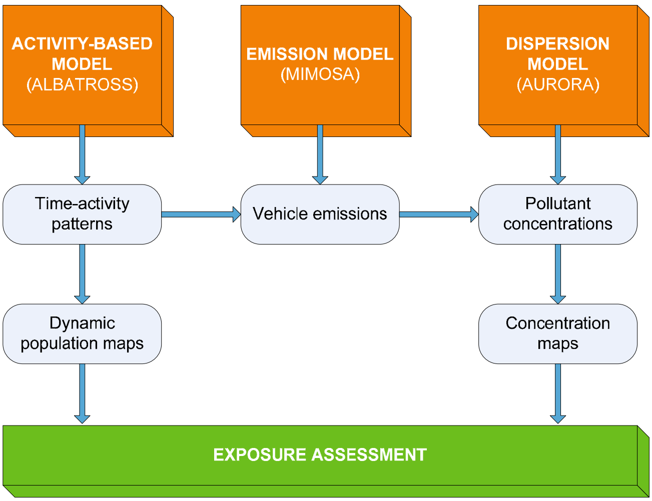

Fig. 1. The exposure modelling framework.

Each step in the process is evaluated to assess modelling power. As it appears, a comparison between the modelled emissions and reported emission values demonstrate good correspondence. When comparing the simulated concentrations for the base case with measured values at Dutch monitoring stations, the index of agreement varies between 0.40 and 0.70 for NO_2 [6], which demonstrates that the activity-based air quality model chain is able to simulate the hourly concentration patterns in the Dutch study area with sufficient accuracy.

Results and discussion

The scenario considered involves a widening of shop opening hours for daily and non-daily shopping. The new opening hours are from 6 a.m. until 10 p.m. on weekdays and Saturdays, which allows shopping earlier in the morning and later in the evening. As a tendency, this is a relevant policy scenario keeping in mind that liberalization of shopping hours in the Netherlands is relevant continuously already since 1996.

As a case study we model the exposure difference for NO_2 of the adult population of the Netherlands (approximately 10.5 million individuals).

Results

Effects on the activity pattern

As a consequence of the scenario, the ALBATROSS model predicts approximately 6% more non-daily shopping hours and 0.5% more daily shopping. The change in activity pattern results in more transport hours; an increase of 0.5%. This increase is translated in more kilometres driven by car. In a study of Jacobson [14] an augmentation in weekly shopping time was observed as well, both from a simple model and from empirical findings.

On an average weekday, shifts are seen during the day (Figure 2). As expected, there will be more shopping in the morning and in the evening. This is offset by less time spent on in-home activities and on leisure. Between 8 a.m. and 5 p.m. there will be somewhat less shopping compared to the reference situation.

In the reference situation and in the scenario women execute more shopping-activities than men. Both men and women adapt quite similarly to the new opening hours and the same temporal pattern can be observed.

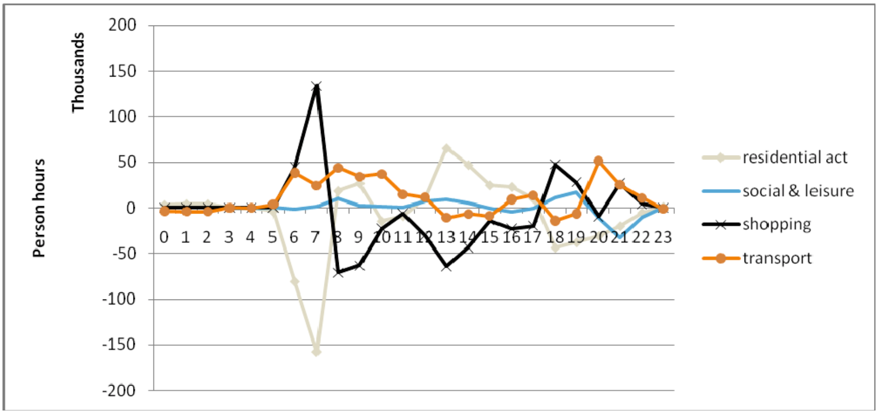

Fig. 2. Difference in activity pattern between scenario and reference situation on an average weekday per hour of the day (on the x-axis the hours of the day are represented).

Effects on concentrations

The ALBATROSS model showed more kilometres driven by car; as a consequence emissions of road transport will rise (output of the MIMOSA-model) as well as the derived concentrations in ambient air (output of the AURORA-model). The activity pattern is assumed the same for every week of the year, but the meteorological conditions change so concentrations will differ from week to week.

The concentration maps use a raster overlaying the Netherlands, one grid cell being 9 km² (a total of 11439 grid cells) (Figure 3).

Generally concentrations are higher in densely populated and in industrialized areas. Around major highways elevated levels of NO_2 are observed. In the North See a wedge of higher concentrations can be observed on shipping routes. The direction of plumes is clearly affected by the wind direction; in the illustration (Figure 3) wind direction was southwest, the most prevailing wind direction in the Netherlands.

On Figure 3 the differences in concentration levels between the scenario and the reference situation are presented. Absolute differences are small as expected (maximum difference of 0.40 µg/m³). Above the North Sea remarkable differences exist between the current situation and the situation where shop opening hours are widened. We did not yet find an explanation for this and it may be caused by assumptions made in the AURORA-model. The sensitivity of AURORA to scenarios with little or no impact is rather limited, caused by uncertainty introduced by each of the models.

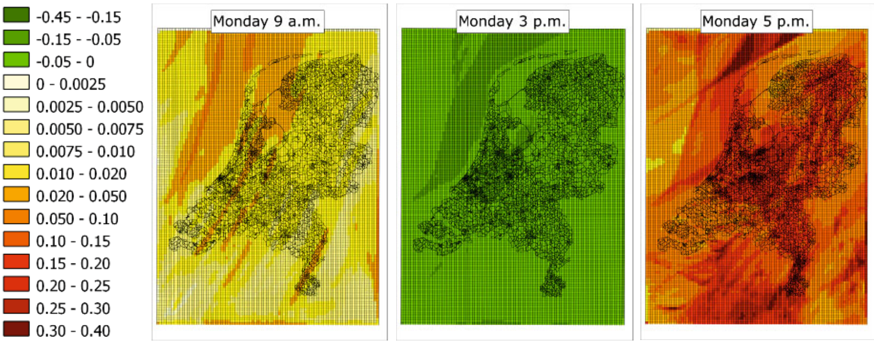

Fig. 3. Difference in concentration levels (µg/m³) between scenario and reference situation on three moments on Monday the 4th of April 2005.

Effects on exposure

Total exposure is calculated multiplying dynamic population in the postal code area with concentrations in the grid cell. All calculations were performed for April 2005.

Results show an increase in population exposure to NO_2 (Figure 4). On an average weekday in April exposure will increase with 0.15 µg/m³. This number is relatively stable across the days of the week. At night-time and on Sundays the difference between the base-situation and the scenario is negligible, which is a reassuring result. Tested over several weeks the relative change in exposure on an average weekday is 0.4%.

In certain neighbourhoods and on certain hours a more substantial increase can be observed (Figure 5).

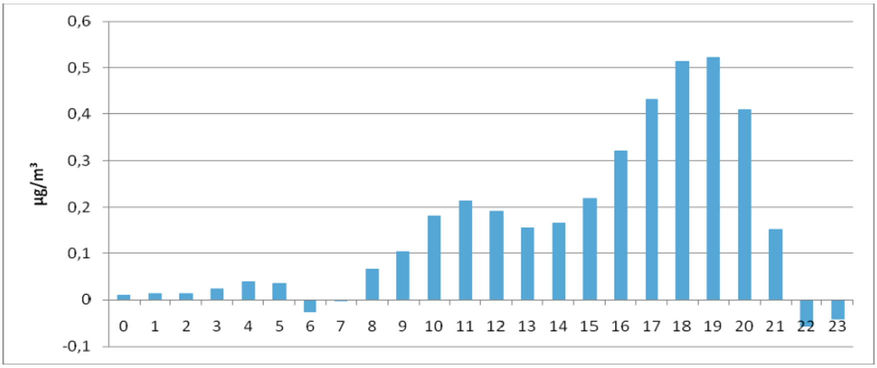

Fig. 4. Difference in exposure between scenario and reference situation on an average weekday per hour of the day for April 2005 (on the x-axis the hours of the day are represented).

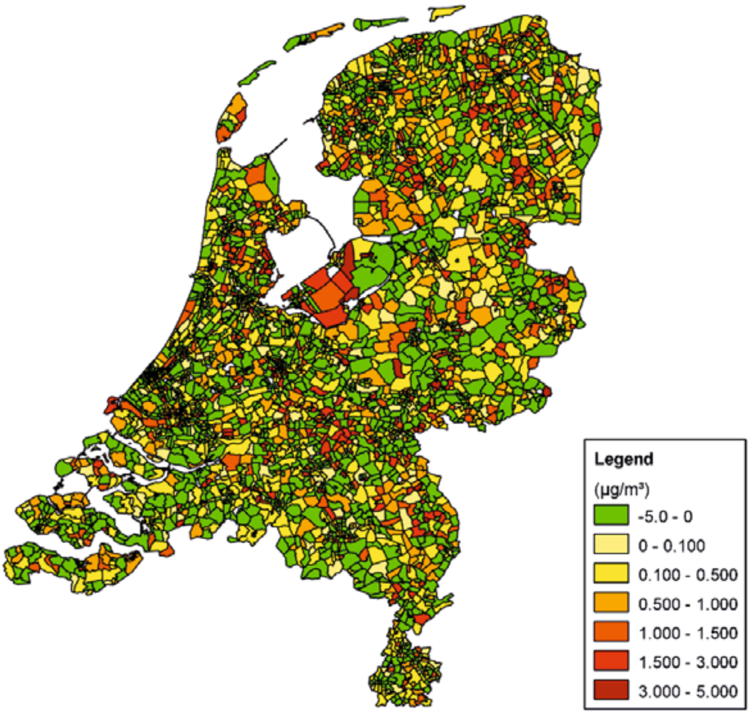

Fig. 5. Difference in exposure between scenario and reference situation on an average weekday geographically represented per postal code area.

The increase in population exposure is statistically significant. The numbers in Table 1 are corrected for postal code areas with a low number of people residing there (less than 100) because a ratio of small values can cause inconsistent results.

Table 1. Difference in exposure between scenario and reference situation (April 2005) [µg/m³].

April	Week 1	Week 2	Week 3	Week 4
Monday	0.1079	0.1599	0.1223	0.1441
Tuesday	0.1163	0.1421	0.1323	0.1487
Wednesday	0.1168	0.1662	0.1174	0.1232
Thursday	0.1122	0.1741	0.1935	0.2165
Friday	0.2113	0.1154	0.1852	0.2371
Avg weekday	0.1329	0.1515	0.1501	0.1739
Saturday	0.1592	0.0881	0.1472	0.1696
Sunday	0.0614	0.0657	0.0604	0.0677
Avg week	0.1265	0.1302	0.1369	0.1581

Discussion

Since the early 1970s, the EU has been working to improve air quality and much progress has been made since then. However air pollution continues to be a matter of concern. Several European, national or regional policy measures explicitly aim at lowering concentrations of harmful pollutants (e.g. through legislation or vehicle technology). There is a lack of awareness that other measures, not explicitly focussing on air quality, will significantly impact exposure and health both in a positive and in a negative way. It may happen that expensive measures to cut emissions and lower concentrations are offset by initiatives from other policy makers who are not aware they are affecting air quality. Prolonging shop opening hours is such a measure that has an unintended negative side-effect on population exposure, although the effect is only limited.

Activity-based transportation models are proven to be better in evaluating the effect of TDM (travel demand measures) and integrated policies because of their ability to incorporate secondary effects [1]. Examples of TDM are traffic restraint measures, pricing mechanisms, telecommunication or high-occupancy vehicle lanes. Next to transportation measures, activity-based models are able to calculate the effects of certain scenarios having no obvious relation with transport or air quality. Institutional changes (e.g. changing working hours, changing shop opening hours) or demographical changes (e.g. ageing of the population [15], changing percentage of part-time workers, more one-adult households) can be assessed with an AB-model, all being evolutions relevant to policy nowadays.

So far and to the best of our knowledge, there are hardly any papers on the modelled quantitative effects of policy measures on air pollutant concentrations, population exposure and health for larger geographical areas. One of the first papers of this sort looked at the effect of the Congestion Charging Scheme (CCS) in London [16] which affected an area with approximately 7 million inhabitants. This study only considered emissions and not concentration or population exposure. The health effects of the London CCS were assessed by Tonne *et al.* [17] and they found a decrease in population exposure to NO_2 in the Greater London area of 0.10 μg/m^3; in the congestion charging zone the effect was larger (-0.73 μg/m^3). A similar study on the Stockholm congestion charging found effects of -0.23 μg/m^3 (population exposure to NO_x) [18]. These resulting exposures are observed ex post; after the introduction of a policy measure. Our simulation is valuable in a way that measures can be assessed ex ante. This gives priceless information to governments who want to assess costs en benefits before a policy measure is introduced.

Conclusion

This paper presented the first analysis of a scenario generated with the activity-based model ALBATROSS and the effects on population exposure to NO_2. We demonstrated that by using this approach the effect of a policy measure on population exposure can be assessed. In this paper, we showed that an increase in population exposure of 0.15 µg/m³ is associated with the widening of shop opening hours. Examples of other measures or scenarios that can be evaluated by such an approach are ageing of the population, teleworking, introduction of congestion charging, etc.

As discussed above the sensitivity of the MIMOSA-AURORA chain to scenarios with little or no changes in concentrations is rather limited. Taking into account the complexity of the modelling framework, the required computer runtime and the lack of flexibility, we suggest replacing the emission and dispersion model with a land use regression model. The challenge here will be to adequately incorporate a temporal dimension into the land use regression model.

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Lung deposited dose of UFP and PM for cyclists and car passengers in Belgium

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Abstract

Commuter cyclists experience short episodes of high exposure to traffic born air pollution that have potential adverse health effects.

We have compared respiratory parameters and exposure to Ultrafine Particles, PM2.5 and PM10 for 55 persons who cycled and drove identical trajectories in three Belgian locations in a pair-wise design. Differences in lung deposited doses are large and consistent across locations. Physical activity significantly increases exposure of cyclists to traffic exhaust. Additional analyses of physiological parameters reveal changes in exhaled NO, serum BDNF and % blood neutrophile cells, but we hypothesize that these effects do not offset the overall health benefits of cycling.

Introduction

Adverse health effects of exposure to air pollution have traditionally and consistently been associated with ambient measurements at fixed monitoring stations [1, 2]. The exact contribution of different compounds and fractions of Particulate Matter (PM) to specific health endpoints has not been fully elicited but emissions of internal combustion engines and traffic have been suggested to be more toxic than the general mixture. Proximity of the residence to major roads has been used as a surrogate for exposure to traffic exhaust [3]. Close proximity to traffic leads to peak exposure

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when trailing vehicles or cyclists cross the tailpipe plume. Exposure during commuting therefore makes a significant contribution to total exposure [4, 5]. At this moment it is not clear what the health effects of short bursts of high exposure are relative to the effects of chronic exposure which are well known from epidemiological studies. Nevertheless some observations suggest that short episodes of high exposure can potentially account for some of the observed health effects [6-9]. Hence, accurate assessments of exposure in different vehicles are necessary to validate model predictions so that future studies can take dynamic exposure during commuting into account.

Here we present measurements of particulate matter inside a car and on a bicycle. Ventilatory parameters are simultaneously measured to assess the amount of pollutants actually inhaled during each trip. Only a few studies [10,11] have taken into account that cyclists have an increased minute ventilation that influences their inhaled dose of air pollutants.

Experimental – Method

The study described in this paper was conducted within the framework of the SHAPES and PM²TEN projects (www.shapes-ssd.be). The working hypothesis was that PM concentrations would be higher in the car than on the bicycle. Three routes were chosen in three different Belgian regions. Brussels (BxL) is the capital located in the centre of Belgium (pop. ~1.5 million, ~ 4900 inhabitants/km²), Ottignies-Louvain-la-Neuve (LLN) is a university town (~30000 inhabitants, ~ 900 inhabitants/km²) in Wallonia about 20 km southeast of Brussels and Mol, a small rural town (~34000 inhabitants, ~300 inhabitants/km²) in Flanders about 70 km northeast of Brussels (NIS, 2009). All routes are in the form of loops and include sections with on-road cycling, cycle lanes and grade separated cycling paths parallel to the lanes for motorized traffic.

The Brussels route (4.8 km) loops through the European district. Its southern leg includes part of the Rue de la Loi, a busy 4 lane street canyon. The routes that were chosen in Louvain-la-Neuve (5.5 km) and Mol (6.8 km) included very quiet residential areas as well as a busier street with mostly local traffic and few heavy duty vehicles. The route in Louvain-la-Neuve includes some slopes, similar to Brussels, whereas the route in Mol is flat.

Test persons were recruited through the 'SHAPES injury registration system'. Commuter cyclists were asked to report weekly on their bicycle usage and related traffic injuries. The inclusion criteria are: (1) age between 18–65 years; (2) having a paid job outside the home; (3) cycling to work at least twice a week; (4) living in Belgium. 55 healthy non-smokers were randomly selected (stratified by their place of residence relative to one of the three case-study locations). Mean age and BMI were similar in all 3 locations. Mean age and sex ratio are similar to those of frequent commuter cyclists in Belgium.

During the experiment, each test person was first driven over the route as a passenger in a car. Immediately after the car trip the subjects rode the same route by bike. Subjects were asked to cycle at the same average speed as during their trips to and from work which were recorded in web-based diaries prior to this experiment. The bike trip always followed the car trip to avoid an effect of the bike ride on the ventilation and heart rate during the car ride.

P-Trak (TSI Model 8525, USA) and DustTrak instruments were used for each pair of trips to sample air within the breathing zone (i.e. approximately 30 cm from the mouth) at 1-second resolution. Simultaneous respiratory measurements were made during each trip and synchronized with the P-Trak, DustTrak and GPS datasets. Breathing frequency, tidal volume and oxygen uptake were measured using a portable cardio-pulmonary indirect breath-by-breath calorimetry system (MetaMax 3B, Cortex Biophysik, Germany) fixed into a chest harness. Minute ventilation (VE) was calculated by the MetaSoft software (VE = breathing frequency x tidal volume). Inhaled amounts were calculated by multiplying PNC and PM mass with VE. The lung deposited fraction was determined based on deposition factors (DF). To relate the lung deposited dose to cycling intensity we used the data presented in [12] who report that DF strongly increases with exercise. An average PNC DF at rest of 0.63+/- 0.03 was used in our calculation of dose for car trips compared to 0.83 +/- 0.04 for cycling. Chalupa and colleagues reported fractional UFP deposition as a linear function of tidal volume [13]. For comparison this function was evaluated for each breath taken during the car trips (only car trips, because it was only defined at tidal volumes <1 liter which was exceeded during all the cycling trips). We took a similar approach for the calculation of PM2.5 and PM10 doses based on [14] (mass based DF= 0.23 +/- 0.03).

Results and discussion

PM data were missing on some trips due to equipment failure, especially in Mol. Ventilation data was excluded whenever breathing frequency fell below 7 sec for cycling trips and below 12 seconds for car trips. Whenever there were anomalies in the measurements of one trip, the entire trip was excluded from the analysis (and since we use a pair wise design, the associated car-or bike trip was excluded too). Only subjects with valid air pollution and respiration data for both the bike and the car trip were taken into account. After applying those cleaning criteria, the final dataset includes 24 cyclists in Brussels, 6 in Louvain-la-Neuve and 13 in Mol.

Concentrations

Concentrations of UFP in traffic are subject to very short but high peaks. PNC values are more chaotic on the bicycle and peak values are frequently higher whereas numbers inside the car are more stable. Average concentrations for the three case studies are shown in Figure 1. PNC are approximately three times higher in Brussels than in both other locations. In general, observed PNC for Brussels are similar to published numbers for other cities such as Copenhagen [15] and a number of Dutch cities [16] but lower than those observed in very large cities such as London or Los Angeles.

We have identified an inconsistency in the measured average PNC values which are significantly higher in the car in Mol, but not in Brussels (at relatively higher concentrations) or LLN (with relatively lower concentrations). This apparent inconsistency indicates that there are differences between locations, or vehicle characteristics, or the time at which they were sampled, which remain unexplained. Similar findings were found by [16] who concluded that the overall mean PNC in the car is 5% higher than the overall mean of cyclists in 11 Dutch cities. Nevertheless they did find important differences in either direction. Mean PNC was higher in the car in 4 Dutch cities and higher on the bicycle in 4 other cities and small differences were reported in the other locations. Earlier studies found 6% higher UFP concentrations in cars compared to cyclists [17].

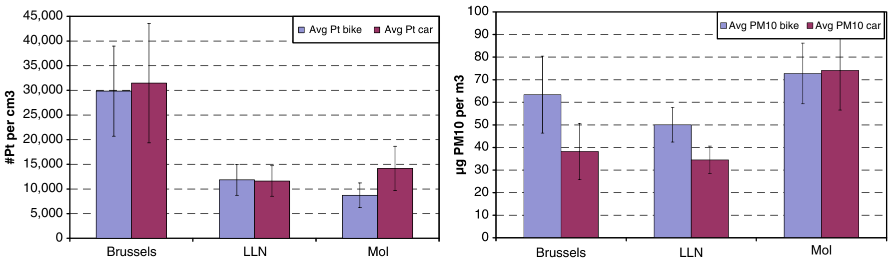

Fig. 1. PNC while cycling and driving identical trajectories in three locations (left: UFP PNC/cm3; right: PM10 in µg/m3). Values are mean and SD.

The opposite result was found for particulate mass. Average PM2.5 and PM10 levels were found to be significantly lower inside the car in Brussels and LLN, but not in Mol. Average levels of PM were elevated during the experiment in Mol due to specific meteorological conditions which did not occur during sampling at the other locations. Although a pilot study had indicated that in car concentrations were higher than on the bike in Mol, this difference was not found to be significant in this study. In Brussels and LLN the opposite was even observed: lower concentrations in the car. In contrast with our results, most other studies [10, 17-19] have reported higher PM concentrations for car drivers. Some authors however reported results which are in line with our results for cyclists in Brussels and LLN [20,21].

Lung deposited dose

Women breath significantly more frequent (t-test $p<0.01$) and at lower tidal volumes than men ($p<0.0001$). As a result men inhaled about 17% more air while cycling ($p<0.01$). Ventilation frequency is about 1.5 times higher and tidal volume increases by about 2.6 while cycling compared to sitting in a car. Combined, this leads to an increase in total inhaled volume by a factor of 4.0 in women and 4.5 in men. The inhaled dose was therefore calculated for each breath as the average pollutant concentration multiplied with the volume of inhaled air and then summed over the entire trip. Quantities of particles inhaled by cyclists are between 400 and 900% higher compared to car drivers on the same trajectory.

The amount of particles that is estimated to stay in the respiratory tract after being inhaled (dose) is shown in Figure 2 for the number of ultra fine particles and Figure 3 for the mass of PM10. The DF for UFP is higher when performing physical exercise but there is little difference whether the average DF from [12] is used or a specific DF for each breath is calculated based on [13]. The fact that the latter is slightly higher may be due to the fact that all of our test persons are commuter cyclists and hence have a higher than average level of fitness.

These results suggest that the discussion on which transport mode is associated with the highest *concentrations* is not relevant because there are obvious differences in *exposure* between cyclists and car drivers, an aspect that has often been ignored for lack of measured data. Three differences influence the exposure of cyclists to air pollution. The most important one is a large increase in ventilation frequency and tidal volume. Secondly, for the same inhaled quantity, the amount of particles that remain in the respiratory tract is higher while exercising. Finally, the

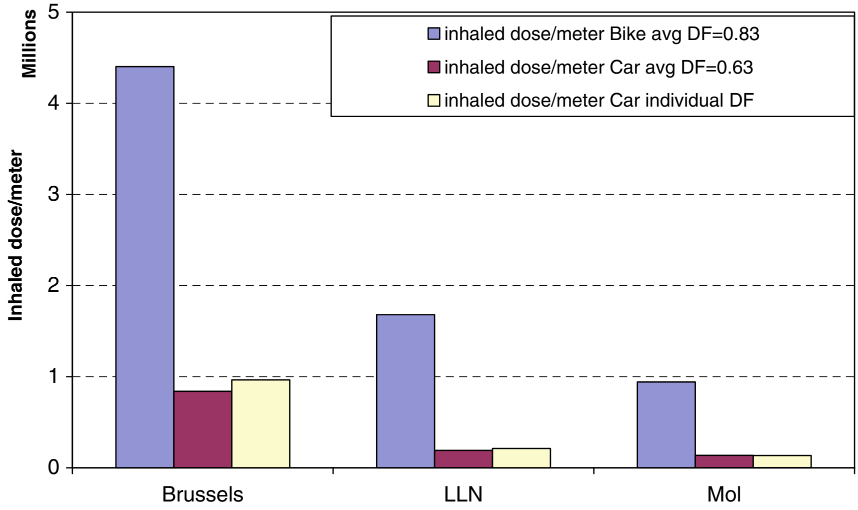

Fig. 2. Lung deposited dose of UFP. Paired trips with bicycle and car.

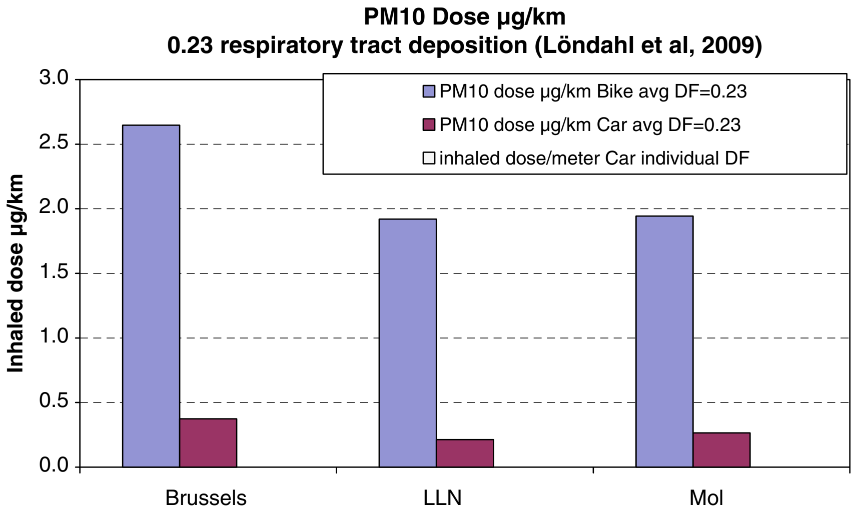

Fig. 3 Estimated lung deposited dose of PM10 per kilometer.

time needed to complete the trajectory is often longer for the cyclist although average speeds in cities are similar. It is mainly the differences in ventilation (and associated deposition) that matter. Earlier studies mostly assume a factor of 2.3 increase in exposure by copying the value proposed by [22].

Zuurbier and colleagues recently suggested a slightly lower value of 2.09 for trips at a low cycling speed (~12 km/h) [11]. We suggest that a value of 4.3 is the best estimate available to date because the average speeds in this study were self selected and in line with everyday speed choice over a long period of time.

The remaining question however is whether this difference in exposure is associated with any significant health risks? Observations by [23] and direct evidence from [7] suggest that short episodes of high UFP exposure can potentially account for some of the cardiovascular effects. Most epidemiological evidence of the health risks related to near-traffic pollution gradients is derived indirectly through PM2.5 but UFP is considered to be a likely candidate to contribute to cardiovascular health effects, due to its characteristics, and its potential to induce inflammation. UFP is part of the diesel exhaust, that is likely carcinogenic and may therefore cause oxidative DNA damage [15]. While evidence points into the direction of respiratory and cardiovascular effects of UFP, some of the evidence is still circumstantial and there is need for more targeted and convincing research to link UFP from traffic to health endpoints [24].

Preliminary analysis of a physiological data collected in the framework of this project suggests that PM0.1 inhalation during exercise causes an immediate reduction in alveolar exhaled NO and a rapid differentiation of blood cells into a pro-inflammatory state (an increase in blood neutrophils) [25]. In addition our observations revealed that exercise induced plasma BDNF (Brain-Derived Neurotrophic Factor, a neurotrophine that is upregulated by exercise) was suppressed after cycling in traffic polluted air but not in filtered air conditions. Although active commuting is considered to be beneficial for health, this health-enhancing effect could therefore be negatively influenced or partially offset by exercising in an environment with high concentrations of PM.

On the other hand there is incontrovertible evidence from observational and randomized trials that regular physical activity contributes to the primary and secondary prevention of cardiovascular disease and several other chronic conditions and that it is associated with a reduced risk of premature death [26].

Conclusion

We conclude that the size and magnitude of differences in concentrations depend on the location, confirming similar inconsistencies reported in literature. In Brussels and LLN, the PM2.5 and PM10 concentration was significantly higher for the bicycle compared to the car. In Mol PNC were higher for the car. Minute ventilation while cycling is on average 4.3 times higher compared to driving a car. Inhaled µg PM2.5/km and µg PM10/km is significantly higher while cycling compared to driving in a car. The bicycle/car ratio ranges between 5.92 (2.06) – 8.99 (1.03). Hence it is necessary that future exposure measurements use realistic cycling speeds that reflect the physical activity and respiration associated with typical speeds in commuter cycling (in line with self selected speed over a long period of time). Likewise it is important to take minute ventilation and deposition fraction into account.

The number of complete observations (pairs) in LLN and Mol is relatively small and the analysis of our results is therefore subject to caution. Further research should be performed on larger samples, in more locations and during other seasons in order to confirm the results. Given that the results for concentrations are apparently inconsistent, care should also be taken to include in-car measurements of relative humidity in future studies.

Our results should be seen as an opportunity to improve cycling conditions. Given the fact that people who choose to cycle contribute to better air quality, policies and measures should first target the motorized traffic that is a cause of air pollution and focus on reducing exposure (rather than reducing average concentrations) to prevent unwanted health effects. The results of this study will likely have interesting implications for the planning of cycling infrastructure. Any measure that increases the distance between cyclists and tail-pipes will help to reduce exposure. Identifying and implementing separated and dedicated routes for cyclists and motorized traffic will go a long way in decreasing exposure but will likely not offset the entire exposure difference. However we hypothesize that it is unlikely that this would completely offset the health benefits of cycling.

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Emissive behaviour of two-wheeler vehicle category. Methodologies and results.

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Abstract

The main objective of the study was to conduct measurements on powered two-wheelers to improve knowledge about emissive behaviour of this vehicle category on various types of test cycles. The study presents a method to analyse, during different driving cycles on motorcycles, the influence of speed profile on the emission factors, through the basic parameters of elementary kinematic sequences. These sequences were obtained through suitable fragmentation of complex urban driving cycles. The method was applied to evaluate the regulated exhaust emissions (CO, HC, NO_X) and to characterize the emissive behaviour of two Euro3 motorcycles (a 250 and a 1000 cm^3), tested on a dynamometer bench.

Introduction

Transport activities contribute significantly to air polluting emissions in all the world. In order to quantify the environmental impact attributable to road transport and to define effective policy to improve air quality, analytical and experimental studies are indispensable for policy makers, consultants and researchers.

Throughout southern Asia and Europe, and particularly in Italy, mopeds and motorcycles are widely used and represent a large proportion of motorized vehicles. Because of their prevailing use in urban environments, determination of emissions from 2-wheel vehicles is important for estimating their relative contribution to total emissions attributable to road transport. They are important means of transport especially in large cities, helping to meet daily urban transport needs. In these countries, the use of mopeds is facilitated by the mild climate and is linked to several factors including parking ease, agility in traffic jams, inexpensive maintenance.

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In the past, the focus was placed on exhaust emissions of passengers cars and Heavy-Duty vehicles (HDV). Due to the introduction of legislation together with tightening the applicable limits on CO, HC, NOx, and PM emissions, the absolute emission level of these types of vehicles have been reduced significantly. As a results, the relative contribution of the emissions of powered two-wheelers to the total air pollution increased since no tighter limits were defined for this category (ECE R 40). The necessity to introduce more stringent limits for this category has also been noticed by the European Commission. For that reason Directive 97/24/EC was developed and it became effective in June 1999 introducing more stringent limits (Euro 1) compared with ECE R40. In 2003, stage 2 (Euro 2) of 97/24/EC entered into force and reduced the applicable limits again without changing the Type Approval test cycle. For 2006 (Euro 3) the emission limits are decreased significantly again and in addition the Type Approval test cycle is replaced by a combination of UDC and EUDC.

Starting from the above considerations, an experimental investigation on 2-wheel vehicles emissive behaviour was being jointly performed by Istituto Motori and the Department of Mechanical and Energetics This experimental activity was performed in order to characterize the emissive behaviour during different driving cycles of two Euro 3 motorcycles: a 250 cm^3 and a 1000 cm^3. Average emission factor models commonly used in Europe, are based solely on the average trip speed to predict emissions, thus they are not sensitive to variations of vehicles instantaneous speed and acceleration, which have a strong effect on emissions and fuel consumption. In this study, the influence of speed profile on the emission factors of these two motorcycles is analysed through two kinematic parameters: the mean speed and the mean product of speed and acceleration of elementary kinematic sequences. These elementary sequences were obtained through appropriate fragmentation of complex urban driving cycles.

Background

In 1999 the ARTEMIS project (Assessment and Reliability of Transport Emission Models and Inventory Systems) was defined within the 5th Framework Programme of the European Commission (Directorate General - Transport and Environment). The objective of the ARTEMIS project was to develop a harmonized emission model for all transport modes which aims to provide consistent emission estimates at the international, national and regional level. The ARTEMIS project consisted of 13 work packages of which WP 500 [1] dealt with 2-wheel vehicle emissions (powered two-wheelers). The ARTEMIS WP500 model is based on a large number of bag and online emission results both from the ARTEMIS WP500 main measurement programme and from several other studies conducted over the years. In all, about 2700 emission results were available to serve as a basis for emission modelling which is - compared to the results that were used for previous emission models - an enormous amount of data. Therefore, the main objective of ARTEMIS WP500 was to develop a set of representative emission results for CO, HC, NO_x and CO_2 of in-use powered two-wheelers that would be the basis for the emission factor model of this vehicle category. The approach to determine emission factors for motorcycles was copied from methodology applied to update the powered two-wheelers part of the Handbuch emission model (HBEFA) [2]. The measurement data (both online and bags) were employed to derive, by regression analysis, emission functions in terms of mass per time in relation to vehicle speed, for each powered two-wheeler vehicle category.

Experimental activity, methodologies and results

Vehicles

In these experimental tests two motorcycles were employed; their technical characteristics are reported in Table 1.

Table 1. Technical specifications of the tested motorcycles.

Vehicle	*Nexus Gilera 250 i.e.*	*Brutale Agusta 1000*
Engine principle	4-stroke	4-stroke
Cubic capacity [cm^3]	244	982
Compression ratio	11.0:1	12.2:1
Power system	electronic injection	electronic injection
Cooling system	liquid	liquid and oil
Max power [kw]	16.2 @ 8250 rpm	104 @ 10900 rpm
Maximum speed [km/h]	125	265
After-treatment system	catalytic converter	catalytic converter
Legislative category	Euro 3	Euro 3

Experimental apparatus

In the Istituto Motori emission laboratory the motorcycles were tested on a chassis dynamometer (AVL Zollner 20” - single roller) that enables simulation of vehicle weights from small mopeds up to heavy two-wheel vehicles (range 80-450 kg). This bench is designed to simulate the road load, including vehicle inertia, and to measure the exhaust emissions during dynamic cycles. Using this system, it is possible to carry out tests in constant speed mode, constant tractive force mode and constant acceleration mode. The chassis dynamometer was set by using the running resistance table according to the procedures laid down in Directive 97/24/EC. A variable speed cooling blower was positioned in front of the vehicles so as to direct the cooling air in a manner which simulates operating conditions. The blower speed was such that the linear velocity of the air at the blower outlet was within ±5 km/h of the corresponding roller speed.

During the tests the exhaust gases were diluted with purified ambient air by a Mixing Unit connected to a Constant Volume Sampling with Critical Flow Venturi (AVL CFV-CVS) unit. During the tests a continuous sample flow of the mixture filled one or more bags so that concentrations (average test values) of CO, HC, NO_X and CO_2 were determined. Average test values and continuous diluted emissions were measured with an exhaust gas analysis system (AVL AMA 4000), according to the procedures laid down in the Directive 97/24/EC. The exhaust pollutants were collected in the dilution tunnel and analyzed at 1Hz. The signals were corrected for the time delay respect to the speed. No other compensation (i.e. mixing dynamics) or signal treatment was applied [3].

Results on Type-Approval driving cycle

Three bags have been filled during the type approval driving cycle (UDC+EUDC): the first during the conditioning phase (the first two elementary urban modes), the second during the following four elementary modes, and the third during the EUDC mode. In this study, emission rates of CO, HC and NO_X were evaluated both during the ‘‘warming-up’’ phase and the ‘‘hot’’ phases of this cycle. The results are reported as mean values of UDC+EUDC five test repetitions to verify compliance with Euro 3 emission limits and to evaluate the variability of tests in the laboratory. In Table 2 and 3 mean experimental emission values of the motorcycles, expressed as mass emitted per kilometre travelled, are reported with the percentage variance coefficient (%CoV) for each phase and for the whole cycle, with the Euro3 emission standard limits.

These motorcycles comply with the Euro3 limits for CO, NO_X and HC. For the *Nexus*, CO emission factors in the whole cycle varied between 0.96 and 2.26 g/km, NO_X between 0.11 and 0.22 g/km, while HC ranged between 0.12 and 0.16 g/km. For the *Brutale*, CO values varied between 0.86 and 1.21 g/km, NO_X between 0.10 and 0.12 g/km, and HC between 0.06 and 0.09 g/km.

Table 2. *Nexus* emission factors, fuel consumption and their variability in UDC+EUDC phases, and Euro 3 standard limits.

UDC +EUDC phases	CO [g/km]	HC [g/km]	NOx [g/km]	CO_2 [g/km]	Fuel consumption [l/100km]
UDC_warm phase	2.27	0.57	0.07	105.66	4.65
%CoV UDC_warm phase	*6.02*	*8.62*	*5.72*	*1.14*	*1.23*
UDC_hot phase	0.80	0.06	0.02	87.95	3.74
%CoV UDC_hot phase	*13.21*	*9.81*	*20.78*	*0.78*	*0.81*
EUDC phase	1.55	0.06	0.19	81.75	3.53
%CoV EUDC phase	*71.57*	*16.46*	5.04	*0.96*	*1.94*
UDC +EUDC	**1.**43	**0.14**	**0.**12	87.37	3.77
Euro3 Standard limits	**2**	**0,3**	**0,15**		

Table 3. *Brutale* emission factors, fuel consumption and their variability in UDC+EUDC phases, and Euro 3 standard limits.

UDC +EUDC phases	CO [g/km]	HC [g/km]	NOx [g/km]	CO_2 [g/km]	Fuel consumption [l/100km]
UDC_warm phase	5,48	0,42	0,12	320,50	13,93
%CoV UDC_warm phase	*10.26*	*7.28*	*4.35*	*1.69*	*1.64*
UDC_hot phase	0,27	0,03	0,14	318,15	13,44
%CoV UDC_hot phase	*83.20*	*56.23*	*9.56*	*1.97*	*2.06*
EUDC phase	0,22	0,01	0,10	158,31	6,69
%CoV EUDC phase	*35.09*	*52.56*	*3.71*	*1.79*	*1.84*
UDC +EUDC	**1,06**	**0,08**	**0,11**	233,09	9,*91*
Euro3 Standard limits	**2**	**0,3**	**0,15**		

The most evident effect of the use of improved engines, in combination with catalytic technology, can be observed in the tested motorcycles in the differences between emissions during the cold-start and the hot phase of the type-approval driving cycle. For the *Brutale* under hot engine and fully operating catalyst, emission factors of CO and HC were even about 90÷95 % lower than those observed at warm engine conditions. During the cold start, the engine and catalytic converter are not at their optimal operating conditions: given the rich gasoline content in the air–fuel mixture, with the catalytic converter failing to reach the light-off temperature, the motorcycles emit higher concentrations of CO, HC and lubricant [4]. The effect of the start condition is certainly less strong on NO_X.

For newly sold motorcycles equipped with a catalytic converter and electronic mixture control, cold-start emissions, then, may represent an important proportion of total emissions, with an obvious repercussion on air quality. Since motorcycles are mostly driven in residential areas, the cold-start influence should be taken into account in assessing CO and HC emissions from this source.

For the *Nexus*, the variation relative to the mean values was always lower than 20%, with a higher variability (~72%) for CO emissions during the EUDC phase, as clearly shown in Fig.1, due to open loop operating conditions that occurred during the Tests III and IV. The values of experimental emissions and of real speed for the Test IV are shown in Fig. 2 where, unlike other cycles, it is clear that the instantaneous emission factor reaches a sudden and very high peak at around 0.6 g/s. In order to provide certain driving dynamics, characterized by high acceleration and speed, the electronic control, in fact, fixes a very rich air-fuel mixture, outside the optimum range of catalyst efficiency, eliminating the lambda sensor control, and thus implying a significant increase in emissions.

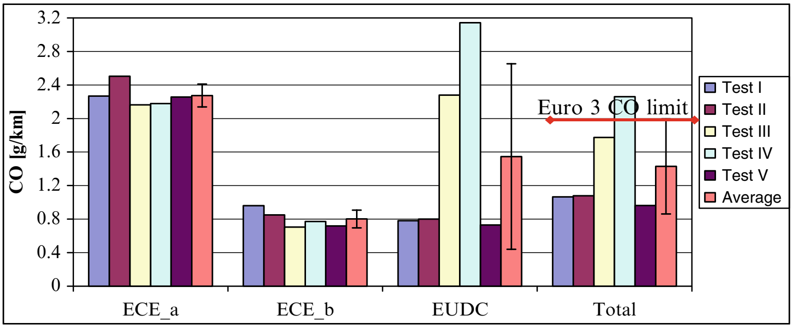

Fig. 1. *Nexus* CO emission factors in ECE+EUDC phases and their covariance.

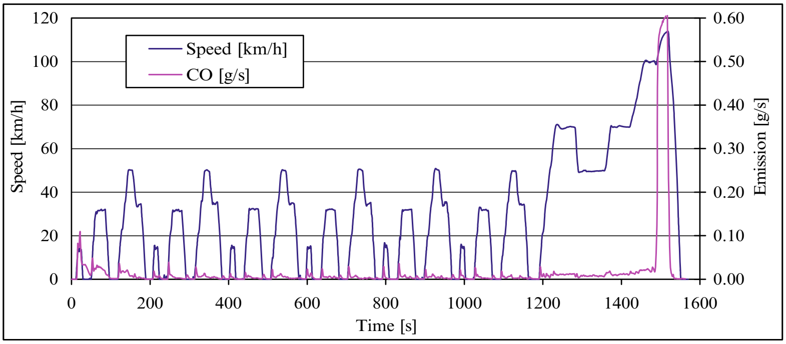

Fig. 2. *Nexus* CO emissions and real speed during ECE+EUDC test IV.

The percentage variance coefficients for the *Brutale* were always much higher than those measured with the *Nexus*, because of both the effects of open loop operating conditions and, mostly, the difficulties found in the repeatability of tests in laboratory with a high-power motorcycle.

Emissive behaviour under real urban conditions

Mean experimental emission values of regulated pollutants, expressed as mass emitted per kilometre travelled, are available (Table 2 and Table 3) for cold and hot phase of the Type-Approval driving cycle. However, such emission factors might not be sufficiently representative of real-world motorcycle riding, significantly different from the speed-time pattern of the test cycle on which they were measured. Besides, while there are many data published on emissions factors from motorcycles belonging to old standards approval, little is known about the emissions of new motorcycles in real-world situations. It is thus very interesting to assess the development of motorcycle emissions in these conditions.

For this purpose, additional measurements during real-world test cycles were therefore indispensable in order to evaluate motorcycle performance under real driving conditions. In this study three real urban driving cycles were considered: the WMTC, the Urban Cold, and the Road Cold. Experimental emission factors of each driving cycle considered (and their mean speed) are shown in Table 4 and in Table 5.

Table 4. *Nexus* emission factors for the driving cycles considered and data of ARTEMIS Project WP 500.

Test Cycle (mean speed [km/h])	CO [g/km]	HC [g/km]	NOx [g/km]	CO_2 [g/km]
ECE+EUDC (m.s.=29,7)	1,42	0,139	0,119	87,36
WMTC (m.s.=37,5)	0,73	0,110	0,102	70,04
Urban Cold (m.s.=19)	1,67	0,146	0,086	89,77
ECE 47 (m.s.=25,1)	0,89	0,151	0,044	80,97
ARTEMIS WP 500	4,04÷6,17	0,26÷0,46	0,094÷0,11	69,39÷102,19

Table 5. *Brutale* emission factors for the driving cycles considered and data of ARTEMIS Project WP 500.

Test Cycle (mean speed [km/h])	CO [g/km]	HC [g/km]	NOx [g/km]	CO_2 [g/km]
ECE+EUDC (m.s.=29,7)	1,06	0,08	0,11	233,09
WMTC (m.s.=37,5)	0,90	0,06	0,10	186,88
Road Cold (m.s.=41)	1,72	0,16	0,09	188,55
Urban Cold (m.s.=19)	2,01	0,20	0,08	314,99
ARTEMIS WP 500	0,89÷2,63	0,18÷0,47	0,049÷0,087	114,80÷188,6

It's evident that application of the Type-Approval driving cycle to motorcycles has the shortcoming of underestimating cycle dynamics. These tables also show the emission factors as calculated from the

ARTEMIS WP 500 emission functions, employing the coefficients of the *Nexus* and *Brutale* vehicular categories, and entering with speed values equal to the mean speeds of the considered cycles. The ARTEMIS WP 500 emission functions provide CO and HC emissions higher than those measured on tested motorcycles in real-urban situations, for the effect of improved engines, in combination with very efficient catalytic technology used on the vehicles.

After these elaborations, each urban driving cycle test was split into elementary kinematic sequences (speed-time profiles between two successive stops) under warmed-up engine conditions and analysed by employing the online emission results. The sequences so identified, that cover different situations present on urban road, were characterized with two kinematic parameters in order to better distinguish their driving behaviour [5]: the average speed and the mean product *v*a* of instantaneous speed and acceleration. This product, in fact, represents the tractive power per unit mass necessary to overcome the vehicle inertia, then it's a parameter strictly related to the emissions and fuel consumption.

In Fig. 3-6, CO and HC experimental emission factors, obtained during several repetitions of the elementary test sequences, are related to mean product *v*a*. Analysis of these results, and of the relevant trend lines, can indentify the driving sequences (then the driving patterns) that, for quite a similar level of average speeds, produce higher CO and HC emissions.

This statistical approach takes into account the differences in the emissions for test cycles with similar levels of average speed but relatively high differences with respect to other driving dynamics. It thus significantly improves vehicles emission assessment in comparison with the modelling approaches that are only based on average speed.

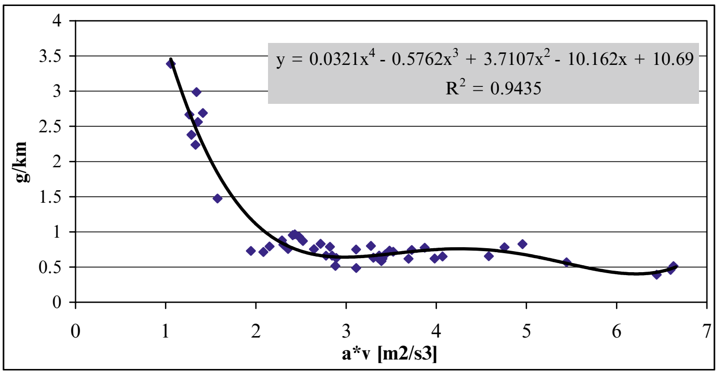

Fig. 3. *Nexus* CO hot emission factors measured against mean product "v*a" of each elementary test sequence.

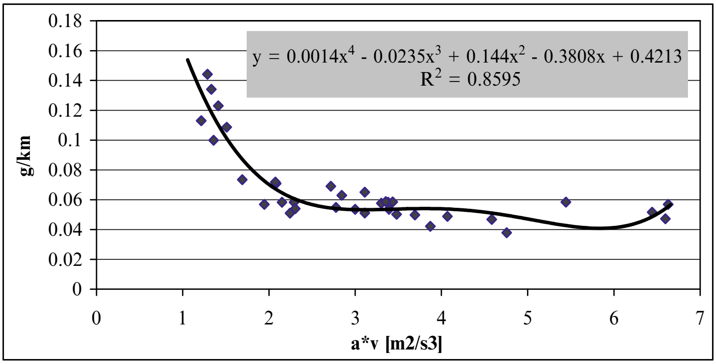

Fig. 4. *Nexus* HC hot emission factors measured against mean product "v*a" of each elementary test sequence.

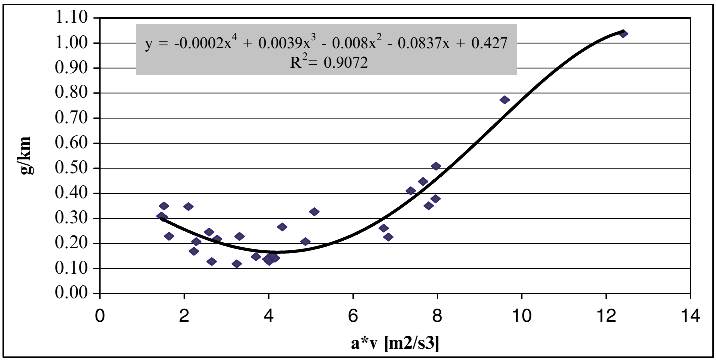

Fig. 5. *Brutale* CO hot emission factors measured against mean product "v*a" of each elementary test sequence.

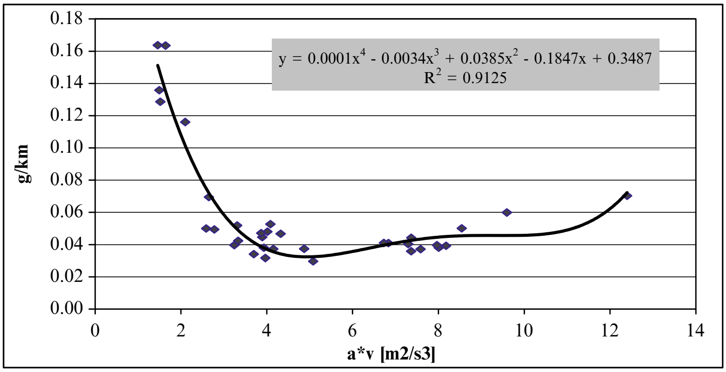

Fig. 6. *Brutale* HC hot emission factors measured against mean product "v*a" of each elementary test sequence.

Conclusion

While mean emission factors from motorcycles measured during the Type-Approval test cycles have been extensively documented, not much is known about the emissions of motorcycles in real-world situations. The main aim of the study was to collect information on the emissions of in-use motorcycles during real driving conditions, contributing significantly to extend knowledge of 2-wheel vehicles emission behaviour.

CO, HC and NO_X emissions were evaluated in the exhaust of two motorcycles belonging to the Euro 3 legislative category, equipped both with catalytic converter and with a displacement of 250 and 100 cm^3. Sampling was performed on a dynamometer bench on a type approval cycle and on real-world cycles both during cold-start and hot phases. In order to evaluate the performance of the motorcycles under real-world driving conditions, elementary kinematic sequences were identified (through suitable fragmentation of complex urban driving cycles) and analysed by employing the instantaneous emission results.

Besides, additional experimental investigations on other 2-wheel vehicles, belonging to other legislative categories (with different after-treatment systems and different technical specifications), will be indispensable. In this way, the output of the emission models will improve significantly and then the emission factors, so obtained, will be more representative of the general 2-wheel vehicles fleet under real driving conditions.

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Mobility of trace metals in urban atmospheric particulate matter from Beijing, China

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Abstract

Total suspended particles (TSP) and particulate matter ≤ 2.5 µm ($PM_{2.5}$) from Beijing, China, were studied for a time period of three years (2005–2008). Beside the total mass and the element concentrations, also the chemical fractionation and bioavailability of various elements was investigated by applying a four-step sequential extraction scheme.

Potential toxic metals like Zn, Cd, As, Pb, and Mn occurred to a high percentage in the mobile fractions and are, thus, especially harmful to the environment and exposed people due to an assumed high bioavailability. Arsenic, Pb, and Ni were even more mobile in the fine fraction ($PM_{2.5}$ samples), which is of special concern with regard to human health because smaller particles are considered to be more health relevant.

Introduction

Air pollution is of great concern in densely populated urban areas. Beijing, a Megacity with about 15 million inhabitants, accounts to the most polluted cities in the World. Sources for atmospheric particulate matter (APM) in Beijing are abundant, and particles originate from numerous anthropogenic activities as well as from geogenic sources.

It is generally accepted that APM is harmful for human health with regard to morbidity, mortality, respiratory and cardiovascular diseases [1,2]. Smaller particles are considered to be even more health relevant [3],

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because they can enter deep into the human respiratory system. Therefore, this study considers both total suspended particles (TSP), and additionally fine particles ≤ 2.5 µm ($PM_{2.5}$). Beside the total mass concentrations of APM, also the chemical composition and especially the concentration of potential toxic metals play a decisive role for the assessment of atmospheric pollution and its hazards to human health [4-6]. However, for the estimation of potential health effects, it is crucial to know not only the total amounts of toxic substances, but also their chemical speciations, because the bioavailability of elements depend on characteristic surface properties of atmospheric particle, the strength of the chemical bonds, and on the properties of solutions in contact with APM [7]. Chemically, elements mobilised by weak acids are much more available to organisms as elements, which can be mobilised only by very strong acids. Therefore, leaching procedures, which determine the mobility of metals with respect to different chemicals, provide a good indication of their bioavailability. In geochemical studies, sequential leaching procedures have been applied since several decades, especially for soil and sediment samples (e.g. summarised by [8] or [9]). For dust filter samples, however, the small amount of sampling material poses a special challenge and only few studies from different cities were published, which include Veszprém, Hungary [10], Karlsruhe, Germany [11], Sevilla, Spain [12], San Nicolás, Argentina [13], Santiago, Chile [14], Rome, Italy [15], and Tirupati, India [16]. In Beijing, no comparable study was carried out yet. Therefore, this study aimed at elucidating the metal concentrations and their bioavailability in a Chinese Megacity and, thus, to provide detailed knowledge for a sustainable urban planning.

Experimental

Sampling

Weekly samples were collected on quartz fibre filters. For TSP, a low-volume TSP-sampler (flow rate of 1 m^3/h) and for $PM_{2.5}$, a Mini-Volume-Sampler (flow rate of 200 L/h) was used. From July 2005 to August 2007, TSP samples were collected at site “TSP”, and starting from September 2007 the site was labeled “TSP-2” (Fig. 1) since it was moved to the northern part of the city. $PM_{2.5}$ samples were collected at both TSP sites and at one additional sampling site (labeled “CUG” in Fig. 1).

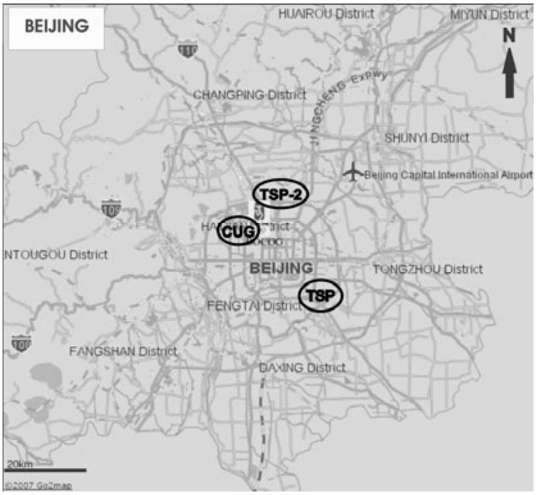

Fig. 1. Map of Beijing (source of the map: beijing2008.go2map.com) showing the location of the TSP sampling sites and the $PM_{2.5}$ sampling site at the Chinese University of Geosciences (CUG).

Analysis

Gravimetric analysis was carried out with a microbalance (Sartorius) for five times after at least 48 h equilibration with room conditions at 40 to 45% relative humidity. For each month of the sampling period, one TSP and one $PM_{2.5}$ filter sample, representing one complete sampling week, was selected for chemical sequential extraction (n=35, and n=32, respectively). A four-step leaching procedure according to [17] was applied (Table 1).

Table 1. Chemical fractions, mobility of each fraction, and reagents used (leaching procedure after [17]).

Fraction		Mobility	Reagent
f1	Water-soluble	Highly mobile	15 mL Milli-Q
f2	Bound to carbonates, oxides, reducible metals	Mobile	10 mL $NH_2OH*HCl$ (0.25M)
f3	Bound to organic matter, oxidisable metals and sulphides	Less mobile	7.5 mL H_2O_2 (30%) + 7.5 mL H_2O_2 (30%) + 15 mL NH_4AcO (2.5 M)
f4	Residual	Not mobile	10 mL (HNO_3:HCl:$HClO_4$) (6:2:5)

Elemental concentrations of 18 elements (Al, As, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, Rb, Sr, Ti, V, Zn) were analysed for each of the four leaching steps by high-resolution inductively coupled plasma mass spectrometry (HR-ICP-MS, Axiom, VG Elemental).

Results and discussion

TSP mass concentrations for the selected 35 weeks from July 2005 till May 2008 at site TSP and TSP-2 varied from 170 to 630 µg/m^3 with a median concentration of 370 µg/m^3. Mass and metal concentrations for TSP samples of the whole period, including the weeks not selected for sequential extraction, are reported in [18] and those for $PM_{2.5}$ samples in [19]. The analysis of water-soluble ions was discussed in [20]. This paper focuses on the eco-toxic elements As, Cd, Cr, Cu, Mn, Ni, Pb, and Zn.

Mobility of highly toxic elements from predominantly anthropogenic sources

For elements, which are potentially toxic for human health, the percentage in the mobile fractions f1 and f2 is of special interest because it determines the bioavailability of those metals. For TSP (Fig. 2) as well as $PM_{2.5}$ (Fig. 3) samples from Beijing, the mobility decreases in the order Zn > Cd > As > Pb > Mn > Cu > Ni > Cr.

All these elements typically originate partly or predominantly from anthropogenic sources. Enrichment factors (EFs) were calculated[3] to determine the anthropogenic influence. EFs were highest for Cd (EF = 2670) and decrease in the order As (EF = 730) > Pb (EF = 590) > Zn (EF = 440) > Cu (EF = 120). Mn, Ni, and Cr had only low EFs (EFs of 10, 5, and 3, respectively). EFs of $PM_{2.5}$ were even higher than of TSP samples, but showed the same sequence. That indicates that the influence of anthropogenic sources is especially high for the fine particles.

[3] EFs were calculated referring to Ti according to the equation:
$EF_{Ti} = (Element/Ti)_{APM}/(Element/Ti)_{crust}$ (1.1)
Average crustal concentrations were taken from [21].

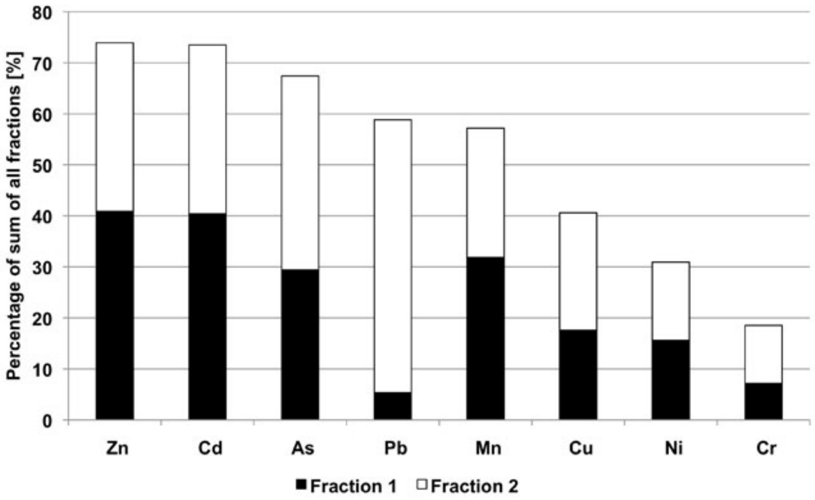

Fig. 2. Median element concentrations of TSP samples (n=35) in percentage for leaching steps f1+f2 ordered by their percentage in the mobile fractions.

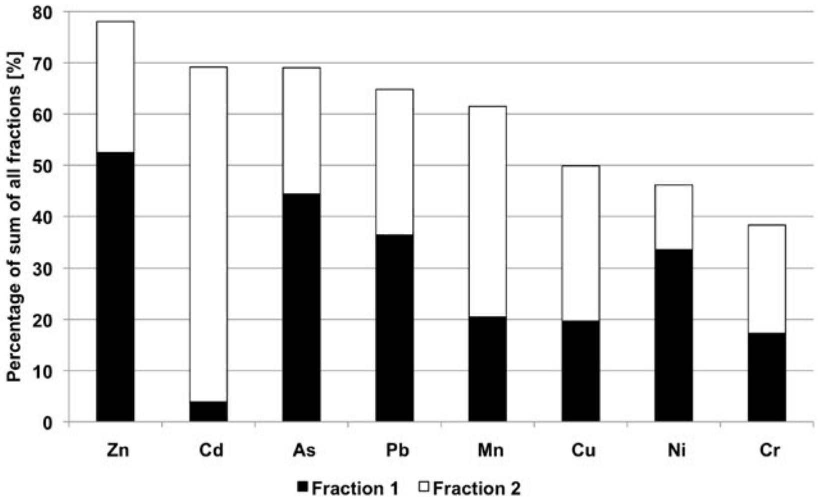

Fig. 3. Median element concentrations of $PM_{2.5}$ samples (n=32) in percentage for leaching steps f1+f2 ordered by their percentage in the mobile fractions.

Seasonal variations of toxic metal concentrations and their mobility

Metal concentrations showed pronounced seasonal variations. The element concentrations for TSP (sum of all four fractions) are illustrated for the distinct seasons in Fig. 4. For all elements considered, summer was the

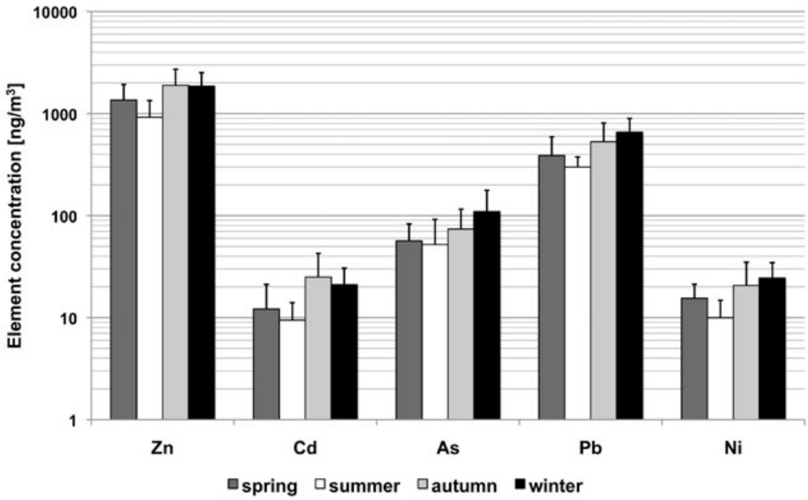

Fig. 4. Element concentrations (sum of all four fractions) for TSP samples (n=35) for the distinct seasons.

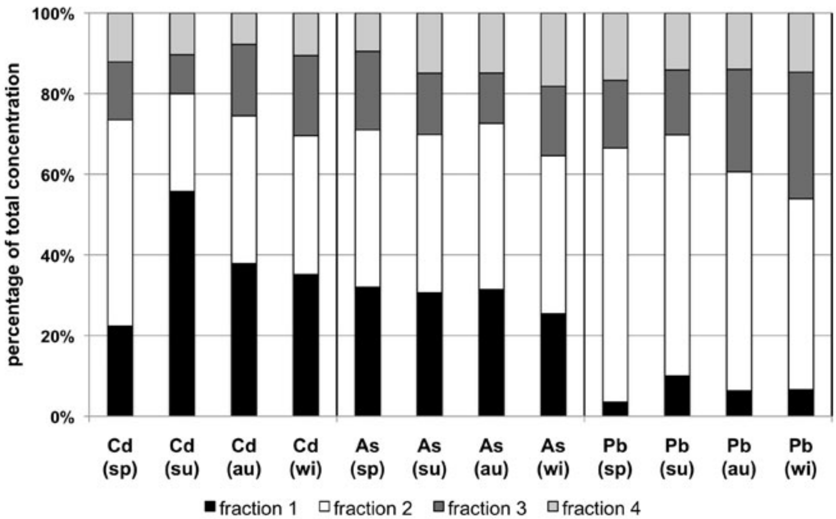

Fig. 5. Percentage of each fraction of total element concentration of TSP samples (n=35) for the distinct season (sp: spring, su: summer, au: autumn, wi: winter).

season with lowest concentration. Arsenic, Pb, and Ni had their maximum concentration in winter, while Zn and Cd concentrations were highest in autumn. Furthermore, also the relative abundance in the mobile fractions varied during the annual course (Fig. 5). For Cd and Pb, summer was the season with the highest percentage in the mobile fraction and spring had the lowest relative abundance in the mobile fraction.

Toxicity of metals in fine particles ($PM_{2.5}$) compared to total particulate matter (TSP)

Some of the toxic metals, which were highly mobile in TSP from Beijing, had even higher percentages in the mobile fractions in $PM_{2.5}$ samples. This was the case for As, Pb, and Ni (Fig. 6). Therefore, these elements are of special concern with regard to negative health effects because the bioavailable fraction in $PM_{2.5}$ was higher and those particles can enter deeper in the human respiratory system. Therefore, special attention has to be paid to those elements when planning mitigation measures in Beijing.

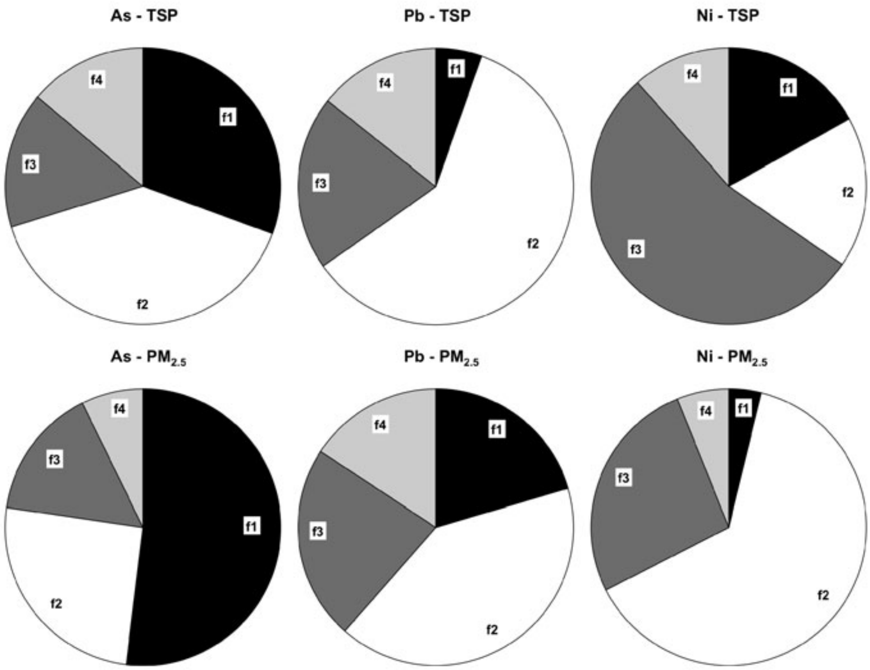

Fig. 6. Percentage of each fraction of total As, Pb, and Ni concentrations of TSP samples (n=35, upper row) and $PM_{2.5}$ samples (n=32, lower row) for each of the four fractions.

Conclusions

Strong seasonal variations of APM in Beijing were observed. The seasonal trend for Zn, Cd, As, Pb, and Ni showed lowest concentrations in summer, which is also the time with lowest APM mass concentrations caused by wet deposition due to the high precipitation. In spring, potential bioavailable metal loads had lower concentrations than in autumn or summer and also a lower relative abundance in the mobile fractions. This means that unless spring has high concentrations of APM mass, the air pollution is less critical with regard to toxic metal concentrations. Winter, on the contrary, is the season most problematic regarding potential negative health effects because of maximum total concentrations of some toxic metals, as well as a high bioavailability of toxic elements, such as As, Pb, or Ni.

The study showed that a cause of concern is the high mobility and, thus, assumed bioavailability of toxic metals like Zn, Cd, Mn, As, Cu and Pb in atmospheric particles in Beijing. Therefore, health studies should consider especially the mobile and bioavailable fraction of those elements. In general, the approach to use sequential extraction procedures for APM characterisation, which was applied for the first time for atmospheric particles in a Chinese Mega-city, proved to be a good tool in order to gain important additional information needed for a future sustainable urban planning.

Acknowledgments

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Health risk from air pollutants, an epidemic in Western Java Indonesia

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Abstract

Complaints from irritation of skin, cough, bad odor, and nausea was suffered by 100% of a village (KS) inhabitant, in Western Java. The location of KS being upon a hill, just across factories along the coast. The KS inhabitants blame the factories for emitting gaseous pollutants, which cause them to suffer the mentioned symptoms. This study was done to investigate the possible relationship of health effects and air pollutants and to estimate the health risk. It designed as a retrospective case-control study and the health risk measured as odd ratio. The results then verified by the air quality study. KS inhabitants who were exposed more likely suffered the health effects as compared to KC who were not exposed, but the concentrations of pollutants were all below the standard except PM. In conclusion, the health risk from air pollutants in KS as the exposed area were much higher than the unexposed area, and the study presents premature evidence of health effect and air pollutant exposure association particularly PM.

Introduction

Complaints from irritation of skin and eyes, cough, bad odor and nausea were experienced by nearly 100% of KS inhabitants, located in Gerem District, Cilegon. Cilegon has been the home of heavy industry and a major coastal industrial city in Banten province, Indonesia. The location of KS being upon hill, just across the industrial site for emitting gaseous pollutants. The inhabitants blamed these industries which caused them to suffered the mentioned symptoms.

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Industries are major source of air pollution, which emit air pollutants, namely nitrogen oxides (NO_x), volatile organic compounds (VOCs), carbon monoxide (CO), carbon dioxide (CO_2), sulfur dioxide (SO_2) and particulate matter (PM). Exposure to ambient air pollutants influenced by industrial emissions has been linked to a number of different health outcomes, starting from modest transient changes in the respiratory tract and impaired pulmonary function and continuing to restricted activity/reduced performance, visits to the hospital emergency department, admission to hospital and death [1].

Therefore, an objective study was needed to evaluate whether there was a correlation between air pollutants exposure in the ambient air and the health effects. This paper present further investigation of the health risk from air pollution exposure in KS. In this study, a retrospective epidemiological study was clearly the first step in assessing the adverse health effects that could result from contact with environmental pollutants. In addition, local air quality study was conducted to verify the results of the epidemiological study, since the health outcome is directly related to air concentration of the pollutants [2].

Monitoring of ambient air quality could provide data to ascertain air pollution concentrations in a specific area [3]. However, air quality monitoring system in this area of study was not available because of economically unfeasibility. Hence, air quality model was used in this study to estimate ambient concentrations as a crucial step in assessing potential risk to human health due to inhalation of ambient air [3].

Methods

Area of Study

KS is a village in Gerem District, Cilegon. Cilegon is located in the western of Java island, Banten province, Indonesia (Fig. 1). According to the geographical location, Cilegon have tropical climate, consist of the rainy season (January to May) with average rainfall is 50-150 mm and dry season (June to December) with average rainfall is 100-150 mm. Daily temperature is about 31.4°C and 25.4°C at night time, and average wind speed range to 3.7 m/sec until 4.8 m/sec [4].

Fig. 1. Banten Povince Site Map.

Epidemiologic Study

The epidemiologic study was carried out to estimate the health risk from air pollutants in the area of study. This study was conducted after the symptoms receded, the people at the time of study were healthy and no longer suffer any symptoms. Hence, the epidemiologic study design was a retrospective case-control study. Basicly, there were two comparable groups which consist of an exposed group (defined as the cases) and unexposed group (defined as the controls). The two groups should similar particularly in social-economic level, so that the differences found between the two groups only in terms of exposure to environmental pollutants.

The data were obtained from questionnaire and interviewed face to face, as well as observation of exposure status in subject without assignment and without following individual subjects for change in disease status. The collected data be composed of variables related to socio-economic and health status. The obtained data then would be summarized in contingency matrix 2-by-2 as illustrated in Table 1 [5].

Table 1. Matrix 2-by-2 of Case-Control Study.

Variables	Case	Control
Exposed Factor	a	b
Unexposed Factor	c	d

In this study, the cases were KS inhabitants who exposed by air pollution and experienced the health symptoms. The selected control were KC village inhabitants, which similar in social-economic level and assumed not exposed by the ambient air which influenced by the industrial emissions since it was located just behind the hill where KS was.

Sampling was based on purposive sampling. The subjects consist of 72 cases and 62 controls, determined by Slovin Equation as follows [6,7]:

$$n = \frac{N}{\left(1 + N\left(e^2\right)\right)} \tag{1}$$

Where:

N = Population number

n = Samples number

e = Error tolerance = 1 –confidence interval (desired)

Risk is classically defined as the probability that a specific outcome or disease will develop within a stated period of time [8]. In this study, the health risk was quantified by Odds Ratio (OR), which estimated indirectly by comparing disease frequency for the exposed population and unexposed population and calculated as the number of exposed individuals divided by the number of unexposed individuals in each group [9]. OR defined how much more likely it is that someone who is exposed to the factor under study will develop the outcome as compared to someone who is not exposed [10]. Based on Table 1, mathematical equation for calculating OR as follows [10]:

$$OR = \frac{a/b}{c/d} = \frac{a.d}{b.c} \tag{2}$$

Air Quality Study

The Gaussian Dispersion Model was applied to predict air concentration at receptors. The model assumes any release from source disperse in a steady-state manner from the time of release until the time it reaches a receptor [2]. It's for describing the mixture of air pollutants in the atmosphere at the vertical and crosswind direction of the source as a result of turbulence [11]. The model calculates the concentration X at any receptor location (X, Y, Z) from a continous source with an effective stack height [12].

The input for the Gaussian Dispersion Model consists of emission information, meteorological data and receptor information. The predicted pollutants consists of HC, NO_x, CO, SO_2 and PM. Emission information were obtained directly from single stack measurements and the results were evaluated against the emission standard (Table 2). Other parameter setting for the model were exhaust gas velocity (4.7 m/sec), exhaust gas temperature (217°C), inner stack diameter (1.20 m), stack height (40 m) and pressure (75.979 mmHg).

Table 2. Source Parameter Used in Modeling.

No	Parameter	Unit	Magnitude	Emission Standard*
1	HC as CH_4	mg/Nm^3	18	35
2	CO	mg/Nm^3	458	100
3	NO_2	mg/Nm^3	75	300
4	SO_2	mg/Nm^3	80	250
5	PM	mg/Nm^3	259	50

Meteorology parameter, i.e wind speed and directions, ambient temperature, ambient pressure, and atmospheric conditions were obtained from local Meteorology Agency for July – December 2002 period. The wind direction and speed data were classified according to speed and direction, then summarized in the form of a polar diagram called windrose. It shows the direction from which the wind was blowing, the length of various segments of the spokes show the percent of time the wind was of the designated speed [12].

A receptor grid for off-site receptor was set – up using Cartesian grid with a 500 m grid spacing out to distance of 2.0 km from the center of the industrial site. The distance from the stacks to the KS was ± 1,5 km and to the KC was ± 2,0 km .

All modeling output was collected in plot files that contained geographical coordinates (i.e., X and Y coordinates) for each receptor, then applied in Microsoft Excell. The predicted air concentrations were expressed as $\mu g/m^3$. The result was illustrated as a contour map of concentration distribution of pollutant released from a point source called isopleths [12]. The uncertainty of the modeling result was assessed by direct comparison with the measurements results. The measured pollutants comprise HC, NO_x, CO, SO_2 and PM as well.

Results and Discussions

Epidemiologic Study

For this study, 72 cases and 62 controls were interviewed face-to-face. Table 3 shows descriptive characteristics of the cases and the controls based on questionnaires and interview. The table shows that once unpleasent odor appeared, percentage of exposed population whom suffering eye irritation were 100%; breath shortness were 29%; and skin irritation were 4%; and cough 8%. In contrast, 100% of unexposed population were not suffering any symptoms except cough for almost 26%.

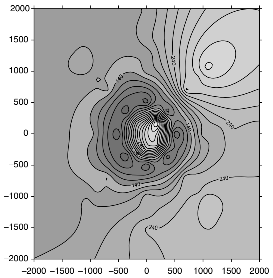
Table 3. Descriptive Analysis of Controls and Cases Characteristics.

Variables	Cases (total = 72)	Controls (total = 62)
Reside time periods:		
0 – 5 year	9	10
6 – 10 year	5	7
11 – 15 year	7	8
>15 year	51	37
Air comfortness:		
Yes	4	62
No	68	0
Time of unpleasent odor:		
morning	12	0
day time	64	0
afternoon	25	0
night	12	0
Health complaints:		
Eye irritation	72	0
Breath shortness	21	0
Skin irritation	3	0
Cough	6	16

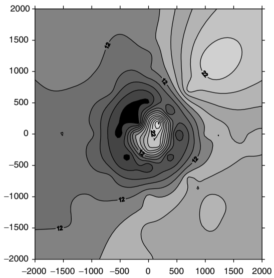
The obtained data then were summarized in contingency matrix 2-by-2 as presented in Table 4 to calculate the OR. The OR was estimated indirectly by comparing disease frequency for the exposed population and unexposed population. Derived from the calculation result, the estimated OR for eye irritation; cough; breath shortness; and skin irritation sequentially were 17712; 3.14; 173.65; and 128.35. Thus, it is clearly that the risk of illness in the case area (KS) was very high compared to the control area (KC).

Table 4. Summary of Health Risk Estimation.

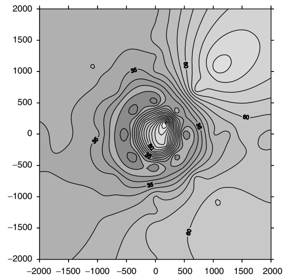
Variables	Cases		Controls		OR
Cough:					
Yes	6	8.3%	16	25.8%	0.26
No	66	91.7%	46	74.2%	
Skin irritation:					
Yes	3	4.2%	0.5	0.8%	5.35
No	69	95.8%	61.5	99.2%	
Breath shortness:					
Yes	21	29.2%	0.5	0 8%	50.65
No	51	70.8%	61.5	99.2%	
Eye irritation:					
Yes	72	99.3%	0.5	0.8%	17712
No	0.5	0.7%	61.5	99.2%	



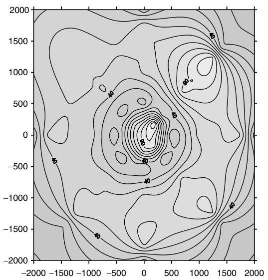
(a) PM



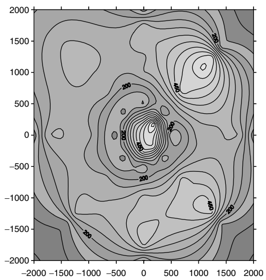
(b) HC



(c) NO_2



(d) SO_2



(e) CO

Fig. 2. The Prediction of Air Pollutants Dispersion.

Air Quality Study

The model was applied to predict the total period average air concentrations at receptors. Result of the dispersion model was contour map of concentration distribution of all pollutants released from the point source called isopleths, as shown in Figure 2 above. From the figure, the concentrations of pollutants were decrease as it travels along the site. The highest are at the centre and at a distance of 700m - 2000m towards the Northeast and the lowest concentration were outskirts of the map.

Table 5 summarizes the predicted and measured concentration of all pollutants namely SO_2, NO_2, CO, HC and PM at both sites with a distance from the source 1.5 km to KS and 2.0 km to KC. The predicted and measured concentrations were all below ambient standards, except for predicted and measured concentration of PM which exceeded the standard. Overall, the predicted air pollutants concentration were much higher than the measured concentration either in KS or KC.

Table 5. Predicted and Measured Concentration of Air Pollutants.

Parameter	Ambient Air Concentration (μg/Nm^3)				Ambient Air Quality Standard* (μg/Nm^3)
	The Cases Area (KS)		The Control Area (KC)		
	Predicted	Measured	Predicted	Measured	
SO_2	142.12	0	42.34	0	365
NO_2	99.97	0.002	88.02	0.001	150
CO	936.36	1.261	387.99	0.890	10,000
HC	37.79	28.49	21.29	28.81	160
PM	356.24	574.23	331.99	215.38	230

*Ambient Air Quality Standard Based on PPRI No.41/1999 about Air Pollution Control [15].

The predicted and measured concentrations data may provide some insight into the potential health burden associated with air pollutants exposure. Eyes and skin irritation and breath shortness showed there were

Fig. 3. Coconut Trees Condition in KS.

irritant substances in the ambient air [15]. In addition, the leaves of coconut trees within the KS were brown (Figure 3). But the concentrations of all gaseous pollutants were below ambient air standard and in contrast the concentrations of PM were exceeded the ambient air standard. There was likely that the health effect suffered by inhabitants in the exposed area were caused by irritants gas which absorbed in PM.

Conclusion

The health risk from air pollutants in the exposed area were much higher than the unexposed area. The environmental epidemiologic study presents premature evidence that there was association between health effect and air pollutant exposure particularly PM. Further research would be needed to investigate the atmospheric PM in the area of study. It was recommended that the factories on one hand, should reduce their emissions. On the other hand, an appropriate continuous ambient air pollution monitoring device should be installed to be able to evaluate better the ambient air and provide some warning of impending health problem. Further environmental epidemiologic research would be needed to accurately measure the health risk from air pollutants.

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URBAN WATERS

Emission control strategies for short-chain chloroparaffins in two semi-hypothetical case cities

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Abstract

The short-chain chloroparaffins (SCCP), ($C_{10\text{-}13}$ chloroalkanes) are identified in the European Water Framework Directive, as priority hazardous substances. Within the ScorePP project, the aim is to develop emission control strategies that can be employed to reduce emissions from urban areas into receiving waters. Six different scenarios for mitigating SCCP emissions in two different semi-hypothetical case cities representing eastern inland and northern coastal conditions have been evaluated. The analysis, associated with scenario uncertainty, indicates that the EU legislation, Best Available Technologies (BAT) and stormwater/CSO management were the most favorable in reducing emissions into the environment.

Introduction

The short-chain chloroparaffins (SCCP, CAS registry number 85535-84-8) have been identified as a priority hazardous substance by the European Union [1] and as such have been targeted to be phased-out [2] of discharges to surface waters. The SCCP group consists of an anthropogenic mixture of $C_{10\text{-}13}$ chloroalkanes with varying chlorine content which are used as plasticizers, flame retardants and lubricants in various articles such as metal-working fluids; textiles and rubbers; paints, coating and sealants/adhesives; as well as in the leather and PVC industries [3].

Within the EU funded project "Source Control Options for Reducing Emissions of Priority Pollutants" (ScorePP), an important aim has been to develop comprehensive and appropriate emission control strategies (ECSs) that relevant stakeholders (such as governments, water utilities

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and industries) can employ to reduce emissions from urban areas into the receiving water environment. The ECSs are based on a "near to the source" approach limiting releases at the sources and "end-of-pipe treatment" to attenuate already released substances. This dual approach is based on findings within the ScorePP project revealing that all substances can neither be mitigated at the source [4] nor removed with conventional treatment to a sufficient extent [5]. The aim of the presented work was to evaluate the efficiencies of different ECSs for SCCP in two semi-hypothetical case city archetypes (SHCCA).

Approach

The semi-hypothetical case city archetypes

Working with selected case cities may hamper or bias the outcome of the study (e.g. due to the lack of key data, differing climatic or socio-economic conditions), hence the concept of SHCCA was developed, virtual cities which contain both generic and specific information to enhance robustness diversity, and facilitate completeness [6].

Table 1. Business-as-usual city indicators and characteristics.

City indicators	EI	NC
Population (mio.)	1.2	0.51
Population growth (5 y; %)	2	0.5
City area (km^2)	500	450
Precipitation (mm/y)	530	650
Receiving water flow (m^3/s)	700	50
Industries		
-heavily polluting	70	30
-moderately polluting	279	119
Wastewater		
-treatment type	Secondary	Secondary
-dwellings connected (%)	90	99
-volume to overflows (%)	18	10
Stormwater		
-in combined sewer (%)	50	90
-in separate sewer (%)	50	10
--stormwater flow to BMPs (%)	20	20
--surface area for BMPs (m^2)	2500	721

Two SHCCA have been defined and are identified (in Table 1) as an Eastern European Inland (EI) city and a Northern European Coastal (NC) city. EI has a growing economy, an increasing population, many heavy industries and is situated by a large river. NC is a city with a consumer-oriented industry and is located on the coast by a brackish water body and at the mouth of a medium sized river.

Emission Control Strategies

Within the ScorePP project, six ECSs (Table 2) have been developed and used to evaluate a number of different priority substances. Since different substances have different properties and source patterns, different source control measures and different treatment processes can be the most feasible and appropriate for them. Therefore, specific ECS scenarios have been developed for each substance defining which source control and treatment options have been included. ECS1 is a business-as-usual strategy, i.e. the future scenario will not involve any deliberate changes, whereas ECS2 assumes that all relevant existing EU directives will be fully implemented. ECS3-6 build on ECS2 with the addition of voluntary options (ECS3), industrial onsite treatment by Best Available Technologies (BAT) (ECS4), stormwater and combined sewer overflow (CSO) management (ECS5) and advanced wastewater treatment plant (WWTP) (ECS6) (Table 2).

The ECSs are effective on different levels in the urban catchment: pre-application (source control e.g. substitution, legislation, and voluntary initiatives); pre-release (treatment before emissions to the environment, e.g. WWTP and CSO treatment); and post-release (attenuation after release into the environment such as stormwater Best Management Practices (BMPs) and dredging of contaminated sediments) (Figure 1). This means that various ECSs address the relevant actions of households, municipalities, governments and industries. For example ECS3, involves the introduction of voluntary initiatives aimed at ceasing the use of or substituting for articles containing SCCP. ECS3 Education campaign can promote the use of eco-labeled articles to industries, municipalities and households. ESC5 stormwater BMPs can be used on household properties, industrial sites, or public open spaces.

Table 2. Definition of ECS1-6 for SCCP. Percentages indicate efficiency of release reduction or removal.

ECS number	Description
ECS1: Business-as-usual	Not all dwellings connected to the sewer system, no BAT, composition based on article Material Safety Datasheets (MSDS) (e.g. 4-20%), limited management of stormwater.
ECS2: Full implementation of relevant EU directives	Generic BAT applied to heavily polluting industries by adsorption [5, 7], efficiency 59%, 100% of dwellings connected to a WWTP, article composition based on legislation, e.g. Directive 2002/45/EC with leather working and leather fat (article content max 1%).
ECS3: Household and municipality participation by voluntary options	ECS2 implemented. Article composition voluntary changed for paint and putty to max 1%. Industrial initiative to substitute SCCP by 50%. Choice of eco-labeled products (rubber, textiles and sealants) reduces the release by 25%. Education campaigns for painters and paint removers reduce societal releases by 50% and collection and recycling of PVC decreases the related emission by 50%. Loosely based on [4]*
ECS4: Industrial treatment by BAT and beyond	ECS2 implemented. Generic BAT (adsorption) applied to both heavily and moderately polluting industries. Heavily polluting industries applies also advanced oxidation processes (AOP; efficiency 24% [8]).
ECS5: Stormwater treatment and Combined Sewer Overflow (CSO) volume reduction and treatment	ECS2 implemented. 75% of the stormwater is treated in the most feasible BMPs; infiltration (25%) and retention ponds (75%) [9]. The volume of CSOs is halved due to storage in tanks and the remaining CSOs are treated by adsorption [5] by coagulation/flocculation (59%)
ECS6: Advanced WWTP end-of-pipe processes	ECS2 implemented. WWTP treatment is enhanced by coagulation/flocculation in dual tanks by 93% [10], the effluent is polished by AOP (24% [7]) and the sludge is subjected to anaerobic digestion (dechlorination, 35% [11]).

*Assumed data in tiers of 25/50/75

Substance flow analysis

The release, flow and environmental emissions of SCCP in each SHCCA have been evaluated though a substance flow analysis (SFA) approach. SFA is “a tool for analyzing the societal metabolism of substances” [12], i.e. for evaluating how substances are used and disposed of. The SFAs have been based on data extracted from an Emission String database (ES-DB) based on a comprehensive literature search [13], additional data from the relevant EU risk assessment reports (EU-RAR) [3]. The ES data is in the form of release per unit with the units originating from the definition of the SHCCA. For example, the EU data were extrapolated according to city size (e.g. population) or industry activities (Standard nomenclature for economic activities, NACE codes). The releases into different compartments can be seen in Table 3 and originate from the ES-DB and EU RAR [3,13]. The releases were divided into discharges to surface waters (if applicable), to water indirect (i.e. stormwater or wastewater systems), to air and to urban surfaces (mainly urban soils).

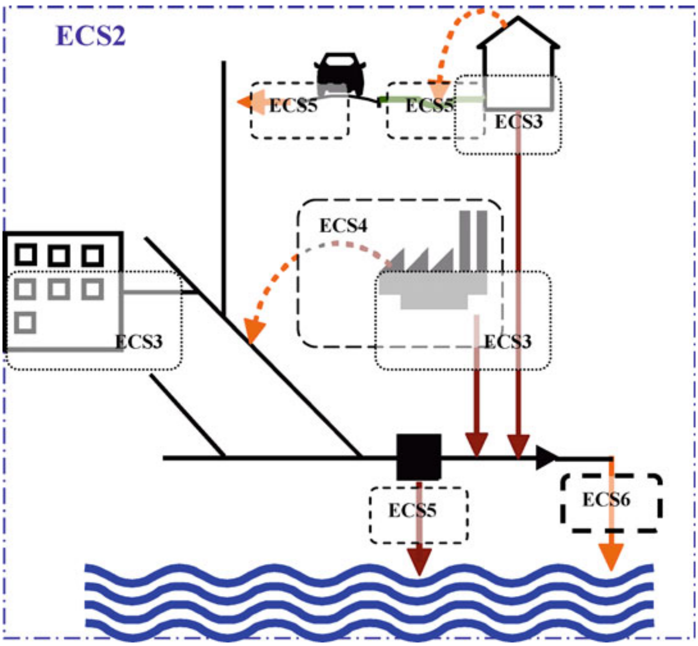

Fig. 1. Illustration of the location of the ECSs in the urban context, based on Eriksson *et al*. [6].

Table 3. Distribution of releases (%) based on literature data [3,13] or expert judgment (when independent information is not available).

ES-ID [13]	Description of ES	Storm-water	Waste-water	Air	Urban surfaces
10931	production of basic organic chemicals		97		3
10954	use in metal working -formulation		100		
10958	use in metal working -formulation		100		
10984	use in metal working and extreme pressure lubricating fluids		100		
10986	use in metal working and extreme pressure lubricating fluids		100		
10995	use in metal working and extreme pressure lubricating fluids		100		
10996	use as flame retardant in rubber formulations	25	75		
10997	leather formulations		99.5	0.5	
10999	leather formulations		100		
11001	Formulations		100		
11001	leather formulations		100		
	paint production		75	25	
	painted surfaces	25		75	
	Painting	50		50	
	putty in society	25	50	25	
	PVC in society	25	25	50	
	rubber in society	25	25	25	25
	sealants/adhesives	25	25	25	25

Results and discussion

City EI has several industrial sources for SCCP (SCCP production, leather and metal works) whereas city NC has only a few minor industrial sources (manufacturing of leather commodities) (Table 4). Articles containing SCCP such as textiles, putty, paint, rubber and plastic are considered for both cities as urban sources as they release SCCP during application, during use or when being discarded.

Loads and releases

For EI the minimum, maximum and average predicted loads are presented in Table 4 based on the ranges of article compositions (e.g. MSDS), the ranges in use applications and the ranges in releases. If no ranges have been found, the same value is presented in all three columns. For NC only the average values are shown as the ranges are relatively small compared to EI as fewer industrial sources are present.

Table 4. Total releases from industrial sources and emission from urban sources of SCCP in EI and NC (kg/year).

ES-ID [13]	Description of ES	Min. EI	Average	Max.	Average NC
10931	production of basic organic chemicals	300	4650	9000	
10954	use in metal working-formulation	39	39	39	
10958	use in metal working-formulation	39	39	39	
10984	use in metal working and extreme pressure lubricating fluids				77
10986	use in metal working and extreme pressure lubricating fluids	12	582	1152	7.7
10995	use in metal working and extreme pressure lubricating fluids	12	582	1152	
10996	use as flame retardant in rubber formulations	1.3	1.3	1.3	
10997	leather formulations	5.3	9.3	13	
10999	leather formulations	5.3	9.3	13	2.4
11001	leather formulations	5.3	9.3	13	4.8
[3,14]	paint production	10	10	10	4.3
[3,14]	painted surfaces	97	97	97	43
[3,14]	painting	2.5	2.5	2.5	1.1
[3,14]	putty in society	8.7	8.7	8.7	26
[3,14]	PVC in society	2.2	2.2	2.2	22
[3,14]	rubber in society	23	23	23	2.1
[3,14]	sealant/adhesive				0.1
	Total	**562**	**6064**	**11566**	**189**

Distribution between urban and environmental compartments

The releases from the different sources have been divided into storm-water, wastewater, air and urban surface fractions (Table 3) but re-distribution may occur following release. For example, in stormwater treated within a BMP, the SCCP could sorb to sediment or infiltrate, volatilize to air, be degraded (by removal of parent substance) or be emitted to surface water via the BMP effluent. In Figures 2 and 3 the emissions directly to surface water have been added to the load estimated in the effluents from BMPs, WWTPs and CSOs to provide the total load to surface water. Similarly, direct air emissions have been added to the loads volatilized from BMPs,

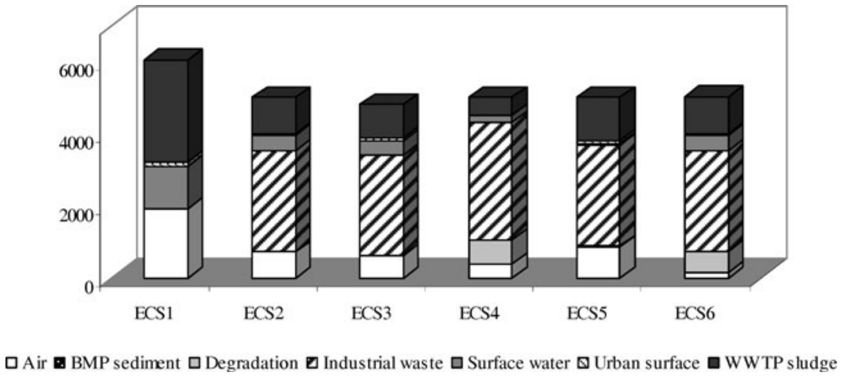

Fig. 2. Emissions of SCCP in EI (kg/year) using a SFA approach.

WWTPs, and CSOs during treatment. As the SCCP substances have medium to high volatility [5] their distribution to air is substantial and for some scenarios, primarily ECS4 and 6, degradation can also be seen to make up a significant part of the mass balance. Industrial onsite treatments generate industrial waste either as sludge or solid waste.

In EI, the only two ECSs that substantially affected the total load being emitted were ECS2 and 4 as the legislation and industrial BAT affect the article composition and treatment onsite, and as the apparent reductions associated with ECS3, 5 and 6 are mainly due to the impact of ECS2. ECS5 was found to yield the lowest emission of SCCP to surface waters, and ECS4 and 6 were found to yield the highest total degradations.

In NC, ECS2 was again effective in reducing the total load being emitted. As very few industrial sources were present in NC, ECS3 consisted of the voluntary reduction in SCCP use by industries and education campaigns to mitigate SCCP during the article use or when the article was treated as waste, however, these appeared to have little impact on the emission of total SCCP loads. As for EI, the lowest loads to surface water in NC are associated with the use of ECS5 and the highest amount of degradation seen for ECS6. The least favorable ECS for both SHCCA was ECS1, i.e., business-as-usual and carrying on without taking any action to limit the release of SCCP.

The total SCCP loads released for EI range from a minimum of 0.56 ton to a maximum of 11.6 tons (Table 4). For NC, the average value (0.189 tons) is considerably lower than the corresponding value predicted for EI (6.06 tons). The calculated loads are fairly good agreement with a SFA study performed for Stockholm (Sweden) [14]. The size of

Stockholm (380 km^2) and the number of inhabitants (1.2 mio) are in the same order as EI and NC, and therefore the total load is in the same order of magnitude. The implementation of the SFA is of course associated with various kinds of uncertainty; statistical uncertainty as illustrated by the concentrations of SCCP varying from article to article as stated in the MSDS; scenario uncertainty associated with values allocated to the SHCCA, ECS and NACE codes. There will also be some level of ignorance as relevant NACE codes may have been overlooked. One flaw is that the release that occurs to air may be wrongly addressed and no data on distribution of SCCP in air/rain could be found.

In the SFA study for Stockholm an estimated annual use of 2.8-26 tons SCCP/year [14] was calculated and the associated annual releases to wastewater and air were calculated to be 3.24 and 0.25 tons. However, based on measurements of SCCP completed in these two compartments, actual loads of 0.036 and 0.75 tons/year were estimated. Hence there is a major difference between the outcome of the SFA and the results obtained from a monitoring campaign. Primarily, the release to air seems to be substantially underestimated and the release to wastewater overestimated. At the same time WWTP sludge were noted to accumulate SCCP (2300 ng/kg dry weight) [15] as also seen for the SHCCA.

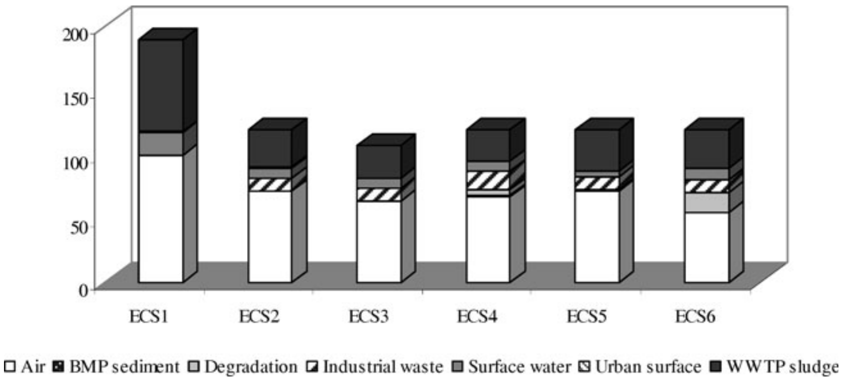

Fig. 3. Emissions of SCCP in NC (kg/year) using a SFA approach.

And in Stockholm as for NC there are no main industrial sources for SCCP. The recommendations in order to limit the releases of SCCP are green procurement in conjunction with information campaigns, efficient management of the SCCP containing articles in city (the stock) as well as implementation of BAT on smaller (modestly polluting) industries [14].

Conclusion

Implementation of the existing EU legislation involving article content limitations, wastewater treatment, and BAT should be encouraged. In EI, which is the more industrialized SHCCA, ECS4 with BAT and beyond was the most efficient strategy to reduce the overall emissions of SCCP but result in a industrial waste fraction, whereas ECS5 (stormwater and CSO treatment) was most successful for mitigating releases to surface waters. For NC, ECS2 produced the greatest reduction in the total SCCP load, ECS5 resulted in the lowest emissions to surface waters and ECS6 yielded the highest SCCP degradation. The total mass balance shows how the ECSs move the SCCP from one compartment to another. These results indicate that the selected ECS needs to be context specific and refer to the sources causing the releases and the pathways the substances take before being emitted into the environment.

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Identification of water bodies sensitive to pollution from road runoff. A new methodology based on the practices of Slovenia and Portugal

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Abstract

The discharge of road runoff water and the requirements for water protection (*e.g.* Water Framework Directive) places a pressure on the need for sound methodologies for environmental sustainability. A bilateral cooperation research permitted a comparison between Portuguese and Slovenian practices related to water protection from road runoff. In recent years both countries were confronted with a rapidly developing national highway network. Based on this study, a new methodology for sound water protection from road pollution was developed and applied to case studies in both countries, being hereinafter presented. The method showed a good potential for application, and the authors believe it to be a step forward to sustainable water protection.

Introduction

Highway runoff pollution has a diffuse pattern and may impact water systems. There are three main types of occurrences: (1) the typical rainfall events that washout the pollutant load accumulated at the paved surface; (2) the washout of chemical and other materials resulting from maintenance activities that are combined with the traffic pollutant emissions causing continuous discharge; and (3) the pollution from road surfaces that may occur after accidental spillages of toxic and dangerous substances, usually as a result of accidents involving vehicles which transport such material.

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Typically, road runoff quality is characterized by a large quantity of suspended solids, heavy metals, petroleum derived hydrocarbons, and organic matter, amongst other pollutants [4, 7]. The characteristics of the continuous discharges and the possibility for acute impacts derived from accidental spillages on the road must be correctly assessed, and its effects in the environment evaluated, taking into account requirements given by the European Water Framework Directive. The conclusions of the studies should indicate the needs for measures for pollution prevention and control [2, 3]. The very different approaches and practices applied in protecting water bodies from pollutants generated from road infrastructures, both at international and European level, show that the absence of transparent procedures in defining and protecting sensitive water bodies can lead to high and unnecessary design and construction costs, which can also later provoke non-transparent and high maintenance costs, or the opposite: the non-identification of a water body that needs protection.

Portugal and Slovenia [1] have constructed an extensive highway network since the 90's and technical know-how has been accumulated, related to the interactions between road and water bodies. In spite of the fact that the two countries have different climate, and therefore different hydrological and hydrogeological regimes, many pertinent questions about water protection from negative influences from the roads seem similar and even universal on the global level.

Based on the multiparty study [8] where comparison of Slovenian and Portuguese practices for the protection of water bodies from negative influences from roads were made, a new proposal for a methodology to define water bodies' sensitivity was developed and is presented in this paper. The "water bodies sensitive to road pollution" are here understood to be the inland surface waters, transitional waters, coastal waters or groundwater which are, either by their uses and/or supported aquatic ecosystems, alone or cumulatively, areas that are more sensitive to road pollution.

Therefore, sensitive water bodies are understood as areas to be protected, where direct and non treated road runoff discharges should not be allowed.

Definition of water bodies sensitive to road pollution

Slovenian and Portuguese methodologies

In Portugal there is no official methodology to define sensitive areas to road pollution. Nevertheless, in 2005, during a study carried out by LNEC to the Portuguese Water Institute (INAG), the participating researchers and technicians from both institutions have proposed such a methodology, in the form of a flow-chart and associated guidelines [5, 6]. It should be understood that it is not a regulation and therefore its use is not mandatory. The methodology to categorize “water bodies sensitive to road pollution” classified the receiving water bodies into three classes: “sensitive”, “non-sensitive” and “requiring site specific analysis”. It was also proposed that non listed situations require further specific site analysis. This includes the identification of key-pollutants and their maximum concentration ranges as well as soil natural background. Above certain proposed levels of key-pollutants in road runoff, water bodies cannot receive road runoff discharges without treatment.

On the contrary, in Slovenian there is a legally defined methodology but it is not directly related to the sensitivity concepts as they are understood in hydrological sciences. The definitions given by the “Decree on the emission of substances in the discharge of meteoric water from public roads” (dated from 2005) are practically oriented, and the definition of the type of drainage systems is based on the traffic characteristics, in combination with the natural characteristics of the water bodies. Roads and their sections are divided into two groups; one with the required treatment of drainage water and the other without treatment. The Decree defines two modes of water drainage from the road surface called the point of dispersion and dispersal. The point of dispersion is represented by the concentration of run-off in different drainage facilities. The dispersal is regarded as unsuitable if the prescribed passenger car equivalent - PCE limit is exceeded. On the karstic aquifers the limit is of 6,000 PCE, on porous aquifers is of 12,000, and on low permeable areas is of 40,000 PCE. In the case that road’s own waters are disposed into the flowing stream or into the coastal sea the limit is set to 12,000 PCE. In special cases, when very sensitive areas are crossed, the environmental impact assessment report can define more strict sensitivity and more demanding treatment.

For the case of surface water bodies, the relation between discharges of road runoff and receiving water is important. This question is not regulated with strict values; however, in practice and in some guidelines, the principle that 10% of the average low discharge of the receiving stream should not be exceeded by the discharge from the road runoff is applied. When exceeding this criterion, retention ponds need to be constructed to treat road runoff before its disposal. Treatment is required only for the critical rainfall event of up to 15 minutes defined with the intensity of 15 l/s/ha.

Proposal of a common methodology to define water bodies sensitive to road pollution

The proposal for a new common methodology to define "water bodies sensitive to road pollution was based on the Portuguese and Slovenian experiences. Envisaging the water bodies' protection, this methodology aims at giving a first approach for identifying the most sensitive areas to road pollution, where control of water discharges from road surface and constructions as well as emissions that may have potential negative effects on water bodies should be made. The spirit of this methodology is within the WFD which implies that member states take the necessary measures to prevent or limit the input of pollutants into different water bodies.

This methodology was developed using a flow-chart (Fig. 1). Therefore, it is represented as a decision tree, and several steps must be considered during its application. In the first step, the user should define what general type of water body is crossed by the road; whether they are inland waters, transitional waters or coastal waters. In the following step a sub classification is given. Following the guidelines, from box to box, in the decision tree the user will arrive to three final categories:

- Sensitive – water body classified as such is, *per se*, sensitive to road runoff pollution. All necessary precaution and protection measures must be used. The best solution, if possible, is to avoid any road construction in these areas. Sometimes this is not possible; such is the case of Slovenia, where nearly half of the country is positioned on karstic aquifers. These are also areas where direct discharge of road runoff to water bodies is not possible.
- Non-sensitive – these areas are not sensitive to road pollution. The main reason for such classification is the presence of a large volume of water or high water flow. Consequently there is a considerable dilution of road pollutants. In such areas there is no restriction for road runoff disposal.
- Study – this category means that it is not possible to decide in advance if the water body is sensitive or not. The decision must be taken based on further data collection, field and desk studies, for each particular case.

The identification of sensitivity factors, as proposed, is based on the intrinsic physical and hydromorphological properties of the water bodies, and is identified depending on their role in controlling the sensitivity of the water to pollution from roads (*e.g.* type of receiving water bodies, flow rate, aquifer material permeability, soil properties, retardation, etc.).

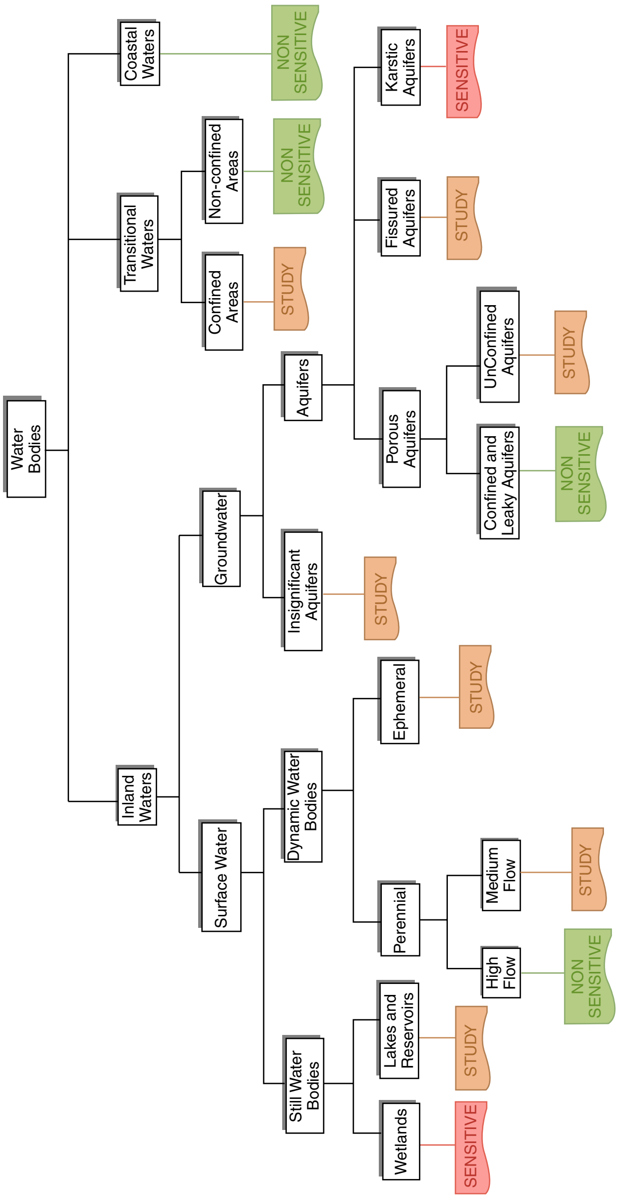


Fig. 1. Flowchart proposal of a common methodology to define water bodies sensitive to road pollution.

Due to the complicated natural conditions and many possible influences of roads on their status, comparing to other general types of waters, the classification of inland waters is complex. This classification considers the receiving surface water bodies as being of three types: still and dynamic surface waters, and groundwater. Still waters can be further classified in wetlands, lakes or reservoirs. The first ones are considered sensitive due to the lower rate of water renovation, and to the ecosystems that they usually support. The latter are considered water masses that need specific site analysis based on their characteristics and uses.

In what concerns the dynamic waters there are two different situations: perennial and ephemeral water bodies. Perennial water bodies are classified in high and medium flow rivers. In the first group, the combined effect of dilution and hydrodynamics should be sufficient to ensure that the discharge of non treated road runoff is not a problem. The definition of a "high flow river" for this methodology is based on the analysis of the monthly averaged flow measurements that represents the river flow. Since seldom this data concern to the study site, it should be done the correction of the flow data to the river section where the road runoff is discharged – in what concerns, for instance, the watershed area, the precipitation and elevation. The flow data should represent a period of, at least, 10 years (the most recent ones). Considering that there may exists seasonal flow variability, which is for instance typical in Mediterranean countries, when the lowest monthly averaged flows calculated is above 20 m^3/s, it is considered for the purpose of applying the present methodology that one is in presence of a "high flow" river – in which it is possible to discharge non treated road runoff.

For the case of medium flow rivers, *i.e.*, rivers for which the lowest of the monthly averaged flows calculated is below 20 m^3/s there is the need for a specific site analysis to access their sensitivity - which has to do with not just the flow but several other processes. The limit of 20 m^3/s was established based on a conservative principle, and taking into consideration the objectives of the WFD. Road runoff may cause both acute and cumulate impacts in river systems, meaning that it is not just a matter of dilution in the river water mass of the dissolved road pollutants; there is also the effect of particulate pollutants that accumulate at the river bed. A high flow enables not only a greater dilution but also increases the sediments transport and resuspension rates.

In the case of ephemeral water bodies there is the need to assess the soil infiltration capacity in the riverbed. In case it is high, the area sensitivity will by defined by the underlying groundwater sensitivity. For the cases of low soil infiltration rate, it is considered that water will flow down gradient, and therefore the runoff discharge impact should be assessed based on the effects on the receiving surface water body, downstream. Therefore, for these cases, further site specific studies are needed.

The methodology to define sensitive groundwater bodies to road pollution was based on the hydrodynamic characteristics of the aquifer. The situations defined are based on the pollutants infiltration capacity and further migration into the groundwater body, as well as in the capacity for pollution retention. Two types of formations were considered: insignificant aquifers and aquifers.

Insignificant aquifers are defined as local and limited groundwater or essentially areas with no groundwater. Although not significant at national scale, these aquifers frequently play an important role in the water supply of local populations, with a small number of inhabitants.

They are considered as areas that need further specific site analysis in the sense that it is important to access the presence of permeable faults and fissured zones, or any other feature that might be responsible for fast pollutant leaching towards groundwater.

Aquifers are the geologic formations being important for potential exploitation (having good storage capacity and transmissivity characteristics). There are three different hydrogeologic situations that can be considered: porous aquifers, fissured aquifers and karstic aquifers.

Porous aquifers can be further divided into confined, leaky and unconfined aquifers. In the first two cases, these areas are considered non sensitive for road pollution. The permeability of soil covering these aquifers is low, the vertical leakage of potential pollutant is very small, and the retention capacity is high. In the case of unconfined aquifers there is the need of further specific site analysis in which several important properties concerning the potential for leaching of the soil accumulated pollutants should be accessed (*e.g.* depth to the water table, recharge, soil type and characteristics, topography, hydraulic conductivity, retardation, etc.).

Fissured aquifers sensitivity to road pollution depends on the degree of fracturation at the surface, since it controls the infiltration capacity, as well as the depth interconnections between these fractures. As a result, there is the need of further specific site analysis for areas located in fractured aquifers. Karstic aquifers are sensitive areas due to its very high porosity and transmissivity characteristics, as well as the very low capacity to retain pollutants, which makes them very vulnerable to pollution.

The sensitivity of transitional water bodies is very much depending on the tidal water renovation within the estuary. As a result, non confined areas are considered non sensitive since a great volume of water enters daily the system, being therefore able to dissimulate the effects caused by the comparably smaller road discharges. Confined areas are usually represented by shallow water bodies that, at the same time, support sensitive ecosystems, shellfish, and salt production, among other. They are therefore considered as areas that require further studies.

Considering the characteristics of coastal waters, namely in what concerns its volume when compared to road runoff discharges, they are considered as non sensitive areas.

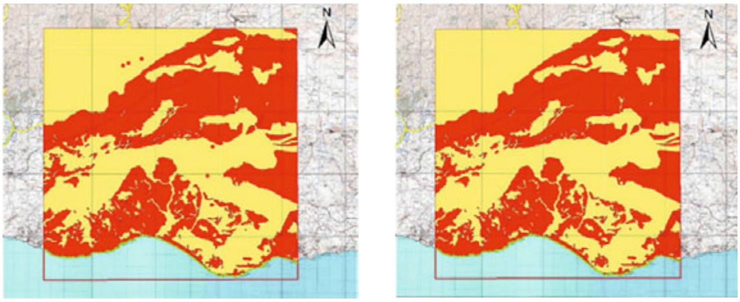
Discussion of the new methodology application to case studies

The existing Portuguese methodology [5] and the proposed methodology (Fig. 1) were applied to a case study in Portugal and another in Slovenia. The sites were selected according to the traffic characteristics of roads, and similar geological and hydrological conditions. In both cases the methodologies were applied in karstic areas where carbonate aquifers are present. The testing site for Portugal is positioned in the Algarve region, between the cities of Silves, Lagoa and Albufeira. The area consists of a large karstified carbonate aquifer system Querença – Silves, which is composed by Jurassic formations. The aquifer system has an opened to confined hydrodynamic character, and it is of great importance for the water supply at the coastal region of Southern Portugal.

The testing site in Slovenia is located in its southwestern part, along the highway from the Razdrto (central east Slovenia) to the direction of the Adriatic Sea coast. Geology of the area is represented by Cretaceous and Palaeocene carbonates with the stratigraphical thickness of more than several thousand meters intervened with low permeable Paleogene flysch deposits. The carbonates form a large karstic aquifer with an unconfined hydrodynamic character. The aquifer system is very vulnerable and sensitive.

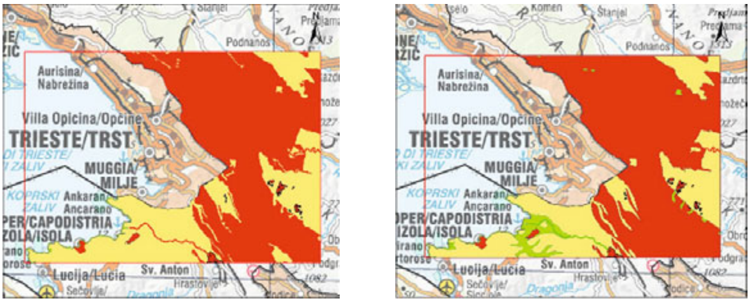
The results from the application are shown in Fig. 2. The methodology allowed the classification of the case-study areas as: sensitive, non sensitive or requiring further studies. The common methodology compared to the Portuguese one, proved to be more expeditious and easier to apply, with information generally available. It is also more easily understood by anyone, because it is not tied up to specific characteristics for Portugal or

Slovenia. As can be observed in Fig. 2, the results are similar for both applications, which is considered to be a good indication of the effectiveness of the new methodology.



(a) (b)

Portuguese case study: Querença – Silves aquifer. Results of the Portuguese methodology (a) and the new methodology (b)



(a) (b)

Slovenian case study: SW Slovenia. Results of the Portuguese methodology (a) and the new methodology (b)

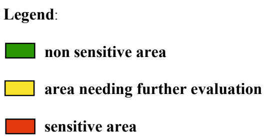


Fig. 2. Results of application of the Portuguese and the new methodology to a case study in Portugal and a case study in Slovenia.

Conclusions

The multiparty study done confirmed the need for sounder methodologies in order to improve the practice concerning protection of water bodies from road runoff pollution. Moreover, it was acknowledged that the approach for defining water bodies' sensitiveness to road pollution, *i.e.* the areas that need to be protected, is different in the two countries, although some approaches may be considered similar. The difference is also a result of differences in the nature of the surface water bodies and groundwater in Portugal and Slovenia.

A new common methodology for identifying water bodies sensitive to road runoff pollution is proposed, based on the evaluation of both methodologies. It aims at defining the water body sensitiveness to road pollution based on the intrinsic characteristics of inland (surface and groundwater), transitional, and coastal waters. The methodology allowed the classification of the case-study areas as: sensitive, non sensitive or requiring further studies. The common methodology proved to be expeditious and easy to apply with information generally available. It is also more easily understood by anyone, because it is not tied up to specific national characteristics. The methodology should be further applied in the future, to different sites and environmental conditions, in order to confirm its consistency and effectiveness.

Acknowledgments

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Modeling nutrient and pollutant removal in three wet detention ponds

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Abstract

Three wet detention ponds receiving stormwater runoff and located in Århus, Silkeborg and Odense, Denmark were studied. The ponds were continuously monitored by measuring turbidity, temperature, pH, oxygen level and water level in the ponds as well as inflow and outflow. Water samples were collected from the inlets and from the downstream end of the ponds and analyzed for suspended solids, total and soluble phosphorous, nitrogen and seven heavy metals. Performance of the ponds was modeled by routing the measured flow though the ponds and simulating pollutant removal by 1st order kinetics with and without temperature adjustment.

Introduction

Stormwater contains a large range of pollutants, e.g. suspended solids, nutrients, heavy metals, pesticides and poly aromatic hydrocarbons (PAHs) [1]. Nutrients can lead to eutrophication and following oxygen depletion in the receiving waters. Furthermore, heavy metals, PAHs and other micopollutants can have toxic effects on flora and fauna in the receiving waters [2,3]. That is, treatment of stormwater is from time to time necessary before leading it back to the environment. An obstacle for stormwater runoff treatment is that hydraulic and pollutant loads are intermittent and highly variable over time. The facility must furthermore be robust and maintenance-free and treat runoff at low pollutant concentrations to even lower concentrations. Wet detention ponds have proven efficient performance in this context and are commonly chosen for treatment of stormwater runoff.

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The performance of a wet detention pond depends on numerous factors such as pond geometry and hydraulic retention time. The retention time of a pollutant – being it a dissolved component or a particle – is affected by the size, depth and length to width ratio of the pond as they influence the mixing and flow patterns [4]. The pollutant removal rate is affected by factors like temperature, which influence on the biological processes but also on the viscosity of the water and thereby the sedimentation of particles.

As a part of the EC-founded LIFE-treasure project three wet detentions ponds were constructed in 2007 and 2008 in the cities of Silkeborg, Århus and Odense, Denmark [5]. An aim of that project was to investigate the performance of wet detentions ponds in Denmark by caring out an intensive monitoring campaign of flow, pollutant load as well as removal together with monitoring the physical and chemical state in the ponds.

The objective of this study is to determine whether removal rates for different pollutants are alike in the three ponds. Furthermore it is investigated whether the rates are affected by temperature. Mixing in the wet detention ponds is investigated to determine the degree to which the ponds in this context can be viewed as fully mixed or following a plug flow principle.

Methods

Site and pond characteristics

The three ponds receive stormwater runoff from three different catchment types and have different physical outlines (Table 1). Precipitation is measured by tipping bucket gauges of the Danish SVK rain gauge system, located a few kilometers from each pond. All three ponds were designed as recommended by e.g. [4,6,7], that is, they were designed to have a retention time of 72 hours with a return period of 4 year^{-1}.

First the stormwater enters a grid chamber. After the grid chamber, the stormwater enters the earthen ponds with boulders to disperse the inflow jet stream. All ponds are equipped with clay membranes. The pond in Århus has three small circular islands with plants in the middle of the pond, to disperse jet streams. In Silkeborg the pond is equipped with two planted earthen barriers, during dry weather dividing the pond in three disconnected sections. The pond in Odense has no additional jet stream management. Schematic drawings of the ponds are presented in Figure 1.

Table 1. Pond and catchment characteristics.

Type of catchment	Odense	Århus	Silkeborg
	Light industry	Residential (blocks of flats)	Residential (detached houses) and highway
Total catchment area [ha]	27.4	57.4	21.5
Impervious catchment area [ha]	11.4	25.8	8.8
Permanent wet volume [m^3]	1,990	6,900	2,680
Retention volume [m^3]	1,992	1,400	3,230
Permanent water depth, max [m]	1.45	1.25	1.00
Length to width ratio	4.5: 1	3:1	3 chambers of app. 1:1 each
Precipitation [mm y-1]	657	661	719
Annual runoff [m3]	55,500	131,900	49,600

On-line measurements

Inlet flows were measured by a smaller and a larger full-flowing magnetic flow meter in each pond (Krone optiflux, DN 150 mm and DN 500 mm in Odense, Århus and DN 400 mm in Silkeborg) coupled in series and parallel to a rectangular weir. The accuracy of the measurement was better than 1% for inflows between app. 3 L s^{-1} and app. 1 m^3 s^{-1}. At flows over app. 1 m^3 s^{-1} the weir begins to convey some of the flow. The flow over the weir is estimated by the water level in the grid camber.

Fig. 1. Schematic drawings of the three ponds, top: Odense; bottom left: Århus; bottom right: Silkeborg.

Sensors for online monitoring of pH (WTW SensoLyt 700 IQ), dissolved oxygen (WTW FDO 700 IQ), temperature (via pH meter) and water depth (Klay Hydrobar) were placed in the ponds. In Odense and Århus the sensors were in the middle of the ponds. In Silkeborg sensors were in both the middle and last sub-compartment. Readings were stored every minute.

Stormwater sampling

Auto-samplers equipped with plastic containers were placed in the inlets after the grid chambers and in the middle of the basin (Odense), in the end of the basin (Århus), and in the last sub-compartment (Silkeborg). Water samples were collected flow proportionally in the inlet and time proportionally in the basins. The auto-samplers were emptied when full or every 14 days. The auto-samplers were placed underground, i.e. dark and cold to minimize changes of the samples during the storage on site. The subsamples from the auto-samplers were pooled and analyzed for total concentrations of copper, lead, zinc, cadmium, nickel, chromium, mercury, total PAH (after USEPA), suspended solids and nitrogen as well as total and dissolved phosphorus. All analyses were performed by an accredited laboratory following ISO 17294m, manual MK2260-GC/MS, DS/EN 872, DS/EN I 6878aut, manual SM17 udg 4500 and DS/EN I 11905 auto. The water samples from Odense cover the time span from mid April 2008 to late September 2009. Århus cover the time span from mid June 2008 to late September 2009 and Silkeborg cover the time span from mid December 2008 to late September 2009.

Wet pond modeling

The water balance in the ponds consists of measured inflow, volume of water in the pond and outflow. Evaporation from and precipitation on the pond surface are neglected as these are minor compared to inflow and outflow. Exfiltration from and infiltration to the pond through the wetted perimeter are set to zero as all three ponds were equipped with an impermeable clay membrane. Some groundwater infiltration was visually observed during wet periods but deemed inconsequential. The volume of water in the pond is calculated continuously with a 1-minute time resolution, corresponding to the frequency of the flow and water level readings.

Removal of a pollutant is simulated simultaneous with the water balance by simple 1st order kinetics with and without temperature adjustment (Arrhenius equation, see eq. 1 and 2). The true removal of pollutants from the water phase in wet detentions ponds are a complex mixture of chemical, physical and biological processes e.g. flocculation, bacterial degradation, adsorption, absorption, resuspension of sediments and sedimentation which again are affected by for example particle characteristics and water viscosity. That is, simple kinetics such as 1st order kinetics has its limitations when simulating removal of pollutants in wet detention ponds [8,9], but they were chosen as the detailed processes and boundary conditions are not well defined or understood [10].

$$\frac{dC}{dt} = -kC \tag{1}$$

$$\frac{dC}{dt} = -k\alpha^{20-T}C \tag{2}$$

C is the concentration of a pollutant	[g m^{-3}]
k is the removal rate	[g m^{-3} day^{-1}]
α is the temperature dependency constant	[-]

Inputs to the model are the measured temperature, inflow and concentration of pollutants in the inlet and in the pond. The model simulates the pollutants in the pond using inlet pollutant loads and 1st order kinetic for pollutant removal. The model is coded in Pascal and iterates the removal rate until a best fit between simulated and measured pollutants in the pond is achieved. Two different fitting procedures were used. First the average mass of pollutants discharged from the pond was fitted to the measured pollutant discharge. That is, the model was fitted to one value, namely the average removal of the whole measuring campaign. Next a least square method was applied to continuously simulate the pollutant removal.

Results and discussion

Mixing of the water phase in the ponds

Examples of online measurements and measured inflow are shown for Odense in Figure 2, for Århus in Figure 3 and for Silkeborg in Figure 4.

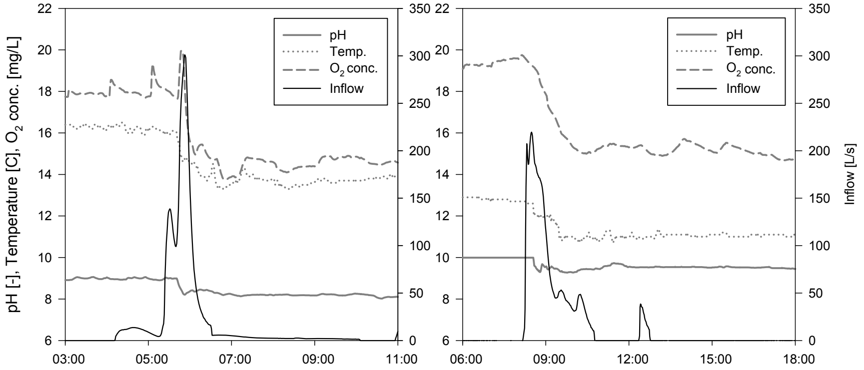

Fig. 2. The influence of runoff events on pH, temperature and oxygen level in the pond in Odense. Right graph: 19-05-2008, left graph: 22-04-2009.

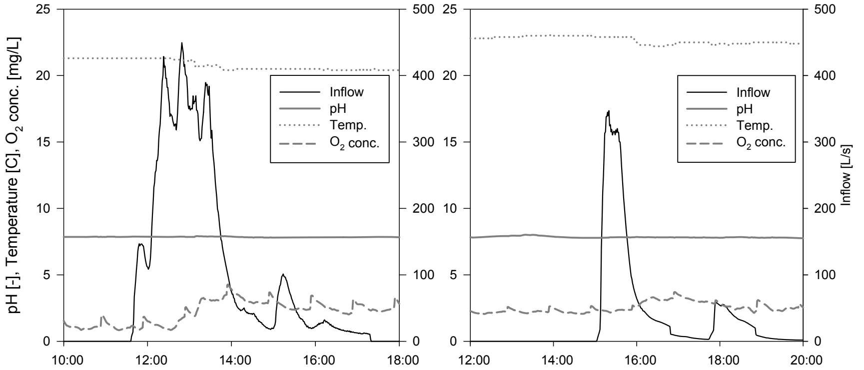

Fig. 3. The influence of runoff events on, temperature and oxygen level in the pond in Århus. Right graph: 18-07-2009, left graph: 06-07-2009.

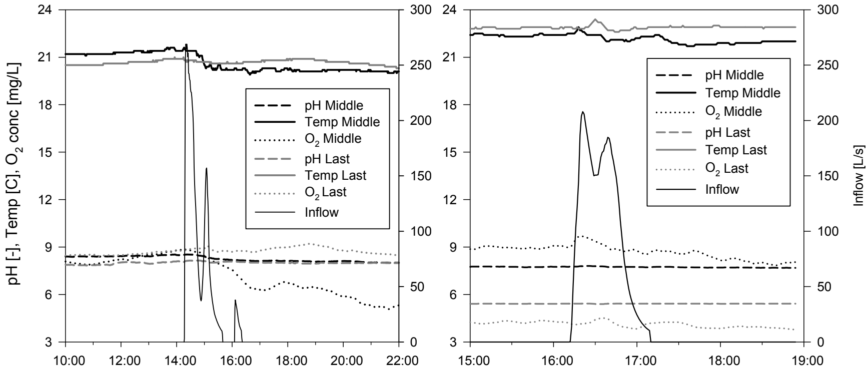

Fig. 4. The influence of runoff events on pH, temperature and oxygen level in the pond in Silkeborg. There are two sets of sensors: in the middle compartment and in the last compartment. Right graph: 08-07-2009, left graph: 08-08-2009.

For the pond in Odense, runoff events caused quick and clear responses in pH, temperature and oxygen levels. After typically 20 minutes the parameters became affected. In the examples of Figure 3, oxygen declined with app. 4 mg L^{-1}, temperature with app. 2˚C and pH with 1 unit.

In the pond in Århus, the response is comparatively smaller and slower than the pond in Odense. In the example of Figure 4, pH is not affected, the oxygen level increases with app. 2 mg L^{-1} (graph to the left) and 1 mg L^{-1} (graph to the right). The temperature declines with app. 1˚C in both examples. The responses occur within 1.5 hours from the start of the event.

In Silkeborg the response to a runoff event is evident in the middle compartment but not in the last compartment. In the example of Figure 5 (graph to the left), temperature in the middle compartment declines with approximately 2˚C, the oxygen level declines fast with 2 mg L^{-1} and continuous to decline. For the event depicted in the graph to the right, the temperature declines a bit and becomes unstable, the oxygen level first increases with 1 mg L^{-1} and thereafter begins to decline. This increase in oxygen level is also seen in the last sub-compartment app. 20 minutes later. The pH is not affected in any of the cases. The responses to the events are seen within approximately 30 minutes in the middle compartment.

As these observations show, the mixing in the ponds is quick compared to the rate with which pollutants typically are removed in wet detention ponds [4]. The ponds are therefore simulated as one fully mixed box and not as a set of boxes.

Fit of models

Each pond has been modeled six times. Once by the procedure of fitting to the average removal efficiency only and without temperature correction (ARE). And five times with the more complex fitting procedure using a least square method (LSM) and with a temperature dependency constant, α, ranging from 1.00 to 1.09. An α of one corresponds to no temperature dependency. An α of 1.03 – 1.05 emulates sedimentation as the dominating removal process as such α-value follows the temperature dependency of water viscosity. When α is in the range of 1.07 – 1.09, biological processes are indicated as being dominant. For each of the six models the error between measured and simulated values was determined by equation 3. The errors were standardized so they can be compared across the different pollutants and ponds, despite differences in pollutant loads and dataset size.

$$\sum \frac{\left(\frac{Modeled\ C - Measured\ C^{\ 2}}{\Sigma Measured\ C}\right)}{Number\ of\ measurements} = Error \tag{3}$$

It was not possible to model the removal of nickel for the pond in Århus as the concentrations of nickel were higher in the pond than in the inlet – properly caused by internal released from e.g. the clay membrane. Modeling the removal of zinc in the pond in Silkeborg was only possible with ARE as LSM went unstable, probably due to the limited number of data. All determined errors are shown in Figure 5.

The smallest errors, that is, the best fit, for the pond in Århus were achieved using ARE for 6 out of 10 pollutants. The other 4 pollutants had the smallest error by LSM with temperature correction $\alpha = 1.07$. Looking at the total error (all errors for each pollutant added together) the best fit was achieved using LSM without temperature correction, that is $\alpha = 1.00$.

In the pond in Odense the best fit was found for 5 out of 11 pollutants when using ARE. The other 6 pollutants achieve the best fit with LSM. One pollutant had the smallest error whit $\alpha = 1.00$, another pollutant had the smallest error with $\alpha = 1.07$ and the last 4 pollutants had the smallest error with $\alpha = 1.09$. The total error was smallest using ARE.

In Silkeborg the picture was more blurry. Four out of the 10 pollutants had the best fit using ARE. The last 6 pollutants had the best fit using LSM with different α-values. Three pollutants without temperature correction, $\alpha = 1.00$, one pollutant with $\alpha = 1.03$, one with $\alpha = 1.05$ and one pollutant with $\alpha = 1.07$. The smallest total error was achieved by using ARE. It was tested whether better fits were achieved using three fully mixed boxes in the model instead of one fully mixed box (i.e. simulating the 3 compartments as 3 boxes), but no tendencies were found for the individual pollutants. However, the total error using ARE with three boxes were 28 % lower than the total error using one box.

For many of the pollutants in Odense and Århus relative good fits were achieved with LSM and an α–value of 1.07 – 1.09, indicating that biological removal was dominating. However, the overall tendency for all three ponds was that the best fit was achieved using ARE, indicating a random temperature dependency. The stochastic behavior of both pond input and removal processes probably overruled the detailed processes like temperature dependency. This independency of temperature was also found in [11]. It was therefore concluded that at the data-level available and with the current knowledge of processes, it is not feasible to simulate detailed removal processes and that only average removal should be modeled.

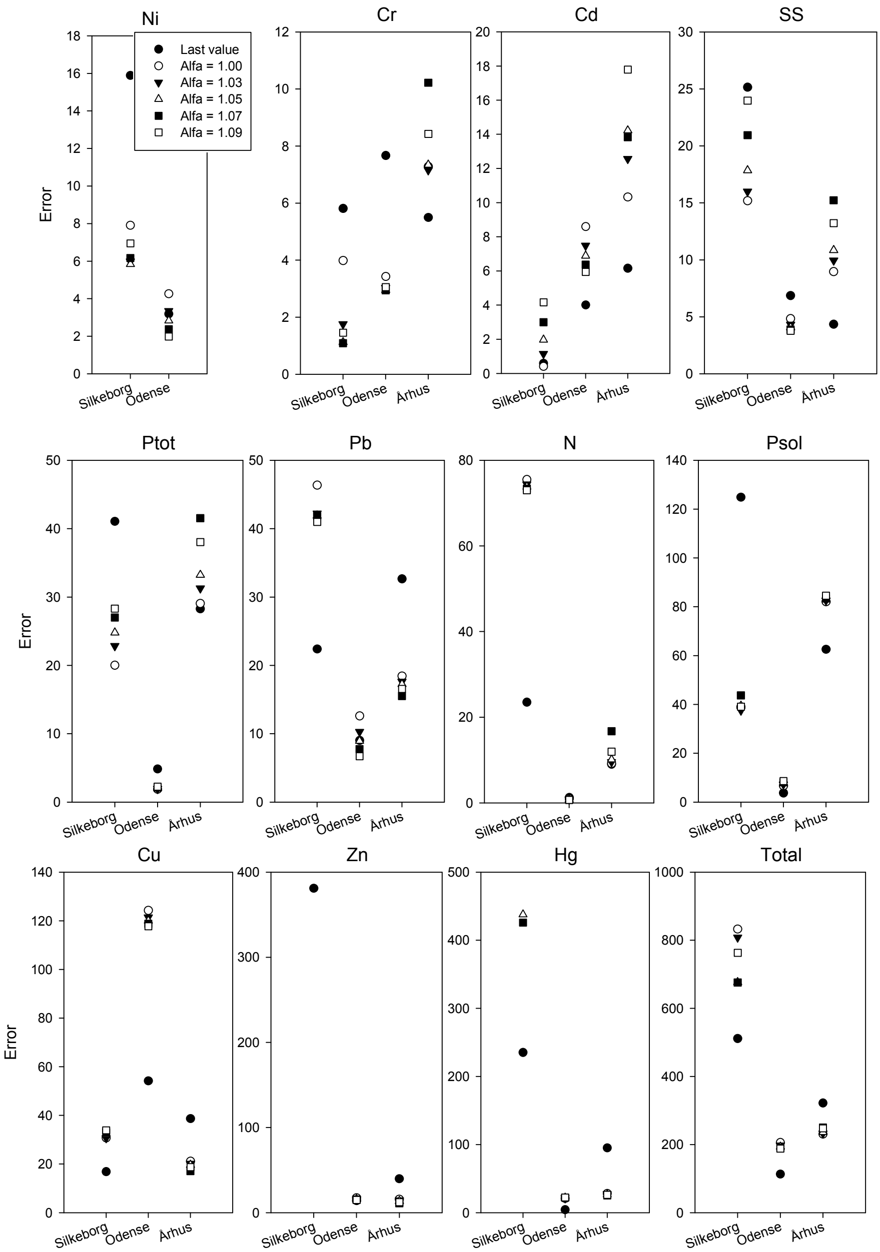

Fig. 5. Error between model and measured data for each pollutant and the total error (sum of the error for each pollutant). The error is calculated for ARE and LSM with α-values from 1.00 to 1.09.

Removal rates

For each model a set of removal rates were found, one for each pollutant. The actual value of a removal rate depends on the model by which it is determined, i.e. which α–value was applied. The rates found for each pollutant differ between ponds but are generally of the same magnitude except for a few pollutants, e.g. dissolved phosphorous in Silkeborg. The removal rates for dissolved phosphorous in Silkeborg ranged from 0.48 to 23.26 when using one box in the models. The rates got significantly lower when using 3 boxes, which was expected [12], but did not give a better fit. In Table 2 the simulated removal rates are shown. The rates for all 11 pollutants in the three ponds were determined by ARE which had the lowest total error. Furthermore, removal rates determined by a similar approach for a wet detention pond in Skullerud, Norway [7] are shown for comparison.

Table 2 shows relative similar removal rates across the four ponds for total phosphorous and total nitrogen. For the rest of the pollutants quite fluctuating removal rates were found, illustrating a general uncertainty in predicting pollutant removal in wet detention ponds.

Table 2. Removal rates determined using ARE. Skullerud is a wet detention pond in Norway which has been simulated by a similar approach [7].

	SS	Ptot	Psol	Ntot	Pb	Cd	Cu	Zn	Cr	Hg	Ni
Odense	0.32	0.14	0.26	0.08	0.90	0.21	0.28	0.15	1.21	0.08	0.20
Århus	0.66	0.23	1.44	0.09	0.43	0.04	0.14	0.07	0.82	0.36	-
Silkeborg	0.13	0.36	3.73	0.01	0.03	0.02	0.03	0.04	0.02	0.02	0.27
Skullerud	1.9	0.12	0.14	0.02	0.65	0.14	0.10	0.52	-	-	-

Conclusion

Changes in pond pH, temperature and oxygen level due to runoff entering the ponds showed that the three wet detention ponds studied should be characterized as fully mixed reactors and not as plug flow reactors. Sensors placed in the middle of the ponds showed mixing of water already 20 to 90 minutes after the onset of storm events.

Two different simulation procedures as well as a range of temperature dependencies were applied in the models describing the wet detention ponds. Generally, the simplest simulation procedure without temperature adjustment gave the best fit to measured data. However, for some pollutants a more complex method with temperature dependencies corresponding to biological degradation being the dominant pollutant removal processes gave the best result. It was concluded that more knowledge of the individual processes is needed before more complex models should be applied; so far only modeling of the average removal seems feasible.

The removal rates for total phosphorous and total nitrogen were somewhat similar in the three ponds studied as well as in a pond reported in literature. For the other 9 pollutants no clear tendency for the removal rates were found and future prediction of removal of these pollutants in wet detention ponds seems difficult.

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Analysis of flow characteristics in a compound channel: comparison between experimental data and 1D numerical simulations

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Abstract

The common configuration of rivers is a main channel flanked by floodplains. During flood events, the main channel is not enough to discharge the flow and the floodplains are submerged. The momentum transfer due to the difference of the velocities between the sub-sections generates a complex 3D flow structure. For the study of the influence of this structure in flow modeling, measurements of the velocities in a prismatic compound channel have been made. Seven 1D methods to compute the sub-section and total discharges were applied. Comparisons between experimental data and modeling reveal good agreement when simple models that indirectly take into account the momentum transfer are applied.

Introduction

The present paper presents a study of the flow in a compound channel. This configuration has extreme importance because in many cases the main channel of the rivers is not enough to discharge the total flow, mainly during flood events. In these cases, the flow inundates the surrounding fields, called the floodplains. Therefore, the common configuration of the rivers during floods is a compound channel flow, where one can observe the interaction between the main channel and the floodplain flows.

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The traditional method to study the flood inundation is based in an old approach that simply divides the total cross section with vertical divisions in the interface of the main channel and the floodplains. Besides that, new 1D approaches can take into account the interaction between the flows in each subsection.

This paper intends to improve the knowledge of the flow in compound channels. So, experimental results were compared with 1D modelling of the flow in this type of channel. The data showed in the present paper corresponds to an upgrade of the work presented in [1].

Theoretical background

The water depths in a single channel are accurately estimated since the equation proposed by Antoine de Chézy [2]. This is not the case for compound channels, because of the velocity gradient between the flows in the main channel and in the floodplains, where the water depth is lower and, in many cases, the roughness is higher. This gradient generates a mixing layer in the interface which creates a 3D flow structure (*cf.* Fig. 1).

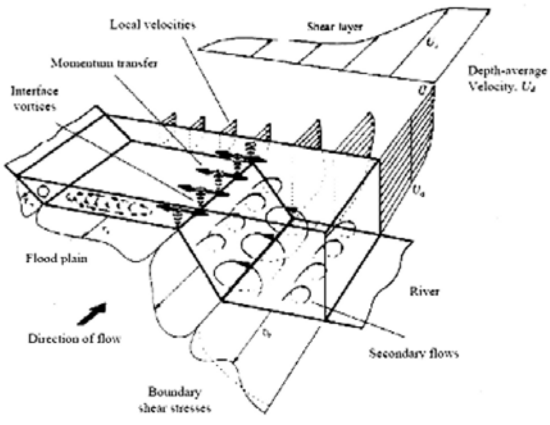

Fig. 1. Flow structure in a compound channel [3].

The discharge capacity of the main channel reduces and the floodplain capacity increases, generating a global loss in the total discharge capacity.

The 2D and 3D methods include some of the characteristics of compound channels. In engineering, due to the amount of data required and the processing time, 1D methods are often preferred. Still, the momentum transfer should be taken into account in 1D modeling [4].

Since [5] presented the first evidences of the flow characteristics in compound channels that there have been attempts to modelling it. [6] referred the difficulty of the developed formulas to be applied universally as, in many cases, they had been set based on a reduced amount of data.

Modelling the flow in a compound channel as a simple channel by applying a formula of resistance to flow does not take into account the sub-section velocity differences. [7] suggested the division of the channel in subsections where velocity and roughness could be considered as uniform. This method, called the Divided Channel Method, is still widely used in commercial models as HEC-RAS [8], ISIS [9], SOBEK and Mike 11 [10].

As pointed out in [9] this treatment of a compound channel assumes that there is no interaction between the subdivided areas despite the existence of mean velocity discontinuities at the assumed internal boundaries. Therefore the simple division of the channel in subsections is not appropriate for modelling the discharge in compound channels [6].

Different methods had been proposed with the attempt to model the interaction processes that occur in this type of flows, including the mass and momentum transfer.

According to [9], these methods can be divided into 5 groups: i) methods that change the sub-area wetted perimeters; ii) methods that made discharge adjustments (with the experimental data, for example); iii) methods that include apparent shear stresses on the sub-area division lines; iv) methods where the lines are located at zero shear stress; v) methods that combine different divisions of the channel.

In this work, seven methods were used to model the flow in the compound channel. Its computation procedures are presented in the Annex 1. Firstly, we used the two traditional methods called Single Channel Method (SCM) and Divided Channel Method (DCM).

From the groups presented before, we used the Coherence Method (CM) and the Debord Method (DM) from the group ii), the Exchange Discharge Method (EDM) and the Interacting Divided Channel Method (IDCM) from the group iii) and the Weighted Divided Channel Method (WDCM) from the group v).

Experimental component

The channel used in the present work is located in the Fluvial Hydraulics Pavilion of the National Laboratory for Civil Engineering, in Lisbon. The channel has about 10 m length and 2 m wide. The slope of the channel is $1{,}1 \times 10^{-3}$ m/m. The cross-section is symmetrical and it is composed by a

0.4 m wide and 0.1 m high main channel, flanked by two floodplains 0.7 m wide. The transition between the subsections is made by banks with 45° slope. The channel bottom is made of polished concrete. Fig. 2 shows a photograph of the channel and a schematic cross-section.

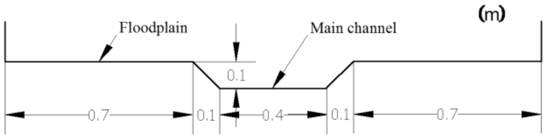

Fig. 2. Compound channel.

Following the recommendations of [12], separate inlets were available in order to avoid the mass transfer between subsections. The discharges for the main channel and floodplains were monitored by two flowmeters and controlled by two different valves. Honeycomb diffusers and polystyrene plates were located at the beginning of the flume to stabilize the flow.

The flow regime is subcritical and the water depths were controlled by three horizontal axis tailgates located at the downstream end of the channel. It was possible to define two different water levels, one for the main channel and the other for the floodplains.

Water levels were measured with three hydrometers, two of them fixed at the upstream and downstream sections of the flume and the other is located in a movable trolley.Velocity measurements were made using a Pitot tube with a 3.2 mm diameter. The difference between static and dynamic pressures was measured with a differential pressure transducer.

Experimental procedure

For the presented compound channel, the distribution of the discharge between the main channel and the floodplains was not known. The procedure used to obtain an uniforme flow starts with the distribution given by the Weighted Divided Channel Method [13]. With this first discharge distribution, the water levels were controled with the tailgates in order to achieve an uniform water depth along the channel. Reached the uniformity, the discharge distribution at the downstream section is compared with the upstream distribution. If the upstream and downstream distributions match unless 0.1 l/s, the uniform regime has been achieved. Otherwise the measured discharge distribution is imposed upstream and the procedure is repeted (normally 2 or 3 iterations).

The velocities were measured in 45 verticals with 5 or 6 points each for the floodplains and main channel, respectively (*cf.* Fig. 3).

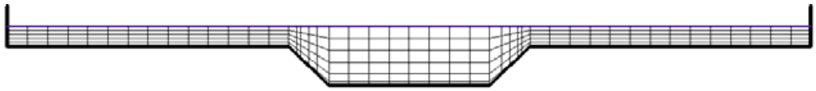

Fig. 3. Mesh for the velocity measurements.

For each vertical, the depth-averaged velocity was computed from the velocity measurements in 5 or 6 points using the following equations.

$$U_{fp}^{ave} = \left(7{,}5 \times U_{bot} + 15 \times U_{20\%} + 20 \times U_{40\%} + 20 \times U_{60\%} + 30 \times U_{80\%}\right)/100 \quad (1)$$

$$U_{mc}^{ave} = \begin{pmatrix} 2{,}5 \times U_{bot} + 10 \times U_{10\%} + 15 \times U_{20\%} + 20 \times U_{40\%} + \\ + 20 \times U_{60\%} + 30 \times U_{80\%} \end{pmatrix}/100 \quad (2)$$

In which $U_{\#\%}$ stands for velocity measured at a height equal to #% of the water depth; U^{ave} for average velocity; *"mc"* for main channel; *"fp"* for floodplains and *"bot"* for bottom. The eqs. (1) and (2) were obtained using measurements of vertical profiles of velocity with 18 points. For these profiles the average velocity was calculated and compared with several equations assuming the knowledgement of the velocity at these 5 or 6 points. The best results were obtained with these equations.

Analysis of results

Results

Four different tests have been made corresponding to relative depth, h_r (relationship between water depths in the floodplain and in the main channel) approximately equal to 0.1; 0.15; 0.2 and 0.3. The discharge distributions are shown in Table 1. The average velocity distributions are presented in Fig. 4.

Table 1. Results of the discharge distributions.

Relative depth, h_r (-)	Flooplains discharge (l/s)	Main channel discharge (l/s)	Total discharge (l/s)
0.1	3.2	34.5	37.7
0.15	6.2	38.6	44.8
0.2	11.2	42.2	53.4
0.3	27.4	53.3	80.7

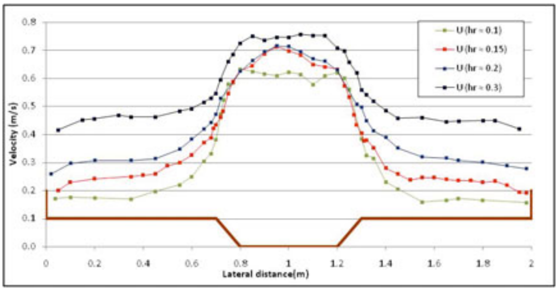

Fig. 4. Velocity distribution in the cross section.

Comparison between experimental data and 1D modeling

The results from the 1D methods presented in the Annex 1 were compared with the results from the experimental tests. This comparison covered the total discharge and the sub-section discharges. Indeed, as refereed by [9], it is essential to perform the analysis for each subsection. The assessment of the accuracy by each method is based on the calculation errors computed by Eqs. (3) to (5).

$$\text{Error}_{mc}\ (\%) = 100 \times \left(Q_{mc}^{Measured} - Q_{mc}^{Calculated}\right) / Q_{mc}^{Measured} \quad (3)$$

$$\text{Error}_{fp}\ (\%) = 100 \times \left(Q_{fp}^{Measured} - Q_{fp}^{Calculated}\right) / Q_{fp}^{Measured} \quad (4)$$

$$\text{Error}_{Total}\ (\%) = 100 \times \left(Q_{Total}^{Measured} - Q_{Total}^{Calculated}\right) / Q_{Total}^{Measured} \quad (5)$$

In which $Q^{Measured}$ stands for measured discharge and $Q^{calculated}$ for the discharge calculated by one 1D method.

In Table 2 the errors obtained for each subsection and for the entire channel by each method are presented. The results of the Table 2 are presented graphically in Fig. 5.

From these results, it can be seen that the SCM, assuming an average velocity for whole cross section tends to underestimate the total discharge. The opposite happens with the results obtained with DCM. The simple division of the channel, without considering the interaction between the subsections, leads to an over estimation around 10%. The overestimation

Table 2. Errors obtained by applying the different 1D methods.

Method	SCM	DCM	CH	DM	EDM	IDCM	WDCM
Relative depth (-)	Errors in the main channel discharge evaluation (%)						
0.10	38.1	–10.6	–2.9	–5.7	13.2	–3.1	–2.0
0.15	36.3	–8.2	1.1	–0.7	15.8	1.5	2.4
0.20	31.6	–10.4	0.7	–0.3	13.9	1.8	2.5
0.30	22.3	–13.9	–2.0	–2.5	9.1	2.3	2.0
Relative depth (-)	Errors in the floodplain discharge evaluation (%)						
0.10	–94.6	6.3	–1.2	–6.8	–42.7	–12.9	–6.3
0.15	–58.2	11.1	5.2	–1.9	–25.4	–6.2	–0.8
0.20	–34.4	14.8	10.1	2.7	–12.8	–0.8	3.4
0.30	–19.1	11.2	11.2	2.9	–7.3	–2.6	–0.7
Relative depth (-)	Errors in the total discharge evaluation (%)						
0.10	26.8	–9.2	–2.8	–5.8	8.5	–3.9	–2.3
0.15	23.2	–5.5	1.6	–0.9	10.1	0.4	2.0
0.20	17.8	–5.1	2.7	0.3	8.3	1.2	2.6
0.30	8.2	–5.4	2.3	–0.7	3.5	0.6	1.1

occurs also for the calculation of the discharge in the main channel and the opposite in the computation of the discharge in the floodplains. This result is due to the non consideration of the deceleration that the flow of floodplains causes in the main channel flow and vice versa.

With the exception of EDM, all proposed methods improve the results obtained with DCM for total and sub-section discharges. For the calculation of the total discharge, the method with better performance is the Coherence Method, with errors below 2%.

The WDCM is the method that shows a better overall performance presenting errors around 2% for the majority of the situations and for the three flow discharges studied.

As the water depth becomes higher, the results of almost all methods improve, revealing a gradual reduction of the interaction between sub-sections, i.e. a reduction of the effect of large-scale vortices in the momentum transfer.

(a)
(b)
(c)

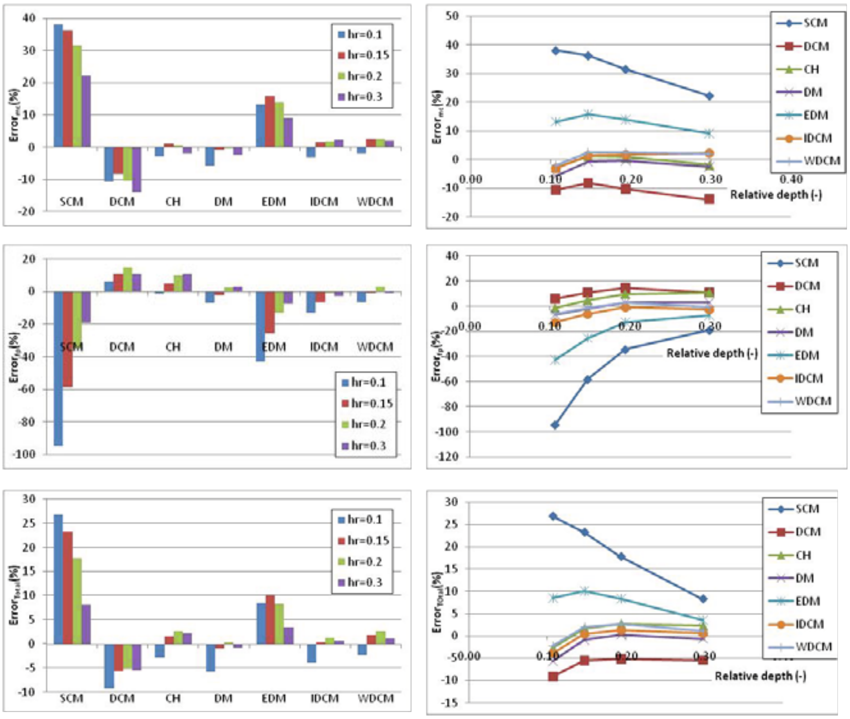

Fig. 5. Error in the calculation of the discharges (a) Main channel; (b) Floodplain and (c) Total.

Conclusions and recommendations

In this paper an experimental study of the flow in a compound channel was presented. The collected data was used to evaluate the performance of several 1D methods available in the literature.

The modelling of the flow was done using 1D methods and the results were compared with the experimental data. The results point out that when the flow overflows the main channel and inundates the floodplains, the effects of the interaction between the main channel and floodplains should be taken into account. Errors when one uses a simple division of the channel are up about 10%. With a greater range of data, [14] points to average errors of around 20%. The errors in the flow distribution between main channel and floodplains are higher and show the need to examine individually the subsection discharge. A relevant aspect is the fact that

DCM, commonly used in commercial models, overestimates the discharge for a given water depth, which goes in the opposite direction of safety. Alternative methods to DCM take into account the interaction between the discharges in each subsection namely the momentum transfer and improve the results of both the calculation of the total and sub-section discharges. For performed tests, the CH and the WDCM showed the better performance. For this reason and because they are simple to implement, their use in engineering case studies should be assessed when the channel or river has a compound configuration.

Acknowledgments

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ANNEX 1. 1D METHODS

Single Channel Method (SCM)

This method does not divide the channel and considerer it as a single channel assuming an average velocity for the whole channel. Using a global roughness coefficient, this method computes the total flow through a flow resistance equation (*e.g.* Manning-Strickler).

$$Q = K\ R^{2/3} A\ S_0^{1/2} \tag{6}$$

In which Q stands for the discharge; K for the roughness coefficient; R for the hydraulic radius; A for the cross section area and S_0 for the slope of the channel.

Divided Channel Method (DCM)

This method proposes the division of the channel in three sub-sections, namely the main channel and the lateral floodplains. The typical division is through vertical lines, where the total flow is given by the sum of sub-section discharges (*cf.* Eq. 7).

$$Q = \sum_i Q_i = \sum_i K_i R_i^{2/3} A_i S_0^{1/2} \tag{7}$$

where the index i indicates each subsection.

Coherence Method (CM)

The Coherence Method was developed by [15] and it improves the results of the DCM, making it the most appropriate for compound channel flows. This method uses two coefficients for the adjustment of the sub-section discharges. The coherence (COH) is the relationship between the discharge given by the SCM and the DCM (*cf.* Eq. 8).

$$COH = \frac{Q^{SCM}}{Q^{DCM}} \tag{8}$$

The closer to 1 is this coefficient, the more appropriate is to treat the channel as a single one. When this coefficient is significantly less than 1 it is necessary to apply a different coefficient, called DISADF in order to correct the discharge in each subsection. An analysis of the experimental results has split the flow in 4 regions according to the relative depth of each one (*cf.* Fig. 6).

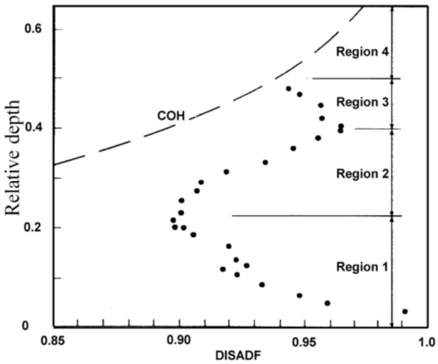

Fig. 6. DISADF coefficient.

Reference [15] presents the formulas for computing the DISADF in each flow region. The discharge is then obtained by the following equations.

$$Q = Q^{DCM} - DISDEF \qquad \text{For flow region 1} \tag{9}$$

$$Q = Q^{DCM} \times DISADF \qquad \text{For flow region 2 to 4} \tag{10}$$

Where DISDEF is a factor called discharge deficit which calculation procedure can be found, for example, in [16].

Debord Method (DM)

The Debord Method (Formulation simplifiée Debord in the original french designation) proposes the correction of the results obtained by the DCM [17]. The basis of the correction is a set of experimental tests conducted with 16 different configurations. In those tests, the flow in the compound channel was compared with the flow in the independent sections (vertical separations were placed in the interface). [17] concluded that the relationship between these flows depends mainly on the relationship between the roughnesses of each subsection. This method models the discharge in each subsection with the Eqs. (11) and (12).

$$Q_{mc} = \varphi K_{mc} R_{mc}^{2/3} A_{mc} S_0^{1/2} \tag{11}$$

$$Q_{fp} = \sqrt{1 + \frac{A_{mc}}{A_{fp}}\left(1 - \varphi^2\right)}\, K_{fp} R_{fp}^{2/3} A_{fp} S_0^{1/2} \tag{12}$$

In which φ stands for the experimentally coefficient given by:

$$\varphi = \varphi_0 = 0{,}9\left(K_{mc}/K_{fp}\right)^{1/6} \qquad \text{for } \mathrm{R_{lc}}/\mathrm{R_{lp}} > 0{,}3 \tag{13}$$

$$\varphi = \frac{1}{2}\left[\left(1-\varphi_0\right)\cos\left(\frac{\pi\ R_{fp}/R_{mc}}{0{,}3}\right)+\left(1+\varphi_0\right)\right] \qquad \text{for } 0 < \mathrm{R_{lc}}/\mathrm{R_{lp}} \le 0{,}3 \tag{14}$$

Exchange Discharge Method (EDM)

This method is based on the concept of the apparent shear stress. The basis of this method is the integration in the cross section of the equation of momentum conservation. After some simplifications and mathematical operations this equation could be written for the main channel and for the floodplains as showed in Eq. 15 and 16.

$$\rho \cdot g \cdot A_{mc} \cdot S_o + \left(h_{\mathrm{int},rig} \cdot \tau_{\mathrm{int},rig} + h_{\mathrm{int},lef} \cdot \tau_{\mathrm{int},lef}\right) - \tau_o \cdot P_{mc} = 0 \qquad \text{Main channel} \tag{15}$$

$$\rho \cdot g \cdot A_{fp} \cdot S_o - h_{\mathrm{int}} \cdot \tau_{\mathrm{int}} - \tau_o \cdot P_{fp} = 0 \qquad \text{Floodplains} \tag{16}$$

In which ρ stands for the density of water; g acceleration due to gravity; h_{int} – interface height; τ_{int} – Boundary shear stress; τ_0 – Apparent shear stress; P – wet perimeter; "rig" – right; "lef" – left.

Modelling the boundary shear stress it is only necessary to know the value of the apparent shear stress to calculate the rating curve of a compound channel.

EDM models the "momentum transfer due to turbulence" through a model similar to the mixing layer model [18], obtaining Eq. (17) for apparent shear stress.

$$\tau_{\mathrm{int}} = \frac{1}{2}\psi\rho\left(U_{mc} - U_{fp}\right)^2 \tag{17}$$

In which ψ stands for an experimental parameter and U stands for average velocity in a single subsection.

EDM also models the momentum transfer associated with the geometry (including enlarging or converging main channels), what is outside the scope of this work.

Interacting Divided Channel Method (IDCM)

This method was developed by [10] and it is also based in the apparent shear stress concept (Eq. 15 and 16). This method uses the formulation of [19] to model the momentum transfer in the interface, obtaining the Eq. (18).

$$\tau_{\text{int}} = \frac{1}{2}\gamma\rho\left(U_{mc}^2 - U_{fp}^2\right) \tag{18}$$

In which γ corresponds to a coefficient, having been obtained from experimental results collected in literature ([10] suggest 0.02).

Weighted Divided Channel Method, WDCM

The Weighted Divided Channel Method was developed by [13] and it is based on the observation of the velocity distributions in the main channel and floodplains. This method consists in a correction of the DCM in order to integrate the effects caused by the momentum transfer by a weighting in the results of velocities obtained with vertical and horizontal divisions between the subsections. The equations for the main channel and floodplains are presented below.

$$U_{mc} = \xi U_{mc}^{DCM-V} + (1-\xi)U_{mc}^{DCM-H} \tag{19}$$

$$U_{fp} = \xi U_{fp}^{DCM-V} + (1-\xi)U_{fp}^{DCM-H} \tag{20}$$

In which *"DCM-V"* stands for the results of DCM with vertical divisions; *"DCM-H"* with horizontal divisions and ξ for the weighting coefficient for the WDCM (from the experiments of the authors for equal roughness of the subsections the value of this coefficient is 0.5).

Comparison of the pollutant potential of two Portuguese highways located in different climatic regions

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Abstract

The accomplishment of the Water Framework Directive requires a good understanding of the impacts of the different pollution sources. In the framework of G-Terra study, two Portuguese highways located in different climatic regions from Portugal have been monitored. The role of climatic variables in controlling the presence of 6 selected pollutants in the road runoff was evaluated. The results showed the relevance of the rain depth and the antecedent dry period in the discharge of higher concentrations of pollutants. Highways located in more arid areas, are more likely to produce acute impacts. The Total Suspended Solids and the Chemical Oxygen Demand appear to be important target pollutants to be controlled in Portugal.

Introduction

The accomplishment of the Water Framework Directive in terms of a good ecological status for all water bodies, by 2015, requires a good understanding of the impacts of pollution sources and the control of the most relevant ones. For the case of roads (and other diffuse sources of pollution) it is most relevant that the evaluation of pollutants' concentrations and loads are assessed taking into consideration both the road and the climate characteristics.

Parameters such as the Event Mean Concentration (EMC), Site Median Concentration (SMC) and pollutant load characterize road runoff quality and are useful to understand the potential impacts of road runoff discharged into the environment. Nevertheless these calculations may hide the occurrence of peaks of concentrations. It is known that under different conditions highway runoff may cause not only chronic impacts but acute effects on the chemical quality and ecological status of the receiving water [4, 7].

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When assessing impacts and risks in receiving water masses, attention should be paid to distinguish between acute and cumulative impacts. Typically, short term impacts occur at time scales of less than 1 hour up to 1 day, and are related to hydraulic effects; discharge of biodegradable organic matter or of suspended solids [6].

Road runoff in Portugal showed values of Total Suspended Solids (TSS) and Chemical Oxygen Demand (COD) that surpass the permitted level for discharge of point effluents (Decree-Law 236/98) in 15% and 50% of the samples, for COD, and in 62% of the samples for TSS [1, 2]. Therefore, these pollutants are on focus in Portugal and a good understanding of their genesis and removal process by rainfall is of most importance.

The final goal of an ongoing research project named "Guidelines for Integrated Road Runoff Pollution Management in Portugal", G-Terra, is to characterize road runoff and improve the understanding of the interrelationship between pollutants and specific site variables [3]. In the framework of G-Terra, two Portuguese highways have been monitored in 2008 and 2009: A1 site is located in central Portugal, and A22 is placed in southern Portugal. The two monitored highways are located in very different geographic and climatic regions from Portugal, as shown in Fig. 1.

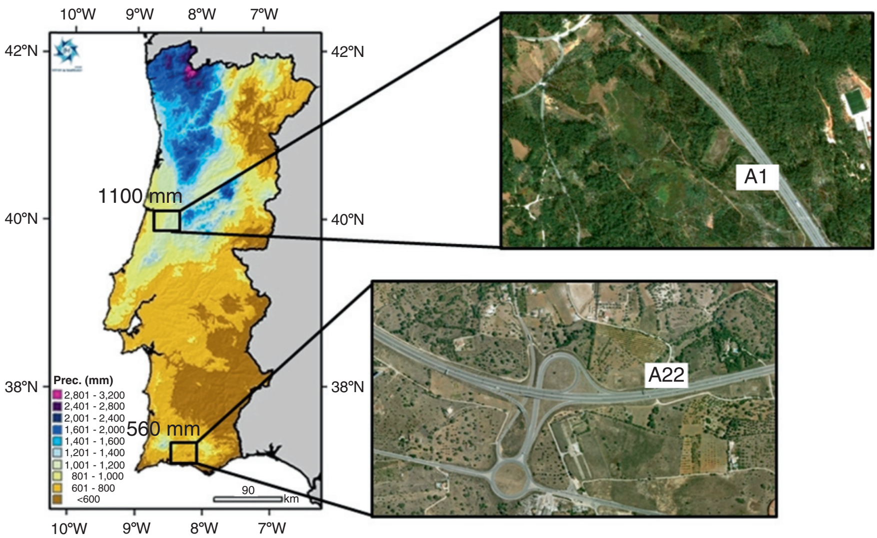

Fig. 1. Location of the two sites and average annual precipitation.

For the monitoring period A1 presented an average annual daily traffic (AADT) of 27746 and A22 of 24000. The studied catchment area is of 22800 m^2 (100% impervious) for A1, and of 15422 m^2 (85% impervious) for A22.

The objective of this paper is to compare the two roads using the results of the monitoring study, and further understand the role of climatic variables in controlling the pollutants presence in road runoff. The sort of impacts that may be expected is evaluate as well.

Methodology

An automatic equipment consisting of a rain gauge, automatic sampler and flow meter, associated to a V-notch weir, were placed at each site, at different times. The monitoring period was of 4 months for A22, and of 2 months and 10 days for A1. The collected data consisted of rainfall and flow (recorded each 5 minutes) and between 6 to 8 discrete samples along 10 or 11 events. The samples were analyzed by contracted laboratories for a total of 18 quality parameters. The laboratories followed standard and certified procedures for analysis and quality control of results. The results of the quantity and quality data were analyzed and the common characterization for the runoff was produced [3].

For the present analysis a set of 6 quality parameters were selected: TSS; COD, Total Organic Carbon (TOC), copper (Cu), iron (Fe) and Kjeldahl Nitrogen (N-Kjel). The selection was based on the following criteria: parameters that represent different sorts of pollution; that were, for both roads, detected and quantified in a significant number of samples, and were observed in concentrations higher than the limits of the referred Portuguese law. Fe is not among the road runoff metals with more toxic effects, but was selected for the last motive.

The variables selected to represent climatic factors were, for each monitored event: Rain depth (Rain Dp); Rain duration (Rain Dr); Rain intensity (Rain Int) and antecedent dry period (ADP). For each quartile of each event (based on the % of runoff volume) the average flow (l/s) and the % of each pollutant mass transported were calculated.

The data analysis was conducted with Excel® and Statistica® tools. The latter was mostly utilized for the multivariate exploratory analysis using Clusters and Principal Component Analysis. To perform the Cluster analysis the variables were standardized, in order to avoid the distortion of the results by the values characteristics.

Results

Characterization of the sites based on climatic variables

To distinguish the climate characteristics of each monitoring site, two meteorological stations were chosen (a minimum series of 20 years was considered for all the parameters) [9]. Fig. 1 showed the variability of the average annual precipitation. The annual precipitation volume for A1 is 1100 mm and for A22 it is 560 mm. For the months of the monitoring period when samples were collected (April, May and June for A1 and January, February and March for A22), the average monthly evaporation was 144 mm and 88 mm for A1 and A22, respectively. During those periods, the average temperature was 17.6ºC for A1 and 12.3ºC for A22.

Table 1 shows the climatic variables for all events; with the exception of event 4 for A22, and event 7 for A1. For the event 7 of A1 a problem occurred with the equipment and the precipitation data was not well characterized. Event 4, in the case of A22, was disregarded because the collection of samples did not cover all the flow. It is known that the definition of a rainfall event has to be in accordance to the use of the data; it can be of few hours to 3 days (e.g., [8]). For this study, the rain events were defined as independence when the period between rain events was longer than the concentration time of the watershed.

Fig. 2 presents the results for the calculations of the average flow (Qm) for each quartile of each event – defined based on 25 % fractions of the event volume.

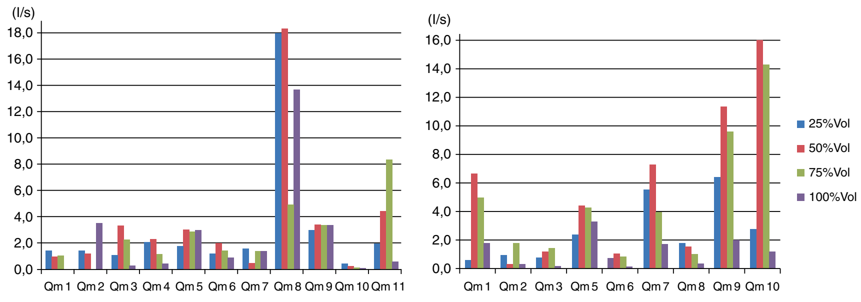

Fig. 2. Average flow (Qm) in each quartile of each of the monitored events at A1 (11 events) and A22 (9 events, due to the exclusion of event 4).

Table 1. Characterization of the precipitation events.

Site	Event	Rain Dp (mm)	Rain Dr (hours)	Rain Int (mm/h)	Run. Coef.	ADP (days)
A1	1	4.6	2.0	2.3	0.04	3*
	2	2.2	0.6	3.8	0.05	4.4
	3	2.8	1.3	2.2	0.15	0.2
	4	2.8	0.8	3.4	0.08	0.8
	5	2.4	0.8	3.2	0.22	0.2
	6	11.2	9.1	1.2	0.29	0.3
	8	16.2	2.8	5.7	0.24	14*
	9	4.2	1.1	3.9	0.15	12.7
	10	3.2	0.3	12.8	0.28	0.7
	11	2.8	0.9	3.1	0.12	1.5
Average (n=10)		**5.2**	**2.0**	**4.2**	**0.16**	**3.6**
A22	1	5.2	3.1	1.7	0.30	10.5
	2	1.8	2.7	0.7	0.14	1.9
	3	1.0	0.8	1.2	0.29	3.6
	5	4.8	2.4	2.0	0.56	1.8
	6	1.4	1.0	1.4	0.18	0.7
	7	8.0	2.8	2.9	0.42	0.9
	8	1.0	0.2	6.0	0.71	0.2
	9	1.4	2.1	0.7	2.00**	2.7
	10	3.4	3.1	1.1	1.00	20.4
Average (n=9)		**3.1**	**2.0**	**2.0**	**0.45**	4.7

* Value estimated based on national data [9] and monitoring records.
** Value inconsistent (probably due to wrong flow measurement) and not considered for the average calculation.

Characterizations of the pollutant concentrations and loads at the two sites

Table 2 presents a characterization of the SMC, pollutant load and maximum observed concentration for the selected parameters. The flow of the event 7 for A1 was estimated based on comparisons of the rainfall data from the local meteorological records [9] with similar rainfall events, with correspondent flow measurements monitored during the study. Table 3 presents the percentage of samples with concentrations of TSS, COD and Fe that exceed the level for discharge of the Decree-Law 236/98. It states also the number of samples (n) for each case.

Fig. 3 and Fig. 4 show the evaluation of the first-flush effect for the TSS, COD, Cu and Fe, for A1 and A22 highways, respectively. Fig. 5 and Fig. 6 illustrate the percentage of mass of TSS, COD, Cu and Fe that is transported in each quartile of the volume for each event, respectively for the case of A1 and of A22.

Table 2. Characterization of A1 and A22 road runoff for selected pollutants.

	A1 (11 events)			A22 (9 events)		
Parameters and roads	SMC (mg/l)	Max. (mg/l)	Poll. Load (kg/ha/yr)	SMC (mg/l)	Max. (mg/l)	Poll. Load (kg/ha/yr)
TSS	22.2	350.0	5956	52.4	220.0	2937
TOC	22.7	72.0	2624	18.4	38.0	1028
COD	81.9	330.0	9475	38.3	226.0	2147
Cu	0.02	0.051	2.3	0.03	0.046	1.4
Fe	0.35	7.192	40.2	1.9	6.627	108.7
N-Kjel	2.0	5.0	230	2.7	10.0	152

Table 3. Percentage of samples of TSS, COD and Fe that exceed the level for discharge of the Decree-Law 236/98.

% and number samples for each case	A1	A22	Level for discharge Decree-Law
TSS	11% (n=71)	30% (n=65)	150 mg/l
COD	13% (n=71)	1.3% (n=75)	60 mg/l
Fe	6% (n=73)	41% (n=76)	2 mg/l

Multivariate Statistical Analysis and Linear Regressions

Cluster analysis showed that the Rain Dp and the ADP are the climatic variables closer to the variables describing the pollutants, for both highways.

The TSS and the Cu, especially their mass, evidenced a closer connection to the Rain Dp; the COD and the TOC EMC and maximum concentrations were closer to the ADP. N-Kjel was the only pollutant included in the same cluster as the Rain Int, for both case studies. The Principal Component Analysis performed did not add further information.

Simple linear regressions were established between the climatic variables and the variables describing the pollutants. The results were in agreement with the evidences from the multivariate exploratory analysis. The presence of pollutants was more correlated with the Rain Dp for A1 and, for A2, with the ADP – especially for COD and TOC. The coefficients of determination (r^2) ranged between 0.507 and 0.833. The lowest respected to N-Kjel correlation with the Rain Int at A1; the highest r^2 concerned ADP and TOC, at A22.

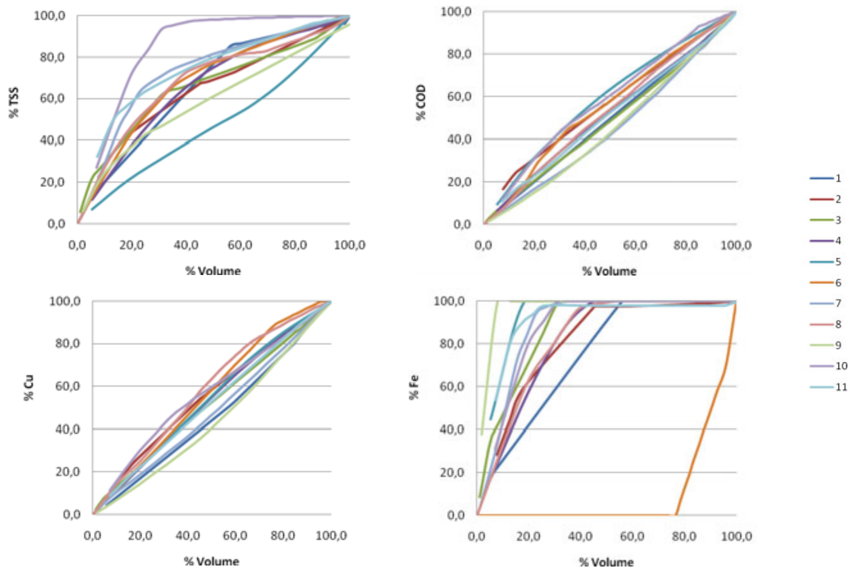

Fig. 3. First flush evaluation for TSS, COD, Cu and Fe for A1 highway.

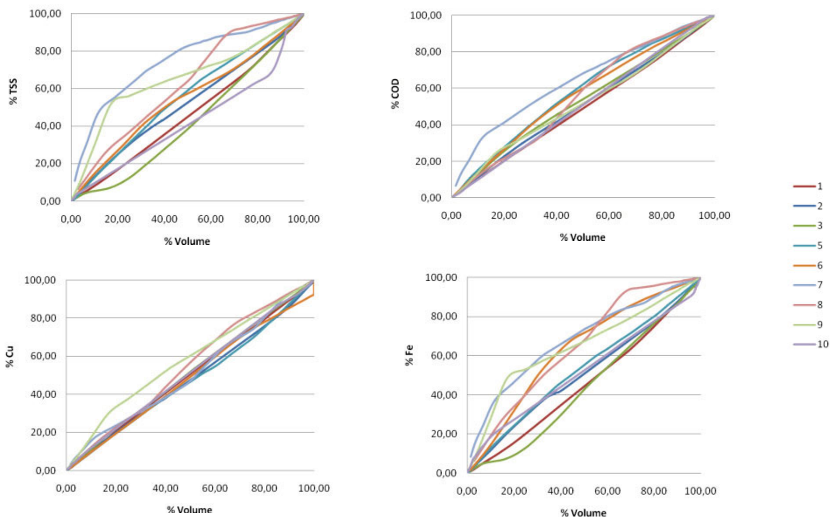

Fig. 4. First flush evaluation for TSS, COD, Cu and Fe for A22 highway.

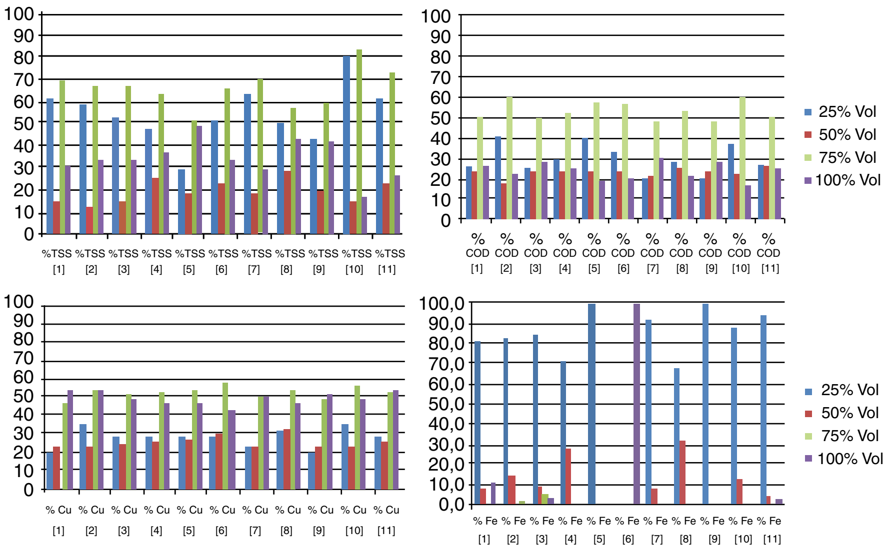

Fig. 5. % of mass of TSS, COD, Cu and Fe transported in each quartile of the monitored events at A1.

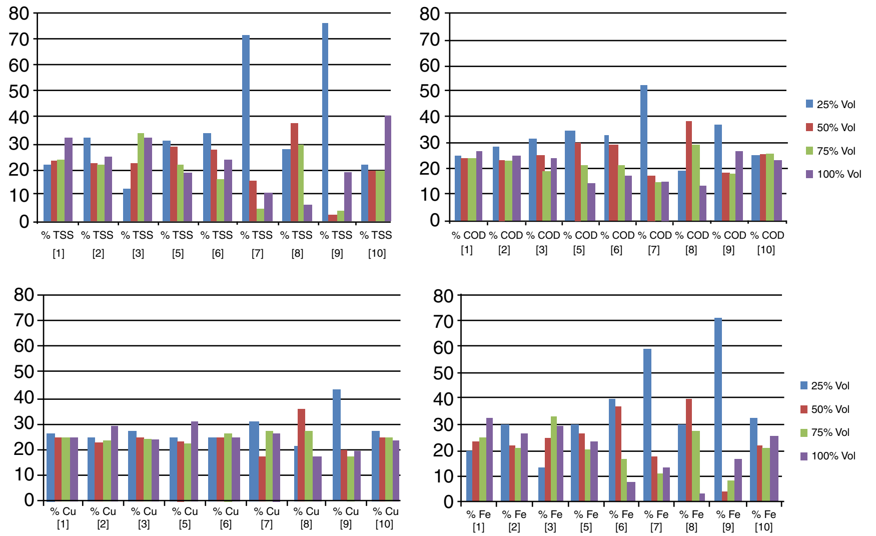

Fig. 6. % of mass of TSS, COD, Cu and Fe transported in each quartile of the monitored events at A22.

Discussion

It is clear the different climatic characteristics for the two sites analyzed (Table 1). The ADP in average is higher for A22 (4.7 days, compared to 3.7 days in A1). The average Rain Dp, on the contrary, higher at A1 site (5.2 mm against 3.1 mm).

These were the two variables showing a closer association to the presence of pollutants in highway runoff for both cases, in agreement with the variable that is stronger for each site. Other authors reached to exactly the same conclusions [5] or these variables are among the ones showing relevance [7]. In average the runoff coefficient is much higher for A22 than for A1 which should be a result of a smaller drainage area and lower temperature for the monitored period. Table 2 data gives evidences that, although for most pollutants A1 is responsible for higher annual loads than A22, the latter may have maximum concentrations almost as high as A1 (or higher, for the case of N-Kjel). Table 3 supports this assumption.

The results concerning the first-flush evaluation (Fig. 3 and Fig. 4) showed for COD and Cu similar curves for each case study, with a slight first-flush effect. For the case of the TSS there are more variations among the events, and the first-flush effect seems more enhanced. The curves for the Fe case are similar to the TSS, for highway A22; for A1 the pattern is sharper; most of the events show a quick first-flush effect.

Comparing the average flow (Fig. 2) and the relative mass of pollutant transported in each quartile of each event (Fig. 5 and Fig. 6), it is clear the absence of direct relations among these variables, for any of the case studies. For instance, A22 has a % of transport of Cu and TOC homogeneous along the event, not showing any response to the higher average intensity for the second and third quartile of the event’s volume.

The characteristics of the catchment (area, slope and concentration time) should be, at least in part, responsible for these observations.

Conclusions

The objective of the study undertaken was not to further explore the analysis of two case studies or quantifying relationships among variables, but to gather evidences concerning the role of climatic variables in controlling the pollutant potential of roads. It is also considered that the amount of data is not sufficient for characterization of all seasons, which may provide rain depths, intensities and interevent dry periods with different characteristics. Therefore, the conclusions that can be made are of a broader sort – but considered to be very relevant in assessing and controlling the impacts of road runoff.

There is evidence that in Portugal a special focus should be placed on controlling TSS, COD and Fe in road runoff, due to at least two of the following reasons: high EMC; high maximum concentrations and potential for first-flush occurrence. Highways located in more arid areas of the country (with longer ADP) are more likely to produce acute impacts in short time durations, because of their potential to discharge higher concentrations of these pollutants.

It should be referred that the runoff from the two studied road catchments are discharged into treatment systems; therefore none of them should cause environmental impacts.

Acknowledgments

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Road runoff characteristics on costal zones – Exploratory Data Analysis based on a pilot case study

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Abstract

Previous studies have demonstrated that the road runoff characteristics in coastal zones show a different pollution pattern, with higher levels of salinity and organic matter concentration.

The specific pollution pattern of these roads is being studied, based on the monitoring of a Portuguese Highway, located at a distance of 5km from the sea.

Based on the preliminary monitoring data, correlations of the main variables where evaluated, in order to select the methodology for statistical analysis of the global monitoring data. The main conclusions of this exploratory data analysis are also presented.

Introduction

Road runoff is a linear source of diffuse pollution that can cause significant environmental impacts. The pollutants result from both mobile and stationary sources. Stationary sources include the degradation of the road platform, erosion of embankments and the degradation of urban infrastructures. Mobile sources are due to the road traffic (including tire and brakes wear, oils and fuels leakages, deteriorating of paints and fuel emissions). Operations of road conservation, including the application of pesticides and fertilizers, and litter, may also contribute also contribute to this type of pollution [1].

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Previous monitoring studies on road runoff [2-4], from two different sites (highways A25 and IP6), located on coastal zones of Portugal have detected high levels of salinity and Chemical Oxygen Demand (COD).

On the other hand, pollutants typical for road runoff characterization, such as total suspended solids (TSS) and the heavy metals zinc (Zn), copper (Cu) and lead (Pb) clearly indicate different levels of pollution.

Such evidences have been related to the proximity of the Atlantic Ocean [3,5], since the non-coastal Portuguese roads did not show this pattern.

In coastal zones, such as found in Portugal, maritime salts transported by the atmosphere are deposited and may remain in the road pavement and crystallize during dry periods between rainfall events and, eventually, lead to the degradation and extraction of the superficial layer of the pavement. Such phenomenon may increase the road pavement potential for environmental pollution, e.g. contributing to increase the refractory organic matter levels.

Marine aerosol can be transported to large distances [6]. In areas with high salinity and where evaporation is systematically superior to the precipitation, the salts crystallization at the top surface of pavement materials tend to provoke a higher degradation of the road pavement [7], with relevant structural consequences to road engineering. The bituminous material adhesion is reduced and small particles may be released to the road base destroyed.

To provide a consistent answer to the question: "*Which are the particular characteristics of road runoff in coastal areas*?" and to establish a conceptual model for coastal roads pollutants are two of the tasks of the project *Guidelines for Integrated Road Runoff Pollution Management in Portugal* (Project G-Terra), a three years study that started in January 2008, funded by the Portuguese Science and Technology Foundation.

The diagram presented in Figure 1 describes the conceptual processes and the leading variables that could be more relevant in understanding the related phenomena.

The analysis of the relations between these special case studies can provide valuable information for the comprehension and understanding of the associated phenomena.

Statistical exploratory techniques will be used in order to evaluate the relationship between the pollutants and the help to understand the most relevant relationships between variables.

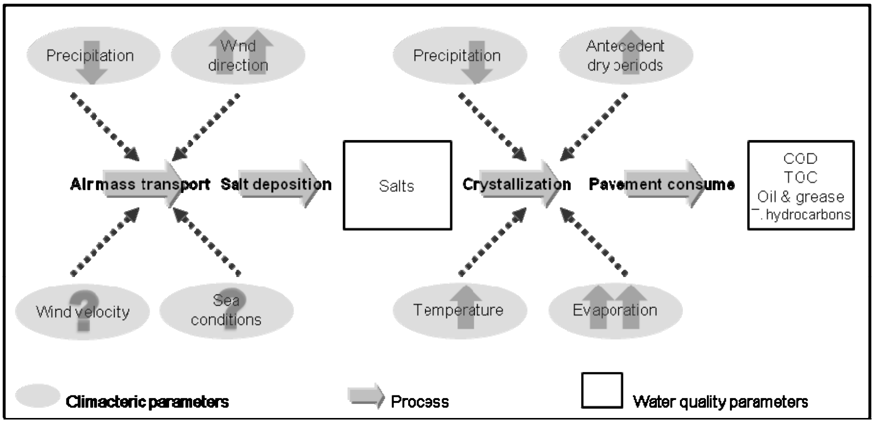

Fig. 1. The conceptual processes and the leading variables.

Multivariate exploratory statistical techniques, as principal component analysis, where applied to a set of coastal and non-coastal Portuguese roads. The result of this analysis indicated different patterns for the two groups, in terms of salinity, conductivity and chlorides, as expected, but also of COD concentration [3].

In fact, possible correlations among road runoff quality parameters namely, salinity, chlorides, TSS, CQO, Total Organic Carbon (TOC), total hydrocarbons and oil and grease and independent variables are being studied. The independent variables belong to three different groups: i) rainfall event characteristics; ii) air masses transport and salt deposition, and iii) road site characteristics.

Within the five case studies of the G-Terra project, the A25 site (Table 1) was chosen as the pilot case study, concerning the effects of atmospheric salt deposition on highway runoff characteristics. Four monitoring periods (in the four seasons) are planned at the A25 site, during 2008 and 2010.

It is expected to gather data from around 40 different rainfall events, under various climacteric conditions and, approximately, 300 runoff samples.

Methodologies

Field Monitoring

In February 2008, the field monitoring system was installed. It consisted of a rain-gauge, a flowmeter (ISCO 730 Bubbler flow module), with a Thel-Mar volumetric weir, and an automatic water sampler (ISCO 6700), working in synchronization. Figure 2 presents this system, at the monitoring site.

A wet candle device methodology [8] was selected to determine the atmospheric chloride deposition rate (amount of chlorides salts deposited from the atmosphere on a given area per unit time).

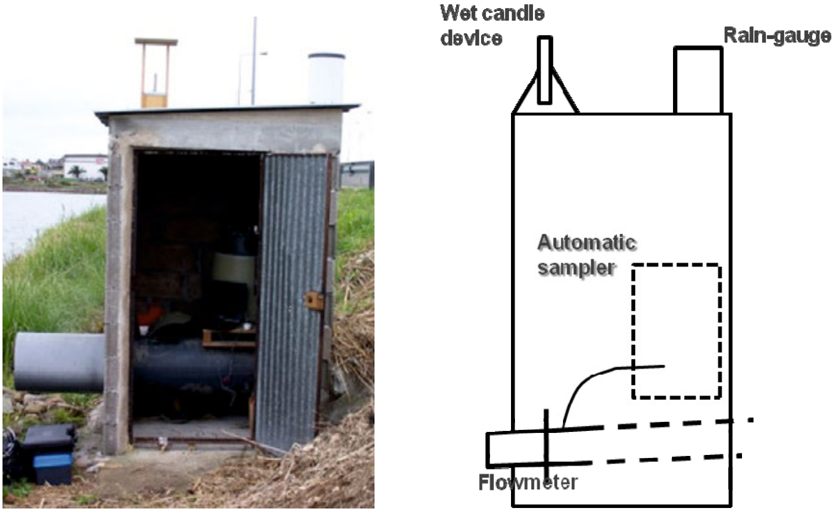

Fig. 2. Photo and scheme of monitoring system at the A25 site.

Table 1. Main characteristics of the highway site studied.

Location	Next to Aveiro city (75,000 inhabitants)
Distance to the Atlantic Ocean	5,6 km
Monitored catchment area	250 m^2 (totally impervious)
Annual Average Daily Traffic	27000 vehicles (2003) 2935 vehicles (May, 2008)
Other site relevant characteristics	The highway crosses, few meters above it, a coastal lagoon (ecological sensitive), with high salinity

A second wet candle device was placed in The Meteorological Station of Aveiro University, located approximately 2 km to the East from the monitoring site. Continuous meteorological and atmospheric data at this site was also collected and analyzed. This data will be useful to explain the air mass transport and salt deposition in the road platform.

A total of 20 different rainfall events were already monitored and 143 runoff samples were collected. The sampling routine was trigged by the runoff flow. About 8 samples were collected during each event, with an average duration of approximately two hours.

Laboratory analysis

The samples were transported to the laboratory as soon as possible and, the preparation, preservation and analysis procedures were followed in accordance with international standards procedures [9], based on an adequate Quality Assurance and Quality Control plan establish for G-Terra Project. The quality parameters analyzed for the runoff studies where: temperature, pH, conductivity, turbidity, Total Suspended Solids (TSS), total hardness, Total Kjeldahl Nitrogen (TKN), total phosphorus, Biochemical Oxygen Demand (BOD_5), total Fe, total Zn, total Cu, total Pb, total Cd and total Cr. Beside these traditional parameters, COD, TOC, oil and grease, salinity and chlorides where also considered.

Exploratory Data Analysis

Based on the qualitative data from the monitored events and the Meteorological Station of the Aveiro University, the following objectives are being analyzed:

- Hydrologic characterization of rain events; water quality parameters concentrations, including Event Mean Concentration (EMC) and Site Mean Concentration; pollutant loads; firsts-flush verification.
- Analyze of the monitored data: minimum and maximum detected values, mean, and standard deviation. For each pollutant, the obtained results where compared with those from the other monitored sites.

In order to understand the relationship between the main variables, and by this way support the understanding of the processes, an exploratory data analysis with different statistical techniques, was done. These correlations can also contribute to future modeling of the phenomenon. The methods that where considered include: Spearman Correlations, *Clusters Analysis*, Principal Component Analysis (PCA); Factorial Analysis.

In different ways these methods highlight the qualitative and/or quantitative possible correlations between parameters (in terms of EMC's).

Beside this analysis, correlation between parameters (in terms of EMC) and the following groups of independent variables where also done:

- Conditions of the event precipitation (e.g. rainfall, maximum intensity of rainfall, cumulative rainfall and antecedent dry period);

- Specificity of the monitored sections (e.g. average daily traffic, contributing watershed area and impervious fraction);
- Air mass transport and other processes related to salt deposition, including the dependency of these with the previous weather phenomena such as temperature, humidity or evaporation.

The data analysis was performed with the use of Microsoft Excel and the Statistical Package *SPSS* for the more complex analysis.

Results and discussion

In terms of G-Terra Project a total of 80 rain events, from five monitoring sites, are going to be monitored. The results from the first 20 monitoring events at the A25 monitoring site are presented and discussed. For these monitoring campaigns, Table 2 presents the summary statistic of the major parameters concentrations (related to the objectives of the research).

Table 2. Summary statistics of highway runoff quality in A25 site, from the winter 2004; autumn 2009 and winter 2010.

Parameter	nº samples	Average	Median	Range of values		St. Dev
				min.	max.	
Conductivity (µS/cm)	143	344.5	257	58.6	970	220.7
Salinity (mg/l)	143	178.6	115	50	520	112.6
Turbidity (FNU)	143	34.3	24.6	2	118	26.4
TSS (mg/l)	143	60.4	33	1.5	642	86.6
Total Hardness (mg $CaCO_3$/l)	143	77.5	70	3	208	36.6
Chlorides (mg/l)	135	74	43.1	1.3	370.9	77.9
Total P (mg/l)	143	0.8	0.3	-	7.4	1.4
COD (mg O_2 /l)	143	92.8	69.4	3.3	375	77.9
BOD_5 (mg O_2 /l)	108	12.6	5.9	-	90	14.4

These results confirm high levels of organic matter in terms of COD, salinity and related parameters.

In terms of general climacteric conditions, that may influence the salinity/organic matter levels, the monitoring periods were performed during the autumn and winter seasons. The salt deposition rate at A25 site is very high, With average values of 49,27 mg/m^2.day, representing higher values than those obtained in a monitoring station (average of

23,95 mg/m^2.day) located 2 km farther from the coast line, as referred by [10]. The atmospheric chloride deposition rate values, when compared to data reported in several studies [11-14], show clear evidences of the presence of salt. These results are justified by the short distance of the monitoring site to the Atlantic Ocean, but also due to the road platform altitude, nearly corresponding to the sea level, and the absence of significant obstacles to the transport of the marine aerosols.

The exploratory data analysis is ongoing and will only be concluded after the end of the monitoring period. In order to evaluate the different data analysis methods an exploratory statistical analysis was done with the available data. This preliminary analysis will help to understand the studied phenomena and relevant process. Therefore, based on the results of the analytical data from 20 of a total of the planned 40 events, the applicability of several statistical methods where evaluated in order to select the most promising techniques for the implementation of this research project. Based on this preliminary analysis, the conclusions where:

The Spearman Correlations are the most useful method to establish a correlation with different data groups.

To identify surrogate constituents within parameters, at first attempt, Spearman correlations where made, selecting pairs with Spearman coefficients elevated. The strongest correlations were observed between parameters associated with dissolved minerals (salinity, conductivity, hardness and chloride); organic carbon (TOC and DOC) and particulate matter (TSS and turbidity). Within the metals category, total iron concentration was highly correlated with most total metal concentrations.

As suspected, the strongest correlation was found between pairs of salinity (or related parameters) with organic matter, especially in terms of COD. The BOD_5 concentration is independent from the salt deposition.

With lack of linearity, Spearman Correlations don’t permit to establish solid correlations between EMC (e.g. salinity or COD) and process related to salt deposition. Although some variables, as event antecedent dry period, could be linked to higher levels of COD, using this method.

Principal Component Analysis (PCA) is a multivariate statistical technique that permits to reveal data patterns, especially when the variables are measured in very different units and scales. This can be applicable to foreseen relations within parameters and between them (in terms of EMC) and other pertinent variables to the phenomenon.

It is seen the Antecedent Dry Period and Rain Depth have a great correlation with salinity and COD EMC’s, as suggested by [3].

Clusters Analysis is a more complex method with a higher reliability, that confirm the correlations observed between parameters.

Factorial Analysis demonstrates to be reliable to evaluate quantitative coefficients between EMC (especially salinity and COD) and other variables (event conditions; site characteristics; air mass transport/salt deposition).

Factorial Analysis also permits to establish links between exploratory data analysis and the modeling by Multivariate Analysis, gathering coefficients to the variables relevant to EMC.

Conclusion

The influence of salt deposition on road runoff from coastal zones is being studied, in terms of the influence of salinity on organic matter concentration.

This study will be based on the monitoring of 40 road runoff events at the A25 Portuguese highway, at a distance o x km from the coastline. Based on the results obtained from the previous 20 events, in the wet period, between October and February, preliminary statistical analysis where done in order evaluate the results, estimate the relationship between the main variables and select the most promising techniques for the project methodology, with all the analytical data.

The highway runoff monitoring procedures included a automatic water sampling equipment, with rain and flow measurements. A wet candle device was selected for the G-Terra studies as a sound methodology to determine the atmospheric chloride deposition rate.

The results for the salinity accumulation rates clearly demonstrate the occurrence of atmospheric salt deposition at the A25 site.

The initial outcomes seem to confirm high levels of salinity, organic matter and related parameters in the A25 highway runoff.

Diverse Exploratory data analysis methods were tested. Simple methods, as Spearman Correlations, resulted as a good method for the verification of qualitative relations between water quality parameters. For example, they confirmed relations between salinity and organic matter.

More complex and reliable methods, as PCA and Clusters Analysis are capable to establish these correlations inter-parameters, but also to correlate EMC's and phenomenon variables associated to event precipitation; site characteristics and air mass transport and salt deposition.

Factorial Analysis also allowed establishing quantitative correlations in terms of coefficients between the considered groups of variables and dependent variables EMC, in terms of salinity and COD. These conclusions could be interesting for further modeling studies, based on multivariate analysis.

It is believed that the adopted methodologies are adequate for the study purpose. Therefore, it is expected that the data of the monitored events, completed with the data from the next three monitoring periods at the A25 site will provide enough data for simple and advanced statistical analysis that will support the identification of the particular characteristics and processes, concerning road runoff in coastal areas.

If the phenomena associated to the high salinity in road runoff of coastal areas are better understood and quantified, this would represent a new and valuable contribution to the present knowledge, and by this way provide valuable information for highway runoff management in countries like Portugal, with a significant coast line.

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Characterization of road runoff: A case study on the A3 Portuguese Highway

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Abstract

Road runoff is a linear non-point source of pollution with a significant impact on water receiving bodies, but whose importance has been neglected in the past. The pollutants in road runoff can be responsible for both acute and chronic effects.

Due to the processes involved in its formation, but also due to its intermittent nature, the characterization and prediction of road runoff quality is still a subject with limited scientific knowledge. Since the European Water Framework Directive demands a good ecological status for natural water bodies, there is an urgent need for controlling this source of pollution. The study of the highway runoff pollution load mass, the inter-event distribution and the estimation of event and site mean concentrations could contribute to increase the knowledge of the related phenomena, estimate its impacts and, therefore, contribute for the definition of mitigations measures for water resources protection or the enhancement of their quality.

In Portugal there is still a reduced knowledge on road runoff pollutants and also a lack of established procedures for the control of this source of pollution. A research project "Guidelines for Integrated Road Runoff Pollution Management in Portugal (G-Terra)" is evaluating, at a national level, road runoff pollution loads and distribution, in order to establish guidelines for road runoff assessment and management.

This paper describes the methodology and monitoring procedures of a case study on the A3 highway, at Santo Tirso, in the north of Portugal. At this site the average daily traffic is 43,000 vehicles. A total of 10 runoff events where monitored through discrete samples, taken along the

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duration of the runoff. The road site characteristics and the pollutants concentration in the different runoff events are presented and discussed.

Introduction

The degradation of soil quality and receiving waters (surface and groundwater) caused by highway runoff is a matter of growing concern and motivates the need to establish discharge limits and standards for their protection.

Previous research has demonstrated that road runoff pollution is related to the following sources [1,2]: (i) combustion of fuel, (ii) wear of vehicle components, (iii) dumping of substances in normal vehicle use; (iv) wear and degradation of the road components; (v) wear and degradation of the platform materials, (vi) application of chemical substances in repairs and / or maintenance operations, (vii) accidental spills and leaks, (viii) drag of pollutants present in the same drainage basin.

As result, a high number of pollutants are found in road runoff. Due to their nature and environmental importance, the most important groups of pollutants are sediments, heavy metals, organic compounds and nutrients [1]. These pollutants can originate in the receiving environment, acute or chronic effects [3]. The pollutants effects are dependent on the specific characteristics of the receiving environment, a complex system that varies with each site.

The flow discharged should also be predicted, since it may be the responsible for downstream damages.

In particular, heavy metals and organic micropollutants appear associated with car traffic. Cu, Pb, Zn and Cd - sometimes also Ni and Cr - are considered the most important heavy metals associated with either the mobile sources or stationary, these metals may come in the form dissolved or particulate bound [2,4].

The monitoring of road runoff is complex and, the use of different monitoring methodologies does not allow to compare the data at different sites and, therefore, to understand the relations between different sites.

In Portugal, the disposal of sewage in water and soil is regulated by standards and criteria. Quality objectives were established, in order to protect the aquatic environment and improve water quality. Sensitive areas were also defined and specific discharge rules are applied for this environment scenario.

Although the legislation already considers this road runoff as wastewater, no mandatory standards have been defined for their discharges. The impacts on ecosystems from these effluents have been primarily associated with their quantitative nature such as floods and erosion, although a brief reference to the deposition of sediments can be found.

The reason behind the lack of specific disposal regulations for this type of runoff is that, at the date of the legislation, the country was still trying to complete the urban water supply and wastewater drainage networks and, therefore, these subjects were the main focus of the legislation.

After nearly two decades, road runoff has established itself as a not negligible source of diffuse pollution and new standards must be defined in order to protect the receiving waters and public health. Recent monitoring studies have found that, for some pollutants, highway runoff concentrations can be higher than the standards allowed for urban wastewater discharges.

In order to study the phenomena associated with road runoff, a three year research project, funded by the Portuguese Foundation for Science and Technology, has began in January of 2008.

The Project "Guidelines for Integrated Road Runoff Pollution Management in Portugal (G-Terra)" has the participation of three research institutions: National Laboratory of Civil Engineering, University of Minho and Escola Superior de Tecnologia de Viseu.

The specific goals for this project are:

- Analyze the origin and presence of pollutants in road runoff and establish relationships with the specific characteristics of the roads, at regional and national levels;
- Understand and model the phenomenon of pollution runoff from coastal road;
- Provide a database with a consistent set of national data on roads and their runoff. This database can be used for forecasting, research and finding new solutions to control this type of pollution;
- Establish and foster a national network of researchers and strengthen the national knowledge in this particular research topic;
- Provide guidelines for better management of road runoff in the attainment of National and European legislation.

Five roads where selected as case studies for the monitoring and study of a wide range of pollutants. These data, as well as all other national knowledge will be processed within the G-Terra project, using statistical tools to better understand the monitored data and to find relationships between pollutants, the road characteristics and the site climatic characteristics. The results of this analysis will also be used to establish methodologies for pollution control in Portugal, which will be presented in a book at the end of the study.

The major motivation behind this study was to gather a more representative set of data characterizing the water runoff and lead to a national analysis of these results, this will provide a comprehensive and overall study, at a higher level than what has been done to the present date, allowing a better and sustainable management of this type of pollutant source. Furthermore, there are still issues in innovative research with potential to generate new scientific and technical knowledge.

The project deliveries also include a open database containing the results of monitoring of 5 sites in Portugal; a project website and a book containing guidelines for a better management of road runoff in terms of compliance with the Water Framework Directive and the Portuguese Water Law objectives.

Methodology

Site Characteristics

The monitoring site is in the A3 highway, near Santo Tirso, in the North of Portugal. This site had, in 2008, an average of 43,000 vehicles per day. The drainage basin is exclusively from the highway pavement, with an area of 2000 m^2. It is also important to detail that the catchment is only on one side of the highway; witch has a decreasing longitudinal slope.

Figure 1, depicts the case study site and details of the monitoring installation.

Fig. 1 Monitoring site location and details of the monitoring site.

Monitoring equipment and procedures

The monitoring system included a rain gauge, a ISCO 730 bubbler flow module, a Thel-Mar volumetric weir and an ISCO 6712 automatic water sampler who controlled all the information obtained from the other devices.

A wet candle device was also used, in order to estimate the atmospheric chloride deposition rate (amount of chlorides salts deposited from the atmosphere on a given area per unit time). This information will be compared with other monitoring sites, at different distances from the Atlantic Ocean, for future analysis of the chloride effect on highway runoff characteristics.

A rain gauge was installed and the sampler trigger was done by a flow meter. The ISCO 6712 automatic sampler collected 8 different samples, with 1.8L, for each event. A minimum dry period was 2 day was considered and, before each sample was collected a rinse of the influent pipe was made. During each event, the flow was measured by means of a weir and a bubbler flow meter. In order to characterize the different events, 8 samples of each event were collected. Since several authors report a higher pollutant load in a first stage of the event [5,6], a higher sampler frequency was considered at this period, to better characterize the first flux phenomena. A GSM modem was used to send alarms to cellular phones and by this way minimize the number of travel to the monitoring site and minimize the time between the monitoring and the deliver of the collected samples to the laboratory.

Between October of 2008 and January of 2009, 10 runoff events were monitored. For each event, in order to better characterize the inter-event phenomena, a total of 8 samples where taken. After the first sample, at the trigger conditions, the other samples where obtained at 10, 20, 40, 60, 80, 100, 120 minutes.

The samples where collected in October (14^{th} and 24^{th}); November (7^{th} and 28^{th}); December (3^{rd}, 5^{th}, 31^{st}) and January (12^{th}, 18^{th} and 23^{rd}).

The following water quality parameters where analyzed: temperature, pH, conductivity, turbidity, total suspended solids (TSS), total hardness, salinity, chloride, total Kjeldahl nitrogen (TKN), total phosphorus, chemical oxygen demand (COD), total organic carbon (TOC), oil and grease, biological oxygen demand (BOD), total Fe, total Zn, total Cu, total Pb and total Cr.

The collected samples were transported to the laboratory as soon as possible, and the preparation, conservation and analysis procedures were followed in accordance with international standards procedures [7], based on an adequate Quality Assurance and Quality Control plan establish for G-Terra Project.

Results and discussion

The analytical results of the 80 samples obtained from the 10 runoff events are presented and discussed. Oil and grease present a maxim value of 12.97 mg/L while the maximum content for total hydrocarbons was 6.80 mg/L (Table 1). These maximum values do not correspond to the same event. Table 1 presents the values obtained in the different events. It has been noticed that the first five events have consistent high values than those for the other events.

Table 1. Oils & Grease and Total Hydrocarbons.

Event	Hydrocarbons (mg/L)			Oil & Grease (mg/L)		
	Min.	Max.	Average	Min	Max	Average
1				0.00	0.00	0.00
2	0.40	3.87	1.02	0.67	6.80	1.64
3	0.52	1.22	0.83	0.79	2.38	1.30
4	0.00	12.97	3.66	0.10	1.78	0.74
5	0.26	1.34	0.54	0.32	2.15	0.80
6	0.17	0.35	0.23	0.18	0.51	0.29
7	0.20	0.60	0.48	0.20	0.88	0.61
8	0.30	0.50	0.39	0.30	0.70	0.45
9	0.20	0.40	0.30	0.03	0.60	0.33
10	0.20	0.30	0.29	0.20	0.40	0.30

The following total heavy metals where analyzed: Zn, Cu, Pb and Cr. Zn was the metal with higher concentrations, followed by copper, confirming the results obtained in other monitoring studies. The average values of these metals were from 0.04 to 0.45 mg Zn/L and 0.01 to 0.05 mg Cu/L, respectively. The concentrations of Lead and Chromium in the collected samples are lower than the obtained for the previous metals, with mean values of 0.40 to 26.58 µg Pb/L and 0.05 to 1.61 µg Cr/L.

Table 2 presents the minimum, maximum average values for these heavy metals in all events. In all events the pH varied from 5.90 to 7.12 confirming the low acidific nature of these effluents. The total hardness varied from 6.73 to 45 mg $CaCO_3$ /L (Table 3).

The total suspended solids (TSS) determined at the laboratory present low values (Table 4). The maximum concentrations for the 10 events varied between 6.80 and 44.40 mg/L. The average event values are always less than 10.80 mg/L. The reason for the low values is being evaluated and may result from the decreasing slope of the highway platform, where low traction is required.

Table 2. Heavy Metals.

Event	Zinc (mg/L)			Copper (mg/L)		
	Min.	Max.	Average	Min	Max	Average
1	0.04	0.61	0.27	0.00	0.08	0.04
2	0.28	0.48	0.33	0.04	0.06	0.05
3	0.24	0.44	0.32	0.03	0.05	0.04
4	0.34	0.66	0.45	0.03	0.05	0.04
5	0.14	0.42	0.20	0.01	0.03	0.02
6	0.01	0.02	0.02	0.01	0.03	0.02
7	0.14	0.37	0.20	0.02	0.04	0.03
8	0.00	0.63	0.19	0.01	0.04	0.02
9	0.00	0.30	0.04	0.01	0.01	0.01
10	0.10	0.14	0.12	0.01	0.01	0.01
Event	Lead (μg/L)			Chromium (μg/L)		
	Min.	Max.	Average	Min	Max	Average
1	0.00	24.60	4.44	0.00	3.10	0.63
2	2.00	122.00	26.58	0.00	4.60	1.03
3	1.00	7.40	3.98	0.00	2.10	0.42
4	0.00	4.95	1.03	0.00	1.20	0.56
5	0.50	8.90	2.31	0.00	2.80	0.79
6	1.40	5.20	3.10	0.10	9.20	1.61
7	0.10	17.70	3.66	0.30	0.80	0.48
8	0.20	27.70	7.59	0.10	1.50	0.59
9	3.60	11.50	5.90	0.00	0.40	0.05
10	0.00	1.80	0.49	0.00	0.60	0.21

Table 3. Physical/Chemical parameters.

Event	pH			Total Hardness (mg/L $CaCO_3$)		
	Min.	Max.	Average	Min	Max	Average
1				6.73	6.90	6.80
2	6.11	6.56	6.37	36.00	44.00	39.33
3	6.87	7.02	6.98	32.00	45.00	39.14
4	5.90	6.93	6.63			
5	6.94	7.03	6.99			
6	6.99	7.07	7.03	16.00	23.00	20.13
7	6.88	7.05	6.97	11.00	36.00	23.25
8	6.38	6.96	6.71	9.00	32.00	19.88
9	6.94	7.12	7.02	19.00	29.00	23.38
10	6.37	6.73	6.55	18.00	25.00	21.38

Table 4. Solids.

Event	TSS (mg/L)		
	Min.	Max.	Average
1	0.00	76.00	10.88
2	0.00	37.30	5.84
3	0.00	5.20	1.18
4	0.00	26.40	4.76
5	0.00	44.40	10.80
6	0.00	6.80	0.85
7	0.00	42.90	9.15
8	0.30	6.80	2.41
9	0.00	0.00	0.00
10	0.00	0.00	0.00

Table 5. Organic Matter Indicators.

Event	COD (mg/L O_2)			BOD (mg/L O_2)		
	Min.	Max.	Average	Min	Max	Average
1	47.00	70.50	56.44	1.42	6.86	2.52
2	32.00	62.00	46.25	0.00	5.28	2.63
3	24.50	47.50	34.81	0.00	1.08	0.14
4	25.00	43.00	32.38	2.96	7.51	3.97
5	0.00	32.00	10.26	0.59	6.08	1.79
6	0.00	16.50	6.35	0.00	1.38	0.18
7	1.09	4.28	1.91	1.09	4.28	1.92
8	6.49	68.19	21.31	1.39	6.97	2.64
9	0.00	11.20	6.34	0.00	2.78	1.66
10	0.00	11.00	6.47	0.05	1.54	0.39

Event	TOC (mg/L O_2)		
	Min.	Max.	Average
1	13.75	16.65	15.50
2	11.40	16.50	14.24
3	7.70	11.90	9.86
4	9.37	13.05	10.64
5	2.45	6.84	3.72
6	2.35	3.19	2.55
7	1.63	6.40	3.64
8	2.50	5.80	3.38
9	1.60	2.00	1.74
10	0.86	1.48	1.21

The Chemical Oxygen Demand (COD) presents a maximum value of 70.50 mg O_2/L and in half of the events the mean value is above 43 mg O_2/L (Table 5). On the other hand, the maximum Biological Oxygen

Demand (BOD) found was 6.97 mg O_2/L. This confirms the low fraction of biological degradation matter in road runoff. The Total Organic Carbon (TOC) has the same variation of COD and BOD, with a decrease of its values with the increase of the monitoring period.

Table 6. Nutrients.

Event	Nitrate (mg/L NO_3)			Nitrite (mg/L NO_2)		
	Min.	Max.	Average	Min	Max	Average
1	8.20	18.30	16.65	0.02	0.14	0.08
2	13.30	18.40	16.58	0.09	0.35	0.18
3	12.50	18.60	16.06	0.03	0.22	0.08
4	18.40	22.60	20.82	0.06	0.16	0.08
5	1.90	3.80	2.99	0.00	0.00	0.00
6	1.70	1.80	1.71	0.01	0.02	0.02
7	2.70	4.70	3.59	0.01	0.07	0.02
8	4.30	11.40	7.79	0.02	0.15	0.04
9	0.60	0.70	0.63	0.00	0.02	0.01
10	0.60	1.00	0.78	0.00	0.01	0.00
Event	Ammonia (mg/L NH_4)			Kjeldhal (mg/L N)		
	Min.	Max.	Average	Min	Max	Average
1	0.15	2.41	0.88	0.00	9.40	2.56
2	0.14	0.47	0.25	0.00	2.10	0.43
3	0.09	0.30	0.16	0.14	2.66	1.09
4	0.48	0.58	0.52	0.00	2.50	1.24
5	0.05	0.35	0.14	0.00	1.70	0.74
6	0.16	0.20	0.18	0.30	1.10	0.71
7	0.08	0.32	0.13	0.00	0.80	0.44
8	0.04	0.13	0.07	0.00	0.60	0.23
9	0.05	0.07	0.06	0.30	0.80	0.48
10	0.03	0.04	0.04	0.03	0.36	0.21
Event	P (mg/L)					
	Min.	Max.	Average			
1	0.05	0.23	0.08			
2	0.05	0.14	0.07			
3	0.03	0.10	0.05			
4	0.05	0.11	0.07			
5	0.03	0.11	0.06			
6	0.04	0.07	0.06			
7	0.00	0.12	0.03			
8	0.00	0.08	0.02			
9	0.00	0.02	0.02			
10	0.01	0.02	0.02			

The average nitrate found, in the 10 events, varied from 0.70 to 22.60 mg NO_3/L, with average values between 0.63 and 20.82 mg NO_3/L (Table 6). Small concentrations of Ammonia and Nitrite found. Ammonia average concentrations were always below 0.88 mg NH_4/L, while the average Nitrite was also below 0.18 mg NO_2/L. Both have consistent lower values in the last five events. The Phosphorus in the analysed samples was also very low, with a maximum value of 0.08 mg P/L in all the events.

Final remarks

These results obtained at this monitoring site, at a Portuguese highway in the North of Portugal, confirm high levels of heavy metals and organic compounds in the monitoring events.

The first four events, at the beginning of the monitoring period have consistent higher concentrations for almost all the pollutants. The proximity of the sampling date to the dry season, with a higher build-up period, seems to be the reason for this distribution. Therefore, with the beginning of the wet season and the increase of runoff events, the concentrations of pollutants decrease.

The monitoring site has a decrease longitudinal slope where vehicles do not require acceleration and traction. These phenomena may contribute to some small concentration found for some pollutants. This fact will be confirmed by comparing of the results of this study those found in the monitoring of other sites of this project, but with different characteristics.

An inter-event analysis is in progress, in order to evaluate the existence of the first flux phenomena and to understand the pollution load distribution on the different monitored events.

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The impact of highway runoff on the chemical status of small urban streams

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Abstract

The aim of this study was to determine changes in the chemical quality of small urban stream sections directly affected by highway runoff (HR) during a rain event compared to unaffected locations as well as dry weather conditions. The study was conducted in three small streams. Parameters of basic chemical quality of water were assessed, and concentrations of specific pollutants in water and bed sediments were evaluated. No direct impact of HR on a deterioration in water quality was seen in these three already-polluted creeks. In addition, no significant difference in water quality between rain events and dry weather flow was observed either. Loading of the bed sediments by specific pollutants was observed, but with no evident relation to HR discharges.

Introduction

Urban streams are impacted by many types of anthropogenic activities, with road traffic playing one of the most important roles. Water quality, from the chemical point of view, is affected mainly by road/highway run-off, and by air contamination caused by emissions of various pollutants from vehicle exhausts. Nitrogen compounds such as ammonia, nitrites and nitrates are indicative pollutants produced by combustion processes [1], [2] and [3]. Other pollutants directly associated with road traffic are mainly the toxic metals Cd, Cr, Cu, Hg, Ni, Pb and Zn [4] and polycyclic aromatic hydrocarbons (PAHs) [5]. Therefore, it can be expected that highway runoff will contribute to the deterioration in water quality of urban streams, making the achievement of good chemical status (as the Water Framework Directive 2000/60/EC requires) difficult. In addition to chemical changes, highway runoff can also cause morphological changes in receiving waters, especially in the case of small urban streams, where

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during rain events storm water drains (SWD) and surface discharges from highway runoff can significantly increase the natural discharges. Changes in the chemical quality and morphology of streams consequently affect aquatic biota, especially benthic communities [6].

An evaluation of the impact of highway runoff on three small urban streams was initiated on the directive of the Road and Motorway Directorate of the Czech Republic. As part of this study, two sampling campaigns were required: during a rain event and during dry weather conditions, performed at two sampling sites on each stream, one situated directly below highway runoff discharge and the second above, to compare the chemical quality, morphological status and quality of benthic communities. This paper is focused on a part of this study, the assessment of chemical quality.

Methodology

Description of study area

The study area is situated in the northeastern part of Prague (the Czech Republic), where a main four-lane bypass highway leading to Mlada Boleslav (R1) intersects with the Hradec Kralove motorway (D11). This highway has a heavy load of both truck and passenger-car traffic, with an annual average traffic volume of more than 25,000 vehicles per day.

The first studied stream is the Chvalka creek, with a length of 2.2 km and catchment area of 4 km^2. This small creek flows through residential areas in its upper section, and has an average discharge of about 0.02 m^3s^{-1}. The section studied, where the creek crosses the highway, is situated in the middle of the creek length. Originally, two sampling sites on this creek were chosen: C-A, situated above the highway 1 km from its confluence, was chosen as a reference profile unaffected by highway runoff; and C-B, the affected locality receiving surface highway runoff discharge, 0.9 km from the confluence.

The second stream studied is the Svepravicky creek. It runs through residential areas and a recreational area near its source and upstream sections, where several small ponds are located. The total length of this stream is 6.8 km and the catchment area is 11 km^2. Its average discharge is similar to the Chvalka creek, about 0.02 $m^3.s^{-1}$. This stream crosses the highway 2 km from its confluence. Two sampling sites, a reference S-A, and that affected by highway runoff S-B, are situated at km 3 and 2.1, respectively (Fig. 1). The highway runoff drained into the Svepravicky creek is a combination of surface highway runoff discharge (draining a flyover crossing) and a SWD from sections of the D11 and R1 motorways that is directed to the creek through a sedimentation tank. The total length of the drained section is about 3.4 km.

The last studied stream is the Rokytka creek, one of the biggest tributaries of the Vltava River in Prague, with a length of 36.2 km and catchment area of 134.85 km^2. The average discharge ranges from 0.3 to 0.4 $m^3.s^{-1}$. This stream runs through industrial as well as residential and recreational areas, and several SWDs and combined sewer overflows are drained into it along its length. The Rokytka creek crosses the highway 16 km from its confluence. Sampling sites R-A (above the highway) and R-B (below the highway) are situated at km 16.3 and 15.7, respectively (Fig. 1). The highway runoff from a 0.5 km-long bridge section is discharged into the creek close to the Pocernicky pond.

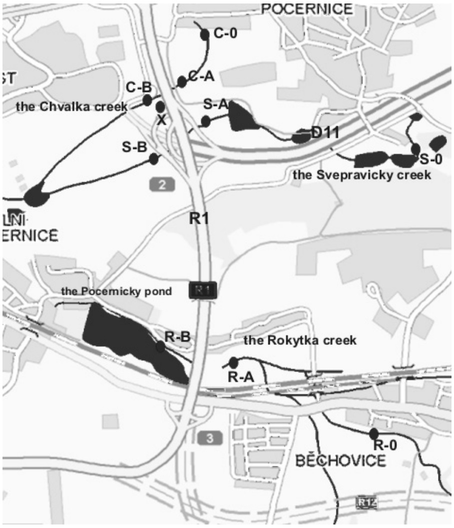

Fig. 1. Map of the study area

This assessment of the impact of highway runoff on the aquatic environment of the studied streams was further associated with hydrological monitoring of SWD discharge into the Svepravicky creek.

To better understand the overall impact of the highway on the studied creeks, an additional sampling during dry weather was performed in August 2009 at the upstream profiles of all three creeks (C-0, S-0, and R-0). Finally, a soil sample was taken at a site close to the highway (X), in order to evaluate the level of contamination by specific pollutants originating from the highway.

Materials and methods

The following basic chemical indicators of water quality were monitored: pH, electrolytic conductivity, temperature, dissolved oxygen (DO), ammonia nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N), biochemical oxygen demand-5 days (BOD), chemical oxygen demand (COD), total P, and suspended solids (SS). Moreover, specific pollutants associated with road traffic were analyzed in water, sediment, and soil samples: the trace metals Cd, Cu, Cr, Hg, Ni, Pb and Zn, and polycyclic aromatic hydrocarbons (PAHs). In solid samples (sediment and soil), total organic carbon (TOC), with which specific pollutants are usually associated, was analyzed as well. Sediment samples for metals analysis were pretreated by Aqua regia according to Czech standards CSN EN 13346 [7] and CSN EN 13657 [8]. PAH analyses comprising 15 compounds were performed according to EPA Method 610 [9]. In addition, a group of six high molecular weight (HMW) PAHs (benz(a)anthracene, benzo(a)pyrene, chrysene, dibenzo(a,h)anthracene, fluoranthene and pyrene) were separately evaluated as well.

For an evaluation of concentrations in water, the Czech standard CSN 757221 [10] was used. This standard classifies water according to the values of different indicators into five classes: class 1 – clean water, class 2 – slightly polluted, class 3 – polluted, class 4 – strongly polluted, and class 5 – heavily polluted. For an evaluation of the dangerousness of specific pollutants in sediment for aquatic biota, Toxicity Reference Values (TRV) from the USEPA [11] were used. Czech criteria given by Decree No. 13 of the Ministry of the Environment of the Czech Republic [12] were used mainly to evaluate the soil sample.

Results and discussion

The "wet weather" sampling was performed on May, 6, 2009. According to the hydrological monitoring, no significant rainfall occurred in the period prior to this sampling. During the sampling day, a series of rainfalls with total precipitation ranging from 4-4.4 mm was measured. The highest SWD discharge into the Svepravicky creek on this day was 8 $L.s^{-1}$.

It was unexpectedly problematic to carry out dry weather sampling during an extremely wet May and June 2009, with total precipitation of about 80 mm recorded in Prague, which was four times higher than in March when the average total precipitation was about 20 mm. After 4 days of dry weather in July, sampling was finally performed. The water quality of small urban streams is easily influenced by acute events, including highway runoff discharges. Changes in soluble pollutant concentrations occur quickly, and a return to primary status after an impact ends is also relatively quick. As Kim [13] points out, the highest concentrations occur during first minutes of a rain event because of the "first flush" phenomenon. It was thus expected that concentrations of the pollutants monitored would increase during rain events, with the most significant increases occurring in profiles below highway runoff discharges.

The results of basic chemical water quality analyses (as the appropriate water quality classes) are summarized in Table 1. Anthropogenic impact is evident in all three creeks and at all sampling sites, even in upstream profiles farther from the highway. All streams have sites that are strongly or even heavily polluted (classes 4 and 5). The worst quality was found at R-A and R-B of the Rokytka creek, where BOD, COD, total P and NH_4^+-N had critical values. For BOD, classes 4 and 5 correspond to the range 8-15 mg.L^{-1} and values >15 mg.L^{-1}, respectively. Classes 3 and 4 correspond to the following ranges: for COD, 25-45 mg.L^{-1} and 45-60 mg.L^{-1}; for total P, 0.15-0.40 mg.L^{-1} and 0.4-1.0 mg.L^{-1} ; and for NH_4^+-N, 0.7-2 mg.L^{-1} and 2-4 mg.L^{-1}. The other two creeks, Chvalka and Svepravicky, can be classified as strongly polluted, as shown by conductivity, an indicator of the overall loading by ions of different chemicals, in classes 3 and 4 (ranges of 70-110 mS.m^{-1} and 110-160 mS.m^{-1}, respectively). In general, it can be summarized that considering the basic indicators monitored, there is no evident deterioration of water quality in the sites directly impacted by highway runoff (B profiles) compared to sites situated above outlets (A profiles) or even to upstream sites (0 profiles). There is also no significant difference between values measured during the rain event (in May) and during dry weather (July and August).

In contrast to the basic chemical parameters, concentrations of specific pollutants (PAHs and toxic metals) in water at all sampling sites had levels corresponding to the class 1 water quality standard (clean water): PAHs (HMW) < 0.01 μg.L^{-1}, Cd < 0.1 μg.L^{-1}, Cr < 5 μg.L^{-1}, Cu < 5μg.L^{-1}, Hg < 0.05 μg.L^{-1}, Ni < 5 μg.L^{-1}, Pb < 3 μg.L^{-1}and Zn <15 μg.L^{-1}. Again, however, no differences between sampling sites affected and unaffected by highway runoff were observed.

The results of both basic parameters and specific pollutants in water did not confirm our expectation that concentrations would increase during rain events with the most significant increase occurring below SWDs and highway runoff discharges. The question might arise whether the rain event we chose for sampling was suitable. However, according to Kayhanian [14], event mean concentrations of pollutants in road runoff tend to be higher for smaller rainfall events than for larger ones, which supports our choice of sampling timing.

Table 1. Basic indicators of chemical water quality of the three monitored creeks, expressed as water quality classes according to CSN 757221 [10].

		cond.	DO	NH_4^+-N	NO_3^--N	BOD	COD	total P	SS
C-A	May	3	1	1	3	3	3	4	2
	July	3	1	1	2	2	3	3	2
C-B	May	4	1	2	3	4	3	3	2
	July	4	1	1	1	1	3	3	2
C-0	Aug	4	1	1	3	2	1	NA	NA
S-A	May	4	1	2	3	2	3	2	2
	July	3	3	2	3	3	4	3	2
S-B	May	4	2	2	3	3	3	3	2
	July	3	4	2	2	3	3	3	5
S-0	Aug	3	4	2	1	2	2	NA	NA
R-A	May	3	1	4	2	5	4	4	4
	July	3	3	3	2	4	3	3	2
R-B	May	3	1	4	2	4	3	4	2
	July	3	4	3	2	4	3	3	2
R-0	Aug	3	1	1	1	3	2	NA	NA

NA – not analyzed

Whereas PAHs and dissolved toxic metals in water were observed at levels signifying no direct danger for aquatic biota, even during the rain event monitored, their concentrations in the stream sediments are much more alarming. Fig. 2 shows the concentrations of PAHs in sediments of all sampling sites, with comparison to both the TRV [11] and the Czech standard (CzStnd., [12]) criteria for soil. There is an increase in PAH concentrations in sediments impacted by highway runoff (B profiles) compared to sediments above the discharges (A profiles) in the Chvalka creek in July and in the Svepravicky creek in both May and July. However, it cannot be unambiguously concluded that these increases are caused by highway runoff, because PAH concentrations in the upstream site of Chvalka creek (profile C-0) are even higher than below the discharge. In the Svepravicky creek, another probable source of high PAH concentrations in the sediment at the S-B profile could be an extensive nearby construction area with frequent track traffic. In all three creeks, total PAHs exceeded the TRV criteria at least at some sites and dates (and exceeded the CzStnd. soil criteria in almost all cases). High molecular weight PAHs exceeded the TRV criteria in even more cases. These levels can be dangerous for aquatic biota, especially benthic organisms, and can cause irreversible changes to their communities. The analysis of the soil sample (X), performed to consider non-point-source pollution from the highway, showed lower concentration of PAHs compared to sediment concentrations, with a value below the Czech standard for soil.

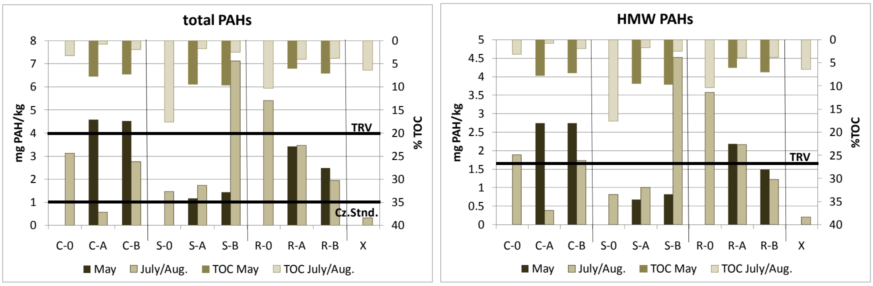

Fig. 2. Concentrations of PAHs (total on the left and high molecular weight on the right) and total organic carbon in the sediments of the monitored creeks.

Concentrations of toxic metals in sediment are shown in Fig. 3. Similarly as for PAHs, metals do not show definite evidence of a higher load in sampling sites affected by highway runoff. Only in Rokytka creek were higher concentrations of Cd, Cr, Cu, Hg and Ni measured below the discharge. According to TRV criteria, sediments of all profiles monitored are contaminated by Hg, Cu, Ni and Zn, and the Rokytka creek profiles R-A and R-B are also contaminated by Cd, Cr and Pb. The soil sample (X) had less metal loading than sediments, and according to the standard for soil concentrations, these levels are not dangerous. While concentrations of dissolved chemicals in the water of small streams can be highly variable in time, sediment serves as a long-term reservoir of pollutants such as PAHs and toxic metals, which tend to bind to the solid phase [15]. It is generally accepted that both PAHs and toxic metals are related to the organic matter content in sediments [16]. Therefore, the TOC content in sediments was also analyzed, with notable differences between the two different sampling conditions. In profiles where both "wet-weather" and "dry-weather" sampling were performed (the A and B profiles), a higher TOC content was found in May (during the rain event). One explanation could be that the May sampling followed a relatively dry weather period (with total precipitation in the previous month of about 10 mm) during which fine organic matter particles may have been stored in bed sediments. In contrast, the July sampling was done after an extremely wet period (total precipitation in May and June of about 80 mm), during which these fine organic matter particles may have been washed out. Despite this, no evident pattern of association of PAHs with TOC in sediments can be seen (Fig. 2). On the contrary, the toxic metals Cu, Ni, Pb, Zn and partly also Hg were positively correlated with TOC. Fuentes

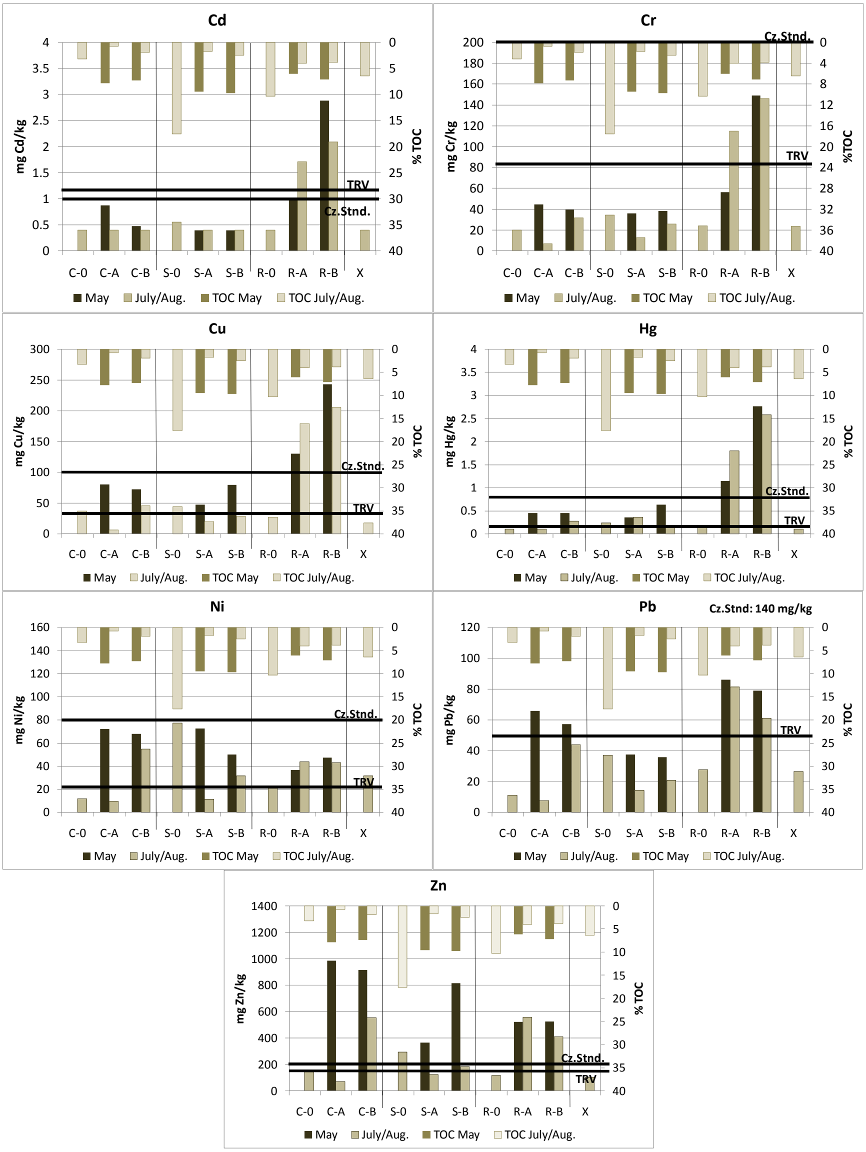

Fig. 3. Concentrations of toxic metals: Cd, Cr, Cu, Hg, Ni, Pb and Zn and total organic carbon in the sediments of monitored creeks.

[17] showed that Cu and Pb have strong affinity to organic matter, while for Ni and Zn this affinity is not as evident. In general, the binding behavior of specific pollutants in sediments is still not clear, and the combination of many different factors in the aquatic environment of small urban streams plays an important role. In the cases of the streams studied here, those factors may play a more significant role than the highway runoff.

Conclusion

This study on the impact of highway runoff on the chemical status of three small urban streams shows that a deterioration of water quality in sampling sites directly impacted by highway runoff is not always evident. Based on basic chemical analyses, all three studied creeks can be characterized as strongly or even heavily polluted, even in upstream sampling sites farther from the highway. In addition, no significant difference in water quality was observed between the rain event and dry weather flow. Sediments of the studied creeks are polluted by PAHs and toxic metals, but no demonstrable direct relation with the highway runoff was found. The amount of organic matter in the bed sediment may be predictable based on hydrological data (rainfall intensity and discharges during the period prior to sampling). This could allow sediment loading by Cu, Ni, Pb, Zn and partly also Hg to be predicted, since these metals were positively correlated with TOC in this study. Though an association of PAHs and TOC in sediments is also often mentioned, results of this study did not confirm such a relationship, emphasizing that there is still much uncertainty in the binding behavior of specific pollutants in sediments. Various other factors of the aquatic environment of small urban streams may play a more important role than highway runoff in the streams studied here.

Acknowledgment

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Changes of toxic metals bioavailability in urban creeks as a potential environmental hazard

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Abstract

The impact of different urban drainage structures (combine sewer overflow, stormwater drain and waste water treatment plant) on fate of toxic metals in urban creeks was studied on three creeks in the Prague area. The results show that bioavailability of toxic metals is affected by the type of urban drainage and environmental conditions. The bioavailability is significantly influenced by a type of urban drainage, while combine sewer overflows cause decrease of biological availability, storm water drains and a waste water treatment plant cause increase of bioavailability of toxic metals.

Introduction

Toxic metals occur naturally in the environment, but may also be introduced as a result of human activities. A natural occurring as well as anthropogenically introduced concentrations of metals in aquatic ecosystems can vary significantly due to different physical and chemical processes involved in the particular water body as well as due to different anthropogenic impacts. The fate of various metals, including chromium, nickel, copper, cadmium, lead and manganese, and metalloids, including arsenic, in the natural environment is of a great concern, particularly near former mine sites, dumps and recently also in urban areas and industrial centres [1]. Soil, sediment, water and organic materials in these areas may contain higher than average concentrations of these elements.

In order to estimate effects and a potential ecotoxicological risk associated with elevated concentrations in the aquatic environment, the fraction of total elemental abundance in water and sediment that are bioavailable must be identified. Bioavailable fraction is the proportion of total metals that are available for incorporation into biota (bioaccumulation). Total metal concentrations do not necessarily correspond with metal bioavailability.

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The degree of a contaminant bioavailability is determined by the reactivity of each contaminant with the biological interface, the presence of other chemicals that may have antagonistic or synergistic effect on uptake and by external factors such as temperature that affect the rate of biological or chemical reaction [2,3]. Bioavailability and bioaccumulation of contaminants in aquatic environment is mainly dependent on the partitioning behavior or binding strength of the contaminant to sediment [4-7]. Dissolved or weakly adsorbed contaminants are more bioavailable to aquatic biota compared to more structurally complex mineral-bound contaminants which may only become bioavailable upon ingestion with food. Metals in aquatic phase are the most available to aquatic biota compared to particulate, complexed or chelated forms [2,8,22].

The environmental conditions in aquatic ecosystems in urban areas are very dependent on the type of urban drainage [10]. While an outflow from a waste water treatment plant (WWTP) impacts the receiving water body continuously by water of approximately same quality, the impact of combine sewer overflows (CSO) and storm water drains (SWD) is not permanent and the quality of receiving water changes due to the level of local pollution of impermeable surfaces, length of the rain and also in the case of CSO due to dilution ration of sewage overflowing to the recipient.

The main goal of this paper is to identify changes in bioaccumulation of toxic metals (TM) in small urban creeks impacted by different types of urban drainage objects.

Method

The area of interest

Three small urban creeks impacted by three different types of urban drainage systems were monitored in Prague area. The first creek is impacted by WWTP (localities are marked P, P2 above and P3 below WWTP). The WWTP treats not only sewage but also technological wastewater, industrial wastewater and rain water (surface runoff from impermeable surfaces of an airport). While the contribution of sewage and industrial waste water is quantitatively and qualitatively stable during year, the surface runoff shows significant seasonal variations. During summer the surface runoff is minimally contaminated and it can be discharged to the recipient after mechanical treatment, during winter the water is highly contaminated and biological treatment needs to be applied. The second creek is affected by CSO and SWD (localities are marked B; B0 above, B2 below CSO, B3 below SWD). The combine sewer system drains area with heavy traffic and industrial and residential development.

The SWD drains residential area. The last creek is impacted by SWD from residential area (localities are marked Z; the creek is impacted by SWDs from its spring area). The creek is quite small and the amount of water entering the creek during rain episodes causes strong erosion of the creek bed as well as resuspendation of the sediment.

The geological background of all watersheds is composed by sedimentary rocks, mostly by slate.

Sampling and analyses

The aquatic ecosystem was monitored by sampling of water, sediment and aquatic biota, mostly macroinvertebrates and attached algae. The samples of water and sediments were collected during the whole year in a month respectively three months interval. Samples of aquatic organisms were collected three times during vegetation season (spring, summer, fall).

The water samples were stabilized by HNO_3 (s.p grade) and kept in refrigerator till later analyses on FAAS or ETAAS (Atomic absorption spectrometer -Solaar S with flame and electrothermic atomization).

The sediment samples were freeze and then freeze dried. The dry samples were sieved and the total fraction smaller than 620 µm were used for TM analyses. The TM were extracted by sediment microwave digestion using HNO_3 and H_2O_2 [11]. The amount of TM in different geochemical fractions was identified by sequential extraction [7,12]. The content of organic material in sediment was identified as ignition loss. Finally, the sediment samples where studied for their mineralogical composition.

The benthic organisms were collected using samplers with artificial substrate [13]. The samplers were exposed on the study locations for period approximately 1 month. After one month the samplers were collected and transported to the laboratory, where organisms were picked up, determined and separated according species and size. Then the benthic organisms and attached algae were freezed and freeze dried. Dry samples were digested by HNO_3 and H_2O_2 [8,13] and then analysed for TM content.

The concentrations of TM (Cu, Cd, Cr, Pb, Ni, Zn, Fe, Mn, Al) were detected by the atomic absorption spectrometer Solaar S with flame and electrothermic atomization.

The assessment of toxic metals hazard

The first step to evaluate the hazard related to pollutants in the aquatic environment is an identification, which phase, liquid (water) or solid (sediment or suspended matter), is more significant for the binding of pollutants. Thought, it is expected that a direct toxicological effect is related mostly to liquid phase, than to solid phase. The pollutants bound to the sediment have potential risk, because there are still a number of factors which may cause release of pollutants from sediment back to water, and they have not been completely understood yet especially in urban creeks, where the conditions often change and mutual effects of different factors may be observed [10]. The distribution coefficient (Kd) allows to identify, which phase is more significant for assessment of the hazard. The distribution coefficient expresses the level of pollutant sorption on solid material and is calculated as a ratio of the concentration in sediment to the concentration in water in equilibrium. The coefficient changes by changing environmental conditions, therefore it indicates acute hazard for biota [14]. The coefficient is often presented as log Kd, where value around 3 indicates occurrence in dissolved form (Cd), while log Kd around 5 indicates preference to bind in sediment (Fe, Pb, Be, etc.) [15].

The assessment of the hazard can be done by the Hazard Quotient (HQ). The Hazard Quotient compares actual concentrations in the environment with environmental quality standards or benchmarkers. The ecological hazard is identified in the case when HQ is bigger than 1 for one element [16].

Another factor very similar to the Hazard Quotient is the Cumulative Criterion Unit (CCU) [17]. CCU is calculated as:

$$CCU = \sum \frac{mi}{ci} \qquad [1]$$

where *mi* is concentration in the environment and *ci* is the environmental quality standard. [17] has used the CCU to classify sampling sites to four categories according to the level of pollution (geochemical background, low, medium, high pollution) and to predict the impact of toxic metals on aquatic macrozoobenthos.

The analyses of TM in biomass of aquatic organisms are very important especially on sites where the environmental concentrations are very low. One of the primary routes to organisms' contamination is through contaminated food; hence an organism living in sediment, which is the local trap for pollutant, is exposed to higher level of pollutants in the environment as well as in the food. There are different ways how to predict load of an organism by pollutants. The simplest method of assessing the load of biota is application of biota accumulation factor (BAF). BAF is calculated as a ratio of the concentration in biota to the concentration in the environment, biota sediment accumulation factor (BSAF) is the most frequently used [18].

Results and discussion

The concentrations of TM in water were compared with EQS (Czech legislation 229/2007). The monitored concentrations were below EQS for all sites and sampling dates. The low concentration of TM in water can be explained by their high affinity to sediment and rapid binding to solid fraction [19,20]. The concentrations of TM in water are not hazardous for the organisms from the long term perspective, they are neither hazardous below WWTP, SWD or CSO.

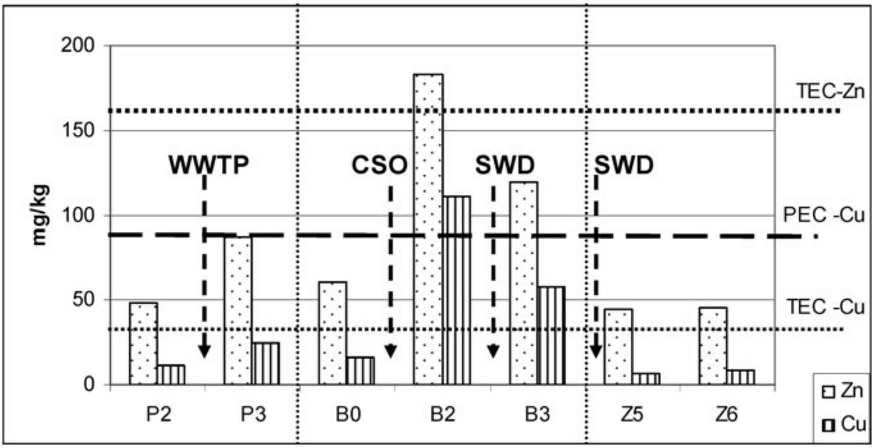

Fig. 1. The concentrations of zinc and copper in sediments and assessment of ecotoxicological hazard by TEC and PEC benchmarkers.

The concentrations of TM in sediments were compared with eco-toxicological benchmarkers PEC (Probable effect concentration) and TEC (Threshold effect concentration) [21]. The highest concentration of all analyzed metals were monitored on the site below CSO (site B2), where the toxicological benchmarker TEC was exceeded for Zn, Cu and Pb (Figures 1 and 2). The benchmarkers for Cu and Pb were also exceeded on the site B3 (directly below SWD, but also impacted by CSO located approx. 400m above B3). The concentrations of TM on other study sites were below TEC and PEC benchmarkers and do not signify any eco-toxicological hazard for aquatic biota from the long term perspective.

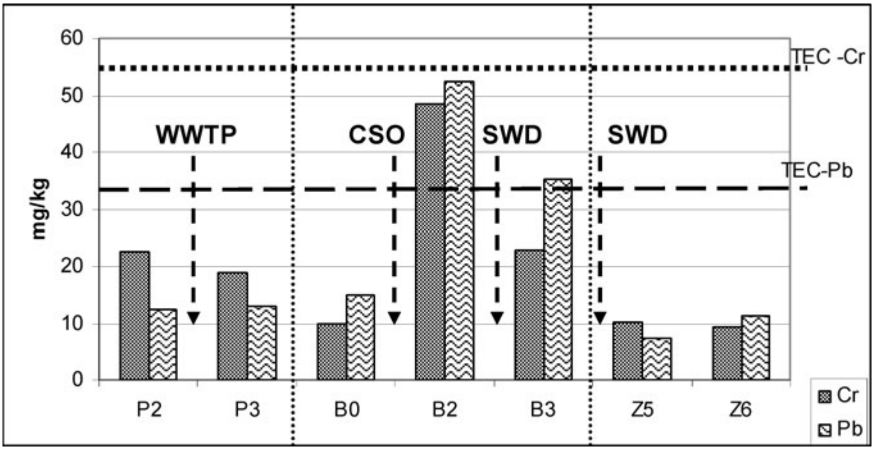

Fig. 2. The concentration of chromium and lead in sediment and assessment of ecotoxicological hazard by TEC and PEC benchmarkers.

A binding strength of TM in sediment and consequently their bioavailability can be described also by proportions of TM concentrations in particular geochemical fractions determined by sequential extraction. Comparing Cd, Cu, Pb and Zn, the most available metals are Zn and Cd, because their concentrations in two most available fractions (exchangeable and carbonates) is significantly higher than the other two metals. The least available is Pb, which is bound by the highest proportion into residual (the least available fraction). Concentration of Cu in two most available fractions is similar to Pb, but Cu is bound by highest proportion into organic matter, the second lest available fraction. Availability can be summarized in order: Pb<Cu<Zn≅Cd. Comparing different types of urban drainage, the most obvious is higher availability of Cu from Z sediments (SWD impacted), where proportion in the two most available fractions is three times higher than in the other creeks. Another evident difference is observed in the case of Pb in B2 profile below CSO, which is bound into two most available fractions significantly less than in other studied profiles. Hence, CSO seems to have positive impact on decreasing availability of Pb, on the other hand SWD increases bioavailability of Cu.

The distribution coefficient indicates which phase is more significant for occurrence of TM in the aquatic environment. The partitioning behavior (expressed as distribution coefficient) and spatial distribution of contaminants are highly regulated by hydrodynamics, biogeochemical processes and environmental conditions (redox, pH, salinity and temperature) of the individual system [2]. While the lowest coefficient for most metals was observed on the creek impacted by WWTP, the highest was observed on the third creek impacted by SWD only. The distribution coefficient indicates, that monitored metals are often present in dissolved form or bound on suspended material (log Kd 3-4). These metals may cause ecological hazard for aquatic biota because they are easy bioavailable for organism. During the monitored period only one metal (Cr) was observed to be present mostly bound into sediment on all creeks. According to the results the average values of distribution coefficient show some trends, the WWTP slightly increases values of Kd for all observed metals, as well as CSO does.

The assessment of ecotoxicological hazard was also done by identification of Cumulative Criterion Unit [18]. According to CCU the macrozoobenthos community is the most in danger on the site B2 (below CSO) (Figure 3), where a top boarder of the medium level pollution class was reached, also on the sites P3 (below WWTP) and B3 (below SWD and close to CSO) the medium level was monitored. The impact caused by medium level of pollution (2<CCU<10) may result in increasing mortality of sensitive species of benthic organisms and decreasing bio-diversity.

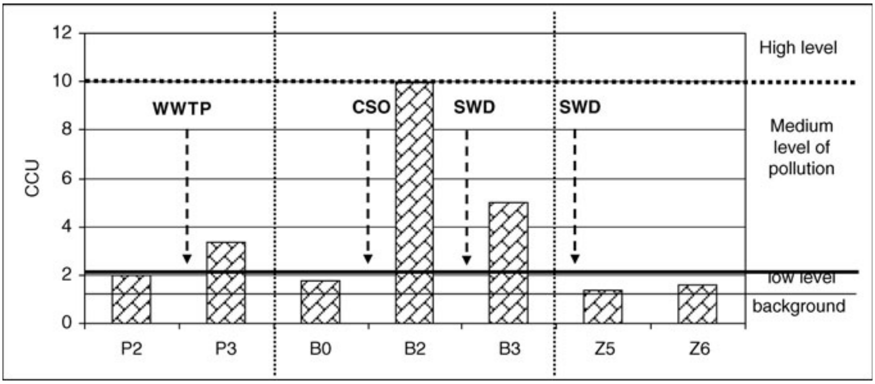

Fig. 3. Assessment of ecotoxicological hazard by CCU.

The ecological hazard is indicated for some metals (especially Zn, Ni, Cu) also by value of biota sediment accumulation factor (BSAF). Figure 4 shows significant differences in BSAF among creeks and types of urban drainage, organisms below WWTP accumulate TM more than organisms above the outlet. It can be assumed that the wastewater entering the creek from WWTP causes changes to bioavailability of metals and an increased accumulation is observed. The opposite effect on bioavailability was observed below CSO, where the sediment is highly contaminated, but the BSAF shows low values. The decrease of bioavailability below CSO can be linked with AVS (acid volatile sulfides) [6], nutrients and chelating substances [22] which enter the creek together with wastewater and may react with TM and create insoluble substances and therefore decrease the bioavailability of TM. On the other hand SWDs cause increase of bioavailability and the biota is highly loaded by TM. The impact of SWDs on increasing bioavailability may be related to the lowering pH during first flush [13], the creek impacted by SWD is quite small and the amount of water entering during rain from SWDs is often higher than the original discharge in the creek, therefore a decrease of pH may occur. The other reason for increasing bioavailability is resuspendation of sediment during rain events and release of TM back to water [2].

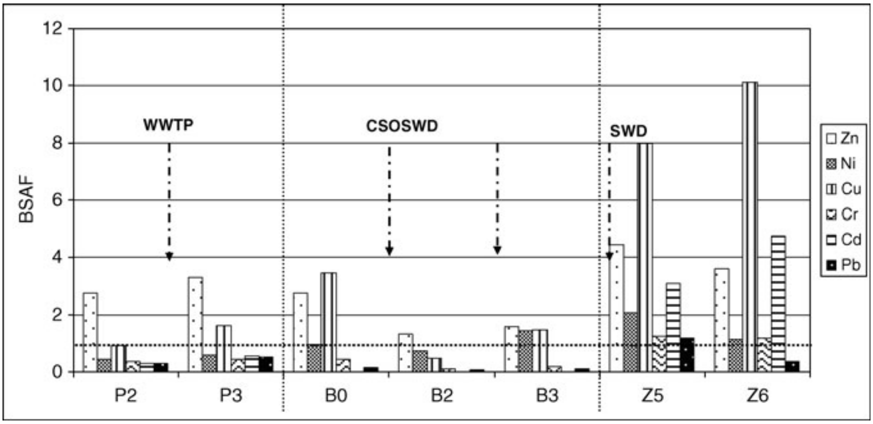

Fig. 4. Biota sediment accumulation factor (BSAF) on study sites.

Conclusion

Pollution of rivers by TM is one of the most serious global issues. A good understanding of ecological effects of TM on aquatic ecosystems is an important pre-requisite to decisions on sustainable use and management of water resources in urban and industrial areas, and in the identification of environmental quality standards.

The paper demonstrates the necessity of incorporating monitoring of aquatic organisms into routine monitoring programs. While concentrations of HM in water do not present hazard for the aquatic environment, concentrations in sediment signalize possible hazard caused by Cu and Pb according to the TEC benchmarker in studied creeks.

The results show that bioavailability of TM is affected by the type of urban drainage and environmental conditions. Consequently, the emission standards identified for creeks impacted by CSO are not applicable to creeks affected by SWD. There is a necessity to accept an individual approach to the identification of emission limits.

The results also show that there are significant differences among age stages of organisms in their ability to cumulate TM and to resist stress connected to occurrence of TM in their environment. These differences, together with the adoption of the most sensitive life stages of the organisms, need to be taken into account when identifying environmental quality standards.

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Possible sampling simplification of macroinvertebrates for urban drainage purposes

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Abstract

Admissible simplification of field sampling of macroinvertebrates compared to the standard AQEM method was searched for. Both the effect of the subjectivity of multihabitat sampling and the effect of the reduction of the number of sampling points and their uncertainties were studied. A substantial reduction of the number of sampling units (from 20 to 7) is possible only for ASPT and Saprobic index and diversity (to 9). A certain reduction (to 14) is also possible for the number of individuals, % EPT and IBI index but no reduction can be applied in case of number of taxa and BMWP, where already the replicate 20 unit samples were biased by an unacceptable uncertainty.

Introduction

European Union legislation (Water Framework Directive -WFD [1]) requires achievement of good ecological status of European streams and rivers until the year 2015. For this reason, the ecological status of all streams has to be assessed and evaluated. The assessment is based on several indicators, macroinvetebrates being one of them. For the needs of the WFD, AQEM protocol (A Comprehensive Method to Assess European Stream Using Benthic Macroinvertebrates) was developed [2]. The AQEM protocol prescribes a detailed sampling procedure and the way of sample processing.

However, the same procedure cannot be applied for the identification and localization of main disturbances caused by urban drainage as it is very time consuming and expensive [3,4]. Monitoring of urban drainage impacts must be detailed in space (tens of profiles needed within a city) as well as

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in time (2-3 samples a year in each location). Thus, a simplified biomonitoring method with known uncertainties must be applied.

The biomonitoring process contains several steps, which can be simplified to save time, and hence money: field sampling, laboratory processing and taxonomical identification. Most studies of possible simplification have focused on the laboratory processing by subsampling when only a part of the sample is processed. However, the minimum number of individuals that has to be taken into account to obtain a valid result varies widely between 100 and 700 [5,6].

Methods of field sampling differ substantially in various countries. The methods can be divided into those taking samples from a defined surface area of the streambed (e.g. AQEM prescribes about 1.25 m^2) and those sampling a certain reach length (e.g. 25 m in UK [7]). A further classification of the methods considers types of habitats sampled. Some methods require multihabitat sampling where a sample consists of units (points) taken from all microhabitats at the sampling site. The sampling units are distributed according to the share of the microhabitats occurrence. Sampling of the whole site is performed for a certain time (e.g. 3 minutes AQEM [2], PERLA [8]). Other methods concentrate on one or several typical habitats (e.g. RBA US [9]. In Australia, the rapid bioassessment is performed from two specific habitats at least. For routine monitoring, a 10 m sample is recommended whereas a 10-20 m reach should be sampled when edge habitats are important [10].

The comparison and intercalibration of the national methods within EU with the AQEM one was performed in the STAR project [5], which found that some methods were compatible with AQEM and their variance was < 10% (e.g. Czech method PERLA) [11].

However, little attention has been paid to the possibilities of the field sampling simplification. Only Vlek *et al.* [4] studied the effect of the sample size on individual benthic metrics and final bioassessment of the streams. They collected samples with a pond net in 20 sampling units over a length of approximately 0.25 m per unit and tested the decrease of the sample from 5 m to 0.25 m. Metrics dependent on the absolute abundance were more sensitive to the sample size than those dependent on the relative abundance. These results correspond with the results of authors focusing on subsampling [6]. Also the effect of the variability of multihabitat samples has not been investigated systematically. Lorenz a Clarke [12] studied the similarity of replicate samples on one sampling site and found out that it varied 83 – 100%.

Thus, the lacking systematic investigations of the field sampling simplification motivated us to carry a project with the goal to assess the effect of the sampling simplification compared to the reference AQEM and to determine how many sampling points are needed to meet a required accuracy of basic benthic metrics used for the evaluation of the stream ecological status and of urban drainage impacts. Three main topics were studied:

1. The effect of the subjectivity of multihabitat sampling, i.e. of a random selection of 20 sampling points from a dense sampling grid, and its uncertainties.
2. The effect of the reduction of the number of sampling points and its uncertainties compared to the reference dense grid.
3. The influence of the stream morphology on the possible sampling simplification. We hypothesize that degraded morphology (and biocenosis) might lead to less sampling effort.

Method

Seven streams of different morphological quality were sampled in 2008 and 2009. Three of the streams were in urban areas affected by combined sewer overflows (Hloucela, Rokytka, Botic), four were natural streams (Tritrubecky, Cerveny, Obecnice, Sazavka).

The stream morphological status was assessed according to the Swiss methodology [13]. In the field survey data were taken on the variability of the water surface width, stabilization and character of the riverbed and banks, width and character of the riparian zone and disruptions of longitudinal connectivity (steps, weirs, dams etc.) The measure of the deviation of individual parameters from the natural status was evaluated on a point basis. Depending on the total number of points obtained, reaches were assigned to the ecomorphological classes I (natural/near-natural), II (little affected), III (strongly affected) or IV (artificial). In addition, flow velocity, its diversity and substrate character were evaluated.

AQEM procedure (reach length 20 m at least, multihabitat sampling, 9 seconds per unit) was applied with the exception of the number of units. Contrary to 20 units required by AQEM, 34 to 41 units were sampled in individual streams (Table 1). It allows both to determine the uncertainty of the subjective selection of 20 sampling units in the AQEM method and to study the possibility of a further simplification in a screening method.

Organisms from each sampling unit were collected, kept separately and preserved in 90% ethanol. In the laboratory all organisms were identified to the lowest practicable level (usually species) except Oligochaeta, Nematoda, Turbellaria, Acarina, Chironomidae, Ceratopogonidae, Psychodidae which were not identified any further.

Monte Carlo simulations

In order to identify uncertainties introduced by the reduction of sampling units, Monte Carlo method was used (Figure 1). In each simulation (randomization) a desired number of sampling units x was randomly chosen and benthic metrics were calculated. The randomization was repeated y times (y = 500). It resulted in y possible values of each benthic metrics, which were statistically analyzed to obtain probability density functions (PDF) (log-normal distribution was considered).

The process described was applied to x values ranging from 8 to 20 in order to find a relation between the number of sampled units and the uncertainty of individual metrics. The values of metrics from the original sample (i.e. from all sampled units) served as a reference.

In order to ensure that selected sampling units are not similar (as a biologist selects different representative habitats) criteria restricting the selection were implemented. The criteria considered the side of the stream (left, right, middle), depth of water (shallow, deep, middle), type of flow (riffle, run, glide, pool) and prevailing substrate (blocks, cobbles, coarse gravel, gravel, sand, fine sediment and phytal, algae, xylal, roots) to be different habitats. A maximum difference of 40% from the original sample was set for all criteria (e.g. in case 30% of sampling units in the original sample had flow velocity of 1 m/s, then 18-42% of units had to be in the

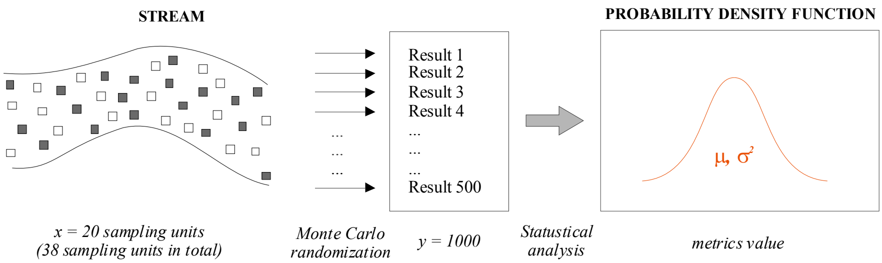

Fig. 1. Schematization of Monte Carlo simulations for x = 20.

same category in the reduced sample). To allow a comparison of this restricted selection with a non-restricted one, a second run of simulations was done for the random selection without restrictions.

Metrics evaluation

Eight metrics were evaluated: number of individuals, number of taxa, metrics describing general degradation: % of EPT taxa and diversity, and metrics characterizing organic pollution: Si (Saprobic Index), ASPT (the Average Score per Taxon) and BMWP (Biological Monitoring Working Party) [2]. Also B-IBI (Benthic Index for Biological Integrity) describing general and morphological degradation was calculated [14].

Requested uncertainty

The simplified sampling method must be able to reveal stream disturbances caused by discharges from the urban drainage system. A site is considered to be significantly disturbed by urban drainage if the difference in total abundance from reference conditions (usually a site upstream of the disturbance source) is higher than 30% or the difference in number of taxons greater than 20% or 3 or more species with at least mean abundance or one highly abundant species do not overlap [15]. For other metrics the uncertainty of about 15% reflects the natural variability [12]. Thus, the relative error of the simplified sampling method compared to the standard method must be lower than these values.

Results and discussion

Stream biological quality

Basic characteristics of the streams studied are in Table 1. Streams of all morphological classes are present, i.e. ranging from natural to artificial. Also water quality differs from non-polluted (class I) to highly polluted (class V). Rokytka, Hloučela, Botič a Sázavka belong among lowland streams whereas Tritrubecky, Obecnice a Cerveny are submontane streams. Table 2 summarizes the biological quality of the streams. The main cause of the disturbance of the urban streams is their poor morphology.

Table 1. Basic stream characteristics.

Characteristic	Tritrubecky	Obecnice	Cerveny	Sazavka	Hloucela	Rokytka	Botic
Number of sampling units	41	34	36	35	39	34	35
Altitude m above sea	570	600	540	430	215	230	210
Morphology	I	I	I	II	II-III	III	IV
Average depth (cm)	25	19	24	28	35	22	9
Average flow velocity	3	3	2	4	2	2	2
Average flow diversity	high	high	mean-high	mean	mean	low	low
Water quality	I	I	I	II	I-II	V	III-IV

Explanation:

<u>Stream velocity</u>:
4 riffle
3 run
2 glide
1 pool

<u>Ecomorphological class [13]</u>
I - natural/near-natural
II - little affected
III - strongly affected
IV – artificial

Table 2. Stream biological quality.

Metrics	Tritrubecky	Cerveny	Obecnice	Sazavka	Hloucela	Rokytka	Botic
Individuals	5633	2245	1230	5547	6092	10537	5986
Taxa	40	50	41	64	58	40	31
%EPT	62.5	54.0	41.5	51.6	51.7	25.0	38.7
Si	2.1	2.8	2.3	2.4	1.8	1.2	1.9
ASPT	6.7	7.2	6.5	6.4	6.6	5.0	5.4
BMWP	254	344	254	405	356	191	150
Diversity	2.1	2.8	2.3	2.4	1.8	1.2	1.9
IBI	21	21	24	22	21	17	19

Subjectivity of sampling

The uncertainty of the metrics based on samples containing randomly selected 20 sampling units is summarized in Table 3 for all 7 streams. Simulations showed that the restriction criteria applied on the sampling units selection had only a minor influence on results as they caused less than a 3.5% decrease of uncertainty (less than 2% in most cases) (see also Figure 2). Therefore, the results of random selections are presented.

A further reduction of the number of sampling units is possible for metrics, for which the calculated relative uncertainty based on the 20 units sample is lower than the requested uncertainty. Thus, it is possible for the number of individuals, ASPT, Saprobic index, diversity and % EPT. However, in case of the number of taxa and BMWP the average uncertainties are higher than the requested ones even for replicate 20 units samples.

Reduction of sampling points

Figure 2 shows two typical patterns of behavior of benthic metrics: 1. nearly no effect of the systematic error, and 2. a pronounced affect of the systematic error (i.e. a systematic under- or overestimation of the metrics due to the insufficient number of sampled units). Number of individuals, Saprobic Index, diversity and ASPT belong to the first group as they are not affected by a systematic error larger then 2%. Percentage of EPT taxa is systematically overestimated by up to 4% and IBI shows no systematic error until 15 sampling units (if the number of sampling units is further reduced, the systematic underestimation increases). Number of taxa and BMWP are representatives of the second group as they are highly affected by the systematic error caused by the increased probability of missing rare species when number of sampling points is reduced.

Table 3. Relative uncertainty (%) of a 20 units sample with 90% confidence.

Metrics	Tritrubecky	Cerveny	Obecnice	Sazavka	Hloucela	Rokytka	Botic	Mean value	Standard deviation	Variation coefficient	Requested uncertainty
Individuals	20.4	39.0	16.2	18.5	19.6	22.7	14.4	21.5	7.6	0.35	30
Taxa	24.0	28.3	20.4	19.4	26.2	25.4	20.8	**23.5**	3.1	0.13	20
%EPT	10.4	12.4	7.8	8.3	9.4	23.7	15.1	12.4	5.2	0.41	15
Si	1.6	9.4	2.6	1.4	2.5	0.1	1.0	2.7	2.9	1.08	15
ASPT	3.3	4.1	2.1	2.8	5.0	5.3	4.3	3.8	1.1	0.28	15
BMWP	24.4	29.2	22.0	21.1	28.3	29.2	20.6	**25.0**	3.6	0.14	15
Diversity	4.6	21.1	7.1	3.8	7.2	5.1	3.3	7.5	5.7	0.77	15
IBI	1.8	14.5	5.7	0.0	3.5	17.2	13.7	8.1	6.4	0.80	15

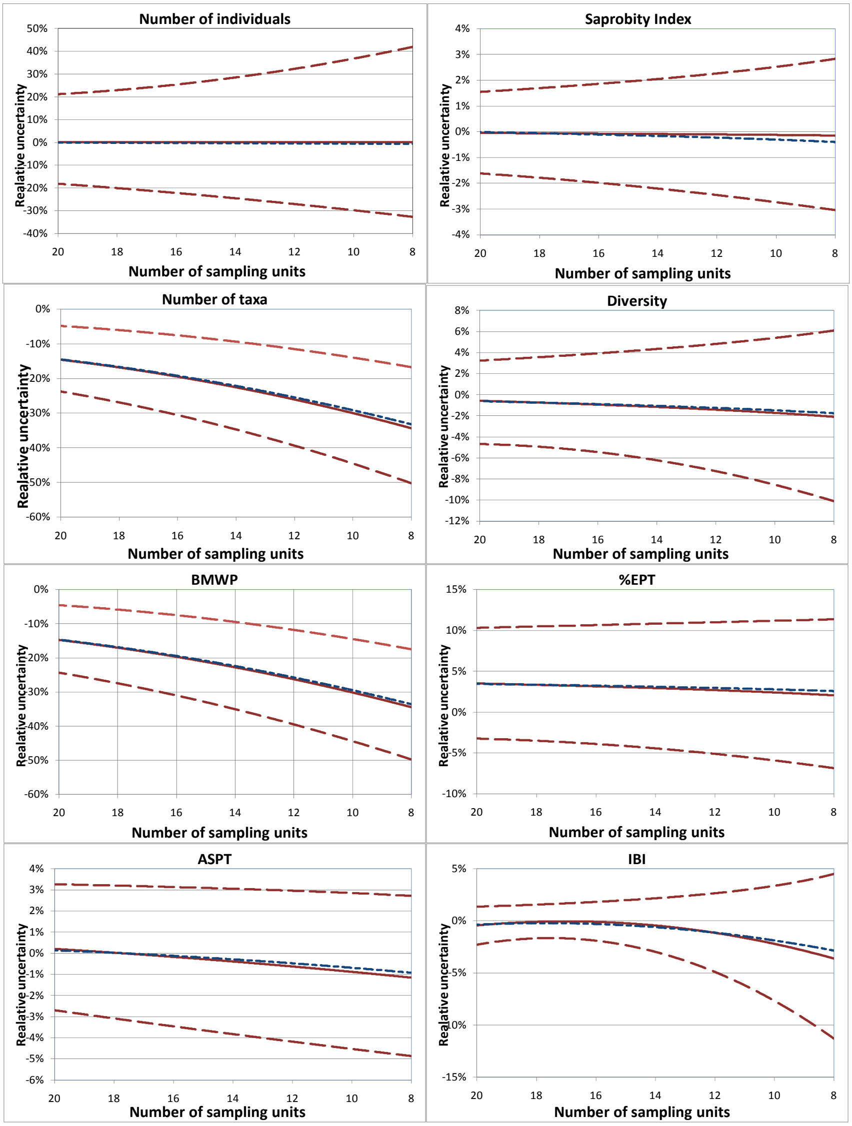

Fig. 2. Course of metrics uncertainties in relation to the decrease of the number of sampled units for Tritrubecky Cr. (full line – mean value of metrics in unrestricted simulations; dotted line – mean value of metrics in restricted simulation; dashed lines – random error as 90% uncertainty of unrestricted simulations). Relative uncertainty of 0% corresponds to the reference value of metrics (i.e. from all sampled units).

Necessary number of sampling points

The minimum number of sampling units needed to meet the criteria for the relative uncertainty of individual metrics can be derived from the courses of uncertainties of the individual metrics (Table 4).

Table 4. Number of sampling units needed to meet criteria for relative uncertainty.

Metrics	Tritrubecky	Cerveny	Obecnice	Sazavka	Hloucela	Rokytka	Botic	Mean value	Standard deviation	90% conf. interval
Individuals	13	20	11	12	13	16	10	13.6	3.2	19
Taxa	20	20	20	20	20	20	20	20.0	0.0	20
%EPT	6	16	6	8	11	20	20	12.4	5.8	~20
Saprobic in.	6	11	6	6	6	6	6	6.7	1.7	10
ASPT	6	6	6	6	6	8	6	6.3	0.7	8
BMWP	20	20	20	20	20	20	20	20.0	0.0	20
Diversity	6	20	6	6	9	7	6	8.6	4.8	17
IBI	7	20	6	6	6	20	19	12.0	6.7	~20

It is apparent that no sampling simplification is admissible for the number of taxa and BMWP, where already the replicate 20 units samples were biased by an unacceptable uncertainty. On the other hand, only 7 sampling points are in average necessary for ASPT and Si and 9 points for the diversity. 14 sampling points should be in average sufficient for the number of individuals, % EPT and IBI. However, standard deviations of the necessary number of sampling units are quite high for some metrics (especially IBI, % EPT and diversity).

Influence of the stream morphology

No correlation between possible sampling simplification and stream morphological status was found. Thus, the hypothesis that degraded morphology (and biocenosis) might lead to less sampling effort was not confirmed.

Conclusion

Benthic metrics studied exhibited different dependency on the samples size as also confirmed by [4,5,11]. A substantial reduction of the number of sampling units is possible only for ASPT and Saprobic index, which are very robust. A certain reduction is also possible for diversity, number of individuals, % EPT and IBI index but no reduction can be applied in case of number of taxa and BMWP.

Therefore, no screening method of the field sampling reducing the number of sampling units from the original 20 unites used in the AQEM method can be recommended. Time saving potential in laboratory processing must be searched for, which is a second part of the project carried out.

Acknowledgement

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Bioaccumulation of heavy metals in fauna from wet detention ponds for stormwater runoff

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Abstract

Stormwater detention ponds remove pollutants e.g. heavy metals and nutrients from stormwater runoff. These pollutants accumulate in the pond sediment and thereby become available for bioaccumulation in fauna living in the ponds. In this study the bioaccumulation was investigated by fauna samples from 5 wet detention ponds for analyses of heavy metal contents. Five rural shallow lakes were included in the study to survey the natural occurrence of heavy metals in water-dwelling fauna. Heavy metal concentrations in water-dwelling fauna were generally found higher in wet detention ponds compared to rural shallow lakes.

Introduction

Stormwater runoff from cities, roads and highways contain high loads of environmental pollutants, e.g. heavy metals and nutrients. Untreated stormwater runoff from separate sewer systems can therefore be a significant source for elevated concentrations hereof in the receiving waters [1]. In order to meet environmental objectives such as formulated under the EU Water Framework Directive, proper management of stormwater discharge is required. One approach is low-structural management practices for example wet detention ponds [2], which retain stormwater runoff long enough to allow physical, biological and chemical processes to proceed; hereby purifying the stormwater prior to discharge [3]. Over time the heavy metals accumulate in the bottom sediments of the pond, which can lead to ecotoxicological risk for the aquatic environment depending on the quantity and the chemical form of the metals [4].

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Uptake of heavy metals in fauna occurs either directly from the surrounding aquatic medium or through feeding. The relative distribution between these routes vary with type of organism and bioavailability of the metals in the diet and water [5][6]. When uptake exceeds excretion, the metals become bioaccumulated. Several studies indicate that aquatic living organisms can bioaccumulate heavy metals. For example, a study in 1994 found that fish living in wet detention ponds in Orlando, Florida, had 2-10 times higher levels of cadmium, nickel, lead and zinc compared to fish in natural lakes [7]. Similarly, biota in a shallow Danish twin-lake receiving highway and stormwater runoff had elevated contents of certain heavy metals and PAH closest to the stormwater inlet. Growth experiments with bivalves showed reduced growth and increased death rates in the most polluted lake [8]. Furthermore, a study on temporal trends of trace metals in sediments and invertebrates from wet detention ponds in Maryland, USA, demonstrated significant concentrations of copper, lead and zinc in invertebrates [9].

The concentrations of heavy metals may increase with increasing trophic level in the aquatic food web as metals are non-biodegradable. This process is known as biomagnification where metals are transferred from one trophic level to the next with hardly any excretion [6].

Bioaccumulation and biomagnification of heavy metals in fauna of wet ponds is today largely unknown. The objective of this study was to investigate the degree to which bioaccumulation and biomagnification occur in fauna of wet detention ponds. Samples were collected from 5 wet ponds and 5 rural shallow lakes unaffected by stormwater runoff. The sample types of this study comprised invertebrates (e.g. molluscs, dragonflies, mayflies and caddisflies). It was an objective to add to the knowledge of how pollutants allocate in wet detention ponds, and contribute to develop more efficient storm water detention ponds, further reducing the environmental impacts of urban stormwater runoff.

Methodology

This section describes where and how fauna samples were collected and how the preparation and laboratory analysis was performed.

Data collection and preparation

In April and May 2010, the investigated fauna samples were collected from 5 wet detention ponds in Denmark that receive stormwater runoff from various types of catchments (Fig 1). Five small, rural, and shallow lakes not receiving stormwater runoff were included in the investigation to survey the natural occurrence of heavy metals in water-dwelling fauna of the same region and comparable environments.

Fig. 1. Sampling locations of wet ponds (black) and rural shallow lakes (white), Denmark.

Samples were collected in 3-4 plastic containers (2.5 L) and placed on ice immediately after collection, transported to the laboratory and sorted within 24 hours according to groups of species. Subsequently, samples were frozen (-20°C) prior to freeze drying (ALPHA 1-2 LD plus, Martin Christ, Germany) at -55°C at vacuum for 48 hours for determination of dry matter content. All specimen, except for clams, had dry weight less than 0.6 g and were directly after freeze drying transferred to teflon vessels. Clams were, due to larger sample amounts, homogenized prior to freeze drying. Liquid nitrogen was added to samples in a mortar, after which samples were crushed with a piston. Subsamples were then freeze dried and similarly transferred to teflon vessels. Hereafter, samples were digested with 10 ml concentrated trace metal graded nitric acid (67-69% HNO_3, SCP Science, Canada) for 10 min, following EPA 3051 method for microwave assisted acid digestion (Multiwave 3000, Anton Paar, Austria).

After cooling, the digestates were diluted to volume with ultra pure water (NanoPure Diamond UV, Barnstead, Thermo Scientific), transferred to plastic flasks and allowed to settle before analysis of heavy metals plus aluminum and phosphorus.

Analysis and quality assurance

The samples were analyzed for the following elements: copper (Cu), zinc (Zn), iron (Fe), lead (Pb), cadmium (Cd), nickel (Ni), chromium (Cr), phosphorus (P) and aluminium (Al). These elements were analyzed using Inductively Coupled Argon Plasma with Optical Emission Spectrometry detection (ICP-OES) (ICAP 6300 Duo View, Thermo Scientific). All elements were measured axially at two to three wavelengths.

Matrix matched multi-element standards were prepared from single element standards (1000 µg ml^{-1}, PlasmaCAL, SCP Science, Canada) and used for external calibrations for all ICP-OES analyses. They were prepared with the following concentrations: 1, 10, 100, 1000 and 10000 ppb. Both standards and samples were added Yttrium (Y) as internal standard, with ending concentration in samples and standards of 200 µg (L^{-1}). The addition to samples occurred before digestion, and thereby accounts for transfer loss after digestion and change in plasma intensity. Additionally, certified reference material (EnviroMAT - BE1, SCP Science, Canada) was included in the digestion-batches (16 samples). The measured concentrations were within in tolerance interval of the certified reference material for Cd, Cr, Cu, Fe, Ni, Pb and Zn. P and Al had 4 and 2 references out of 6 that matched the tolerance interval of the certified reference material.

Description of the ponds

The wet detention pond in Odense holds app. 2,000 m^3 and is from previous studies known to receive illicit industrial discharges of especially copper, but also other heavy metals [10]. The pond in Silkeborg holds 2,700 m^3, and the same study showed that the pond received less polluted waters than the pond in Århus, which holds 6,900 m^3. In 2009, the pond in Århus has received iron salts to enhance pollutant removal, and the pond in Silkeborg has received aluminium salts [10]. The pond in Lemming holds 1,000 m^3 and receives water of concentrations comparable to those of Århus [11]. The facility in Sæby is app. 600 m^2 and receives runoff from a highway truck centre. The actual pollutant load on the pond is unknown.

The rural shallow lake in Poulstrup is app. 11,000 m^2 and is located in a recreational heath and forest reserve of 170 ha. Organic matter is removed with a couple of year's interval to ensure good condition for predator fish. Similarly, Hundsø is app. 19,000 m^2 and is located in a forest reserve. The lakes in Sjørup and Auning are app. 4,300 and 9,500 m^2, respectively, and are both situated in old plantations, whereas the lake in Dorf is an old mill lake of app. 17,000 m^2, where the water from a small stream is stemmed.

Results and discussion

The invertebrates from wet detention ponds and rural shallow lakes were divided into the following groups: Molluscs (snails and freshwater clams); Odonates (dragonfly and damselfly larvae); Composites (remaining macro-invertebrates). The bioaccumulation of metals in the invertebrates was detected within a wide range. Ni, Cr, and Cd, were generally present in the lowest total concentrations, followed by Pb, Cu, Al, Zn and Fe (Table 1 and Fig. 2). The general tendency was that all metals but Pb were found in clearly elevated concentrations in the fauna of the wet detention ponds compared to the reference group of rural shallow lakes. This observation holds true both when viewing metal concentration per unit of dry matter, but also when normalizing against the phosphorous content of the fauna (Table 1).

Table 1. Mean metal concentration in the invertebrate groups; Molluscs, Odonates and Composite. *Lake* and *Pond* refer to rural shallow lakes and wet stormwater detention ponds, respectively.

Element	Invertebrate group (mg (kg DM)$^{-1}$)					
	Molluscs		Odonates		Composite	
	Lake	Pond	Lake	Pond	Lake	Pond
Frequency[1]	4/5	3/5	3/5	5/5	5/5	5/5
Al	31.0	58.9	42.3	159	49.0	157
Cd	0.1	0.9	0.1	0.2	0.3	4.5
Cr	0.5	0.3	0.2	1.1	0.6	2.5
Cu	8.1	67.4	19.9	33.4	53.8	214
Fe	589	626	154	1254	636	2709
Ni	0.5	2.4	0.0	0.4	0.6	1.2
P	13505	5223	9754	14641	9606	16596
Pb	3.2	1.4	3.3	2.8	6.4	4.5
Zn	160	128	64.0	296	314	625

[1] Frequency state the fraction of sample sites where the group were caught

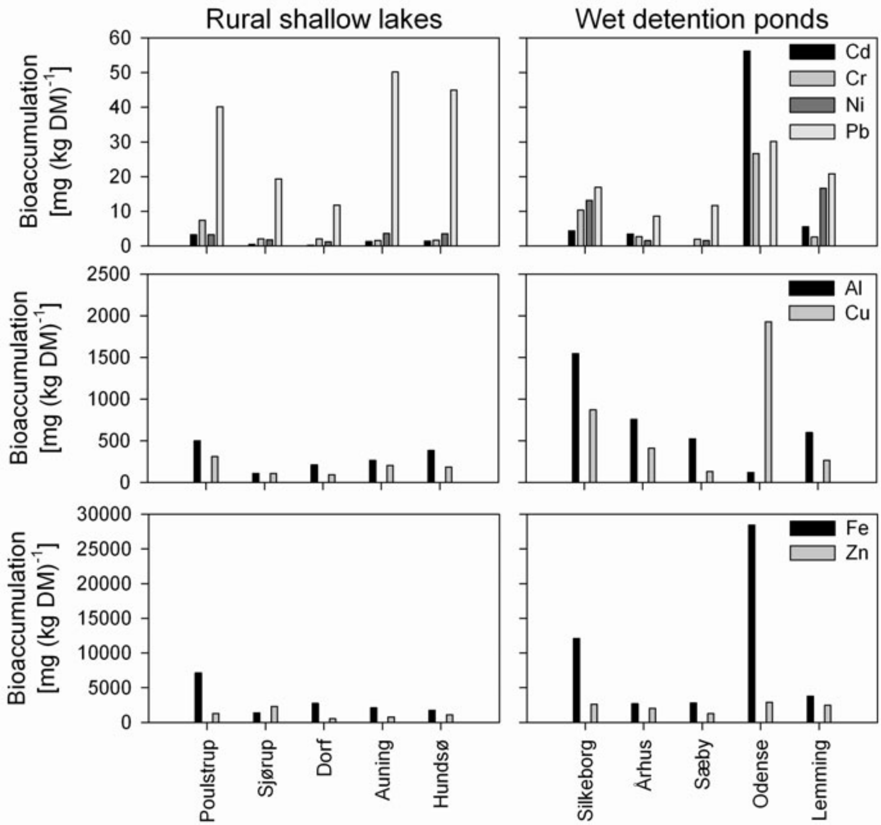

Fig 2. Total concentrations for Cd, Cr, Ni, Pb, Al, Cu, Fe and Zn in 5 rural shallow lakes and 5 wet stormwater detention ponds, Denmark.

Another study of bioaccumulation in similar invertebrate groups and also obtained from stormwater ponds [8] show bioaccumulations of: Cu, 6-45 mg (kg DM)$^{-1}$; Pb, 0.5-1.5 mg (kg DM)$^{-1}$; Zn, 54-198 mg (kg DM)$^{-1}$. That is, the bioaccumulation in both the stormwater ponds and the rural lakes of this study were comparatively high.

Looking at the 5 wet detention ponds, the pond in Odense tended to have highest heavy metal bioaccumulation, followed by the pond in Silkeborg, Lemming, Århus and Sæby. The pond in Odense was also the one most heavily loaded with pollutants, and it received heavy metals from an illicit industrial source, probably discharged on dissolved form. This probably explains why the fauna in that pond showed the most significant bioaccumulation. The pond in Silkeborg, however, had the second-highest bioaccumulation, but received comparably low pollutant loads, indicating that pollutant loads alone could not explain the bioaccumulation observed.

The most significant bioaccumulation was seen for Cu in invertebrates from Odense – nearly 2,000 mg (kg DM)$^{-1}$. This can clearly be linked to the illicit discharge of Cu, which in periods increased the Cu concentration in the pond water to above 1,000 mg m^{-3} [10], i.e. one to two decades above typical stormwater concentrations [1]. Similarly, the illicit discharge in the catchment in Odense probably also explains the elevated concentrations of Cd and Cr. With respect to Pb, the bioaccumulation was highest in Odense, followed by Lemming and the other ponds. This ranging corresponds to what was observed for the stormwater concentrations entering the ponds [10,11]. On the other hand, the bioaccumulation of Ni was by far highest in the invertebrates from Silkeborg and Lemming, however, for these ponds the water phase levels of Ni were not elevated compared to typical stormwater [1,10,11].

The total bioaccumulation of Al was in general higher in wet detention ponds compared to rural shallow lakes, except for the pond in Odense. The elevated Al concentration in Silkeborg can maybe be explained by the aluminium addition that was applied in 2009, which might have entailed increased bioaccumulation of Al in fauna living in the pond.

Surprisingly, the bioaccumulation of Pb in invertebrates from the rural shallow lakes was in general higher than Pb bioaccumulation in invertebrates from wet detention ponds. The reason here for is unknown, but could relate to the wet detention ponds all being less than 10 years old and the elevated Pb-levels in the rural lakes the remnants from the times of leaded petrol.

The invertebrates in the Composite group were further divided into the following subgroups: Aséllus (water louse), Hirudinea (leech), N. Cinérea (water scorpion), Dytiscidae (diving beetles), Trichopteras (caddisflies), Chironomidae (chironomids), Corixa larvae, Ephemeropteras (mayflies) and tadpoles (unknown specie). One should be aware that not all subgroups were found in both wet detention ponds and rural shallow lakes, nor were they found in all ponds or lakes. For example N. Cinérea and tadpoles were only collected in rural shallow lakes, whereas Chironomidae and Corixa larvae were only found in one wet detention pond. Therefore, the following conclusions are subject to some uncertainty, as conclusive evidence would require a much stronger dataset. The general tendencies were, however, still clear.

The mean metal content of the Molluscs, Odonates and subgroups of Composite were ranked according to increasing levels. Here, it appeared that pollutant loads influenced the bioaccumulation to a higher degree than trophic level, as invertebrates from wet detention ponds were primarily located towards the high ranking, especially for Al, Cu and Zn (Fig. 3). For Ni the ranking between wet detention ponds and rural shallow lakes was somewhat even, while the high ranking for Pb mainly was dominated by invertebrates living in rural shallow lakes. The ranking for Pb indicated a general bioaccumulation of Pb in invertebrates from rural shallow lakes, and not only a single species that elevated the mean concentration in the composite group (Table 1).

Generally, the bioaccumulation also seemed to be influenced by other factors than pollutant loads, as the order among the invertebrate groups changed from element to element (Fig. 3). Interestingly, exchange in position happened between Odonates and Trichopteras. Odonates are predators, whereas the diets of Trichopteras vary depending on subspecies and living condition, but they are primarily feeding on plant material. The ranked Al, Cu and Ni diagrams showed that Odonates and Trichopteras had higher mean metal contents in wet detention ponds than in rural shallow lakes, but also that Trichopteras had higher mean metal contents than Odonates. Only for Zn was the bioaccumulation in Odonates higher, and only in the wet detention ponds. Looking at Pb, the concentrations in Trichopteras were still higher than in Odonates, and here concentrations were highest in rural shallow lakes, as started earlier. This observation can be due to many factors that cannot be explained based on the results from this study; like bioavailability, species specific affinity or excretion ability towards some specific elements.

Conclusion

Invertebrates from 5 wet detention ponds were shown in general to have elevated heavy metal contents compared to invertebrates from 5 rural shallow lakes of the same region. For cadmium, chromium, nickel and to some degree also copper, the organisms sampled from wet detention ponds contained up to an order of magnitude more metal compared to organisms from the rural shallow lakes. Only for lead, the picture was reversed as the content of this metal was about twice as high in the rural shallow lakes compared to the wet ponds.

Looking at species groups, the heavy metal content did not correlate with the trophic level of the different groups. That is, even though the study clearly showed bioaccumulation to occur, biomagnification could not be identified.

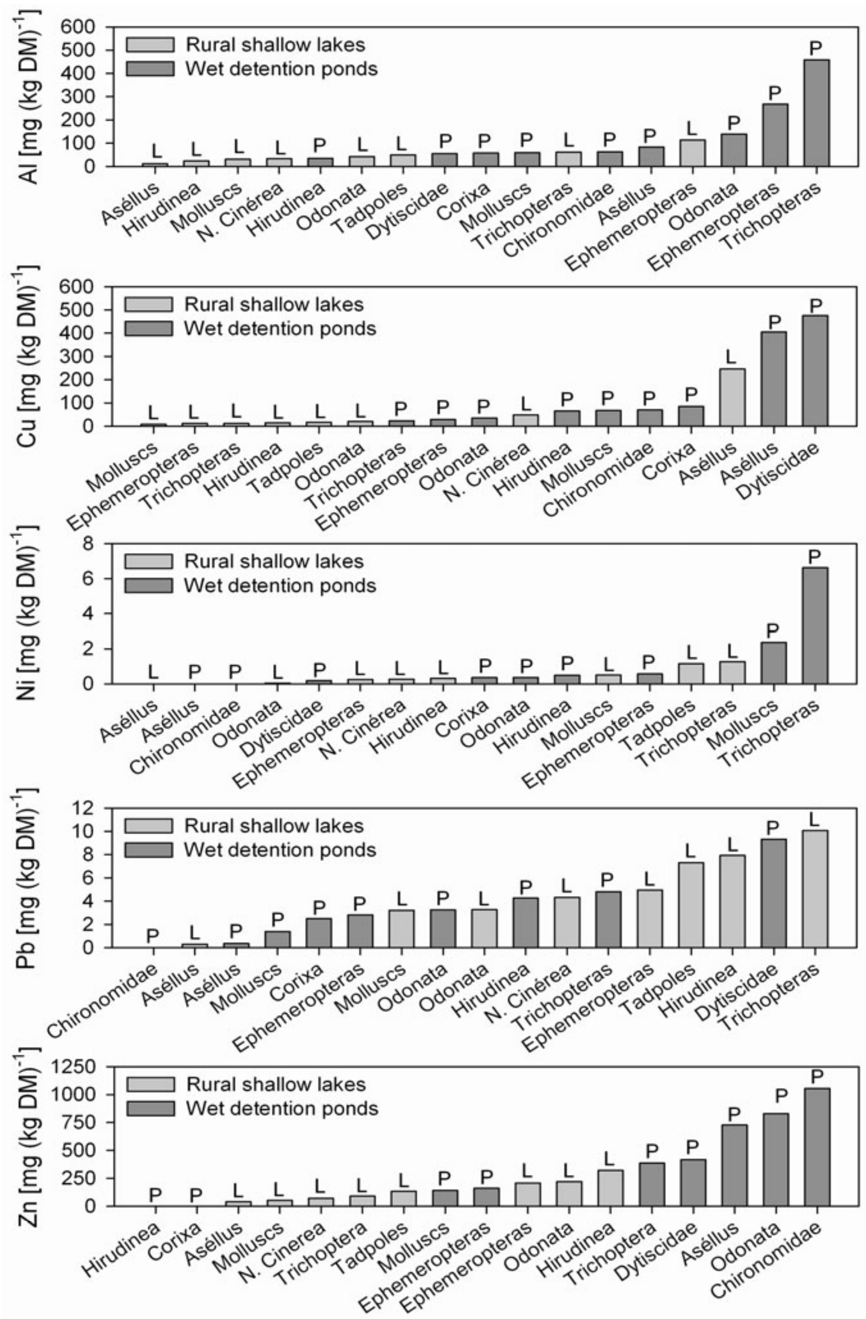

Fig. 3. Mean metal concentration for invertebrate species living in 5 wet detention ponds and 5 rural shallow lakes ranked for the element; Al, Cu, Ni, Pb and Zn.

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Behavior of dissolved trace metals by discharging wastewater effluents into receiving water

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Abstract

Municipal wastewater (WW) discharges induce various environmental impacts on receiving freshwater systems e.g. contribution to metal pollution. Besides the quantity of metal release, metal speciation and possible transformation processes present danger to aquatic biota. In experimental mixtures of WW effluents with receiving waters, discharged dissolved trace metals showed different forms of behavior. Cobalt showed adsorption onto particles, while Cu showed desorption from particulate into dissolved state. In presence of high amounts of river particles, interactions with WW contaminants were elevated.

Introduction

Urban water systems receive liquid and particulate contaminants amongst others by treated and untreated WW discharge. This deteriorates the quality of receiving freshwater systems and induces various environmental impacts, e.g. contribution to metal pollution [1,2]. Besides the quantity of metal release, especially the metal speciation and possible transformation processes are of environmental concern presenting serious danger for aquatic biota [3]. Depending on the chemical species, many heavy metals as well as many essential trace metals in excess quantities can cause toxicity to organisms. Final effluents of wastewater treatment plants (WWTP) are generally dominated by soluble metal forms [4,5], which are known to be the most toxic and bioavailable species [6]. Since WWTP effluents and receiving waters differ in physicochemical and biological parameters, their confluence will probably entail the modification of metal speciation. Many researchers report variations in the partitioning

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between particulate and dissolved metal phases in the dispersion plume of WWTP effluents [1,7,8]. Adsorption and complexation reactions appear to be the main processes controlling the transition of dissolved metal species into particulate phase. Trace metal adsorption onto suspended particulate matter (SPM) is influenced by the particle mineralogy and size as well as the surface area and type of adsorbing surface such as oxide and organic matter coatings [9,10]. However, the role of river particles in the alteration of toxic and bioavailable trace metal species in WW dispersion plumes was not investigated and determined yet. The present study was realised as preliminary investigation of (1) the contribution of discharged dissolved trace metals to urban freshwater pollution and (2) interactions between these dissolved trace metals and river particles. The studied trace metals were dissolved cobalt and copper (Co_D, Cu_D), essential micronutrients for living organisms, which can be noxious or even toxic from a certain concentration on. Study sites were the Garonne and Alb Rivers crossing the cities of Bordeaux, SW-France, and Karlsruhe, SW-Germany, and receiving final effluents from municipal WWTP. The Garonne River receives ~3 m^3/s WW effluent of the WWTP "Clos de Hilde" (CDH) [11] representing 1% of the mean Garonne River discharge. So, the effluent is generally diluted by more than a hundredfold. In times of low river discharge, the proportion of the WWTP CDH effluent can amount up to 3%, thus the dilution decreases. The Alb River has a discharge of only 3.4 m^3/s [12] and receives approximitaly the same amount of WW, namely 2-4 m^3/s, from the WWTP "Klärwerk Karlsruhe" (KK) [13].

Experimental – Method

Water sampling was realized on November 18th and 30th 2009 at Bordeaux and on February 2nd at Karlsruhe in order to observe trace metal behavior regarding fixation and desorption processes with respect to varying physicochemical parameters. Sample water was taken with the aid of a plastic bucket. In this bucket the physicochemical parameters pH, oxygen content, conductivity and temperature were measured. Sample water was filled in acid-cleaned plastic bottles and transported in the laboratory. There, water samples were filtered with polypropylene syringes and syringe filters (cellulose acetate) of 0.2 µm pore size. To one aliquot of the filtered sample water HNO_3 (0.02 mol/L) was added to reduce the pH to a value <2 in order to avoid potential metal adsorption on the labware surfaces as well as microbiological activity. Dissolved nutrient (Na, K, Ca, Mg, Fe, Mn) and trace metal (Co, Cu) concentrations were measured by means of ICP-MS. In a non-acidified sample aliquot, the dissolved anions PO_4^{3-}, SO_4^{2-}, NO_3^- and Cl^- were analysed by means of ion chromatography. A water volume of several hundred milliliters was passed through a 0.2 µm cellulose acetate filter with the aid of a Nalgene filtration unit. The recovered filter was dried at 45°C in a drying cabinet. Afterwards, the filter containing SPM was digested by acids (HNO_3, HF, $HClO_4$). The resulting solution was determined by ICP-MS on concentrations of nutrients and trace metals. For the determination of the dissolved organic carbon (DOC) content, sample water was filtered through a 0.7 µm Whatman GF/F fibreglass filters (pre-combusted at 500°C) performing the filtration with a glass syringe. To maintain the DOC content of the filtrate, bacterial activity was stopped by adding chloric acid (0.3 mol/L HCl). For the determination of the particulate organic carbon (POC) content, sample water was filtered with a Nalgene filtration unit. The used filter (0.7 µm Whatman GF/F filter of 47 mm diameter and known weight) was dried at 45°C in a drying box and then reweight before being stored in a slide until analysis. Besides for the POC determination, the filter served for the calculation of the total SPM content. The chemical composition and topography of SPM on these filters were determined by means of scanning electron microscopy (SEM). In order to observe trace metal partitioning processes taking place at the outlet of WWTP effluents, samples of WWTP effluents were mixed with receiving water and agitated over a time span of five days. Within fixed intervals of time, the mixtures were analysed regarding dissolved trace metal content. To consider the dilution effect of river systems, lab experiments were carried out. Mixtures of WW and river water were prepared in different ratios (1:2, 1:5, 1:10, 1:100 in Bordeaux, only 1:2 in Karlsruhe). This simulates the spatial progression of the WW effluent stream and the influence of increasing dilution by river water on trace metal transformation processes. All WW dilutions were produced one time with untreated and one time with filtered river water and all dilutions were prepared in three replicates. Additionally, test solutions of all sampled waters were prepared. All samples were placed on a mechanical shaker and shaken over a period of five days. After 1, 4, 8, 24, 48 and 120 hours the shaker was stopped. 5 mL aliquots were taken from each sample and analysed by means of ICP-MS. At the beginning, after the first day and at the end of the sorption experiment, the pH, oxygen content and the temperature of the river water and the WWTP effluent sample were measured.

Results and discussion

The WWTP effluents and their receiving waters differed regarding their particle composition, physicochemical parameters and nutrient concentrations. Dissolved Mo, Co and Cu showed further different behavior when WW got in contact with river water. In the following these results will be presented and discussed.

Particles

In the Garonne River 314 mg/L SPM were measured, of which 3.1 mg/L POC. The Alb River contained 9.9 mg/L SPM, of which 1.8 mg/L POC. The WWTP effluents contained 3.9 mg/L (CHD) and 12.5 mg/L (KK) SPM, of which 1.7 mg/L and 5.0 mg/L POC, respectively (Table 1). Thus, at the Bordeaux study site, the SPM as well as the POC content was higher in the river water than in the WWTP effluent. In contrast, at the Karlsruhe study site, the SPM and POC content were higher in the WWTP effluent than in the river water. SPM found in the Garonne and Alb Rivers mainly consisted of silicates, e.g. quartz, feldspar, amphibole and mica. Besides, non-siliceous minerals such as calcium carbonates and dolomite as well as Fe, Al and Mn oxides were present. In addition, organisms such as diatoms and organic matter like plant remains were found. In the WWTP effluents the SPM was of the same composition. Further organisms, which were found very often beside diatoms are illustrated in Fig. 1.

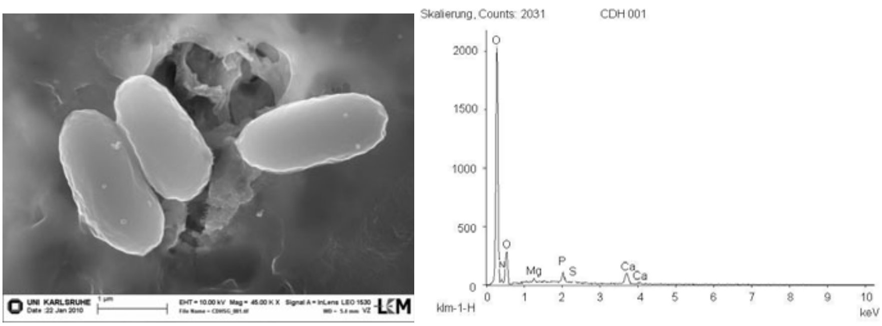

Fig. 1. SEM-image (scale 1 μm) and EDX-spectrum of bacteria.

Due to the oval form of appearance, the size of ~ 1 μm and the chemical composition of carbon, phosphor, sulphur and other nutrients, they were supposed to be bacteria. Besides, other phytoplankton was observed, namely the green microalgae *Chlorococcum infusorium* (Fig. 2).

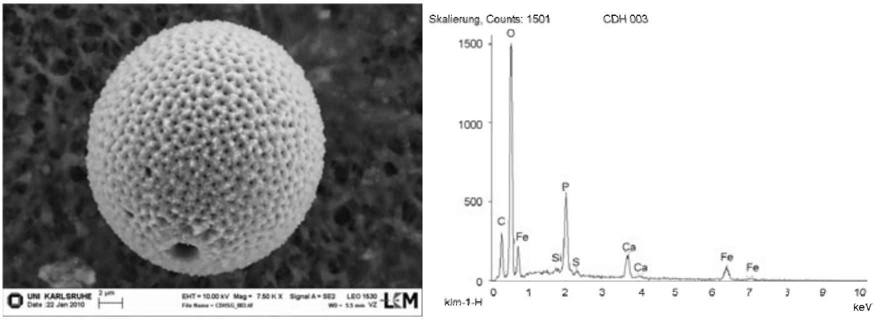

Fig. 2. SEM-image (scale 2 µm) and EDX-spectrum of *Chloroc. infusorium*.

The EDX-spectra of SPM of both WWTP effluents showed almost all the presence of carbon, phosphor, sulphur and calcium indicating that the particles and the filter surface were covered with a C-P-S-Ca-layer. Moreover, small particles with the same composition were detected by SEM.

Different water chemistry of WWTP effluents and river waters

The municipal WWTP effluents under study had a higher temperature (T) and DOC than their receiving water systems. In contrast, the pH was lower. Concerning dissolved O_2 the Garonne River and the WWTP CDH showed nearly the same amount, while the WWTP KK effluent showed a clearly lower amount than the Alb River (Table 1).

Table 1. Physicochemical parameters at the different sites at time of sampling.

Physico-chemicals	Garonne	WWTP CDH	Alb	WWTP KK
SPM [mg/L]	314	3.9	9.9	12.5
POC [mg/L]	3.1	1.7	1.8	5.0
DOC [mg/L]	4.9	5.4	7.1	12.5
O_2 [mg/L]	8.9	9.5	13.1	8.4
pH	7.4	6.5	7.7	7.5
T [°C]	10.8	14.7	3.1	11.2

The comparison of water characteristics highlighted further the relative enrichment of WWTP effluents as against the receiving waters for dissolved nutrients (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NO_3^{-}, PO_4^{3-}, SO_4^{2-}, Cl^{-}) confirming the observations by SEM regarding the nutrient layer. Dissolved Fe and Mn as well as Co and Cu were also higher concentrated in the WWTP effluents. All observations are in agreement with previous studies [1,8].

Behavior of Co_D and Cu_D in the WW dispersion plume

In the mixtures of the WWTP effluents with river water, Co_D and Cu_D showed decreasing and increasing concentrations over time, respectively. While the test solutions of the Garonne and Alb Rivers Co_D showed no concentration change, Co_D decreased in the test solutions of the WWTPs. Thus, the decrease can be attributed to partitioning from discharged Co_D into particulate state. In the WWTP effluents the highly concentrated Fe_D and Mn_D, obviously reacted with dissolved O_2. The observed decrease of Fe_D and Mn_D was going along with O_2 depletion indicating the oxidation into Fe and Mn oxides and hydroxides. These newly build oxides and hydroxides provide supplemental adsorption surfaces and are therefore considered as excellent adsorbers of trace metals like Co [14]. Dissolved Co could also have been coprecipitated with these newly build hydrous oxides [14]. Another possible explanation for the partitioning of Co_D may be the generally high complexation capacity of WW effluents [1,15]. Due to high DOC contents, Co_D is supposed to be mainly present as stable organo-metal complexes, which may have participated in adsorption processes onto SPM [1,15,16]. The Co_D transformation into particulate state took place later in the WWTP CDH effluent than in the WWTP KK effluents. This may be due to the lower content of Fe_D in the WWTP CDH effluent and therefore a lesser formation of Fe hydroxides.

After first being subjected to adsorption processes, too, Cu_D increased over time in the mixture of WWTP effluents with receiving waters indicates the partitioning from particulate Cu into dissolved state. Release of both WW and river particles was observed in the test solutions of the different waters. Copper is known to form strong complexes with organic matter in natural waters and to adsorb onto particulate matter [6], its release can be principally attributed to organic matter degradation by bacterial activity [17,18]. In the test solution of the Garonne River the Cu_D increase began after two days, while it took five days in the test solution of the Alb River. Thus, the microbial Cu release seemed to be higher and/or more rapid in the former. One indicator for higher bacterial activity in the Garonne River is its lower oxygen saturation. The reason may be the temperature influencing the growth of bacteria. Even cold loving bacteria have their optimum temperature at 15-20°C [19]. The Garonne River had a temperature of over 10°C at time of sampling and reached room temperature (20-25°C) within the first day of the batch experiment. The Alb River had a temperature of only 3°C at time of sampling, so bacterial activity was probably inhibited. Bacteria obviously needed more time to adapt to the changing temperature conditions, thus to become active and start organic matter degradation. The higher and/or more rapid bacterial activity in the Garonne River may also be due to the influence of flood tide in the Gironde Estuary. As a result of sea water intrusion in the estuary, the water body in the Garonne River branch is flowing upstream two times a day. So, proportions of municipal WW are supposed to be even present at the sampling point upstream the discharge point of the WWTP CDH. The presence of allochthonous organic matter, nutrients and microorganisms may be the reason for increased bacterial activity when compared to the Alb River [20]. Since the Cu release was higher and began earlier in the WWTP effluent samples, the organic matter degradation and thus the activity of WW bacteria seem to more intense.

Increased sorption of and from Garonne particles in presence of WW

In order to investigate if the sorption processes taking place in the mixtures of the WWTP effluents with river water was a result of the reactions taking place in the separate waters or if additional interaction occurred between discharged compounds and river particles, expectation values were calculated on the basis of the metal concentrations measured in the test solutions. The mixture of the WWTP KK effluent with Alb water behaved conservatively. The Co_D and Cu_D concentrations represent the average concentration values of the two separate waters over time, no interaction took place between discharged WW and river particles. Since the amount of river SPM was more or less equal to the amount of WW particles, the SPM content in the dispersion plume is believed to stay constant. Thus, Alb particles do not display additional surface area for adsorption processes and do therefore not augment the partitioning of Co_D onto SPM. Neither the rivers POC is affected by the presence of WW bacteria. Since the POC of the Alb River is less than half the content of the WWTP KK effluent, the discharged WW bacteria find less degradable substances in the dispersion plume than in the undiluted WWTP effluent. In contrast, the discharge of the WWTP CDH into the Garonne River obviously had impact on river particles and their sorption behavior. On the supposition of conservative mixing of the separate waters the expectation values for Co_D were higher and those for Cu_D lower than the measured concentrations. Apparently, the behavior of discharged Co_D and Cu_D is not only controlled by the dilution with river water. Further partitioning between dissolved and particulate phases takes place within the dispersion plume. At a low dilution of the WWTP CDH effluent Garonne particles adsorb discharged Co_D. At the same time, the riverine organic matter degradation seems to be augmented due to the presence of WW bacteria what leads to further release of complexed Cu_D. However, WW a hundredfold diluted had no effect anymore on Co and Cu transitioning.

The fact, that additional sorption processes could be observed for the Bordeaux, but not for the Karlsruhe study site, lead to the assumption that theses processes were related to the content of river SPM in the dispersion plume. The Alb River contained the same amount of SPM than the WWTP KK effluent, whereas the Garonne River contained nearly a hundred times more SPM than the WWTP CDH effluent. So, the SPM content in the dispersion plume of the WWTP CDH effluent is believed to augment with increasing distance from the effluent outlet. Receiving waters are known to take on the characteristics of the WW effluent below the outlet, resulting in the augmentation of the DOC content and the complexation capacity [1]. So, organo-metal complexes formed in the mixture of the WWTP CDH effluent and Garonne water, encounter not only WW or newly built particles as carrier phases, but also an increasing amount of Garonne SPM in the increasing WW dilutions. In the two-, five- and tenfold WW effluent dilution the Garonne SPM adsorbed 20%, 30% and 24% of the Co_D fraction, respectively, within 24 hours (Fig. 3). This transformation has to go along with an augmentation of the Co charge of the Garonne SPM, which was initially at 25 mg/kg Co_P. The further Co_D adsorption entailed the augmentation of the Co_P charge by 2.8%, 1.3% and 0.8%, respectively.

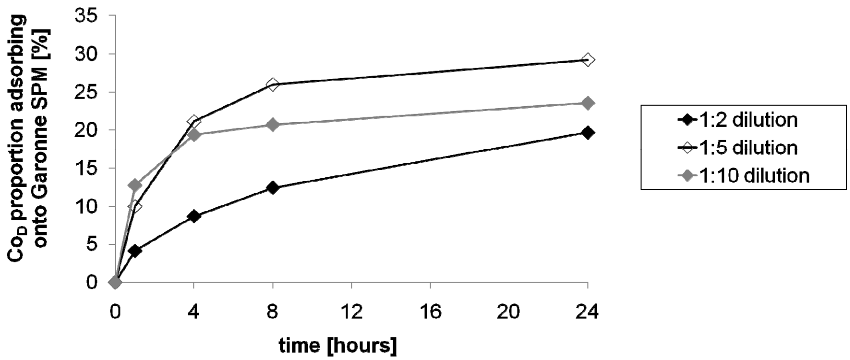

Fig. 3. Proportion of Co_D [%] adsorbing onto Garonne SPM in the different dilutions of the WWTP CDH effluent with Garonne water.

The Cu_D release over time is illustrated in Fig. 4. Between the fourth and 24th hours of agitating the mixture of the WWTP CDH effluent and Garonne water, Garonne particles released 322 ng/L Cu_D in the 1:2 dilution. In the 1:5 dilution, the Cu_D release from Garonne particles amounted to 420 ng/L and in the 1:10 dilution to 248 ng/L. The additional Cu_D release augmented the Cu_D fraction in the two-, five- and tenfold WW dilution by 9%, 19% and 14%, respectively, within 24 hours.

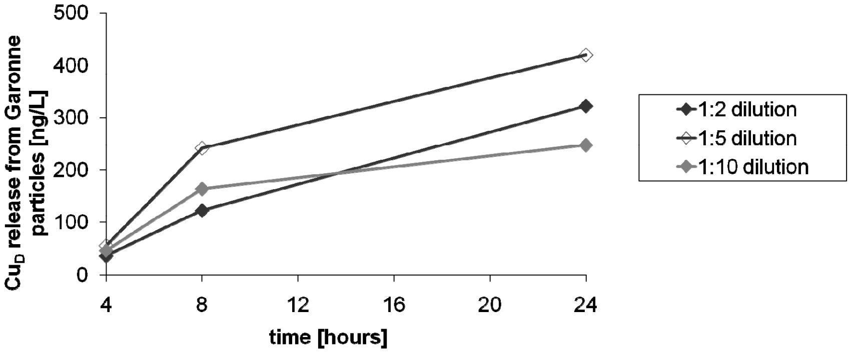

Fig. 4. Increase of Cu_D [ng/L] due to release from Garonne particles in the different dilution of the WWTP CDH effluent with Garonne water.

This desorption has to go along with a decrease of the Cu charge of the river particles, which was initially at 44 mg/kg Cu_P. In the two-, five- and tenfold WW dilution the Cu charge of the Garonne particles may decrease by 4.7%, 3.9% and 2.5%, respectively. The increased release of organically bound Cu from river particles in the WWTP dispersion plume may be due to the introduction of a large amount and high diversity of bacteria. WW has been shown to be an important source of heterotrophic bacteria, which cause significant oxygen depletion downstream effluent discharges, since the WW bacteria are believed to compete with autochthonous bacteria and take part in the degradation of riverine organic matter [21-25]. Since not only the SPM; but also the POC content of the Garonne River is higher than that of the WWTP effluent, the POC increases in the dispersion plume of the WWTP CDH effluent with increasing distance from the outlet displaying nutrition for present WW bacteria. So, in a low dilution of the WWTP CDH effluent the interaction of WW constituents and Garonne particles is highest. While the adsorption of discharged Co_D onto river SPM means a reduction of its bioavailability and toxicity and thus less endangerment of aquatic biota, the Cu_D release augments the environmental risk for organisms.

Conclusion

The present work has shown that the dilution of WWTP effluents into river systems of high SPM and POC content, such as the Garonne River, induces interaction between WW pollutants and river particles. In low WW dilutions, thus in the immediate proximity of WWTP outlets, river particles were identified to represent a source for dissolved Cu and a sink for dissolved Co affecting the transport and fate of both trace metals. Discharged Co_D was shown to get generally adsorbed by WW or by newly built particles as a result of the simultaneous release of complexing agents as well as Fe_D and Mn_D transforming into oxides. Obviously the adsorbing surfaces of river SPM may be used as additional carrier phases. So, while river SPM minimize the concentration of discharged Co_D, their charge with Co may augment by nearly 3% in the dispersion plume of the WW effluent. The bioavailability and toxicity of Co is therefore decreased. At the same time, the release of heterotrophic WW bacteria entails competition with autochthonous river bacteria and augments the microbial degradation of riverine organic matter. The river SPM charge may be reduced by almost 5% releasing adsorbed Cu into dissolved state and augmenting therefore the environmental risk for aquatic organisms. With increasing dilution by river water, thus with increasing distance from the WW outlet, the impact of the WWTP CDH effluent on Garonne SPM becomes obviously negligible (at a dilution of 1:100). However, in case of low water discharge in midsummer, the achieved 1:10 dilution does not seem to be sufficient to avoid interaction between WW and river particles.

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Speciation of trace metals in organic matter of contaminated urban sediments

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Abstract

A methodology based on the coupling of chemical methods (methyl isobutyl ketone (MIBK) and IHSS fractionations) and physical characterization methods (XRD and SEM) was applied to study the relevance of these two fractionation methods on the speciation of trace metals in organic matter of three stormwater sediments. Trace metals distribution among MIBK and IHSS fractions is very different. Except for Cr, 10 to 30% of trace metals are bonded to MIBK-humin fractions, whereas 70 to 100% are bonded to IHSS-humin fraction. Moreover, Cd, Cr, Ni and Zn are strongly bonded to the highly aggregated fraction.

Introduction

The availability of pollutants in sediments and soils depends on their distribution between the different solid phases, which is now usually called speciation. Characterizing pollutants in soils or sediments has therefore to go beyond a simple determination of total content. Speciation is necessary and should be performed using physical and/or chemical methods. Indirect and direct methods of speciation can be distinguished. In case of indirect methods, the pollutants (mostly trace metals) are removed from their structural context by solubilization and their analysis is indirect insofar it is carried out in solution. The main critics of these methods are their non-specificity (incomplete extraction) and the possible re-adsorption of metals [1]. To overcome these critics the speciation of pollutants in soil or sediment has to be considered by fractionation (direct method).

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The IHSS (the protocol recommended by the International Humic Substances Society) [2] and the MIBK (Methyl IsoButyl Ketone) fractionation protocols [3] are two chemical fractionation methods that have been developed to study the organic matter. Both methods allow the fractionation of fulvic acids (FA), humic acids (HA) and humin according to their solubility properties. The asset of the MIBK method is isolation of the mineral fraction (MF). Durand (2003) [4] studied the repartition of trace metals between IHSS fractions on stormwater sediments. It appears that besides chromium and nickel, trace metals are mainly associated to FA. Doick *et al.* (2005) [5] studied the distribution of two polycyclic aromatic hydrocarbons (HAPs) (fluoranthene and benzo[a]pyrene) and two polychlorinated biphenyls (PCBs) (28 and 52) among MIBK fractions on an agricultural soil. The mineral fraction of the investigated soil apparently played a significant and previously unreported role in the sequestration of both studied PAHs and PCBs. However Doick *et al.* [5] recommended to test the hypothesis that the mineral fraction may contain a residual unremoved quantity of organic matter (OM).

The objectives of the present work were: i) to characterize the MIBK and adapted IHSS fractions by physical methods; ii) to study and compare the distribution of trace metals among MIBK and adapted IHSS fractions. The methodology used is based on the coupling of MIBK and adapted IHSS fractionation and physical methods of characterization (Infrared spectroscopy (FT-IR), X-ray diffraction (XRD) and scanning electron microscopy (SEM)).

Material & Methods

Sampling sites

Three stormwater infiltration basins located in Nantes (west of France) and Lens (north of France) were selected:

- the first one, the Cheviré basin, located south west of Nantes, receives runoff waters from a 19,000 m² contribution area of the south part of the Cheviré bridge which is a section of the Nantes ring-road. In use since 1991, the Cheviré bridge now carries an average daily flow of about 100 000 vehicles.
- the second basin, Boisbonne, is located in the east part of Nantes, the drained runoff waters come from a 16,000 m² contribution area of the All highway. The average daily flow is about 27000 vehicles. The Boisbonne basin is in use since the opening of the A11 highway in 1992 and had never been dredged since this date.

- the third basin (G08), located in the east part of Lens, drains runoff waters from a 14,000 m² contribution area of the A21 highway and carries an average daily flow of 80 000 vehicles. Since its opening in 1971, the basin had never been dredged.

A representative sample was composed of a homogeneous mixture of 6 to 13 shots distributed according to a grid system for each basin. Samples were sieved to 2 mm and dried at 40°C.

Sample characterization

Solid phase characterization

Major crystalline phase identification was carried out by X-ray Diffraction (XRD) on powered samples with Brüker "D8 Advance" powder diffractometer operated in Bragg-Brentano geometry. The phase identification was performed using EVA software in conjunction with the Powder Diffraction File (PDF) database.

The SEM (Scanning Electron Microscopy) used for this study is a Hitachi S 570 with LaB6 tip excited by a source that can produce up to 30kV. In addition to the picture, a qualitative analysis of the basic structure is made using an EDX (Energy Dispersive X-ray spectroscopy) probe which can save energies of 1000 to 10 000 eV.

The Fourier transform infrared spectra were recorded by diffuse reflectance using a Perkin-Elmer FT-IR spectrometer (Spectrum 1000). The spectral range is 4000 to 600 cm^{-1}. The samples are crushed and mixed with potassium bromide (KBr).

Chemical analyses

Total element content, especially trace metal concentrations (Cd, Cr, Cu, Ni, Pb and Zn) were determined. The term "total" is used here as the amount of metal dissolved according to the acid attack (NF X 31-147 [6]), the dissolution was performed with HF and $HClO_4$ acids. The quality of the analytical data for the acid attack was assessed by carrying out analyses of the certified reference material BCR-320 (river sediment). Total organic matter content was determined by combustion at 550°C.

Element analysis was performed using ICP-OES (Inductively Coupled Plasma – Optical Emission Spectrometer) (Varian 720-ES). The quantification limits for Cd, Cr, Cu, Ni, Pb and Zn are respectively 0.5, 5, 2, 10, 10 and 2$\mu g.L^{-1}$. In case the concentrations are under the limit of quantification, the sample was analyzed using ICP-MS (Inductively Coupled

Plasma – Mass Spectrometer) (Varian 820-MS). The ICP-MS limit of quantification for trace elements is ranging from 0.02 µg.L^{-1} to 0.3 µg.L^{-1} depending on the element.

Elemental analyses of carbon, hydrogen and nitrogen were provided by flash combustion on a FlashEA 1112 analyzer (Thermo Electron Corporation). Percentages of C, H and N are automatically calculated using the Eager 300 software.

IHSS and MIBK fractionation procedures

MIBK and IHSS methods allow the fractionation of fulvic acids (FA), humic acids (HA) and humin according to their solubility properties. An isolation of the mineral fraction (MF) is enabling with the MIBK protocol.

An adapted IHSS protocol, corresponding to the IHSS protocol [2] without the lipids extraction, was performed on 5g of dried samples, in order to keep lipids in their original structure and also to avoid the addition of HCl on the entire sample (Fig. 1a). Humic acids (HA) are recovered by precipitation at pH 1 and centrifugation (8000 g, 15 min, SIGMA 3-15), Fulvic acids (FA) were concentrated by freeze-drying (Bioblock Scientific, Christ Loc 1 m alpha).

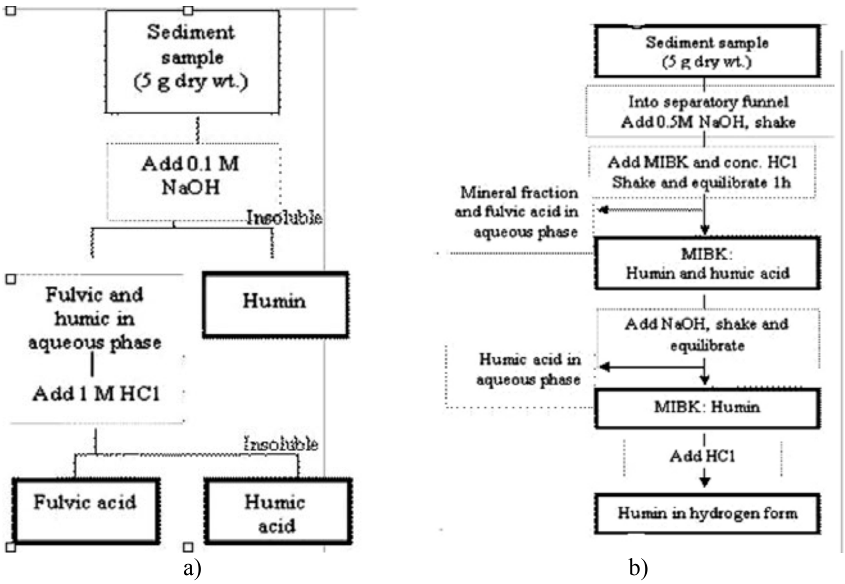

Fig. 1. a) adapted IHSS and **b**) MIBK fractionation procedures.

The samples (5g of dry wt) were fractionated using the MIBK method described by Rice and MacCarthy [1]. Theoretically, four solid phases are recovered (Fig. 1b). The mineral phase was collected from the FA fraction by centrifugation (8 000g, 15-20 min, SIGMA 3-15). The FA fraction was concentrated by freeze-drying. Hydrogened humin (through the addition of 0.1 M HCl) (HS) remains in suspension in the organic phase and can be collected by evaporation of the MIBK solvent.

This is foreseen that HCl solutions added during the IHSS and MIBK procedures could solubilize the trace metals present in carbonated solid phases or weakly adsorbed on solid fractions. Therefore the protocols could be a priori more relevant to study the speciation of strongly adsorbed trace metals on mineral and organic phases.

Results and discussion

Description of bulk sediments

The XRD analyses indicated that quartz, albite (Na feldspar), microcline (K feldspar) and muscovite (mica) are present in the three sediments. Clinochlore (clay mineral) is also present in Chevire and G08 sediment. A complementary carbonated phase (calcite) was also identified in G08 sediment. The FT-IR spectra are similar for the three bulk sediments. The G08 sediment spectrum presents four additional absorption bands at 2514, 1793, 1448 and 879 cm^{-1} corresponding to calcite mineral. An absorption band at 3620 cm^{-1} could correspond to Si-OH vibrations in clays. An important contribution of alcohol OH groups (3350 cm^{-1}) and aliphatic group (2860-2930 cm^{-1}) is observed. A broad band between 900 and 1100 cm^{-1} can be due to C-O stretching band of ethers, esters, alcohol but could also probably essentially to Si-O bonds in these mineral rich samples (quartz, chlorite, muscovite)[7]. For Boisbonne sediment, clay minerals were only detected by FT-IR spectrometry.

Table 1. Elemental analysis of the studied samples.

	%TOM	%Ctot	%H	%N	%CaCO3	%Corg
Boisbonne	16.9	8.2	1.6	0.8	0.5	8.1
Cheviré	16	10.2	1.4	0.4	0.5	10.2
G08	17	13.5	1.4	0.3	13.7	11.7

Total organic matter (TOM) contents are similar (about 17% wt.) for the three sediments (Table 1). After correction for carbonate contribution, the percentages of organic carbon in sediment are 8.1, 10.2 and 11.7 for

Boisbonne, Cheviré and G08 sediments respectively. G08 has the highest organic carbon amount whereas Boisbonne has the highest hydrogen and nitrogen contents. The oxygen and sulfur contents were not determined.

The main mineral phases (quartz, feldspar, mica and clay) have been observed by SEM and EDX analysis which is consistent with XRD analysis. Aggregates composed of minerals and a binding phase which we believe to be organic matter are predominant in the samples. Such aggregates were previously mentioned by Badin *et al.* (2009) [8] in a sediment from a Lyon (France) infiltration pond.

Table 2 displays the total trace metals contents in the three basins. G08 basin is the most contaminated basin in trace metals whereas Boisbonne is the least. Cheviré and G08 sediments are highly loaded with copper, lead and zinc, their contents being above the Dutch intervention thresholds.

Table 2. Total trace metals contents (mg.kg^{-1}) in the three sediments.

	Cd	Cr	Cu	Ni	Pb	Zn
Boisbonne	1.2	54	59	75	41	480
Cheviré	1.2	92	366	37	368	1863
G08	9.6	111	489	54	1064	2587

IHSS and MIBK fractions characterization

The MIBK fractionation protocol applied to the sediments normally allows the determination of four fractions. However additional fractions were obtained by comparison with Rice and MacCarthy [3] or Doick *et al.* [5] results when the MIBK fractionation procedure was performed on our samples. In fact, for the three sediments, instead of having all the humin fraction remaining in suspension in MIBK, a part of the organic matter has decanted in the aqueous phases. This fraction is named heavy humin (HH). For Boisbonne, another additional fraction was obtained. In the second step of the MIBK procedure, HCl solution is added in order to solubilize humic acids (HA) and a part of the suspended humin decanted instead of remaining in suspension in the MIBK solvent. This fraction is named “heavy humin decanted with HA (HHA)”. These results may be due to the nature of the sediments containing anthropogenic organic matter and to their highly aggregated structures [8].

XRD analysis were carried out on all fractions of the three sediments except FA and HA. Quartz, feldspar, chlorite and mica peaks are observed with variable intensities in HHA, HH, HS and IHSS humin fractions. The absence of calcite peak in HHA, HH and HS could be due to the addition of HCl solution (dissolution of carbonates). Only quartz and feldspar were noticed in the mineral fraction (MF). Halite (NaCl) is identified in the mineral fraction and in the humin in suspension (HS). This precipitation is due to the successive additions of NaOH and HCl. The FT-IR spectra are consistent with the XRD analysis for the MF regarding the absence of clay peak at 3620 cm^{-1}. The FA and HA fractions spectra from the two fractionation methods and the three samples have the same shape. Alcohol OH groups are represented by a large band at 3350 cm^{-1} and broad bands between 1650-1455 cm^{-1} correspond to aromatic C=C vibrations. HHA, HH, HS and IHSS humin spectra from the three samples have the same shape and approximate the bulk spectra. The difference rests on the Si-OH (3620 cm^{-1}) and aliphatic group (2860-2930 cm^{-1}) which are more intense than in bulk spectra.

Total carbon and organic matter contents were determined when fractions contain enough material (Table 3). The organic matter content of FA fractions could not determine due to mass uncertainty (FA in sodium salt form). The mineral fraction obtained by MIBK fractionation still contained a small amount of organic matter (total carbon content ranging from 0.5 to 3.5%). Moreover, the total carbon contents in HA fractions from IHSS and MIBK protocols are very different (from 0.3 to 4 times). The total carbon and organic matter contents of IHSS humin fraction correspond to that of HH in the MIBK fractionation.

Table 3. Total carbon and organic matter contents in the obtained fractions ("–" not determined due to low quantity of solid phase).

		MIBK					IHSS	
		MF	HA	HHA	HH	HS	HA	Humin
Boisbonne	%Ctot	0.5	29.5	5.5	5.7	12.2	41.8	5.0
	%OM	2.5	–	15.2	14.7	23.8	–	12.7
Cheviré	%Ctot	1.7	9.8	–	8.7	17.5	43.5	9.0
	%OM	3.3	–	–	15.0	29.0	–	14.9
G08	%Ctot	3.5	8.3	–	11.1	20.0	32.6	12.7
	%OM	–	–	–	16.4	29.9	–	18.3

All the fractions (except FA fraction) were observed by SEM. Similar conclusions could be drawn for the three sediments. The humin fraction observations are very close to that for the bulk sediment. The mineral fraction (Fig. 2a) is mostly composed of crystallized mineral particles from 30 to 500 µm. A few aggregates (100 to 300 µm) can be noted. Crystallized NaCl was identified by EDX analysis. The general aspect of the heavy humin (HH) fraction (Fig. 2b) and the humin in suspension (HS) from the MIBK fractionation is roughly the same: well crystallized mineral particles from 10 to 100 µm and aggregates (up to 100µm) are observed. The difference rests on aggregates sizes which are smaller for

HS. The humic acid (HA) fraction from the IHSS fractionation presents particles from 50 to 100 µm (Fig. 2c). In the MIBK-HA fraction, same particles are recognized but they are covered by NaCl. For the heavy humin decanted with humic acids (HHA) for Boisbonne, mineral particles are recovered and fibrous particles are observed (Fig. 2d). These particles could be poorly crystallized illite.

The study of IHSS and MIBK fractions shows that even if the total carbon contents of a fraction obtained by the two methods are different, the general aspect of its FT-IR spectra is the same. The difference rests on the band intensities. All fractions present mineral phases (except FA) and organic contents. Moreover, heavy humin fractions present large aggregates that can explain the decantation process.

Trace metals speciation

Acid attacks (with HF and $HClO_4$) were carried out on all MIBK fractions (except fulvic and humic acids) and on IHSS-humin fraction to quantify cadmium, chromium, copper, nickel, lead and zinc contents. The IHSS-FA fraction was dissolved in NaOH to assess trace metals content.

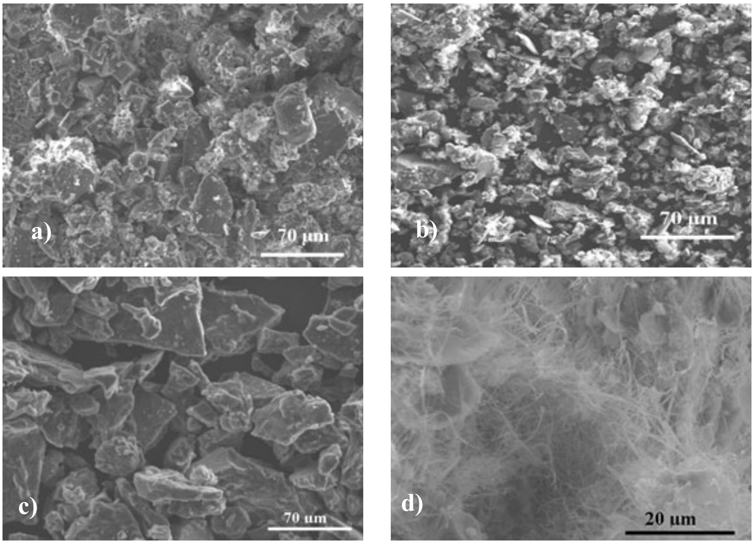

Fig. 2. SEM pictures of a) G08 mineral fraction (x 300); b) Chevirté heavy humin fraction (300x); c) Boisbonne IHSS humic acids fraction (300x); d) Boisbonne heavy humin decanted with HA (x1470).

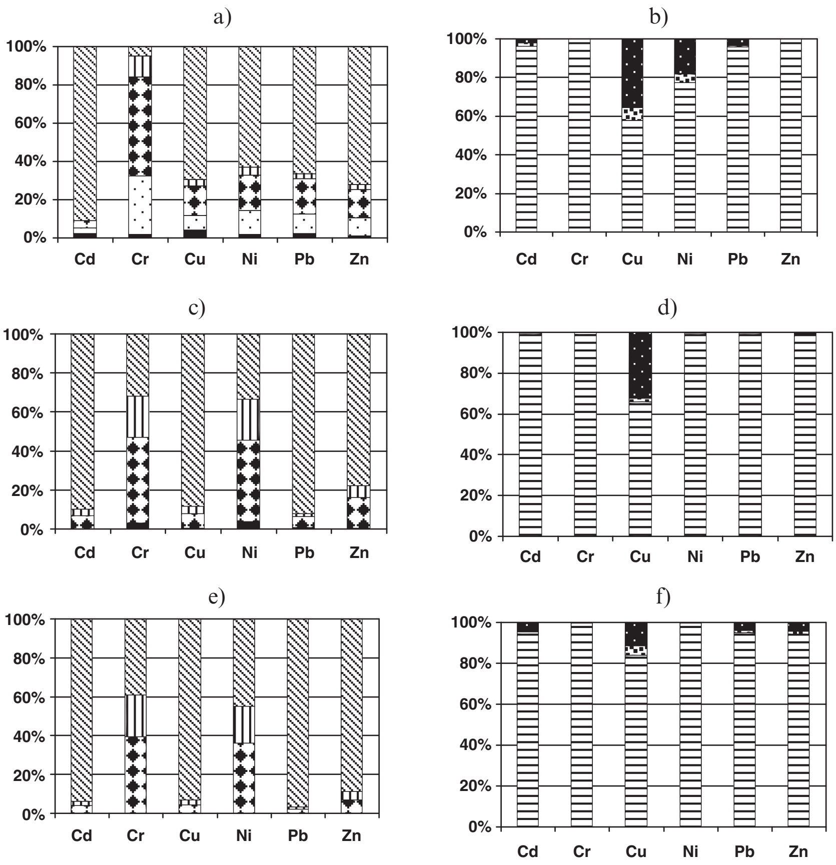

Fig. 3. Trace metal distribution in MIBK (left) and IHSS (right) fractions for Boisbonne basin a) & b); Cheviré basin c) & d); and G08 basin e) & f). Legend for MBIK: MF (black), HH (black diamonds), HS (vertical lines), FA and HA (slantwise lines), HHA (black dots and white background); legend for IHSS: humin (horizontal lines), FA (black points), HA (white dots and black background).

Trace metals concentrations in MIBK-FA and HA fractions and in IHSS-HA fraction were obtained by subtraction from total contents of bulk samples. Fig. 3 displays the distribution of cadmium, chromium, copper, nickel, lead and zinc in the MIBK and IHSS fractions. Cd, Cu, Pb and Zn are mostly present in MIBK-FA and HA fractions (70 to 90%). Solubilization by HCl could explain a major part of the trace metals amount in MIBK-FA and HA fractions. 10 to 30% of these trace metals were detected in humin fractions. Cr and Ni have a different speciation with 40 to 90% of the content in humic fractions. This result indicated that Cr and Ni were more strongly bound to organic matter than the previous trace metals. Heavy humin (HH) is the predominant organic trace metals bearing phase, considering the fraction of metals strongly bound (not adsorbed on FA or HA). The mineral fraction contains very low amount of trace metals (only up to 5%).

The distribution of trace metals in IHSS fractions was really different. More than 90% of Cd, Cr, Pb, Zn amounts are present in IHSS-humin fraction. Ni content in humin fraction was also close to 100% in Cheviré and G08 sediments, but 25% of Ni was present in IHSS-FA and HA fractions of Boisbonne sediment. Cu exhibits also a quite different speciation. As it could be expected from the literature, Cu was present from 20 to 40% in IHSS-FA and HA fractions.

El Mufleh *et al.* [9] carried out sequential extractions (indirect method) on the Boisbonne and Cheviré sediments. The sequential extraction was performed by the three steps BCR procedure (the European Community Bureau of Reference) and a fourth step was added consisting of a mineralization of the final residue by use of a mixture of concentrated $HF/HClO_4$. According to this procedure, the first extractible fraction is the exchangeable phase related to clay, carbonates and amorphous phases; the second one is the reducible fraction related to metal oxides and the third one is the oxidizable fraction related to the organic matter. The speciation of some trace metals (Cd, Cr, Ni, Zn) deduced from sequential extractions and the distribution of trace metals among MIBK fractions for Boisbonne and Cheviré sediments are very similar. Cd, highly solubilized, is mainly found in exchangeable and reducible fractions. The amount of Cr, Ni and Zn in FA and HA fractions is also very close to those in the exchangeable and reducible fractions which corresponds to the fractions in which trace metals are the most easily extractible. The amount of Cr, Ni and Zn in the humin in suspension in MIBK (HS) (higher total carbon content) corresponds to their amount in the oxidizable fraction (organic matter phase in the BCR protocol). The heavy humin (HH), the mineral fraction (MF) and the heavy humin decanted with HA for Boisbonne (HHA) could therefore correspond to the residual fraction. No comparison for copper and lead can be drawn due to their high solubilization by HCl solution.

Conclusion

The MIBK fractionation procedure was performed on three sediments from stormwater infiltration basins. One or two additional fractions were obtained by comparison with previous studies ([3] and [5]) on agricultural soils. The nature of organic matter present in sediments could explained this difference. Moreover the characterization of the MIBK fractions showed that the mineral fraction still contains a residual amount of organic matter. The comparison of the trace metals distribution among MIBK and IHSS fractions confirms the high solubilization of metals by acid solution in the first part of the protocol. The study of strongly adsorbed trace metals on organic matter speciation shows that Cd, Cr, Ni and Zn: i) are bonded to the humin remaining in suspension in MIBK solvent corresponding to the organic matter fraction obtained by sequential extraction; ii) are mostly contained in the heavy humin which presents the largest aggregates. It confirms the important role of aggregates in the trace metals retention.

Acknowledgements

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An urban watershed regeneration project: The Costa/Couros river case study

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Abstract

During the last decades, the City of Guimarães was subject to heavy anthropogenic pressure due to increasing urban and industrial occupation, giving rise to the visible deterioration of existing buildings and the environmental degradation of the Costa/Couros River.

Beside project Campurbis, a urban regeneration project with strategic investment in knowledge, technology and innovation, supporting "Guimarães, European Capital of Culture 2012", a specific project is promoting the revitalization and regeneration of the Costa/Couros river as an enhancement and an essential complement to this plan of urban intervention.

This river has been part of the population's life and a vital element for the development of the former leather industry. Through several decades this river has been subjected to severe pressure from the leather industry that, associated with high urban growth, led to high level of pollution and contamination in the river and the need to control the effects of floods that frequently affect the historic center of Guimarães.

The rehabilitation of water resources in urban environments has an added complexity not only because the multidisciplinary approaches needed but also due to the strong history constraints, the lack of space for the implementation of structural measures and the high value of the available land.

A field survey was realized in order to understand and detail the river characteristics and related infrastructures. Based on these elements, the following objectives where defined for this study: Minimizing the flooding of the urban center; Mitigation of the effects of artificiality conduit of various reaches of the river; Stabilization of the banks with risk of erosion; Creation of recreational areas; Elimination of point source pollution and control of diffuse pollution.

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This paper presents the main tasks associated with the characterization and diagnosis of the existing infrastructures in the Costa/Couros river, and the preliminary results of the hydrological and hydrodynamic studies.

Introduction

The Municipality of Guimarães (CMG) and University of Minho (UM), through its School of Engineering, formalized a partnership to implement the Project Campurbis, aimed at urban requalification of part of the historic town of Guimarães, with the integration of an urban campus in the historical center (Figure 1).

Fig. 1. Historical center of Guimarães.

With this strategic investment in knowledge, technology and innovation, the Project Campurbis emerged as an important supplement to the application of the "Guimarães, European Capital of Culture 2012", as formalized by Guimarães municipality.

The urban area covered by the intervention of Campurbis is crossed by the Costa/Couros river. The City Council intends to rehabilitate and revitalize this river, given the current level of pollution and contamination, and the need to mitigate the flood effects, that repeatedly affect this area in the center of Guimarães.

The river was always present in people's life and a vital element for the development of the leather industry. Today, the leather industry is no longer present but the increased urban pressure has led to the occurrence of sewage and stormwater pollution. Thus, the intention of promoting the revitalization and recovery of this river presents itself as an enhancement and an essential complement to this plan of urban renewal, combining the recovery of the more natural riparian zones, present at the City Park and at Veiga de Creixomil, with the artificialized reaches present in the urban center.

Beside this local strategy, the European Water Framework Directive demands a good ecological status for natural water bodies and, therefore, there is an urgent need for controlling the detected sources of pollution.

The restoration of urban watersheds is a multidisciplinary task involving, hydrology and hydrodynamics, biological and political knowledge.

Since a restoration programme can only be effective when the main processes involves are fully understood, a preliminary study was done, focused on the geomorphology and ecology processes in the river, at a watershed scale, incorporating insights from recent research in fluvial geomorphic and ecology, and developing connections between strategies and actions.

Objectives

The disordered occupation of the territory and the lack of effective mechanism for water resources management in the last decades have contributed to ecological imbalances in this watercourse, and modifications of its hydrodynamic regime, increasing the occurrence of floods, erosion and siltation.

Through fieldwork several situations that contribute to the degradation of the aquatic environment have been identified, including the improper disposal of domestic and industrial effluents, and the excessive soil imperviousness, as a result of an intense and growing urban occupation on this watershed. Such situations have led to the obvious negative impacts on the environmental quality of the aquatic ecosystem, particularly in landscape, aesthetics and water quality.

The methodology for the regeneration of this ecosystem must include the definition and prioritization of the intervention areas, the definition and evaluation of options strategies for medium and long term, based on a decision support system that includes not only the potential for human intervention, but also the capacity for self-purification and regeneration of natural systems.

For this restoration project, the defined objectives are:

- Estimation of average month flows;
- Estimation of flood flows and minimization of their effects in the urban center;
- Mitigation of the effects of artificiality banks of some reaches, responsible for significant changes in the river hydrodynamic;
- Stabilization of the riverbanks in critical conditions or with future risk;

- Modeling the river hydrodynamics and evaluate the changes with the proposed interventions;
- Creation of recreational areas, in order to provide interaction with the population;
- Elimination of point source pollution through the drainage and treatment of wastewater that are present in the riverside.
- Estimation of non-point source pollution loads and identification of control measures.

Site Characterization and results

The Costa/Couros river has its spring in Serra da Penha, and presents a total length of 6.2 km with a 11.23 km^2 catchment area. In its upstream section, the river is developed inside the City Park (Fig. 2a), constituting a natural system.

From the City Park, the stream becomes partially channeled in the urban center of the cityof Guimarães. The former mills, tanneries and public tanks are still visible.

At Veiga de Creixomil, an agriculture area, the river reappears again at the surface (Fig. 2c) before flowing into the river Selho.

Fig. 2. Costa/Couros river: a) City Park; b) Channelized reaches in the urban center; c) Agriculture land at Veiga do Creixomil.

As a start for this project, the river was studied thought "*in-situ*" visits, followed by a cadastral survey of the river banks, with particular attention to the reaches that are underground, and the identification of sewage and runoff infrastructures that are discharging directly into the river.

Based on the collected information, an extensive field survey (Figure 3) was done in order to characterize the river and related infrastructure domestic and industrial sewage networks and runoff drainage). The resulting information was organized as cartographic data and a database with the characterization of all inputs to the river.

An analysis to the existent meteorological data was a support for the hydrological characterization of this basin.

Followed by these preliminary tasks, hydrological studies where done in order to understand the precipitation/runoff process and to estimate the river flows. A hydrodynamic model of the river was built in order to compute the hydraulic characteristics of this river, under different flows, to evaluate the related effects, and to propose corrective measures. The estimation of the pollution load by urban runoff is also being evaluated.

Fig. 3. Survey of riverside infrastructures.

Hydrological studies

The flood flows where estimated through empirical, statistical and cinematic methods.

The empirical methods resulted in very high specific flows. These methods are based on geometrical data and, therefore, this estimation process can only represent a rough estimate of the flood flows.

Since there is no historical data of previous flood flows for this river, statistical methods where applied with the pondered correlation of the flow data of a downstream watershed. The obtained results depend on this simplification.

Several cinematic methods where also computed in order to estimate the flood flows. These methods require precipitation data or the definition of design hietograms. Three design hietogramas where used, based on the intensity-duration-frequency relations for this region.

The results obtained from the different methods where very different and, since they all depended on different parameters, their qualitative comparison was not possible.

In fact, small urban watersheds have a specific runoff formation and the routing through the watershed [9]. The estimation of flood flows, based on methodologies used for much bigger watershed, do not represent these specificities and, for this reason, should be evaluated when applied to small urban watersheds.

The routing of runoff is also specific in small urban watersheds and seems to be controlled by their inclination and soil cover. The simple estimation of the concentration time, by several methods, have resulted in very distinct values.

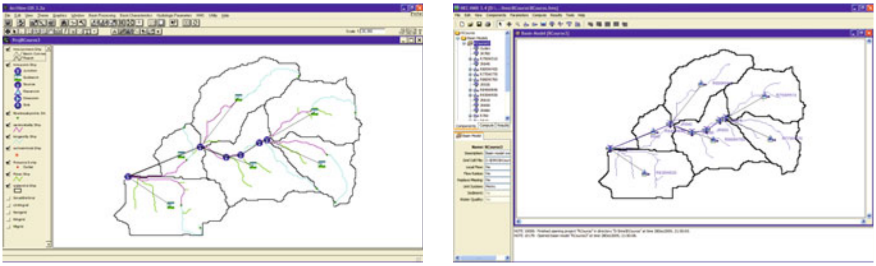


Fig. 4. HEC-HMS hydrological model.

Based on the lacks of the previous methods, a hydrological model was built in order to understand the precipitation/runoff phenomena in the Costa/Couros basin.

The Hydrologic Engineer Center's - Hydrologic Modeling System (HEC-HMS) model was chosen due to the user-friendly interface and worldwide usage [4,5,6,13]. This model user-interface and the definition of this hydrological watershed is depicted in Figure 4.

The resultant hydrograms, as shown in Figure 5, represent the flood flow with time, computed with the specificity of the studied basin. This model also allows future calibrations, based on with real data, and for this reason will be the methodology used for this project.

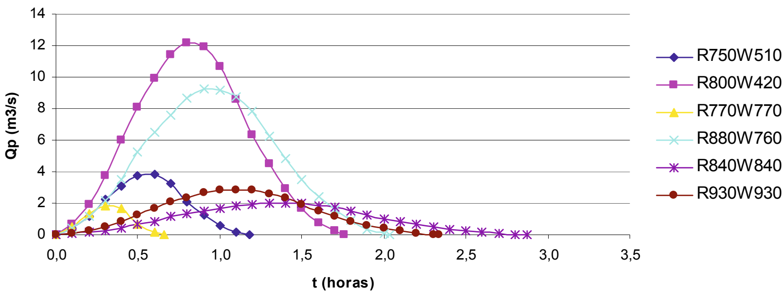


Fig. 5. Resultant hydrograms (HEC-HMS).

Hydrodynamic model

Based on the topographic data a hydrodynamic model of the Costa/ Couros river is being developed. This model will allow estimating the hydraulic parameters for different flows, for each section, and by this way predict the need for protective measures.

For this task the Hydrologic Engineering Centers River Analysis System (HEC-RAS) model was chosen, due to the same reason as the HEC-HMS [8]. The river was defined by section with the characterization of the transversal section, with a spacement of 20 meters. Figure 6 presents the definition of the river sections and a tridimensional representation of the river.

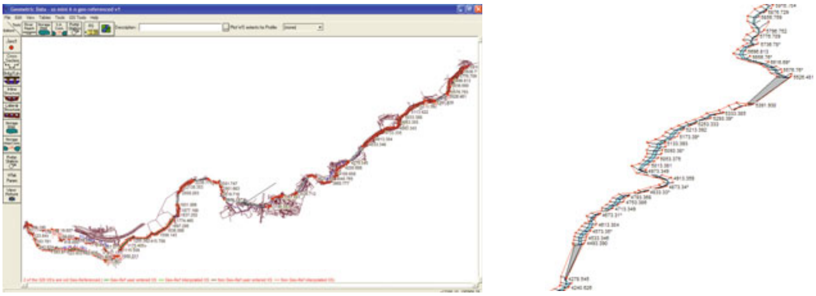


Fig. 6. Hydrodynamic model and tridimensional representation of the river.

Some reaches have already been simulated with the developed model. Figure 7 represents the water level at the city park, with and without the proposed correction measures to the riverbanks.

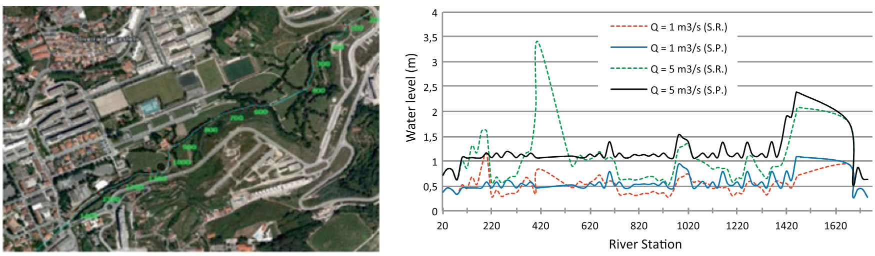


Fig. 7. Hydrodynamic simulation of the upstream reach.

The model is now being completed for the entire river and will allow to prioritize the interventions needed and the related measures.

Pollution load estimates from urban runoff

The pollution load due to urban runoff is also being evaluated by the use of the Storm Water Management Model (SWMM) developed by the United States Environment Protection Agency [11,12,14].

Figure 8 presents the selected study basin and the created model.

Based on the precipitation data used from a nearby rain gauge, with a period of 240 days, the runoff lows and quality was estimated with the model. Figure 9 presents the precipitation data and the correspondent runoff computed by the storm water model.

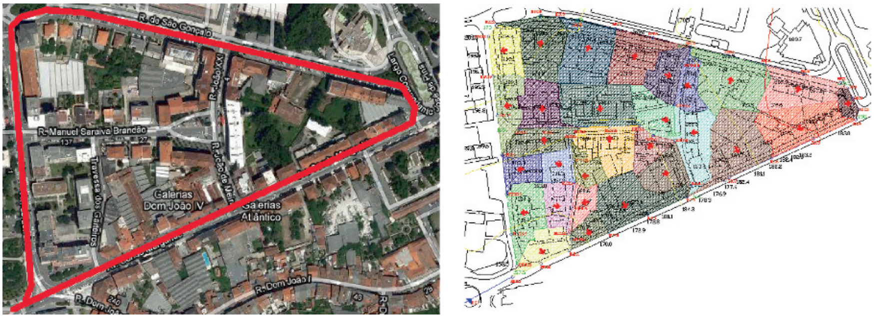
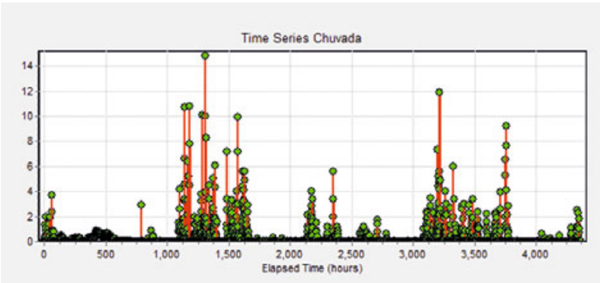
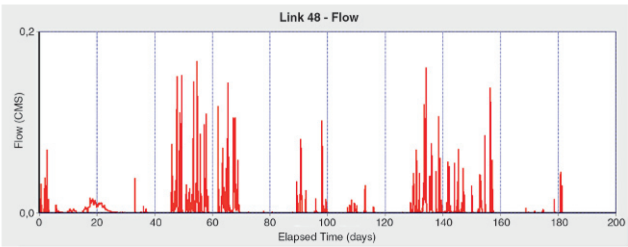


Fig. 8. Case study basin and urban runoff model.



a) Precipitation data



b) Computed runoff

Fig. 9. Precipitation and runoff flows (SWMM).

The pollution concentration and loads for this case study were based on on Site Mean Concentrations (SMC) and Event Mean Concentrations (EMC) found in literature, from other case studies [1,2,3,7,10,12].

Further studies are needed in order to calibrate the model with data from runoff monitoring. These studies will help to understand the phenomena, in order to predict the build-up and the wash-off process, and therefore estimate the pollution loads for this watershed.

Final Notes

The rediscover of the rivers ecosystem importance to the life quality, especially in the urban environment, motivates the implementation of rehabilitation programs to mitigate the errors accumulated from many decades of unsustainable water resources management.

This rehabilitation, especially in the urban environment, presents itself as a demanding and complex mission due to the technical, political and economic issues involved.

This paper presents the main tasks performed to characterize the Costa/ Couros river and to report the preliminary results of the hydrological and hydrodynamic model, in order to predict the nature of phenomena, as precipitation and related runoff, and river hydrodynamics.

The developed models allowed to estimate the flood flows, their effects in the river hydrodynamic, and the quantitative and qualitative nature of urban runoff, and related impacts. Based on these results mitigations solutions are being proposed. The models also allow simulating the response of the case study to future changes in the watershed or the riverbanks.

The implementation of this methodology in the restoration of urban watersheds can contribute to the establishment of effective rehabilitation measures for water resources in urban environment, changing the traditional approach that neglected for decades the values associated with the preservation of aquatic ecosystems as a factor to enhance the quality of life in cities.

Finally, due to the high cost of the implementation of rehabilitation measures and the variability of the phenomena involved, the use of these predictive models can provide an easy an feasible way to evaluate the original conditions, test improvement measures and estimate their effectiveness.

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URBAN SOIL CONTAMINATION AND TREATMENT

The Astysphere, a geoscientific concept for the urban impact on nature

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Introduction

Unquestionably, volcanoes, earth quakes, wind, rain, plant and animal life enormously contribute to form the earth's face by abrupt impacts or permanent ongoing processes. But are they the main forces forming the present earth's face? During the last centuries no geological force has changed the earth's surface as has done mankind. Humans have altered the morphology and element balances of earth by establishing agrosystems first and urban systems later. Globally, geogenic and anthropogenic substance fluxes occur already in the comparable scales [1]. Currently, urban systems happen to become main regulators for fluxes of many elements on a global scale due to ongoing industrial and economic development and a growing number of inhabitants. Already more than half of the world's population is living in urban systems. Additionally, urban systems are constantly expanding and cover more and more former natural and agricultural areas.

For natural history, urbanisation is a new phenomenon never existed in previous geological eras. From the view point of geosciences, urban areas correspond to sediment formations. Urban residues will be a specific of recently formed sediment rocks in the geologic future. The urban impact is also manifested in altered element fluxes on a global scale. The use of fossil fuels in urban systems is an important source of the greenhouse gas CO_2. Via atmosphere, water and human transport activities pollutants and products from urban systems are globally distributed. Especially industries, which are urban subsystems, create element association not existed in nature before, such as specific alloys or organic compounds. Because of the tremendous global impact of urban systems, a new geoscientific sphere is developing: the Astysphere [2]. This sphere comprises the parts

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of earth influenced by urban systems. Within this concept, urban systems are described to act as network influencing each other and altering together the earth's environment.

This concept is shortly described in the following, but because of the limited space, the aspects of astysphere can only be highlighted and not comprehensively explained.

The meanings of urban systems

Urban systems, which become manifested in towns, have manifold meanings for nature in general and for human beings in particular. However, the traditionally made difference between nature and human beings is an artificial one made by human minds but not by nature itself. From the viewpoint of evolution, urban systems are still part of nature and are finally still subject to natural forces. Nature is not only a primeval tropical forest, the deep sea or the remote tundra of uninhabited Polar Regions. Human beings are part of nature and therefore fields, pastures, canalized rivers, mining areas, urban areas and industries are part of nature as well. Even machines or the internet are part of the evolution of nature.

The meaning of urban systems for human beings is for example that these systems are sites of knowledge and education with the schools and universities. Urban systems are furthermore central places for culture including fine arts, architecture, sports or movie industry. They are also sites for recreation with parks, pubs, or the access to medical facilities. Political power is a further specific of urban systems. Some towns act as capitals, some are issues between nations as was Berlin and still is Nikosia or Lefkosia on Cyprus (fig. 1). Also financial and economical powers are characteristics of urban systems. Urban systems are locations of production, as for example Ludwigshafen in Germany with the world largest chemical plant of BASF. Consequently, human beings find work in urban systems and they reside in them.

From the view point of nature, humans are just one of many other species nowadays creating cities to live in. The development of cities (or urban systems) superimposes the former forms of nature in those particular regions, such as forests, river banks (fig. 2) or coastal environments as excellently described for New York [3]. For Geology, urban systems are deposits like sandstone or limestone but consisting of concrete, asphalt, steel and many more materials not produced by nature without human beings so far since the development of cities is based on an enormous material input to construct the buildings, machines and the infrastructure with streets or sewerage.

Fig. 1. UN protected border between Lefkosia and Nikosia on Cyprus.

Fig. 2. Development of the upper slopes of the Yangtze.

Geochemistry is the science on the distribution of chemical elements in the system earth. Cities are systems of element and energy fluxes. Industries are parts of urban systems. Here, many artificial chemical compounds are produced. It is estimated that world wide about 60,000 chemical species are technically produced by humans [4]. Furthermore, specific chemical elements are accumulated in urban systems due to construction and processing activities of humans. In consequence to global urbanization, cities are more and more important factors for the global distribution of elements. The global importance of urban systems is demonstrated by satellite night images (fig. 3).

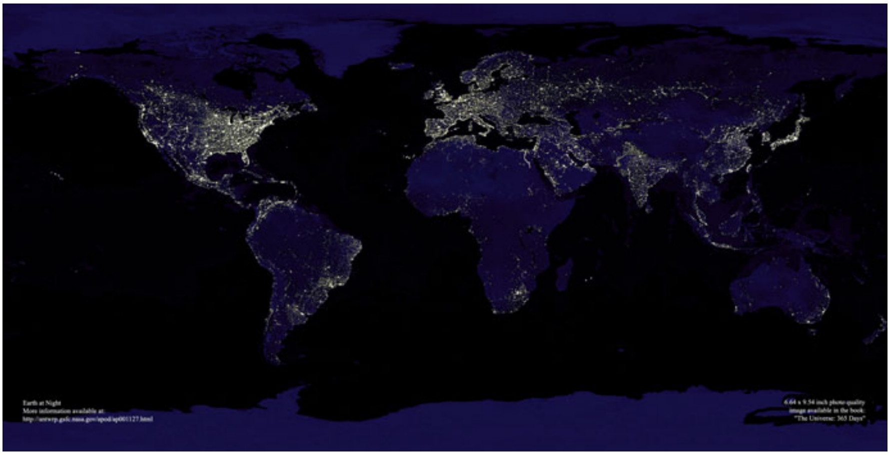

Fig. 3. Earth at night (image credit: NASA).

Cities form a global network of energy and substance fluxes linked via streets, air traffic, water ways, overseas shipment, pipelines and power cables. Furthermore, their emissions of chemical elements into the atmosphere or the rivers are distributed over the whole globe. Thus, cities envelope the globe like a net the football.

The development of urban systems

Urbanization is a global process that accelerated its velocity since the industrial revolution. Countries like Great Britain or Germany show today urbanization rates already above 80 %. Currently, the urbanization of developing countries India and China is increasing dramatically. More and more Megacities with a population above 10 million inhabitants emerge by this process and most of them are located in the developing world.

For 2008 it was estimated that about 50% of the world population live in urban areas and in 2050 around 70% of the world population will live in urban areas [5]. That means, till 2050 the number of urban population doubles and will be similar to today's world population. Concurrently, the urban area is constantly expanding. The global extension of urban areas is under discussion. In 2000, 0.3% of the total land area of countries was urbanized. It is expected that cities grow 2.5 times in area by 2030 or will cover 1.1% of the land surface area [6]. Other calculations [7] claim that already in 2000 up to 2.7% of the total land area was urbanized. The portion of areas developed for traffic and settlement purposes is higher in developed countries. In Germany, already 12.5% of the total area is used for traffic and settlements and every day more than 120 ha are added. Ongoing urbanization causes the sprawl of more or less dense urban systems over the globe [2].

The astysphere

Urban systems are globally cross linked to each other by fluxes of energy, information and matter. The various urban processes do not only affect urban systems within their specific borders but also remote areas by pollution via the air path or by mining activities and even tourists. Thus, a new sphere develops that changes the world's shape, the Astysphere. This Astysphere is a new approach to integrate urban systems into the geoscientific concept of spheres and an initial point for the understanding of urbanization of the earth as natural process [2].

Eduard Suess presented 1875 a concept of geoscientific spheres [8]. He distinguished spheres, such as the lithosphere and the atmosphere to promote a comprehensive understanding of the system earth. Since then, based on the works of geochemists like Clark [9], Goldschmidt [10] and Vernadsky [11], this idea became a dominating concept for the understanding of the distribution of chemical elements in the system earth. Later, due to the importance of human beings on global element fluxes, the terms technosphere and anthroposphere [4] were introduced (fig. 4). Nevertheless, in face of the ongoing urbanization of the earth, this concept is not any more precise enough to develop a comprehensive understanding of global element fluxes. Typical units of urban systems, such as buildings, industrial plants or automobiles show different chemical element ratios representative units of the primeval nature, such as sandstone or volcanic rock. Thus, it seems appropriate to classify the anthroposphere into an agriculturally and an urban dominated sphere. According to the greek terms agros (αγροζ) for agriculture and asty (αστυ[1]) for city or town in opposite to the surrounding farmland, the anthroposphere comprises the agrosphere [12] and the astysphere (fig. 4). The agrosphere represents the rural and agrarian environment, and the astysphere corresponds to the urban environment. Both spheres are intensively connected due to former and recent agricultural activities on urban land and vice versa. Figure 4 displays the concept of the astysphere within the earth's spherical system in a sketch. The astysphere comprises also parts

[1] The author thanks Prof. Dr. G. Kloss, Department of Classical Studies, Universität Heidelberg, Germany for the discussion on the correct Greek expression for urban systems with respect to the geoscientific concept of spheres.

of other spheres such as the atmosphere, hydrosphere and the pedosphere. Like the anthroposphere, the astysphere is always part of the biosphere. Within the astysphere, soils (pedosphere) are important sinks for elements and materials and show specific development properties.

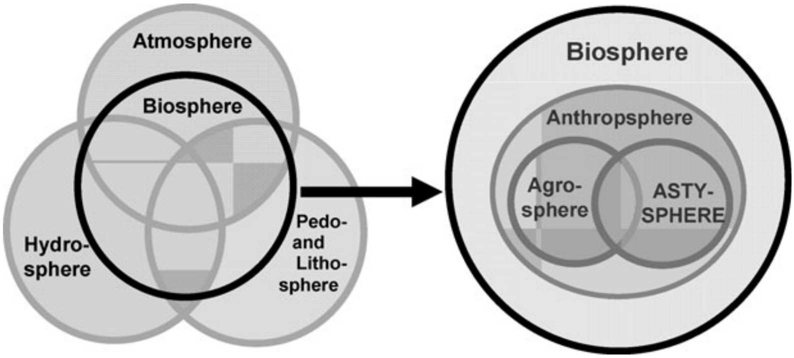

Fig. 4. The astysphere within the geoscientific concept of spheres.

One characteristic of the astysphere, with respect to the human view point is the environmental pollution caused by processes of urban systems. For geochemistry pollution is the enrichment of chemical elements harmful to human beings and other living organisms. Building density and urban structure in combination with emissions from various anthropogenic processes such as combustion, cause a specific urban atmosphere. Alteration of environmental compartments is not only documented in air but also in soil, groundwater, fresh and coastal waters and sediments Urban soils, for example, show specific artificial compositions [13] and are subjected to dramatic and fast transformation contrary to primeval soils [14]. Furthermore, urban soils are main sinks for atmospheric pollutants. These pollutants, such as heavy metals, deposit on soils, where they are relatively immobile. Thus, even land use patterns of urban systems can be identified by the spatial pattern of chemical elements in soils. Like a footprint, urban processes are archived in soil [15, 16]. Urban systems do not only alter the environments in which they are built in, but influence even remote areas via long-distance transport of atmospheric pollutants, overseas shipment or political decisions to establish mining pits or logging. In consequence, human activities in urban systems can be globally detected. Witnesses or urban impact on nature are e.g. the ozone hole over Arctic and Antarctic areas, global temperature increase due to CO_2 emission or lead pollution of Antarctic ice cores [17, 18].

The asytsphere and the agrosphere

The astysphere is part of the anthroposphere, which consists of urban and agricultural systems. Thus, anthropogenic activities can be distinguished belonging to the astysphere or to the agrosphere. This differentiation is not always trivial, since both systems can overlay each other. Agriculture can occur within urban areas and industrial products can be produced in the rural area. Urban systems influence chemical element fluxes of agricultural areas by the needs of urban systems, and their political and economic power determine agricultural processes. Furthermore, both are intensively linked via transport ways. Nevertheless, both spheres help to understand the interaction of input and output variables for both systems. Anthropogenic activities, such as construction, industrial production, housing, traffic and sewage and waste management, are characteristics of the astysphere, whereas forestry, cultivation of field crops, stock-farming and fishing are characteristics of the agrosphere. Often, both spheres compete for the same areas. In those situations, mostly the agrosphere loses against the astysphere. Towns frequently are expanded on valuable agricultural soils. Thus, it might be worthwhile to consider the establishment of protection zones for agricultural systems in cases when urban development becomes too dominant.

Conclusion

Global urbanization, urban growth, sprawl and aggregation cause the expansion of urban pollution plumes, the accumulation of chemical elements in ratios not occurring without human activities in nature or large scale deposits of technical materials. From a geoscientific point of view, urban systems are deposits of basic materials, potentially worth to be reused and conveyed as for example aluminium, asphalt or various heavy metals. The geochemical concept of spheres allocates the chemical elements to specific spheres such as lithosphere, atmosphere or biosphere. Since in its sphere of influence urban systems alter the abundances of chemical elements a new sphere, the astysphere evolved, that forms together with the agrosphere the anthroposphere. The future aim for geoscientific research in the field of the astysphere must be to develop models describing and prognosing the global process of urbanization and the development of the astysphere with respect to fluxes of materials, chemical elements, energy and information as well as to model the impact of urban systems on the forming of the earth's face.

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Traffic related metal distribution profiles and their impact on urban soils

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Abstract

The main sources of soil pollution in urban areas are traffic emissions. Its contribution to the global urban emissions of trace elements is increasing every year. The dispersion of those pollutants is affected by the climatic conditions, placing finally in other areas such as the surrounding soil and water. Trace elements could have different behavior regarding to their vertical and horizontal distribution pattern. Thus, the concern for the distribution profile of trace elements in the urban environment is rapidly growing. In this context, the present study was designed to investigate the horizontal and vertical distribution of traffic related metals in urban soils surrounding a road and to assess their level of contamination.

Introduction

Road traffic is one of the most important environmental problem in many cities and the main source of pollution of urban soils [1]. The contribution of road traffic to the global urban emissions of trace elements is increasing every year at the same time that the problem of contaminated soils is becoming of increasing concern for the environment. Trace elements could show a different vertical and horizontal distribution pattern depending on their own nature, their main form of diffusion or their primary source. Thus, the concern for the distribution profile of trace elements in the urban environment is rapidly growing [2, 3].

Soil is considered contaminated when chemicals are present or other alterations have been made to its natural environment. In this sense, traffic emissions involve an anthropogenic input of heavy metals to surrounding soils, altering their natural composition. There are different methods to

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estimate the pollution level of an altered soil. The geoaccumulation index, I_{geo}, is a simple method for assessing soil quality and provide a simple way of comparing the extent of metal pollution of the urban soils. It has been used since the late 1960s, and has been widely employed in European trace metal studies. Originally used for bottom sediments [4], it has been successfully applied to the measurement of soil contamination [5]. The I_{geo} enables the assessment of contamination by comparing current and background concentrations, although it is not always easy to reach background soil layers. It is calculated using the following Eq. (1):

$$I_{geo} = \log_2 \frac{C_n}{1.5 \cdot B_n} \tag{1}$$

where, C_n is the measured concentration of the element in soil and B_n is the geochemical background value. The constant 1.5 allows us to analyze natural fluctuations in the content of a given substance in the environment and to detect very small anthropogenic influences. According to the geoaccumulation index, soils can be classified as non-polluted ($I_{geo} < 1$), very slightly polluted ($1 < I_{geo} < 2$), slightly polluted ($2 < I_{geo} < 3$), moderately polluted ($3 < I_{geo} < 4$), highly polluted ($4 < I_{geo} < 5$) and very highly polluted ($I_{geo} > 5$).

However, metal background values are not enough for establish soil deterioration state and other methods to assess the risk for the ecosystems are used as reference values within environmental protection policy framework. This is the case of the Basque Indicative Values for Assessment (VIEs) [6], which are scientifically based generic assessment criteria to help evaluate long-term risks to human health and to the ecosystem from chemical contamination in soils from the Basque Country (North of Spain).

The present study was designed to investigate the horizontal and vertical distribution of traffic related metals in urban soils surrounding a road and to assess their level of contamination.

Experimental Method

All plastic and glassware material in contact with samples or ICP-MS solutions were soaked in a 10% HNO_3 bath at least 24 h, then rinsed twice with Elix (Millipore, USA) quality water and finally rinsed with Milli-Q water (18.2 MΩ cm, Millipore, USA). After drying the material in a laminar air flow hood inside a class 100 clean room, it was stored in clean plastic bags until use.

Table 1. ICP-MS experimental conditions.

RF power	*1300 W*
Plasma gas flow rate	15 L/min
Auxiliary gas flow rate	1 L/min
Nebulizer gas flow rate	0.9 L/min
Sample uptake rate	1 mL/min
Data acquisition	Peak Hopping
Dwell time	100 ms
Sweeps per reading	8
Readings per replicate	1
Replicates	3

Soil sample transects of 15-20 cm in depth were collected in an urban area of Bilbao (North of Spain) with elevated traffic density (28.200 vehicles per day from the year 2004 until now [7]) and without the presence of any channel and thus, runoff waters from the road arrive directly to the soil. In order to study the horizontal distribution of metals, soil samples were taken at different distances from the road: at 0 meters (S0m), just immediately after the road and at 1 and 3 meters of distance (S1m and S3m). Samples were mainly clay soils, as this is the typical soil composition of the Bilbao metropolitan area. Samples were divided in subsamples of 2-3 cm of depth (S1m A - S1m H and S3m A - S3m F) and then air dried in a fume hood during 24 h, ground in a planetary ball mill Pulverisette 6 (Fritsch, Germany) and sieved to particle size under 250 μm for their homogenization. The acid digestion of the soil samples was carried out in a microwave digestion system Multiwave 3000 (Anton Paar, Graz, Austria) provided with a 8XF-100 digestion rotor and 100 mL fluorocarbon polymer (PTFE) vessels following the EPA 3051A method [8]. Nitric acid (69%) and hydrochloric acid (36%) were Tracepur grade and supplied by Merck (Darmstadt, Germany).

Elemental analysis was achieved using a Perkin Elmer SCIEX 9000 ICP-MS (Toronto, Canada) in a class 100 clean room. The argon used for the plasma was supplied by Praxair (99.995%, Madrid, Spain). The operating conditions for sample introduction and data acquisition are shown in Table 1. Sample solutions were diluted to 1% HNO_3 concentration prior to analysis. 46 metals, including alkaline, heavy metals and rare earth elements (REE) were determined in the samples measuring the following isotopes: Li^{7}, Na^{23}, Mg^{24}, Al^{27}, K^{39}, Ca^{43}, Ti^{47}, V^{51}, Cr^{53}, Mn^{55}, Fe^{57}, Co^{59}, Ni^{60}, Cu^{63}, Zn^{68}, As^{75}, Se^{82}, Sr^{88}, Nb^{93}, Mo^{98}, Ag^{107}, Cd^{114}, Sn^{120}, Sb^{123}, Ba^{138}, W^{184}, Hg^{202}, Tl^{205}, Y^{89}, Pb^{208}, La^{139}, Ce^{140}, Pr^{141}, Nd^{142}, Sm^{152}, Eu^{153}, Gd^{158}, Tb^{159}, Dy^{164}, Ho^{165}, Er^{166}, Tm^{169}, Yb^{174}, Lu^{175}, Th^{232},

U^{238}. All solutions were prepared using Milli-Q water. ICP-MS standard solutions were prepared from Alfa Aesar (Specpure®, Plasma standard solution, Germany) stock solutions. Sc, Ge, In, Re and Bi were used as internal standards.

The performance of an ICP-MS instrument strongly depends on the operating conditions. The plasma operating conditions such as the nebulizer flow rate, the position of the torch and the ion lens voltages of the instrument were optimized prior to any experiment with a 10 ng mL^{-1} standard solution of Mg, Rh, In, Ba, Pb and U. The nebulizer gas flow rate was optimized to obtain a good compromise between high sensitivity and low oxide levels (lower than 3% for CeO/Ce). For quality assurance purposes soil certified reference materials (SRM 2711, total content; and BCR 142R, aqua regia soluble content) and a freshwater containing trace elements (SRM 1640) were routinely analyzed in each sample batch.

Results and discussion

After digestion and analysis of the soil samples, high concentrations of heavy metals were found in upper soils close to the road (S0m and S1m) compared with those values obtained in deeper soils and at 3 meter of distance to the road (S3m) (Fig. 1). Furthermore, concentrations show a severe decreasing profile with depth and distance to the road, reaching a constant value in the deepest soils (15-20 cm). The fact that element concentrations in depth reach a plateau and that it was similar to the concentration of soils at 3 meters of distance, suggests that these could be the natural values of the elements in those soils. Besides, these values are in agreement with those given by IHOBE, the Basque Environmental Public Corporation, for some metals as background levels in Basque Country (North of Spain) soils (Table 2). So there is a clear input of these elements from the road traffic to the urban soils. The traffic affection on the soils is more important close to the road. Metal concentrations in soils at 3 meters of distance to the road were similar to background values, except in the upper 2 cm (S3m A), where values were slightly higher. Fig. 2 is a 3D representation of the concentration of heavy metals, showing above mentioned decreasing behaviour with depth and distance.

It is possible to see 3 different types of metal profiles in the urban soils. Some heavy metals show a steep decreasing profile between upper and deeper soils (Fig. 1a). That is the case of Ti, Cu, Zn, Mo, Cd, Sn, Sb, W, Hg, Nb and Pb, whereas others like Na, Mg, Ca, V, Cr, Mn, Co, Ni, Sr,

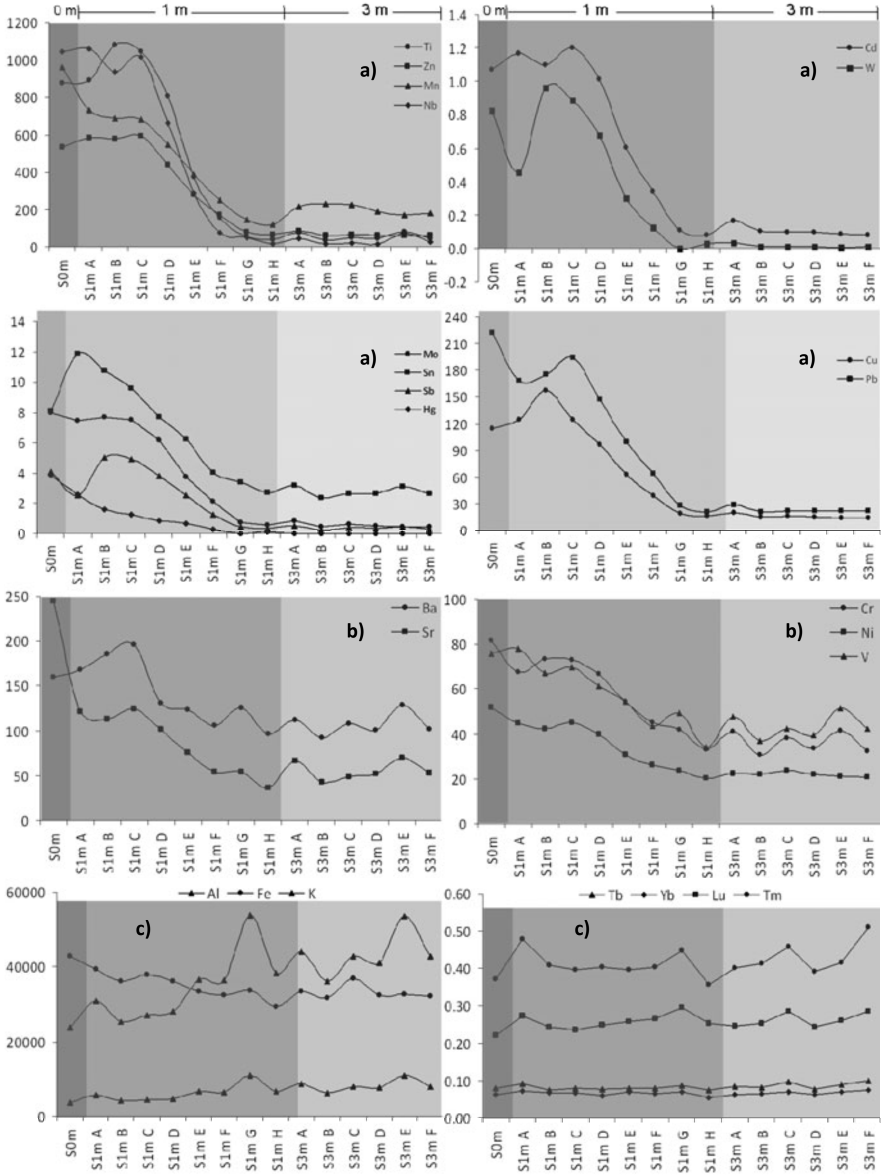

Fig. 1. Metal concentration profile in depth at different distances from the road (mg Kg^{-1}): a) high decreasing, b) slightly decreasing, and c) constant.

Ag or Ba have a less sharper profile (Fig. 1b). All of these elements have a traffic related emission source [1, 9, 10] and most of these elements were already related to traffic emissions in our previous work [11] but in this study other elements such as Na, Mg, Ca, Ti, V, Mn, Co, Ag, Ba, W and Hg also appear. The last group of elements is formed by Li, K, Al, Fe, As, Tl, and REE, and their concentration in soil maintains nearly constant, without a significant trend (Fig. 1c).

Table 2. Background levels of some metals (mg Kg^{-1}) in soils from the Basque Country.

	Cd	Cr	Cu	Ni	Pb	Zn
This study	0.1	38	15	21	22	60
IHOBE [6]	0.2	30	14	19	22	65

The geoaccumulation indexes calculated for the soils are summarized in Fig. 3. The background values used have been obtained from deeper soils, since metal concentrations reach a plateau and these values agree with those given by IHOBE [6] for some metals listed in Table 2 as have been previously stated. As can be seen in Fig. 3a, geoaccumulation index in the upper soils close to the road are higher than 1 for most of the analyzed elements (Ca, Ti, Mn, Cu, Zn, Sr, Mo, Ag, Cd, Sn, Sb, W, Nb, Hg and Pb) whereas at 3 m of distance all of them are below 1, which shows that soils are polluted in those heavy metals. Furthermore, geoaccumulation indexes for Ca, Ti, Mo, Cd, Sb, W, Hg and Nb point out that soils are highly or very highly polluted. It must be noted that some extremely toxic elements such as Cd, Sb and Hg are among those and their geoaccumulation indexes are still higher than 1 in deeper soils (Fig. 3b). It is shown again that significantly affected soils are those close to the road, whereas soils far away from the road cannot be classified as polluted soils based on their geoaccumulation index.

The Basque Indicative Values for Assessment (VIE) establish the risk acceptance limits for the different uses of a soil. There are 3 levels of acceptance. VIE-A is a reference level and below it, it is possible to state that the soil is not affected and there is not risk for the human health. VIE-B marks the lower limit of acceptability of the risk. Values lower than this but higher than VIE-A involve acceptable risk, whereas values higher than VIE-B could give place to unacceptable risk and a more detailed study of the zone must be carried out. VIE-C represents the upper limit of acceptability of the risk and when exceeded, implies a serious risk to the ecosystem and measures to eliminate the risk are required. Table 3 shows these Indicative Values for Assessment calculated by IHOBE [6] according to the use of the soil.

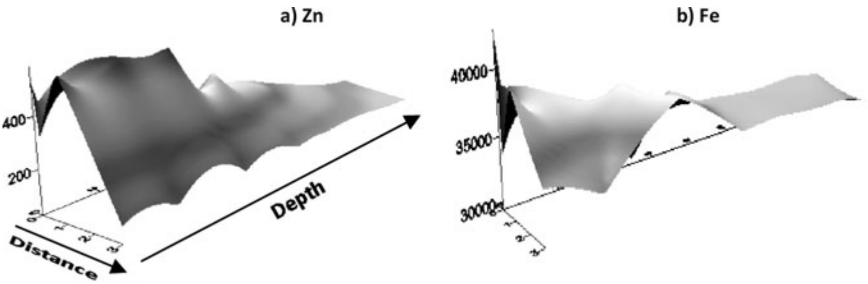

Fig. 2. 3D representation of heavy metal concentrations (mg Kg^{-1}) in urban soil surrounding the road. Zn and Fe have been chosen as representative of the two different behaviours: a) traffic related metals accumulate in top soils close to the road, b) rare earth elements and other elements not affected by the traffic have a uniform distribution.

Table 3. Indicative Values for Assessment (VIE) calculated by IHOBE (mg Kg^{-1}).

Metals	Human Health Protection					Ecosystem Protection	
	(a)	(b)	(c)	(d)	(e)	VIE-B	VIE-C
As	30	30	30	30	200	23	35
Cd	5	5	8	25	50	0,8	18
Cu	(*)	(*)	(*)	(*)	(*)	24	250
Hg	4	4	4	15	40	0.3	3
Ni	110	110	150	500	800	40	280
Pb	120	120	150	450	1000	44	330
Co						20	30
Mo	75	75	75	250	750	1	620
Zn	(*)	(*)	(*)	(*)	(*)	106	840

(*) Obtained value is around some tens of g Kg^{-1}
(a) Children's play area
(b) Residential with garden
(c) Residential
(d) Park
(e) Industrial/Comercial

As can be seen in Fig. 4, concentrations of Mo, Cd, Ni, Cu and Pb are higher than the VIE-B values, whereas Hg and Zn exceed the VIE-C values in surface soils at 0 and 1 meter of distance.

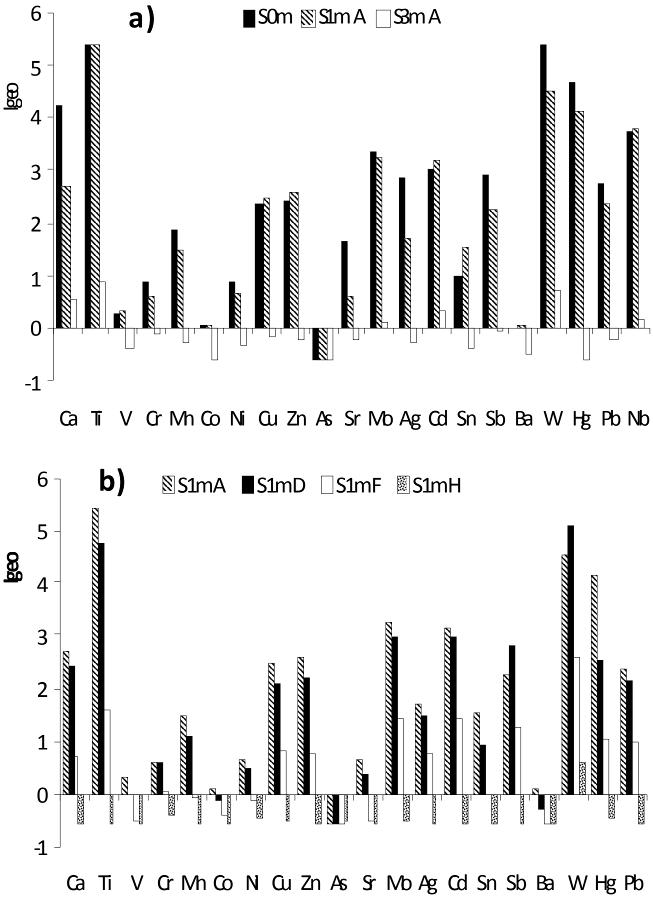

Fig. 3. Geoaccumulation indexes representation: a) at different distances to the road; b) at different depths.

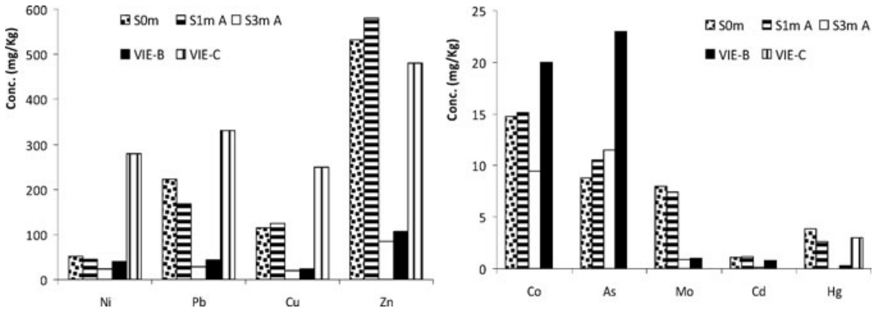

Fig. 4. Concentration of heavy metals and their regulated values for the Basque Country, given by IHOBE (Basque Government Public Corporation). VIE-B and VIE-C are, respectively, the lower and upper limit of acceptability of the risk.

Conclusions

Heavy metals accumulate in upper soils close to the road and show a high decreasing profile with depth and distance to the road. Most of the elements have a decreasing pattern with the depth in the first centimetres in soils at 1 meter of distance to the road and then reach background levels. However, heavy metal concentrations in soils at 3 meters are similar to background levels and show a slightly decreasing profile in the first centimetres. This data suggest an external input of those metals coming mainly from the runoff waters of the road and atmospheric deposition in a lesser extent: Na, Mg, Ca, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, Sr, Mo, Ag, Cd, Sn, Sb, Ba, W, Hg, Nb and Pb. On the other hand, rare earth elements, alkaline metals and major soil components such as Fe or Al did not show this trend.

Geoaccumulation Indexes, as a tool for risk assessment of the soils, are higher than 1 for most of the elements and in many cases higher than 3 in soils at 0 and 1 meter of distance. Therefore, soil samples close to the road are highly polluted by traffic related metals. Geoaccumulation Indexes for Ca, Ti, Mo, Cd, Sb, W, Hg and Nb point out that soils are highly or very highly polluted. It is worth to mention that elements with elevated toxicity such as Cd, Sb or Hg are among the most polluted ones. Pollution of the soil decreases rapidly with distance to the road. Soils at 3 meters of distance have geoaccumulation indexes below 1 which classify these soils as non-polluted.

According to the indicative values that IHOBE, the Basque Environmental Public Corporation, sets up for some toxic heavy metals, concentration of Mo, Cd, Ni, Cu and Pb are higher than the VIE-B (the lower limit of acceptability of the risk), whereas Hg and Zn exceed the VIE-C value (the upper limit of acceptability of the risk) in top soils at 0 and 1 meter of distance to the road. Therefore, gardeners and other municipal maintenance workers should manage these soils as a toxic residue and should take the proper security measurements.

Acknowledgments

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Assessment of total petrol and polycylic aromatic hydrocarbon mobility in soils using leaching tests

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Abstract

The risk assessment of total petrol hydrocarbons (TPH) and polycyclic aromatic hydrocarbons (PAH), two of the most frequent soil and water contaminants, has to include their exposure pathways, especially the soil-groundwater pathway. Threshold values have to be aquired for the amendment to the German Federal Soil Protection Contaminated Sites Ordinance. For this purpose, a batch experiment with a liquid to solid ratio (L/S) of 2 L/kg was developed. The aim of this work was to verify the robustness of this leaching procedure for TPH and PAH in different soil types against varying test conditions. Also, stir bar sorptive extraction (SBSE) was tested as alternative sample preparation of eluates for PAH analysis.

Introduction

Total petrol hydrocarbons (TPH) are a complex mixture of crude oil distillation products mostly consisting of aliphatic hydrocarbons with carbon chain lengths of 10 to 40 (C_{10}-C_{40}). Polycyclic aromatic hydrocarbons (PAH) are byproducts of incomplete combustion of organic material and comprise of fused aromatic rings, usually two to seven. Both classes of substances represent severe environmental pollutants due to their harmful and partly carcinogenic properties and arise from numerous natural and anthropogenic sources. To assess their actual risk potential, their exposure pathways have to be taken into account. Especially the soil-groundwater pathway is of importance, since it affects several compartments of protection (soil, air, water).

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Reasonable thresholds values, based on validated leaching procedures, have to be established in the course of the amendment to the German Federal soil protection contaminated site ordinance (Bundesbodenschutzverordnung, BBSchV [1]). Thus, a batch experiment with a liquid to solid ratio (L/S) of 2 L/kg was developed for organic compounds and a draft standard compiled [2]. Several facts might affect the release of contaminants and thus the test results. Besides the sample properties itself like amount and composition of contaminants, grain size distribution and organic matter content, the test conditions (and their possible effects) are to be considered:

- tumbling duration and frequency (alteration of particle size due to friction)
- centrifugation acceleration (turbidity and pollutant concentration in the eluate)
- sample pretreatment (loss of analyte due to liquid-liquid-separation, sorption on equipment surfaces, clean-up and/or drying)

Objective of this work was to verify the robustness of this batch test procedure for the determination of TPH and PAH applying different test conditions and soil types. The liquid-liquid separation step (LL) is known as one of the critical sample preparation steps of eluates prior to PAH analysis since it may causes loss of analyte. An alternative sample pretreatment for PAH analysis was tested applying stir bar sorptive extraction (SBSE). This method is well described for PAH trace analysis in water [3, 4] but few is known about its suitability for aqueous eluates of contaminated materials involving higher concentrations of PAH and complex matrices. The PAH results obtained with SBSE have been compared to those of the liquid-liquid extraction (LL).

Materials and methods

Selected properties of the soils used for the preparation of test materials are given in table 1. The soils were mixed with an appropriate amount of high contaminated material obtained from a former tank maintenance installation (TPH) and a former railway sleeper preservation facility (PAH) respectively. The test materials' solid matter contents of pollutants are given in table 2. PAH amounts are stated as sum parameter of the 16 US EPA PAH. The PAH source material (S-5) also contained considerable amounts of TPH and was used for additional leaching experiments.

Table 1. Selected properties of applied soils (S).

Soil	S-1	S-2	S-3	S-4	S-5
pH	5.41	6.07	6.00	8.66	8.22
C_{org} [%]	2.84	3.44	0.96	0.64	-
Particle size distribution [%]					
> 2 mm	-	-	-	7	-
2-0.063 mm	78	29	67	92	99
0.063-0.002 mm	14	43	29	1	1
< 0.002 mm	8	28	4	-	-

Table 2. Solid matter contents of contaminated soils (CS).

Contaminated soil	CS-A	CS-B	CS-C	CS-D	CS-E	CS-F	CS-G
Applied soil (see table 1)	S-1	S-1	S-5	S-1	S-2	S-3	S-4
TPH C_{10}-C_{40} [mg/kg]	1123	337	707	-	-	-	-
TPH C_{10}-C_{22} [mg/kg]	211	65	281	-	-	-	-
PAH [mg/kg]	-	-	1885	6.21	8.21	7.69	9.51

The batch tests for both TPH and PAH investigations were performed according to E DIN 19527 [2]. 250 g of the test material was placed in a glass bottle, covered with 500 ml demineralized water and the capped bottle was agitated in an end-over-end tumbler for 2, 5 or 24 h at 3, 7 or 15 rpm, respectively. After tumbling, the suspended solids were allowed to settle for 15±5 min, followed by decantation and centrifugation using stainless steel beakers (5 h at 2,000 g or 30 min at 20,000 g). The eluate was filtered through a glass fibre micro filter using a pressure filtration device.

Samples for TPH analysis were prepared following DIN EN ISO 9377-2 [5]. The eluate was extracted twice, each with 25 ml *n*-hexane and 30 min agitation in an horizontal tumbler. After liquid-liquid separation and drying over 5 g Na_2SO_4, the extract was cleaned by filtering through 3 g Fluorisil®, concentrated and analyzed per GC-FID. Analysis was performed on a Shimadzu GC-2010 Plus with on-column injection, equipped with a Zebron ZB-5-HT column (15 m, 0.32 mm ID, 0.25 µm film thickness).

Samples for PAH analysis were prepared following DIN 38414-23 [6]. The eluate was extracted three times, each with 20 ml *n*-hexane and 1 h agitation in an horizontal tumbler. After liquid-liquid separation the extract was dried over Na_2SO_4, the actual amount of desiccant depends on the moisture content of the extract. Subsequent to concentrating and dissolving in acetonitrile, the extract was analyzed per HPLC-FLD.

The alternative method for PAH sample preparation including SBSE was conducted according to the procedure described by García-Falcón *et al.* [3]. Analysis was performed on an Agilent 1200 Series HPLC system with a Zorbar Eclipse PAH column (4.6x100 mm, 1.8 µm) using acetonitrile/water as eluent.

Results and discussion

To study the effect of tumbling frequency and duration on the release of TPH from contaminated soils, batch experiments with two test materials (CS-A and CS-B) were performed. 3, 7, 15 rpm and 2, 5, 24 h were chosen as test conditions, broaden the specifications given by the draft standard, which are 5-10 rpm and 24±0.5 h [2]. The TPH concentrations in the respective eluates are given in Figs. 1 and 2.

For CS-A, the results show generally lower TPH concentrations with higher tumbling frequency, apart from the 24 h experiment, which releases more TPH at 7 rpm than at 3 rpm (Fig. 1). This might be due to sorption on container surfaces which presumably increase with higher frequency and agitation time. For the test duration, no clear influence on the TPH release can be observed. This is probably because of the complex matrix of the soil which releases much fine particles to the eluates, thus hampering the sample preparation, especially the liquid-liquid separation step as well as the clean-up.

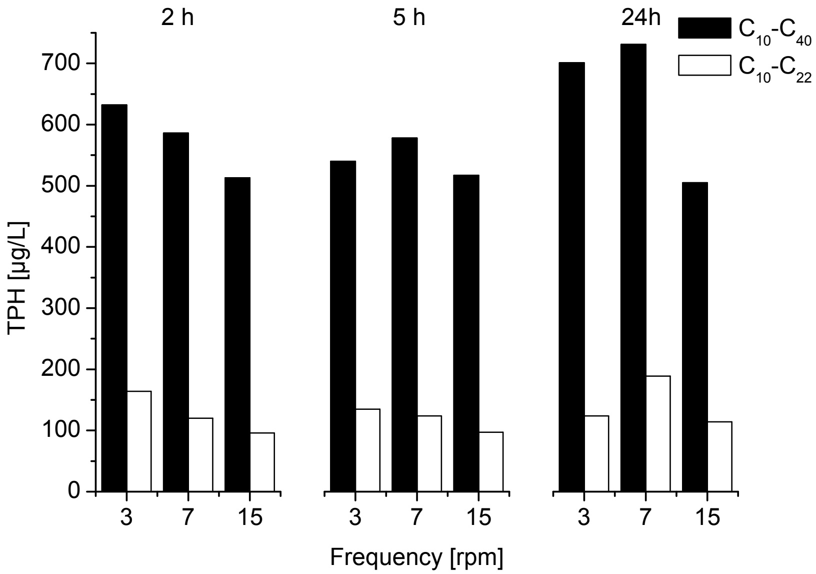

Fig. 1. TPH concentrations in CS-A eluates.

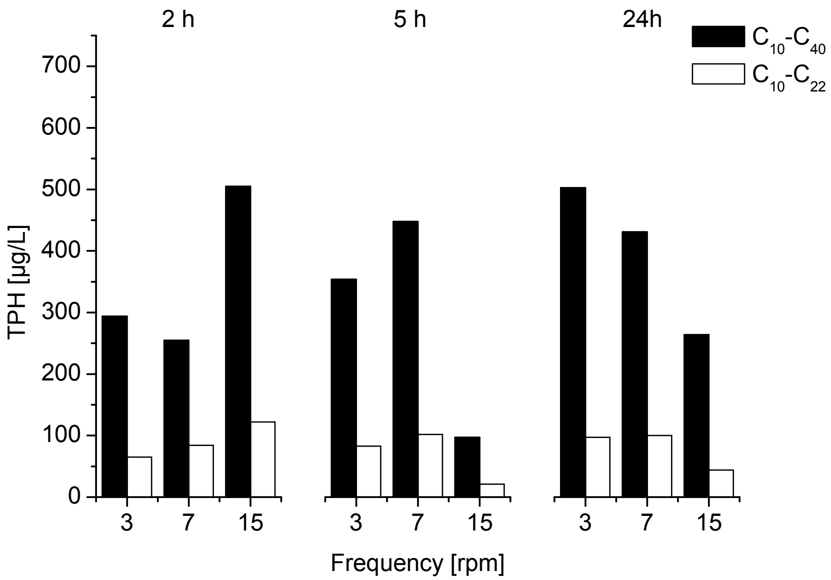

Fig. 2. TPH concentrations in CS-B eluates.

For CS-B no clear trends are observable concerning a possible influence of tumbling time and frequency. Just like with CS-A, this may be caused by the complex soil matrix which hampers the analysis. Furthermore, the low solid matter content of TPH (see Table 2) may inhibit a clear trend.

The test material CS-C was used for batch tests at 7 rpm, varying the tumbling time from 2, 5 to 24 h (Fig. 3). The results show a significant decline of leached TPH over time. This is possibly due to adsorption of TPH on container surfaces. Although the solid matter contents of the tested soils are in the same order of magnitude (see Table 2), CS-C releases up to five times more TPH than CS-A or CS-B which might be due to different soil matrices (see Table 1). In case of CS-A and CS-B the TPH distribution of the eluates is with about 20% of the C_{10}-C_{22} fraction almost identical to that in the solid matter (see Figs. 1, 2 and Table 2), whereas for CS-C this ratio is 40% in solid matter (Table 2) and around 60% in the eluates (Fig. 3). This corresponds to the higher water solubility of the shorter-chained TPH. The effect might not be observable for CS-A and CS-B due to their more complex soil matrix.

The four PAH contaminated test materials (CS-D to CS-G) were tested with various tumbling frequencies (3, 7 or 15 rpm) and centrifugation acclerations (2,000 g or 20,000 g), tumbling time was 24 h constantly. The actual test conditions are specified in the respective figures (Figs. 4-7).

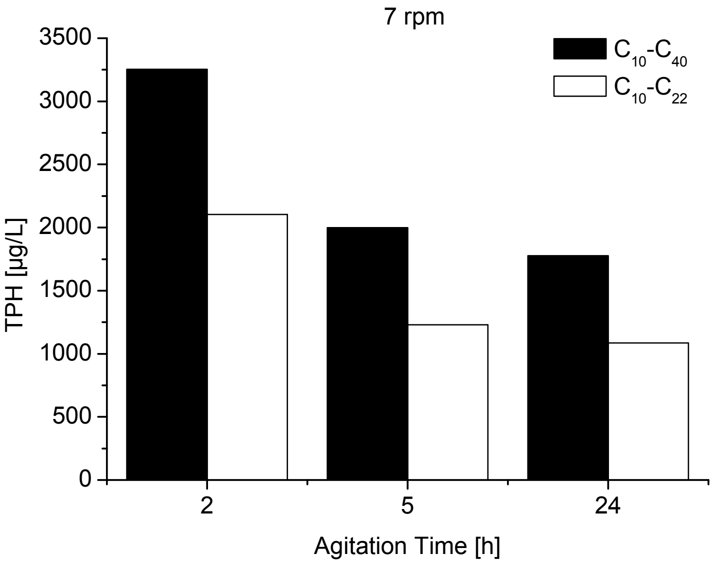

Fig. 3. TPH concentrations in CS-C eluates.

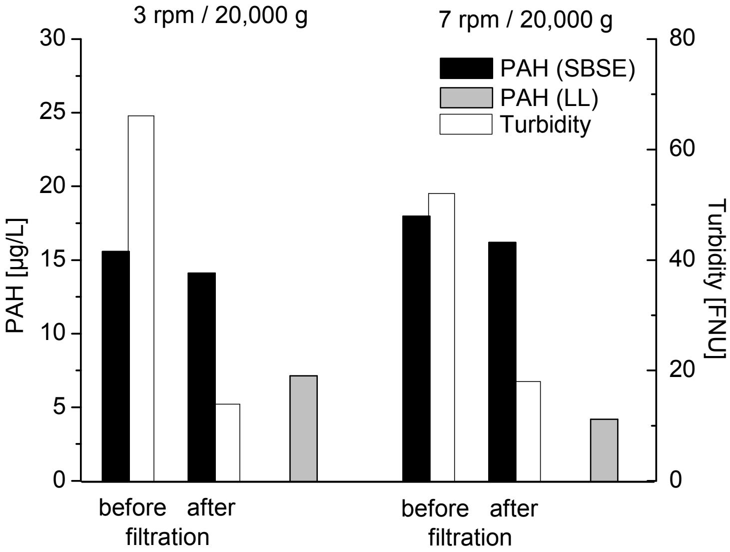

Fig. 4. PAH concentrations and turbidity of CS-D eluates.

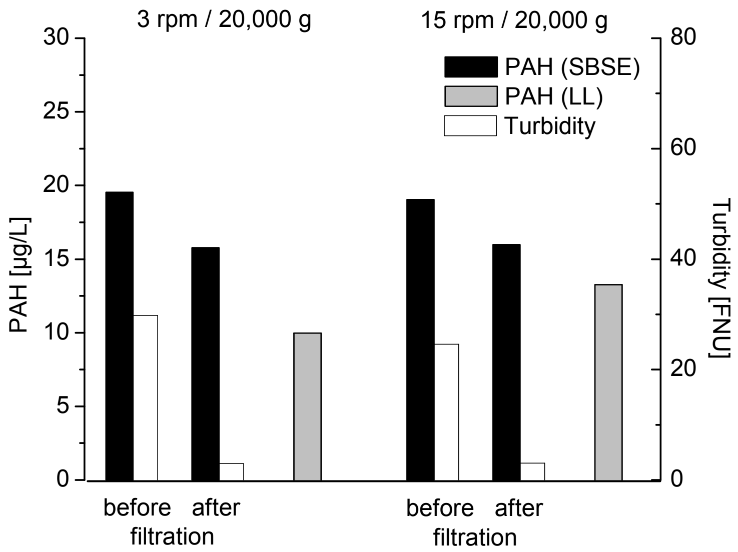

Fig. 5. PAH concentrations and turbidity of CS-E eluates.

The PAH concentrations in the obtained eluates, determined with liquid-liquid sample preparation (LL) and with stir bar sorptive extraction (SBSE) are stated. In case of SBSE, the PAH content was analyzed before and after filtration, hence the turbidity is also given.

The results show that with SBSE generally higher PAH amounts are detected than with LL - with the exception of CS-G, where the LL values are slightly higher (Fig. 7). Wide differences in SBSE and LL values correspond to relatively high turbidity (Figs. 4 and 6), whereas lower differences (Figs. 5 and 7) come along with low (Fig. 5) or very low turbidity (Fig. 7).

High turbidity indicates much particles in the eluate which may complicate the LL sample preparation through hampering the liquid-liquid separation and affording more desiccant thus possibly leading to loss of analyte. This matrix effect of the soil may explain the observed differences in SBSE- and LL-derived PAH values.

The remarkably high PAH amounts found in CS-G eluates, which are up to eight times higher than those of the other investigated soils although the solid matter content is similar in each case, may also be due to matrix effects, especially the grain size distribution. PAH often adsorb to fine particles [7], hence the more fine particles are present, the less PAH might pass into the aqueous phase.

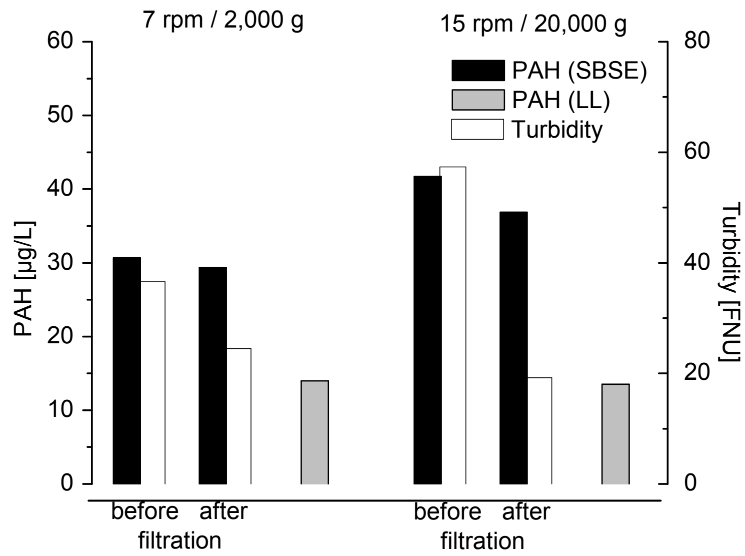

Fig. 6. PAH concentrations and turbidity of CS-F eluates.

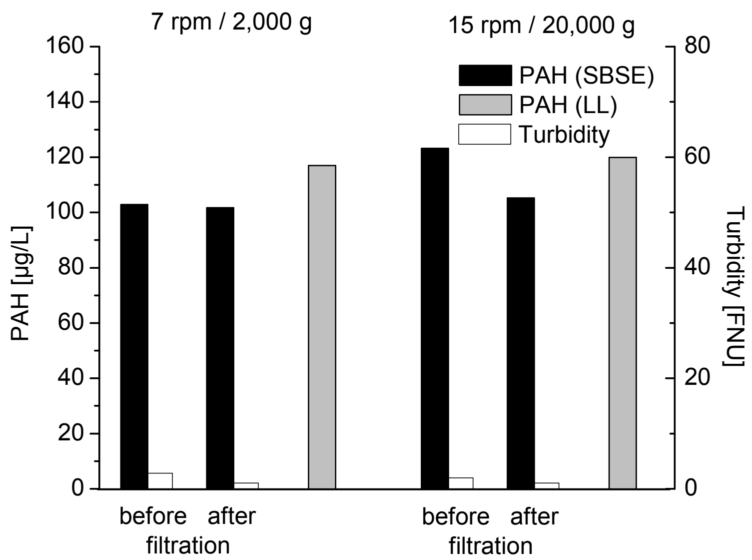

Fig. 7. PAH concentrations and turbidity of CS-G eluates.

Comparison of SBSE derived values before and after filtration shows always less PAH after it, independent of the actual batch test conditions. This accords to the assumption that together with fine particles (colloids) some PAH have been removed from the eluate as well. This effect is more distinctive with higher centrifugation acceleration (Figs. 6 and 7).

Concerning the effect of tumbling frequency and centrifugation acceleration on the amount of released PAH, there is no definite pattern observable. However, with SBSE, slightly higher PAH amounts were found at higher frequency and accelerations (Figs. 4, 6 and 7).

Conclusions

Generally, less TPH have been detected with increasing test duration and tumbling frequency, presumably due to increasing sorption of TPH on equipment surfaces. Complex soil matrices seem to influence the amount of detected TPH not just through affecting TPH release but through hampering the analysis.

The soil matrix seems to have a major effect on the elution of PAH. Soils that generate low or virtually no turbidity seem to release significantly more PAH.

Stir bar sorptive extraction (SBSE) might be a suitable alternative to liquid-liquid sample preparation (LL) in case of PAH analysis. Since LL-derived values were all the lesser compared to SBSE the higher the turbidity of the eluate was, SBSE seems to be less susceptible to complex matrices than LL.

To estimate the effect of test conditions like tumbling frequency and centrifugation acceleration on PAH elution comprehensively, further experiments are required.

Acknowledgements

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Soil-plant relations in an urban environment polluted with heavy metals

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Abstract

In the northwestern part of Romania there is one of the most polluted cities in the country, Baia Mare. Non-ferrous ore exploitation, it's processing in smelting and flotation plants are all sources of environmental pollution by sulfur oxides and heavy metals. Research has revealed that the content of heavy metals in the upper horizon of soils from small vegetable gardens of inhabitants exceed the maximum allowable limits up to: 3.6 (Cd), 7.95 (Cu), 46.3 (Pb) and 11.5 (Zn) times. Also, in the edible part of vegetables grown in gardens of peripheral areas denizens were determined heavy metals contents up to 2.94 mg·kg^{-1} Cd, 71 mg·kg^{-1} Cu, 600 mg·kg^{-1} Pb and 356 mg·kg^{-1} Zn. The mobile heavy metals content of soil and horticultural plants recorded direct proportionality relationships, statistically assured, correlation coefficients having values between 0,413 and 0,829.

Introduction

In the northwestern part of the country is one of the most polluted cities in Romania (Fig. 1), Baia Mare, with a population of 148,263 inhabitants and an area of 35.7 km^2. In the early '90s, American Blacksmith Institute, a rating agency for environmental situation assessment, worldwide, included Baia Mare in the list of most polluted places in the world. At that time, the average expectance life of the inhabitants of this town was ten years lower than in the rest of the country. Major pollution source is the emissions from two smelters of non-ferrous concentrates, two ores flotation (concentration) units, and even ore extraction from mines located around

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the city. In addition, many waste dumps generated from ore mining and their concentration in flotation units and ponds have a significant pollutant impact on the environment. Over all this, adds pollution caused by emissions from fuel burning in vehicles motors.

Heavy metals, especially Cd, Cu, Pb and Zn from poly-metallic sulfides and sulfur oxides formed in sulphides combustion process are the main pollutants of the area. Acid precipitation formed in the atmosphere and returned to land area, contributed to increased soil acidity and the mobilization of native soil chemical elements, including heavy metals and finally their uptake in plants, including those for animal and human nutrition.

This paper highlights the abundance of heavy metals in urban soils used for vegetable growing, located in Baia Mare city, both in total forms, but also mobile forms from soil solution, or in forms bounded to soil components and the accumulation of these chemicals in the edible parts of some vegetables.

Materials and methods

From urban soils, planted with vegetables, located at distances up to 8 km to the west and east and up to 5 km to south from emission sources of pollutants 56 soil samples from the upper horizon (0-20 cm) and 89 samples of vegetables (edible part of lettuce, orach, carrots, radishes) were collected.

Soil samples were analyzed in terms of content of total and mobile forms of heavy metals (Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn) [6]. Also were determined fractions of heavy metals associated with soil components, namely, soil solution content, contents of heavy metals bounded by exchangeable fraction, organic matter, free oxides and hydroxides and fraction bounded in the crystalline lattice of clayed and heavy minerals from soil [7].

The total content of heavy metals (Cd, Cu, Pb and Zn) was measured with flame atomic absorption spectrometer in hydrochloric solution resulted by digestion of soil samples in $HClO_4$-HNO_3 mixture. The heavy metals content in edible parts of vegetables (lettuce, orach, carrots and radish) carried out in hydrochloric solution resulted by plant ash solubilization.

Analytical data were statistical processed, computing the values of the grouping centre (arithmetic mean, geometric mean, median) and the spreading parameters (minimum, maximum, standard deviation and variation coefficient). Also, correlative relationships between some of the chemical elements from plants and soils were established.

Results and discussion

The total content of heavy metals in the upper horizon (0-20cm) of urban land planted with vegetables

Analytical data presented in Table 1 reveals wide range of heavy metals content between the minimum and maximum values. Also, the averages (X, Xg and Me) show close values between these parameters of the grouping center, but the general trend is that the arithmetic mean values (X) to be larger than the other two parameters (Xg and Me).

In any case, it appears that, compared to the maximum allowable limits, the average content of heavy metals, held by the arithmetic mean value is higher: by 1.5 times for Cd, 3.2 times for Cu, 8.9 times for Pb and 2.6 times for Zn. Other heavy metals determined (Co, Cr, Mn, Ni) had lower mean content values compared with MAL.

Table 1. Statistical parameters of total heavy metals content in the upper horizon (0-20cm) of urban land planted with vegetables in Baia Mare municipality, compared with normal content (NC[b]), the maximum allowed limits (MAL[c]) and the alert threshold for a sensitive use (AT[d]).

Parameter[a]	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
				$mg \cdot kg^{-1}$				
Xmin	0.7	14	9	29	507	18	195	282
Xmax	10.7	97	46	795	914	54	4632	3464
X	4.5	32	29	321	732	27	989	787
σ	2.8	18	8	172	209	14	643	613
Xg	4.3	30	27	305	730	25	872	771
Me	4.2	28	24	314	714	24	854	783
NC[b]	0.3	5	30	20	500	20	20	100
MAL[c]	.0	50 3	100	100	–	50	100	300
AT[d]	3.0	30	100	250	1500	75	50	300

[a]Xmin – minimum value; Xmax – maximum value; X – arithmetic mean; σ - standard deviation; Xg – geometric mean; Me – mediane

[b]after Fiedler and Röstler [3]; [c]after Kloke [5]; [d]after Romanian order of Environment Ministry (no 756/1997) for soil pollution [14]

Therefore, polluting chemical elements present in urban soils of Baia Mare, soils planted with vegetables, are: Cd, Cu, Pb and Zn. In fact, these are the main chemical elements generated by the exploitation and processing of ore (galena, blende, pyrite, chalcopyrite), which contain cadmium. If we compare, again, the arithmetic mean values (X) with alert threshold values for a sensitive land use is found that the four heavy metals content were higher than this threshold, namely the same order of magnitude to those found when comparing to the MAL values, except Pb, which recorded an increase of almost 18 times compared to the alert threshold (AT). Also, the threshold at which intervention is required (IT) is exceeded by 8.9 times for this chemical element. It is therefore absolutely necessary to take, for beginning, intervention measures to block gardeners on such soils, and then measures to reduce the levels of Pb in these soils.

The content of mobile forms of heavy metals in the upper horizon (0-20cm) of urban soils planted with vegetables

Concentration of heavy metals soluble in EDTA-CH_3COONH_4 solution at pH 7.0 (Table 2) was evaluated only for the four polluting chemicals (Cd, Cu, Pb and Zn) compared with maximum allowable limits (MAL) established by Lacatusu, cited in [1]. This solution extracts the chemical elements existing in soil solution, but most of those bounded to organic matter and soil exchangeable component. Taking into account, also, the arithmetic mean value is found that in the upper horizon of urban soils planted with vegetables in Baia Mare and surrounding areas, mobile forms of heavy metals are higher than the MAL by 3.4 (Cd), 22 (Cu), 17 (Pb) and 4.8 (Zn) times.

Table 2. Statistical parameters of mobile heavy metals content soluble in EDTA-CH_3COONH_4 solution at pH 7.0, in upper horizon (0-20 cm) of urban soils planted with vegetables in Baia Mare city compared with maximum allowable limits (MAL).

Parameter[a]	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
				$mg \cdot kg^{-1}$				
Xmin	0.3	1.0	0.5	9	127	2	61	130
Xmax	5.1	12.0	16	210	352	14	1293	599
X	3.4	7.0	9	176	193	9	306	207
σ	1.8	3.4	7	84	104	6	124	139
Xg	3.1	6.5	8	163	175	13	285	210
Me	3.0	6.1	8	169	180	12	292	193
MAL*	1.0			8			18	43

[a]idem Table 1; * after Lacatusu in [1].

In addition to the heavy metals total content, the level of solubilization depends by the chemical forms in which these chemicals are present in soil, soil reaction, organic matter content, clay content and interference from other chemical elements, some with role of nutrition, growth and development of plants (N, P, K, Ca, S, and Mg).

Native, urban soils that we analyzed are acidic, but their reaction has been corrected over time by amendment and fertilization, usually organic, so now they have a slightly acid reaction. Soils have a loamy, loamy-clay texture, with a moderate content of humus and nutrients.

Fractions of heavy metals in urban soils planted with vegetables

Fractions of the four chemicals polluting were determined sequentially by extraction with specific reagents. In this way, have been highlighted parts of the total heavy metals content that may be located either in the soil solution and bounded to exchangeable colloidal complex, organic matter, oxides and hydroxides present or may be included in the crystalline structure of soil minerals . The results were presented as percentage mean values (Figure 1).

It reveals that in soil solution cadmium has the highest concentration, followed by Pb and Zn. The phenomenon is due to both high mobility of Cd compared with the mobility of others chemical elements [10] and its high soil content, on average, 15 times more than normal content.

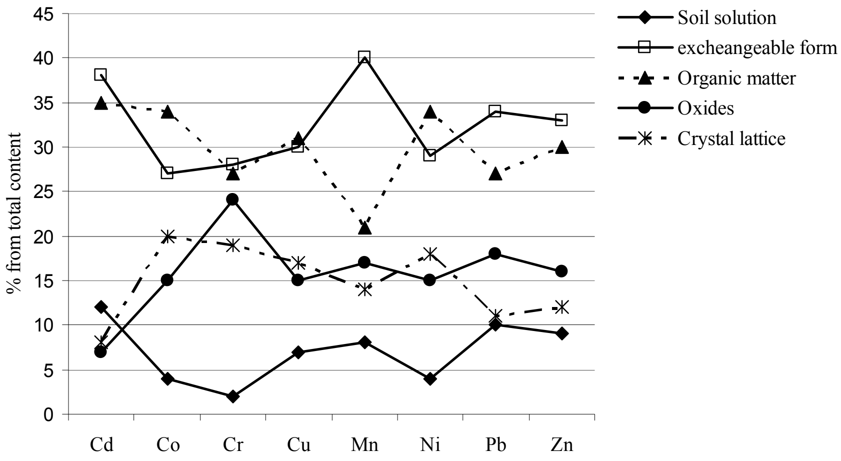

Fig. 1. Variation of heavy metals fractions expressed as average percent from the total content determined in the upper horizon of urban soils from Baia Mare city and the surrounding area, planted with vegetables.

Percentage values of the other fractions of heavy metals to the total content are relatively close in case of exchangeable fractions and those bounded to organic matter. Cadmium is highlighted again by the lower percentage values of its fractions related to oxides and hydroxides or crystalline structure.

Accumulation of heavy metals in the edible parts of vegetables

As a result of high levels of heavy metals in soils, especially in forms related to soil solution, organic matter and exchangeable fraction, vegetable plants were given large amounts of heavy metals that have passively absorbed along with other nutrients from soil. A portion of absorbed heavy metals accumulated in the edible parts of vegetables: leaves (lettuce and orach) and roots (carrot and radish).

Analytical data of the Cd, Cu, Pb and Zn contents in the plants organs examined are presented in Table 3.

Table 3. Statistical parameters of the heavy metals content in edible parts of vegetables grown on urban soils from Baia Mare city, compared with normal values (NV *).

Parameter[a]	Cd				Cu			
	mg·kg^{-1} dry matter							
	1	2	3	4**	1	2	3	4
Xmin	0.9	0.5	0.7	0.5	3.0	1.9	3.8	21
Xmax	8.1	7.9	5.1	4.7	19.1	14.7	18.7	87
X	4.9	4.7	2.7	2.6	11.8	7.8	12.0	44
σ	3.2	2.6	2.5	2.2	14.3	3.4	7.3	21
Xg	4.4	4.3	2.5	2.3	10.5	7.1	10.9	40
Me	4.6	4.1	2.3	2.2	10.7	7.3	11.0	36

Parameter[a]	Pb				Zn			
	mg·kg^{-1} dry matter							
	1	2	3	4	1	2	3	4
Xmin	36	30	21	39	74	134	63	59
Xmax	196	74	81	74	169	301	174	193
yyyX	118	51	55	39	121	197	97	88
σ	251	95	43	42	64	131	66	73
Xg	109	50	49	33	114	187	90	84
Me	106	43	41	32	110	183	88	80

[a] idem Table 1; *NV: <1 mg·kg^{-1} Cd; <5 mg·kg^{-1}Cu; 5-10 mg·kg^{-1} Pb; 20-50 mg·kg^{-1} Zn after [2] and [4]

**1-lettuce; 2-orach; 3-carrots; 4-radish

Keeping the same term of comparison (arithmetic mean value) relative to normal content, it can be easily seen that have accumulated large quantities of heavy metals, higher than normal content values. Thus, cadmium was accumulated, on average, by 4.9 times in lettuce, by 4.7 times in orach, by 2.7 times in carrots and by 2.6 times in radish. Similarly, in the case of copper, which at low content values is an important microelement in plant nutrition, taking as term of comparison the maximum allowable limit (5 $mg \cdot kg^{-1}$) reveal that this chemical element is by 5.9 (lettuce), 1.5 (orach), 2.4 (carrots) and 8.8 (radish) times more concentrated than normal. Under the same rule of comparison, in the edible parts of vegetables analyzed lead accumulated by 11.8 times (lettuce), by 5.1 times (orach), 5.5 times (carrot), 3.9 times (radish) as compared with the maximum allowable value. Finally, for zinc, which also is an important trace mineral for plant nutrition to a certain level, the levels exceeded 2.4 times (lettuce), 3.9 times (orach), 1.9 times (carrot) and 1.7 times (radish), the maximum permitted normal value (50 $mg \cdot kg^{-1}$ dry matter).

Therefore, it is observed without exception that the accumulation of heavy metals in urban soils that were cultivated with vegetables has led to their accumulation in cultivated plants, including edible part of them. In fact, between soluble Cd content in soil and Cd contents in edible parts of lettuce and carrot directly proportional relationship, statistically assured were recorded (Figure 2). Similar relationships were obtained for the other heavy metals, and in all plants analyzed, as reports values (η) and correlation coefficients (r) reveals (Table 4).

Table 4. Values of correlation coefficients (r) and correlation reports (η) between heavy metals content in edible part of vegetables and soluble heavy metals content in urban soils cultivated with vegetables.

Plant species	Cd		Cu		Pb		Zn	
	r	η	r	η	r	η	r	η
Lettuce	0.770	0.771	0.553	0.441	0.829	0.701	0.650	0.519
Orach	0.659	0.703	0.413	0.508	0.723	0.567	0.807	0.759
Carrots	0.472	0.473	0.520	0.667	0.470	0.581	0.623	0.519
Radish	0.419	0.523	0.707	0.614	0.530	0.720	0.724	0.645

Ingestion of such vegetables with high heavy metal loading, is leading to humans and animals healthiness state alteration, causes a number of the digestive and circulatory systems diseases. Are known the carcinogenic effects generated by continuously consumption of vegetables and fruits loaded with heavy metals as Cd, Pb or even Cu and Zn, which contributing, in a significant proportion, in increasing the incidence of gastro-intestinal cancer, cancer of pancreas, urinary bladder or prostate, as was reported in various countries [11, 12, 13]. Life expectancy of people in Baia Mare city and surrounding areas is ten years lower than in other unpolluted zones, due to environmental contamination, especially with heavy metals [11.12, 13]. Life expectancy of people in Baia Mare city and surrounding areas is ten years lower than in other unpolluted zones, due to environmental contamination, especially with heavy metals.

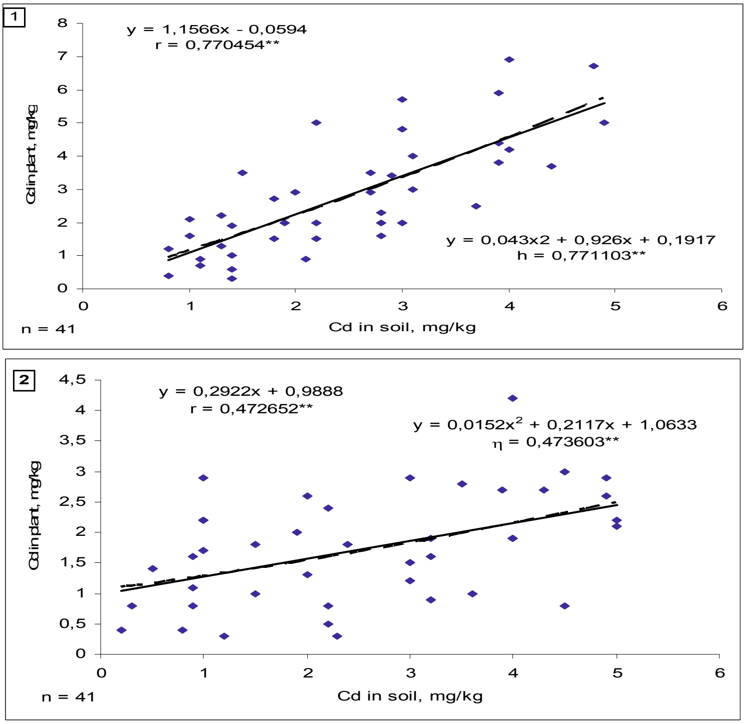

Fig. 2. Relationships between Cd content in edible part of lettuce plant (1) and carrot (2) and content of Cd soluble in EDTA-CH_3COONH_4 solution at pH 7.0 in urban soils cultivated with vegetables from Baia Mare city.

Conclusions

Emissions released from several sources, such as: two-ferrous smelting of mineral concentrates, two units of ore concentration (flotation), tailings dumps of materials resulting from ore flotation, including the extraction of ore led to a strong environmental pollution, including urban soils, mainly with Cd, Cu, Pb and Zn.

Urban soils used for vegetable cultivation have a high pollution degree. In upper horizon (0-20 cm) of these soils, average heavy metals contents exceed maximum allowable limits by 1.5 times for Cd, 3.2 times for Cu, 8.9 times for Pb and 2.6 times for Zn. Highest concentration recorded in these soils exceed the MAL values by 3.6 times for Cd, 7.9 times for Cu, 46.3 times for Pb and 11.5 times for Zn.

Mobile heavy metals contents, soluble in EDTA-CH_3COONH_4 solution at pH 7.0, are, also high and specific to these extracting type, its average values exceeding by 3.4 (Cd), 22 (Cu), 17 (Pb) and 4.8 (Zn) times MAL.

The most part (57-85%) of total heavy metals content belonging to forms available to plants nutrition (soil solution), exchangeable forms, and forms bounded by organic matter.

Vegetables were absorbed from the soil and accumulated in the edible parts appreciable quantities of heavy metals up to contents that, on average, have exceeded from 1.7 times (Zn in radish) to 11.8 times (Pb in lettuce), normal values specific for each chemical element and each plant analyzed.

Between the heavy metal content of the edible parts of vegetables and soil content of heavy metals, soluble in EDTA-CH_3COONH_4 solution, directly proportional relationship, statistically assured, were recorded.

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Risk assessment of contaminants leaching to groundwater in an infrastructure project

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Abstract

Possible risks posed by rebuilding of a road and railway within a contaminated area have been studied by assessing the mobility of metals in soil through analysis of field samples, leaching tests and calculations of distribution K_d coefficients. The results from the standardized leaching tests showed a very high release of Pb, but also high release of Zn, Cu, Ni and As. The site specific K_d values determined showed that Cr and Pb are the metals most strongly bonded in the soil, and that As is the most easily released. Input of humus colloids increased the leaching of Cu, Pb and Zn and show on potential risk with enhanced leaching if flooding of the river or excavation activities at the site. It was concluded that Pb and As constitute the highest risk of leaching from the landfill site.

Introduction

Contamination of land and water is an issue of high environmental concern, and in Sweden more than 80 000 sites are identified as potentially contaminated. Infrastructure projects, as rebuilding of roads in areas with contaminated land, could highly affect the environment through enhanced leaching of contaminants from the soil. In Nol, north of Gothenburg city, there is an old industrial landfill that after rebuilding of the road E45 and railway construction through the landfill area, pose a risk for leaching of metals and polycyclic aromatic hydrocarbons (PAHs) [1]. Parts of the contaminated soil in the landfill have been excavated and sent for deposition and are replaced with new clean filling materials. The landfill was in use until 1984 and contains large amounts of residues from battery production and the soil and groundwater is contaminated with metals as Pb, Cd, Cu, Zn, oils, PAHs and plastic materials. The landfill is situated

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on the river bank of Göta Älv 18 km upstream of the drinking water intake for Gothenburg city with ~500 000 citizens. To be able to make risk assessments of contaminants leaching from soil at contaminated sites, the mobility of the pollutants needs to be quantitatively estimated, and the applicability of laboratory test for the prediction of soil solution and groundwater contamination by metals is under research [2-6].

In the literature there are K_d coefficients available that describe the distribution of metals leaching from soils [7-9]. The K_d concept is a simplification of the often complex processes that govern how a contaminant is distributed in the soil, but needed for the calculations of the mass transport of contaminants; used in modeling and risk assessment of contaminants leaching from soil to groundwater at a contaminated site. For the mass calculations, the estimated K_d coefficients from the literature could be used, but the optimal is to determine the contaminants K_d on soils from the specific site through leaching tests [10]. In a landfill area the soil composition is heterogeneous, and thereby the K_d values will highly vary within the site.

Climate change, with impact as increased temperature and deposition, may affect the leaching of contaminants from a polluted land area by changes in the liquid to solid (L/S) ratio, turbulence and oxygen conditions and thereby redox conditions affecting precipitation/dissolution of contaminant-bearing phases, leaching of more dissolved organic matter (DOC) as humic and fulvic acids [5, 11]. The release of DOC is known to enhance leaching of metals, and the solubility of DOC increases with increasing pH [2, 12]. In this case, the conditions for an enlarged colloidal transport of contaminants from soil to groundwater are possible [13]. Acidification and changes in pH is another example of a human impact that could possibly affect the release of metals from a contaminated soil [3]. Under rapid infiltrations of acidic rain as heavy rainstorm, particle-facilitated transport by silicate minerals is a dominant transport process of Pb and As [14]. Variations in redox conditions induce the precipitation and reductive dissolution of hydr/oxides acting as adsorbent of metals, and affect the formation of metal sulfides [15].

In this project, the leached amounts and partitioning K_d coefficients for the metals As, Cd, Cr, Cu, Pb and Zn has been determined through a standardized two steps leaching tests on soil samples from the landfill area at Nol. The leaching tests were performed on soil samples from different spots and depth in the landfill to show on the heterogeneity impact of the leaching and K_d values. For the investigation of how changes in environmental conditions could affect the leaching of contaminants at the site, also a beaker test with changes in environmental conditions was performed to determine contaminants amounts released and K_d coefficients.

The determined K_d values will be used in scenarios to calculate and assess the risks of contaminants leaching as a consequence of changes in environmental conditions.

Experimental

In the landfill area a total of seventeen soil samples were collected with an auger sample at four locations (1A, 2A, 1B, 2B), and in each spot at different depths from 0 till 5m. All soil samples were analyzed for water, organic pollutants, organic and metal content. Groundwater samples were collected at three occasions from the six wells 1A, 2A, 1B, 2B, 1C, 2C. All samples were analyzed by an accredited laboratory for: organic pollutants, metals in both dissolved and total phases, DOC/TOC, alkalinity, turbidity, conductivity and suspended solids.

Standardized leaching test

A standardized two step leaching test (SS-EN 12457-3) was carried out on two soil samples (1B;2-4m and 2B;1.5-3.5m) at a commercial laboratory, and on three samples (1A;1-2m, 1B; 2-4m and 2B;1-2m) at the Chalmers laboratory. The eluates were divided into one part for total metal concentration analysis and another part filtered through 0.45 μm cellulose acetate filters for analysis of dissolved metals concentration. Samples for metal analysis were preserved with supra-pure concentrated HNO_3 and kept in refrigerator (+4°C) before analysis by ICP-MS. The release of metals after the first step (6 hours) and the cumulative release have been calculated as described in the SS-EN 12457-3. The K_d values in $l \cdot kg^{-1}$ were calculated for As, Cd, Cr, Cu, Pb and Zn as the concentrations of the metal in the soil sample in $mg \cdot kg^{-1}$, divided with the concentration in the liquid solution after the shaking procedures in $mg \cdot l^{-1}$.

Beaker leaching test

In order to determine how changes in environmental conditions affect the leaching of metals, a set of leaching test in beakers in a single step procedure was performed for 24 hours on samples with 100 g dry soil and Milli-Q water added to receive L/S = 6. To simulate how human stresses and impact such as climate change with heavy rain fall, flooding, excavation and acidification affect metals leaching the following conditions were used: 1) input of air bubbles to receive an oxidized environment; 2) input of nitrogen bubbles to receive an anoxic environment; 3) undisturbed environment without mixing; 4) low pH ~4 by addition of HNO_3;

5) high pH 8 – 9 by addition of NaOH; 6) addition of humic acid colloids to obtain a DOC of 200 $mg \cdot l^{-1}$; 7) addition of iron colloids to obtain Fe concentration of 50 $mg \cdot l^{-1}$. The water-soil solutions in the beakers were gently mixed by a magnetic stirrer if not otherwise stated.

In all leachates conductivity, temperature, Eh, pH and dissolved oxygen were measured every hour for the first four hours, and then after 24 hours. Element concentrations were analyzed in the eluates in total and filtrated samples by inductively coupled plasma mass spectrometry (ICP-MS) at the Chemical Environmental Laboratory at Chalmers following a standardized procedure. For TOC/DOC analysis the samples were sent to a commercial laboratory.

Results and discussion

Soil samples

The data for the concentrations of metals and organic pollutants in the soil samples is presented elsewhere. In the majority of the soil samples the lead concentration by far exceed the Swedish guideline values for contaminated sites ("extremely serious contamination", Pb > 800 $mg \cdot kg^{-1}$ DS) [16]. The maximum lead concentration of 9800 $mg \cdot kg^{-1}$ DS was measured in sample 1A at 1–2m. The depth profiles show that the contamination is heterogeneous as in the locations 2A and 1B where the concentration of lead decreases with increasing depth, while in 2B the lead concentrations increase with the depth. Several other contaminants exceeded the limit for sensitive land use: Hg in 4 out of 17 samples with maximum at 2A (1.7 $mg \cdot kg^{-1}$ DS); PAHs in 14 samples with maximum at 1B (10.0 $mg \cdot kg^{-1}$ DS) and Cd in 7 samples with maximum at 2A (5.6 mg $\cdot kg^{-1}$ DS).

Groundwater sampels

In this study not only dissolved but also total concentrations of metals have been compared with the guideline values, because particle transport of metals from the groundwater may be possible in the case of the Göta river flooding into the landfill area [14]. All data for the groundwater concentrations are presented elsewhere. After construction, the groundwater was moderately contaminated (> 10 $\mu g \cdot L^{-1}$) with Pb in dissolved phases in the well 2A, and extremely serious contaminated (> 100 $\mu g \cdot L^{-1}$) with Pb in most of the unfiltered samples. After four and seven month, and for total lead, the concentrations increased significantly in all the wells and reached the level of extremely serious contamination. The increase in total Pb concentration by 20 and 80 times, for the wells 1C and 2C respectively after four month was especially prominent. These results suggest that the construction activity has increased the mobility of metals from soil to groundwater. The increase of metal concentrations was mainly measured in the total phase and suggests that metals are present in a form of particles/precipitation as pH of the groundwater was high and in the range 7.6-9.5 except for 2B where pH = 7.0.

Standard leaching test

Metal concentrations in the soil samples used in the standard leaching test are summarized in Table 1. The results from the two step leaching tests showed a release of trace metals in high concentrations, and amounts in the order: Pb >> Zn > Ni ~ Cu > As > Cd. Lead, Zn and Mn were leached proportionally to their initial concentrations in soil as suggested by the significant correlations between concentrations in soil samples and in the eluates in the dissolved phase ($r > 0.98$). The release of all the trace metals correlated strongly with Al and Fe release that suggests that trace metals are adsorbed to Al and Fe oxides in soil. In addition, there is a possible dependence between TOC/DOC content in the water phase and leaching of metals.

Leaching at changing environment

The pH is known as a governing factor for metals solubility which was also confirmed by the results of this study. The decrease in pH, from 7–8 that was the equilibrium pH of the soil solution, to pH 4–5 in the beaker experiment caused extensive leaching of almost all the metals (Fig. 1). This effect was especially important for Pb, Zn and Cr concentrations. Input of humic colloids increased leaching of Cu, Pb and Zn and the effect was especially important for Pb and Cu that increased by 2.5 and 7 times respectively. Copper and Pb are also the metals that have the highest affinity for organic matter and probably re-adsorb to DOC and therefore found in the water [12]. This effect is important because organic colloids are small and stable, i.e. do not aggregate or precipitate, and can therefore transport metal pollutants on long distances [12]. Nickel and Cd have also high affinity for humic acids however, but no increased mobilization of Cd and Ni was observed.

Table 1. Characteristics of soil samples used in standard leaching test (mg/kg DS).

	1A 1-2m	1B 2-4m	2B 1-2m	2B 1.5-3.5 m
Al	15000	17000	11000	–
Fe	15000	16000	11000	–
Mn	350	270	270	–
Cd	4.6**	3.3*	0.30	0.3
Pb	12000**	2800**	2900**	3300**
LOI[a] (%)	4.1	4.8	3.8	–

[a] LOI= loss of ignition; *very serious contamination; **extremely serious contamination [16]; – not measured

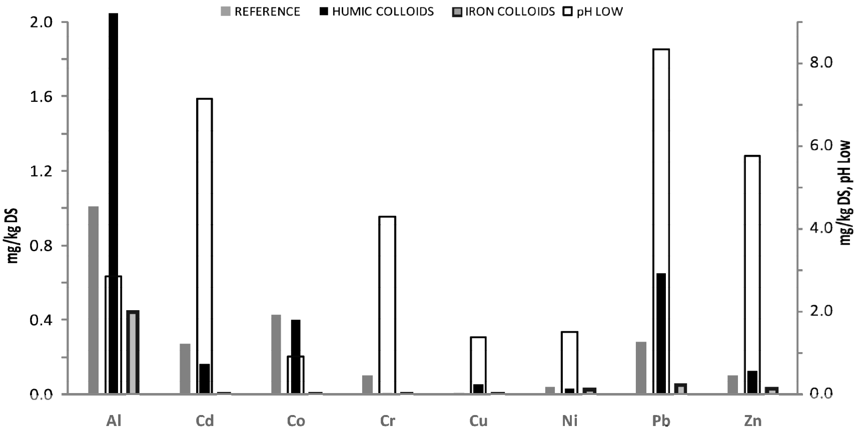

Fig. 1. Release of dissolved metals at different environmental conditions.

Presence of iron colloids had the opposite effect to organic colloids and decreased the water phase concentrations of all the metals. Iron oxides are known to adsorb metals or co-precipitate with them [17]. On the contrary to organic colloids, iron oxides may aggregate together and build bigger particles that may eventually precipitate and sediment [12, 17]. With time precipitated iron oxides may mineralize and therefore immobilize metal pollutants. The decrease of metals in the water phase in this study is probably due to co-precipitation with iron oxides separated from the water phase by filtration.

Distribution coefficients

Site specific K_d coefficients were calculated for both data from field, i.e. soil and groundwater, and for data from the laboratory standard leaching tests. The K_d values were calculated for the metals where the concentrations in soil and groundwater in field were close to exceed the Swedish EPA guideline values for contaminated sites [16].

The K_d coefficients determined for dissolved phases are presented for different soils from the landfill in Table 2, and were in the order (mean in $l \cdot kg^{-1}$): Cr(565000) >> Pb(87000) > Zn(22000) ~ Cu(26000) = Cd >> As(2300), i.e. Cr and Pb are the metals most strongly bonded in the soil and As the most easily released. This order was about equivalent to the order from the reference values, except for As that seems to be more easily leached from this landfill soil than compared with the reference values. The K_d values for both As and Pb were also much lower than in general, and thus showing on a high risk of leaching of these two contaminants from the landfill soils. If including the particulate phase, i.e. total metals, the K_d values for all metals were much lower i.e. showing on a much higher risk of leaching, especially for As, Cr, Pb and Zn.

Table 2. Site specific K_d coefficients in $l \cdot kg^{-1}$, determined in a standardized leaching test[a], calculated on mean concentrations in soil and concentrations in leachate; compared with reference values from literature.

Metal	1A 1-2 m	1B[b] 2-4 m	1B[c] 2-4 m	2B 1-2 m	2B 1.5-3.5 m	Mean	Ref.[d]
As_{dis}[e]	-	-	2600	-	1900	2300	
As_{tot}[f]	-	-	1300	-	80	690	13000
Cd_{dis}	6200	22000	69000	230	8500	21000	
Cd_{tot}	6500	33000	8700	u.d[g]	110	12000	2900
Cr_{dis}	30000	1100000	u.d.	u.d.	u.d.	570000	
Cr_{tot}	4800	11000	2000	u.d.	110	4500	15000
Cu_{dis}	3100	20000	29000	26000	27000	21 000	
Cu_{tot}	2300	3500	3400	28000	140	7500	4800
Pb_{dis}	56000	130000	92000	58000	100000	87000	
Pb_{tot}	11000	7700	5600	12000	110	7300	170000
Zn_{dis}	4000	4500	77000	2600	u.d.	22000	
Zn_{tot}	3700	3300	4000	6900	180	3600	12000

[a] SS-EN12 457–3; [b] Chalmers laboratory; [c] commercial laboratory; [d] mean values from 70 different leaching studies reported in the literature[8]; for each element the data points varies from 4 (Hg) up to 830 (Cd); [e] filtrated through a 0.45µm filter; [f] not filtrated sample; [g] u.d.= under detection limit for analysis methods.

The K_d values calculated from the field data are presented in Table 3 and show for dissolved phases the order (mean in $l \cdot kg^{-1}$): Pb(3.2E+7)>>Cr(74000)~Cu(70000)>Cd(68000)~Zn(58000)>>As(9200), i.e. Pb is the metal most strongly bonded in the soil and As the most easily released. This order was about equivalent to the order from the leaching tests. The more easily release of As in field could be explained

Table 3. Site specific K_d coefficients, calculated on mean concentrations in soil and groundwater.

Metal	Date	K_d ($l \cdot kg^{-1}$)				
		1A	2A	1B	2B	Mean
As_{dis}[a](As_{tot}[b])	20/11/09	5700(1500)	12000(4700)	10000(7700)	1600(130)	7300(3500)
	24/03/10	15000(4700)	16000(540)	13000(5000)	3800(330)	12000(2600)
	29/06/10	9700(4400)	6900(380)	13000(2700)	3800(210)	8300(1900)
Cd_{dis} (Cd_{tot})	20/11/09	47000(2100)	20000(9300)	u.d.(6300)	u.d (210)	34000(4500)
	24/03/10	34000(2500)	5500(530)	u.d.(7000)	85000(610)	31000(2700)
	29/06/10	u.d.(4500)	140000(1.0)	u.d.(14000)	u.d.[c]	140000(6200)
Cr_{dis} (Cr_{tot})	20/11/09	u.d.(16000)	u.d.(11000)	31000(6100)	29000(660)	15000(8400)
	24/03/10	55000(13000)	96000(1100)	13000(4200)	26000(3400)	47500(5400)
	29/06/10	u.d.	u.d.(2900)	160000(8800)	160000(2700)	160000(4800)
Cu_{dis} (Cu_{tot})	20/11/09	25000(5700)	6500(4000)	56000(7400)	20000(340)	27000(4400)
	24/03/10	17000(4200)	10000(880)	4500(2500)	21000(2000)	13000(2400)
	29/06/10	200000(29000)	50000(9100)	280000(72000)	140000(7700)	170000(29000)
Pb_{dis} (Pb_{tot})	20/11/09	7.9E+7(5800)	97000(8600)	u.d.(9300)	4.1E+6(140)	2.8E+7(6000)
	24/03/10	8.9E+6(23000)	87000(580)	2.5E+6(3500)	1.3E+6(1500)	3.2E+6(7100)
	29/06/10	9.0E+7(140000)	2.2E+6(2200)	1.1E+8(17000)	u.d.(1800)	6.6E+7(40000)
Zn_{dis} (Zn_{tot})	20/11/09	33000(4200)	20000(4700)	55000(4300)	6000(440)	29000(3400)
	24/03/10	15000(4000)	33000(700)	44000(1500)	11000(3300)	26000(2400)
	29/06/10	140000(18000)	210000(7400)	190000(40000)	7700(3400)	150000(17000)

[a] dis.= dissolved i.e. filtrated through a 0.45μm filter; [b] not filtrated samples; [c] u.d.= under detection limit for analysis methods.

by low redox potential, anoxic conditions and thereby following enhanced release of As under reducing conditions in the soil and groundwater [18]. The K_d values for all the other metals were much higher, and for lead very much higher, than compared to the laboratory test values, i.e. these contaminants are more strongly bonded to the soil at the landfill site than showed in the leaching tests. In the laboratory tests the samples are shaken which may artificially lead to enhanced contaminant release by abrasion of particles, destruction of aggregates and mobilization of colloids that will enhance the leaching, especially for lead, compared to field conditions in an undisturbed environment [6].

With time, i.e. 4 months from late autumn (November) till early spring (Mars), the K_d values for dissolved Cu, Zn and Pb decreases; thus indicate and enhanced leaching of these contaminants with time. The trend from Mars till June (early summer), on the other hand, showed for all dissolved metals except As a decrease in leaching. The K_d values calculated from the field data when including the particles were following the same order as for the dissolved metals, but these K_d values were much lower, and for As, Cr, Pb and Zn even lower than the reference values.

Conclusion

Soil and groundwater analysis revealed an extremely serious contamination by lead in all the soil and most of the unfiltered, and in a few of the filtrated groundwater samples at the landfill site. Decrease in pH and presence of humic colloids were the factors that increased leaching of all metals. Input of iron colloids had the opposite effect, with decrease in metal leaching, probably due to co-precipitation with iron hydr/oxides. The K_d values obtained in this study are used to calculate the risks of pollutants transport to the river and the newly built railway and road construction. Depending on the high Pb concentrations in soil and groundwater, and for As due to the low K_d and thus easily release, Pb and As constitute the highest risk with leaching of metals from the contaminated site.

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Sewage sludge treatment focused on useful compounds recovery

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Abstract

Phosphorus and nitrogen can be recovered through controlled crystallization of struvite ($MgNH_4PO_4 \cdot 6H_2O$) from digested sludge streams, reached in N, P, Mg (as the result of microorganisms activity) in WWTP. Recovered struvite can be sold as valuable fertilizer. In this paper the results of research on the influence of molar ratio of struvite components, temperature and residence time on the process efficiency were investigated. Various dependences were observed, and as the final result it is supposed to state the process conditions, which allow to obtain product, valuable from economical and ecological point of view.

Introduction

Formation of wastewater sludge provides essential influence on the wastewater treatment process. Total amount of wastewater sludge generated in Poland and in other European Union members increases year by year. The requirements for processing of municipal liquid wastes or industrial wastewater introduced directly into natural water reservoirs (surface waters) are regulated by relevant legal acts which with every subsequent update become more and more restrictive [1-3]. Although the wastewater treatment methods applied in municipal wastewater treatment plants and their regular improvements enable to fulfil current legal requirements, considering state-of-the-art knowledge and development of advanced technologies, they generate considerable amounts of wastewater residues. Wastewater sludge is mainly composed of organic matter with small amount of inorganic compounds, which after appropriate processing could be recognized as a raw material possible to be utilized again. By proper sequence of a number of processes such as anaerobic fermentation,

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co-combustion, pyrolysis, gasification, methane and hydrogen separation, the organic compounds contained in wastewater sludge can be utilized for power generation purposes. The inorganic compounds contained in the sludge, mainly of nitrogen and phosphorus (biogenic) ones, could become, on the other hand, a valuable source of recycled materials, e.g. for the fertilizer industry (agriculture, horticulture). The values of fertilizing substance concentrations in the wastewater sludge derived from municipal wastewater treatment plants are presented in Table 1 where comparison with natural fertilizers was also done.

Table 1. Fertilizer substances in a wastewater sludge derived from the municipal wastewater treatment plants [4].

Fertilizer component	Preliminary sludge	Excessive sludge	Cattle manure	Liquid manure
	% of dry mass			
Nitrogen (N)	2.0-7.0	3.0-10.0	0.45	0.12-0.45
Potassium (K)	0.1-0.7	0.1-0.8	0.425	0.133-0.332
Phosphorus (P)	0.4-3.0	0.9-1.5	0.087	0.017-0.10

Skilful management of these compounds is also especially important for natural environment protection since biogenic elements released in an uncontrolled manner to the natural environment are responsible for its eutrophication. This phenomenon results in disturbance in the natural functioning of water ecosystems.

According to the data of the Head Statistical Office for the year 2007, in Poland there are over 3 thousand municipal wastewater treatment plants, where $567.3 \cdot 10^3$ Mg of sludge has been generated (calculation based on dry mass of the wastewater sludge). About 19.8% of this mass was then used for agricultural purposes (cultivation of all types of crops present in the market, including the crops destined for the production of animal food). 18.5% was used for renewal of the grounds (including arable areas), about 4.9% was subjected to composting, whereas 1.1% was thermally processed and 16.2% was deposited.

To consider the wastewater sludge derived from municipal and industrial wastewater treatment plants a material possible to be utilized again, it must fulfil strictly specified criteria which are regulated by relevant legal acts [6, 7]. The basic criteria governing the recycling of sludge are the contents of heavy metals, their fertilizing properties, hydration, dry mass, reactivity, fulfilment of sanitary standards, etc. Considering both formal and legal aspects the wastewater sludge generated in Poland and in other European Community members can be utilized by [8]:

1. direct application in agriculture, horticulture and forestry: sludge is regarded as an important source of organic matter and nutrients for the plants, thus possible to be re-introduced into the biological transformation cycle; furthermore, wastewater sludge is a valuable material for the reconstruction of appropriate chemical structure of soil in the degraded areas;
2. direct application in rehabilitation and recultivation of landfills and industrial waste dumps; especially in reconstruction of the humus layer of the soil (in the fermented wastewater sludge there is 20 times more humus-generating substances than in the humus soil itself),
3. gasification of sludge – the gas produced may be used for auxiliary heating purposes in the wastewater treatment plants; the co-generated gas components can be also used in various chemical processes.

The problem of the sludge reclaiming defines a considerable challenge for wastewater treatment plants exploiters, who, with the support of part of researchers, seek the most favourable solutions meeting ecological, technical and economical criteria. This is a problem still waiting for optimal solution not only in Poland, but also in most European countries, as well as around the world. During last few years a number of scientific studies have been done, resulting in various technological and equipment solutions, where the nitrogen and phosphorus compounds reclaimed from wastewater and wastewater sludge were fixed in the form of ammonium, calcium, magnesium and potassium phosphates applicable as fertilizer components [9-11]. In the municipal wastewater treatment plant advanced technologies of phosphorus compounds elimination are applied, in which biological methods are used together with the chemical ones. Thanks to simultaneous processes better results of the wastewater treatment plant are obtained compared to the former treatment methods, i.e. lower concentrations of biogenic elements in clear wastewater released into natural reservoirs and more advantageous values of parameters – for any further application – of the occurring excessive wastewater sludge. Phosphates and ammonium compounds are the most often reclaimed from the streams of fermented sludge in the process of precipitation and crystallization of ammonium and magnesium phosphate (struvite), rich in nitrogen and phosphorus. The enrichment takes place in result of sludge thickening and releasing from it the orthophosphates stored in cells of microorganisms. The active sludge is then subject of methane fermentation (repeated release). Concentration of phosphorus increases up to the value of 3-6% mass P_2O_5, which presents the highest value of the phosphates concentration in the conventional technological system of a municipal wastewater treatment plant. This fact also has a negative aspect, too. Presence of phosphate and ammonium ions in this specific location within a wastewater treatment plant spatial structure, in favourable physical and chemical conditions leads to the spontaneous, uncontrolled precipitation of phosphorus compounds. In effect of this phenomenon incrustation on the surfaces of appliances and pipelines is formed. This type of residues tends to appear in pipelines where the flow of wastewater is minimal, as well as in places of turbulent flows and locations where there are significant and frequent pressure fluctuations (pipes, pipe connections, valves, pumps, etc.). Uncontrolled process of phosphates precipitation in the form of struvite produces reduction of the pipelines diameter, which exerts negative influence on the course of the technological process and may even lead to the installation plugging. In such case it is necessary to apply thorough cleaning of the system (acids are usually used to eliminate such type of residues), what in consequence raises the working costs of a wastewater treatment plant and additionally creates a risk connected with use of strongly acidic solutions. This situation could be, however, avoided by the application of chemical control, for example by precipitation of phosphorus and ammonium compounds in a controlled manner. In result the costs connected with the maintenance and operation of the technological system could be reduced. The value of savings covers the costs of chemical compounds applied for the precipitation of struvite, reduced by the revenues generated by the sales of the product obtained, in some cases estimated to equal 25 euro per ton [12].

Experimental and results

The tests of precipitation and crystallization of struvite were carried out in several series, using the wastewater derived from the wastewater sludge processing section of a typical municipal wastewater treatment plant applying the technology of intensified elimination of biogenic substances (equivalent population > 100,000, capacity 50 thousand m^3/day). The wastewater tested contained streams coming from methane fermentation chamber and reflux from the filtration press (filtrate). Before the tests, some series of analytical measurements were carried out for several months in order to identify qualitatively and quantitatively the wastewater components in different seasons of the year. Reflux from filtration presses was selected for testing the precipitation and crystallization process due to the highest contents of key components – the highest concentrations of phosphate and ammonium ions. The required dose of magnesium ions was introduced into the system in the chemical form of a chloride ($MgCl_2 \cdot 6H_2O$). Averaged results of the analyses are presented in Table 2.

Table 2. Filtrate composition from the filtration press.

Compound	Unit	Concentration
$P\text{-}PO_4$	mg/L	130-220
$N\text{-}NH_4$	mg/L	750-1200
Mg	mg/L	15-25
Ca	mg/L	55-70
pH		8.3-8.6

In first stage of experiment some test series were carried out based on model synthetic wastewater, prepared considering the data presented in Table 2. Tests of the batch precipitation and crystallization of struvite from the model synthetic wastewaters were carried out in the laboratory scale (working capacity of the reactor 1.5 dm^3). Their main objective was identification of the optimal parameter values for the further experiments with the application of real wastewater, i.e. pH, temperature, type and dose of the precipitating agent and the wastewater retention time. In order to carry out the precipitation and crystallization of phosphates from the wastewaters, magnesium ions were introduced into the system to fulfil the required molar ratios between the main components, i.e. phosphate, ammonium and magnesium ions. Precipitating agents used in the tests were diversified in terms of their dose used, as well as their quality. Magnesium oxide (MgO) or hydrated magnesium chloride ($MgCl_2.6H_2O$) were used during the tests. Molar concentration of magnesium ions in relation to the phosphate ions was established to fulfil the stoichiometric proportion within the system, as well as to provide 10, 20, 30, 50 or 100% excess compared to stoichiometric demand. Precipitating agent was dosed once, immediately after introduction of wastewater into the reactor. If necessary the pH value was adjusted by NaOH solution. Wastewater retention time in the reactor oscillated between 15 and 120 minutes. Scheme of the laboratory scale plant for the reclamation of phosphorus from wastewaters is presented in Figure 1.

The test results at this stage allowed one to describe the course of the precipitation crystallization by the assessment of the correlation between the degree of phosphorus compounds elimination from the wastewater and the concentration of magnesium ions present in the wastewater (molar ratios of the reagents in struvite formation process), pH of the wastewater tested and the retention time of wastewater in the reactor. The results are presented in the Figures 2 and 3. Quality of the product was evaluated by granulometric composition analysis, chemical composition analysis and microscopic analysis. The results of these analyses are presented in Table 3, as well as in Figure 4 and Photograph 1.

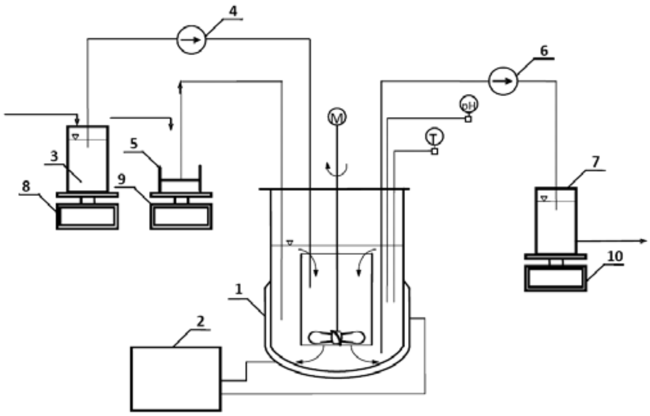

Fig. 1. Scheme of the laboratory installation for the reclamation of phosphorus compounds from wastewater by precipitation and crystallization of struvite, 1 – batch crystallizer, 2 – thermostat, 3 – raw feed wastewater container, 4 – pump dosing the raw material, 5 – dispenser of the precipitating agent: $MgCl_2.6H_2O$, 6 – pump for withdrawal of suspension from the crystallizer, 7 – suspension container, 8, 9, 10 – weights, M – mixer rotation control, pH – pH parameter control, T – temperature control.

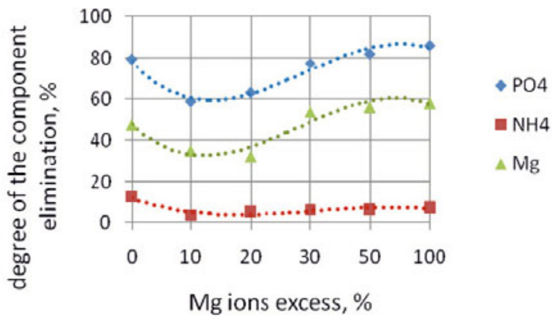

Fig. 2. Degrees of the individual components elimination from the model synthetic wastewater in relation to the quantity of the magnesium ions in the reaction system (pH=8.5, retention time of wastewater in reactor 60 minutes).

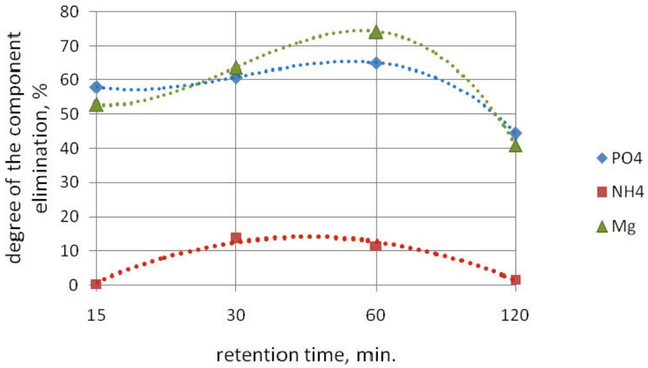

Fig. 3. Degrees of the individual components elimination from the model synthetic wastewater in relation to the wastewater retention time in the reaction system (pH=8.5, Mg/PO_4=1:1).

Table 3. Chemical composition of the product precipitated from the model synthetic wastewater (Mg/PO_4 [mol/mol]=1, retention time 60 min., pH of the wastewater is presented in column 1).

pH of the waste water	PO_4 %	NH_4 %	Mg %	pH of the prod. (1% sol.)
8.0	36.3	6.59	10.15	9.3
8.5	36.0	6.15	10.89	9.0
9.0	36.7	6.64	10.11	9.6
9.5	36.1	6.72	10.04	9.9
10.0	37.0	7.03	9.56	10.0

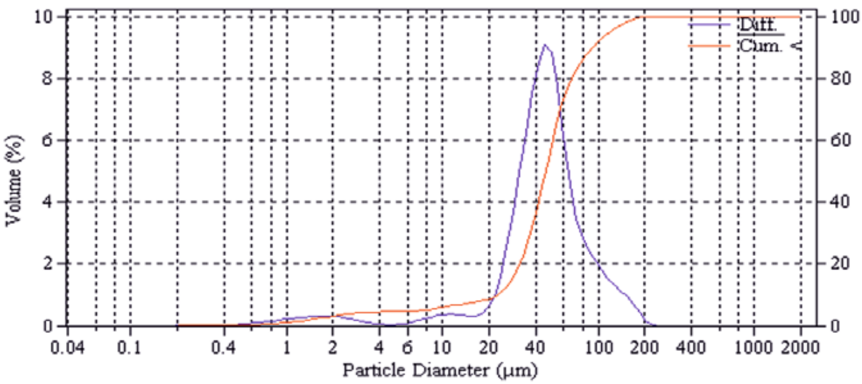

Fig. 4. Cumulative size distribution of the crystal product precipitated from the model synthetic wastewater (pH=9.0, Mg:PO_4=1.5, source of magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 min.).

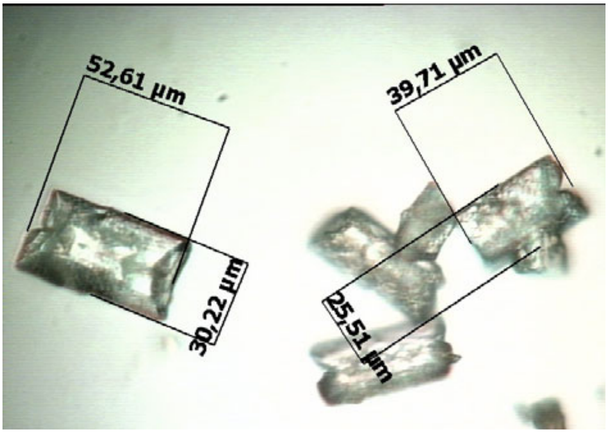

Photo 1. Examples of struvite crystals precipitated from the model synthetic wastewater – image made with the application of optical microscope in the transmitted light, magnification 640x (pH=9.0, $Mg:PO_4$=1.5, source of magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 minutes).

On the basis of the results analysis it was concluded that the maximum effectiveness in the elimination of the phosphate ions corresponded to 100% excess of the magnesium ions in relation to the stoichiometric requirement, when the optimal retention time was identified to be 60 minutes. The source of magnesium ions was magnesium chloride 6-hydrate. These parameter values were thus assumed during testing the process of precipitation and crystallization of the phosphorus and ammonium compounds from the real wastewaters. Process ran in the temperature of 30°C. Since worse results corresponded to the temperature of 20°C, it was decided to assume this value for further research. This higher temperature seems also to be justified with the real process conditions, dominated by the methane fermentation system and the system of fermented sludge dehydration.

The next stage covered the tests on the precipitation and crystallization of the phosphorus compounds from the real wastewaters. The tests were carried out in the system presented in the Figure 1, however working capacity of the reactor was 60 dm^3. Input parameters of the process were as follows:

- pH=8.5,
- retention time: 60 minutes,
- $Mg:PO_4$ ratio = 1.5:1,
- source of the magnesium ion: $MgCl_2 \cdot 6H_2O$.

The product obtained was mechanically separated from the solution with the application of the pressure filtration. Chemical analysis of the -PO_4, -NH_4, -Mg ions contents in the post-crystallization mother solutions was done. The results are presented at the Table 4, in Figure 5 and in Photograph 2.

Table 4. Chemical analyses results of the of post-crystallization mother liquors after the process of precipitation and crystallization of the phosphorus and ammonium compounds from the liquid municipal refuses.

Test	1	2	3
pH	8.0	9.0	8.5
Initial concentration [mg dm^{-3}]			
PO_4	130	160	220
NH_4	870	980	1180
Mg	78	68	85
Final concentration [mg dm^{-3}]			
PO_4	26	44	28
NH_4	783	853	1050
Mg	27	22	24
Component elimination degree [%]			
PO_4	80	85	87
NH_4	10	13	11
Mg	66	68	72

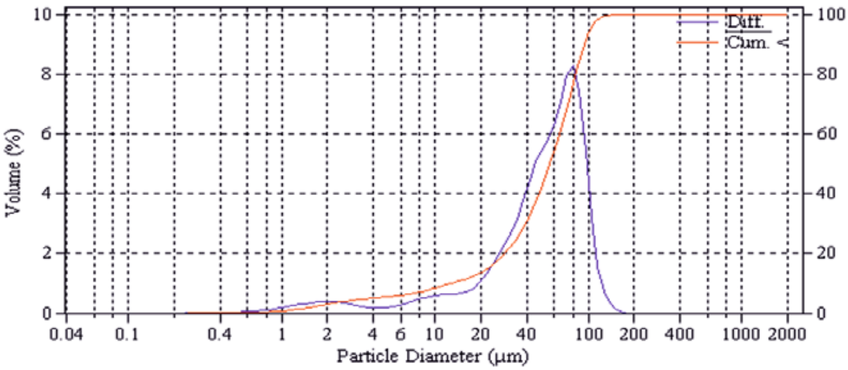

Fig. 5. Cumulative size distribution of the crystal product precipitated from the real wastewater (pH=8.5, Mg:PO_4=1.5:1, source of the magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 min.).

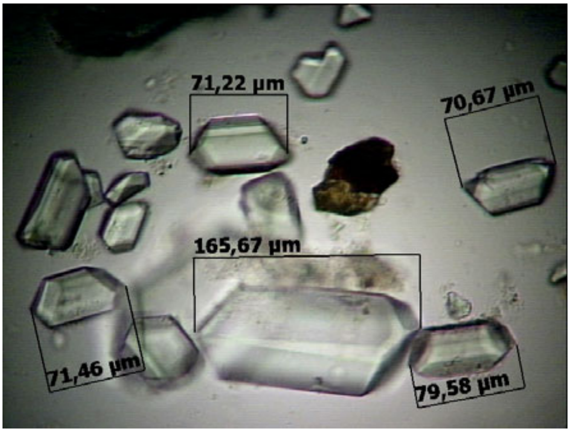

Photo 2. Struvite crystals precipitated from the real wastewater – image made with the application of optical microscope in the transmitted light, magnification 640x (pH=8.5, $Mg:PO_4$=1.5:1, source of the magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 min.).

Discussion

On the basis of test results concerning precipitation of the ammonium and magnesium phosphates from the model synthetic wastewaters in the laboratory scale conditions an critical assessment of the process was carried out by analysis of elimination degree of the phosphate ions from the wastewaters, as well as by determining of the fertilizing aptitude of the product obtained (especially N, P, Mg contents and crystals size). The course of the process of eliminating the phosphate ions from wastewaters depends significantly on the initial pH value of the wastewater treated and on the process environment pH. Literature data were confirmed during the tests. The highest degree of eliminating the phosphate ions from waste-water corresponded to pH value of 8.5-9.5. Considering the pH value of the real wastewater present in the technological system of the wastewater sludge processing, pH=8.5, this value was assumed for the second stage of the research. Depending on other process conditions (molar ratio between $Mg:PO_4^{-3}$, temperature, retention time), effectiveness of the phosphates elimination, determined by elimination degree of $P\text{-}PO_4$ from the wastewaters, was a subject of changes from 59% (pH=8.3, $Mg:PO_4^{3-}$= 1.1:1, source of the magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 min.) to 85% (pH=8.5, $Mg:PO_4^{3-}$=2:1, source of the magnesium ions $MgCl_2 \cdot 6H_2O$, retention time 60 min.). It was observed that with 50% excess of the magnesium ions in relation to the phosphate ions ($Mg:PO_4^{3-}$=1.5:1) in comparative process conditions degree of eliminating $P\text{-}PO^{3-}$ from the wastewater streams reaches ca. 80% (Fig. 2). This observation is important in respect to the process economics since it is advisable to consider to run the process with the addition of a smaller amount of the magnesium compound (input costs reduction) guarantying a satisfactory result in components elimination process. On the basis of chemical analysis results (Table 3) it can be concluded, that the percentage of nitrogen, phosphorus and magnesium in the products resulting from precipitation and crystallization processed are close to the theoretical proportions of these components in struvite ($P\text{-}PO_4^{3-}$ – 38.8%, $N\text{-}NH_4^+$ – 7.3%, Mg^{2+} – 9.8%). Therefore, it is possible to suggest use of them to the soil environment as an additive to the fertilizing mixtures. Utilization method of these products is also determined by the size of crystals. All testing series results confirm that average size of product crystals is strongly influenced by the process environment's pH, as well as by the molar ratio $Mg:PO_4^{3-}$. In case of magnesium chloride 6-hydrate application the increase in the pH value effected in reduction of the average crystal sizes. Furthermore, it was observed that lower value of $Mg:PO_4^{3-}$ ratio corresponded to smaller crystals. Crystal size distribution of ammonium and magnesium phosphate (average crystal size – 42 μm) produced during tests in which the source of magnesium ions was magnesium chloride 6-hygrate, environment pH=8.5 and the initial concentration of magnesium ions exceeded concentration of phosphate ions by 100% is presented in Figure 4. Microscope image of the precipitated product crystals (process parameters shown above) is presented in Photograph 1. Test results from the series using real wastewater confirm the possibility to use the process of the phosphate ions precipitation for the municipal liquid wastes purification, simultaneously recovering usable components. Resulting precipitation products (ammonium and magnesium phosphate) characterize by advantageous chemical composition required for fertilizing compounds. Their application in this area is also supported by the crystal sizes – especially their average size 65 μm (see Figure 5 and Photograph 2). Second test results slightly differ from the first test results using model synthetic wastewater. This discrepancy can be explained by the influence of so-called foreign ions present in various forms in the real complex mixture which the municipal refuse is. Nevertheless, these discrepancies fall within the accepted limits permitted by appropriate normative acts [13, 14].

Conclusion

Production of phosphorus compounds from the municipal refuse, in particular from wastewater sludge, and re-introduction them to the cycle in the form of recycled materials have been examined for many years. In these technologies not only economical aspects of the methods, resulting e.g. from the revenue generated by the products sale, are subject to evaluation. Most of the benefits resulting from their interaction with the natural environment are assessed [15-17]. Elimination of the phosphorus compounds from wastewater, therefore its treatment, provides advantageous changes both in quantitative and in qualitative aspects in respect to the discharged load of pollutants, mainly limitation and prevention of the eutrophication phenomenon. Returning back into cyclic processes running in natural environment some quantities of phosphorus compounds will also contribute to the reduction of the phosphorus ore deposits exploitation, which is in compliance with the postulates of the European policy propagating the principles of the sustainable development. Phosphorus content in the precipitated products was 11.7-12.0%, while nitrogen 4.8-5.5%, and magnesium 9.6-10.9%. On the basis of the products composition, according to the literature data – normative guidelines – they can be classified as fully applicable multicomponent additives to classical mineral fertilizers. Low solubility of struvite can be practically employed in slow releasing of its components into the soil environment. It will also advantageously result in the reduction of losses caused by rapid leaching of the released nutrients from the soil thus contributing reduction of the load of the nitrogen and phosphorus compounds entering natural water reservoirs together with the run-off streams (another aspects limiting the eutrophication phenomenon). The struvite obtained as a result of the tests is undoubtedly usable as a fertilizer or as one component of multi-component fertilizers. However, it must be noticed that in case of utilizing struvite produced from the municipal liquid waste streams it is necessary to make the product sterilization. During the process of pathogens elimination at the temperature of 120°C, at the same time struvite undergoes dehydration by losing 5 molecules of water; $MgNH_4PO_4 \cdot 6H_2O \rightarrow MgNH_4PO_4 \cdot H_2O + 5H_2O$, which has a positive impact on the overall contents of fertilizing components in the final product and on the effectiveness of the elemental components release into the soil environment [18]. The test results correspond to the municipal refuse coming from a municipal wastewater treatment plant where modernization works resulting from connection of subsequent areas of the municipality to the sewerage system are carried out. These activities cause qualitative and quantitative changes in the wastewater streams treated, therefore the tests carried out by the authors and their results seem to be valuable.

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Environmental emission impact from transport during soil remediation

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Abstract

The Swedish Environmental Protection Agency has identified more than 80 000 potentially contaminated sites in Sweden. One of these is the "former Hexion site" in Mölndal, south of Gothenburg. The property was bought by the construction company NCC in order to build a new housing area. On the site industrial production has been performed for almost 200 years. The products have been chemicals, e g binders for the coatings industry and plastics additives like phthalates. Measured concentrations of pollutants exceed the EPA's general guidelines on "sensitive land use" and a remediation is necessary.

The aim of the present study has been to perform a life cycle assessment of the environmental impact caused by excavation, transports and purification in the remediation, comparing three remediation strategies: Insitu, Exsitu and Exsitu in combination with onsite. Four different options for transportation and receiver/treatment have been analyzed.

Exsitu methods were found to cause a much larger environmental load than the insitu. Emissions from the remediation may be reduced by reducing the volume of contaminated soil before transport to landfill. This can be done by pre-treating the soil onsite with sifting or soil washing. Sea transport leads to a high environmental impact. Future reduction of emissions from shipping will make shipping a more competitive choice.

Introduction

Sweden's development as an industrial nation has left its mark in many areas and in many forms. One such remnant of the past can be seen in the shape of 80,000 contaminated sites throughout Sweden [1], a large number of which represent a tangible risk to human health and the environment.

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This situation has given rise to the need for extensive remediation measures. A number of methods for remediating contaminated land have been produced; for an overview, see a list by the US Environmental Protection Agency [2]. Environmental work on the national level in Sweden is based on 16 environmental objectives []. The task of identifying and remediating contaminated land at various points throughout Sweden is being pursued with the aim of bringing about specific improvement in two of these environmental objectives, i.e. "A non-toxic environment" and "A good built environment" [4].

Overall, remediation technologies can be divided into two groups based on the physical location of the remediation [5]: (1). Ex situ remediation, where the contaminated media is removed from the site for subsequent treatment. This can either take place in an above-ground treatment facility (on site) or through treatment or disposal elsewhere (off site). (2) In situ remediation, the treatment of the contaminated media, takes place by employing measures that target contamination in situ in the subsurface. Of all the techniques available, the remediation method used most is dig and dump which is ex situ (off site). Remediation using the in situ strategy is generally a more resource-efficient method than ex situ and on site [6].

Remediation of an area of contaminated land often enhances its value to the community although the actual remediation work also entails greater environmental impact. One example is the increase in the transport of contaminated soil during the remediation process. Consequently, improvement in the local environment can and should be weighed against the load generated during the actual improvement process by making a risk evaluation. The basis of a risk evaluation comprises the overall aim of the measures that are to be taken, the results of the investigations and inquiries carried out, the environmental and health risk assessments and the activity inquiry. In certain cases there could be a need for an in-depth risk evaluation. Several methods are available for implementing such a process [7], the more notable being Cost-Benefit Analysis (CBA), Multi-Criteria Analysis (MCA) and Life Cycle Assessment (LCA) [8].

LCA is a quantitative method for the assessment of the environmental impact of an activity or a system. This method can be applied to calculating, evaluating and comparing the environmental load generated by various remediation methods throughout each phase in their life cycle [9] and [10]. The application of LCA has identified important environmental impact associated with remediation procedures (e.g. [1-3]). A recent literature review shows that the majority of the studies deal with ex situ remediation [5] with focus on site construction work as well as soil treatment and deposition. Landfill emissions are usually not treated [5].

In this study, an evaluation of environmental impact will be conducted as basis for determining the manner and extent to which remediation will need to be carried out at Mölndals Kråka, 500 m east of the centre of Mölndal. This is the site of a large industrial property where production took place from the 1800s through to 2007 when the plant was closed. Industrial activity has left certain parts of the property seriously contaminated. As the aim is to use the land for housing, extensive remediation work will need to be carried out. As part of the evaluation process, LCA will be used to map the environmental impact of potential remediation methods. The aim of the LCA study is to demonstrate the difference in environmental impact of different modes of transport to different recipients.

METHOD

Life Cycle Assessment (LCA)

The method for conducting a life cycle assessment involves a number of stages: goal and scope definition, inventory, classification, characterisation, weighting and interpretation of results [15]. Three of these stages – classification, characterisation and weighting – have been combined under the heading 'Environmental impact assessment', see Figure 1.

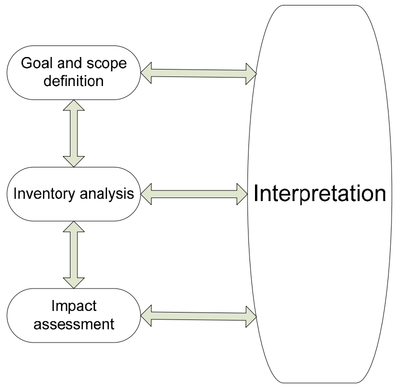

Fig. 1. Flow chart LCA [14].

The goal and scope definition stage comprises a definition of the purpose of the study and how the results will be used. A functional unit is selected, the purpose of which is to create a reference unit to which input and output data can be related. Within this phase the limits of the system being studied are defined as well as the environmental impact categories included in the analysis.

The inventory analysis, the second stage, includes data collection and quantification of the different flows in the system. Based on these results an impact assessment is made by using ‘characterisation factors’, which lead to quantified numbers of impact on the environment divided into the selected environmental impact categories. The concluding part of a life cycle assessment is the interpretation stage, which includes a discussion about data quality, limitations, system limits, validity of the study, a sensitivity analysis, and an assessment of whether the completed study satisfies the objectives formulated at the outset [15].

Description of the site

The property was used to manufacture chemical products for the paint industry. Certain sections of the land within what has been termed the Hexion area (named after the most recent user) are now seriously contaminated. The contaminants identified are lead, the phthalate DEHP and aliphatic hydrocarbons as well as aromatic and polyaromatic hydrocarbons [6]. An extensive risk evaluation was made which will lead to only removing the contaminants that constitute a risk to human beings and the environment. The remediation process will be particularly complex due to the highly varied topography and the resulting geotechnical problems in the form of steep slopes and variations in soil depth. This problem is accentuated by the fact that the contaminants in some places have spread downwards through the easily penetrable landfill media overlying the natural soil layers. The total volume of soil that needs to be remediated is considerable as in certain parts the landfill media are saturated up to 8 m in depth. The remediation will therefore be extensive and will result in a load on the environment.

Case study

The LCA has been conducted with the purpose of evaluating the impacts related to remediation processes. Four different ex situ, off-site scenarios (A-D) were studied (Figure 2). The environmental impact was examined in the form of emissions into air and evaluated in three environmental impact categories: global warming, acidification and eutrophication.

All the scenarios commence with excavation and on-site transport using dumpers. Each scenario is then combined with different recipients and modes of transport. Scenario A differs from the others in the fact that it is only applicable to moderately contaminated media whilst the other scenarios are applicable to highly contaminated media.

The resources that have been identified as inflows into the system are diesel and electricity (Figure 3). The outflows chosen are the emissions carbon dioxide (CO_2), methane (CH_4), dinitrogen oxide (N_2O), sulphur dioxide (SO_2) and nitrogen oxides (NO_x). The functional unit is the treatment of one tonne of contaminated soil.

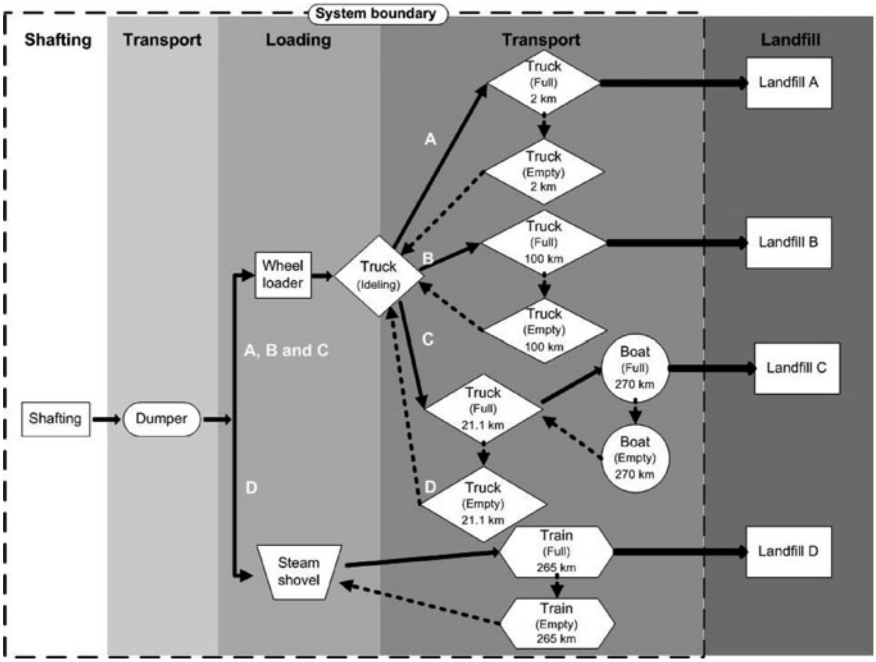

Fig. 2. Flow chart, four different scenarios A-D.

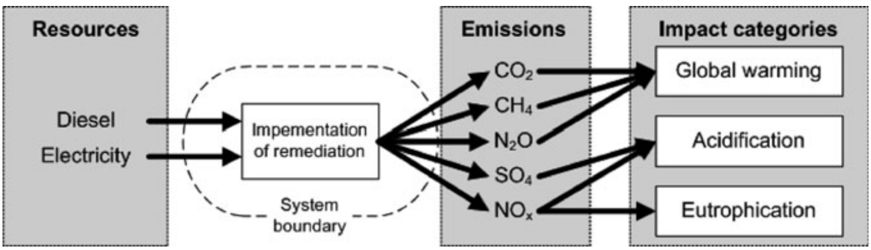

Fig. 3. Illustration of inflows and outflows into/from the defined system.

Results and discussion

The results from the impact assessment for global warming and acidification are shown in Figure 4.

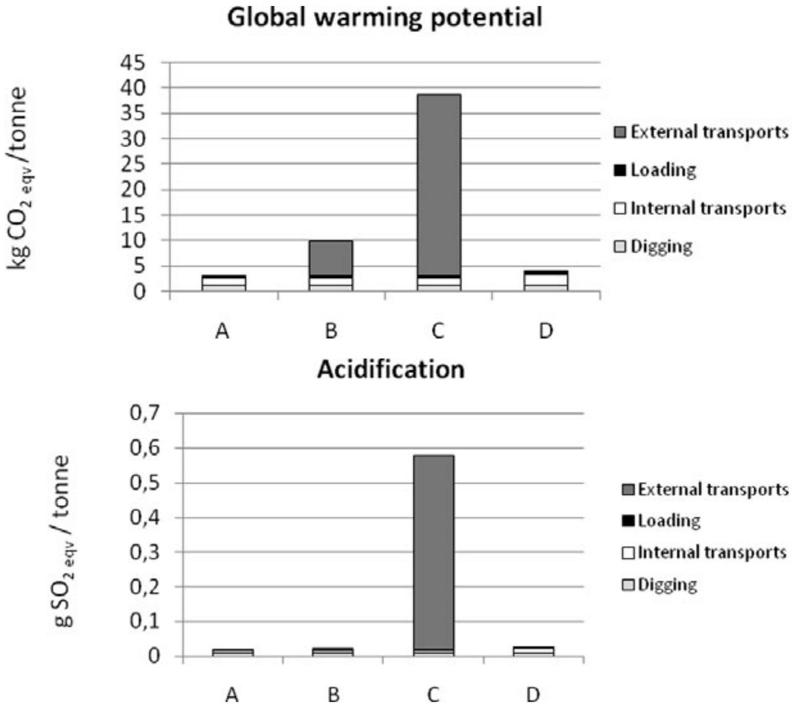

Fig. 4. Emissions from the remediation process for global warming potential and acidification.

The results for eutrophication has similar trend as the other two categories. The LCA conducted shows that scenario C has the greatest impact, with the majority of the emissions related to external transport, which takes place largely by boat. The sea transport contributes to a large degree of the predicted environmental impact, particularly with regard to the contribution to acidification. One reason is that sea transport is the longest transport distance in the study. However, the CO_2 emission data is also higher for boat compared to rail and road; see Table 1.

Table 1. Emission factors used for the three transport alternatives.

Transport alternative	Kg CO_2/tonne-km	Source
Road	0.047	NTM
Boat	0.06	MEET
Rail (Electricity"Good Environmental Choice"	0.000068	NTM

For sea transport, emission factors for small vessels have been calculated using data from the MEET project [17]. These are based on existing vessels without any specific demands regarding emission control. Emission factors for sea transport relating to transport service (tonne-km) are the subject of some discussion owing to a difference of opinion on how to calculate filling factors. The discussion also includes the fact that the load limit can be given by volume or by weight. Slightly different emission factors are proposed, e.g. within the Swedish NTM or the Finnish LIPASTO databases. For instance NTM data for a similar vessel produces an emission factor that is approximately half the MEET value.

Sea transport is generally regarded as having low emissions per tonne-km. This is true for large, slow, long-distance sea transport [17], which is not the case in the present study.

In the future, there will be regulations in place for sulphur and nitrogen oxide emissions. In the North Sea/Baltic Sea regions the permitted sulphur content in marine fuel will be limited to 0.1% from 2015 compared to the most common figure today of 1.5%. In scenario C this would result in a considerably lower impact on acidification, see Figure 5. For nitrogen, the regulations will apply to newly built ships after 2016, giving a reduction of approximately 85% compared to the current average [18]. Regulations or not, this highlights the possibility and importance of the transport purchaser imposing demands regarding emissions.

Alternative D shows the lowest load in all three environmental impact categories. This is particularly obvious for the global warming effect. A strong contributing factor is that the trains operate using electricity generated 100% from HEP with carbon dioxide emissions of 5.6 g/kWh [19]. If the same transport were to be carried out in another part of Europe, where the trains are operated using diesel-powered locomotives or electricity from coal-powered electricity generating plants, the same low emission results would not be obtained. A drawback for rail transport could be limited potential for loading and unloading soil onto/from rail trucks. The chosen landfill of the contaminated soil must, for example, be able to receive the soil by rail. In the case being studied there is the possibility of train loading approximately 800 m from the remediation site.

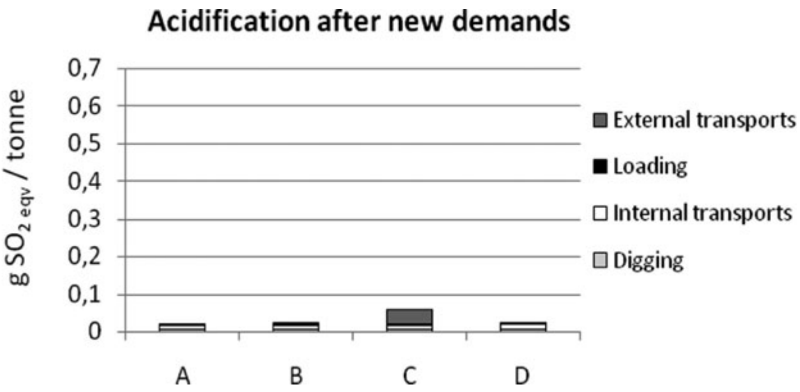

Fig. 5. Acidification after new demands are made on the ocean trade industry.

The fact that transport by road in scenario B is so favourable is due largely to the fact that in the automotive industry national and international regulation of emissions from trucks has been in place for a long time. These requirements often refer to Euro emission standards [20]. In Sweden the most recent changes in emission stipulations took place in 2001 (Euro III), 2006 (Euro IV) and 2009 (Euro V). However whilst heavy vehicle requirements cover carbon monoxide, hydrocarbons, nitric oxide and particulate matter they do not cover CO_2. For diesel-powered machines used within the EU, the first emission requirement was introduced in 1999 in the form of EU Directive 97/68/EC. The Directive involves a gradual tightening up of emission requirements from newly produced vehicles.

In scenarios A and D, where external transport does not account for a high a proportion of the total emissions, it becomes important to study on-site logistics in order to find a way of reducing the volume of produced emissions.

Another interesting angle in the emission problem in conjunction with land remediation is to look at how the national Swedish environmental objectives as a whole are affected. The local improvement thanks to removing contaminants leads to an improvement in the objective “A non-toxic environment”. However there is also deterioration in the environmental objectives “Limited climate impact”, “Only natural acidification” and “No eutrophication”, due to the emissions caused by the remediation work. The best way of reducing environmental impact is to reduce the volume of soil transported and treated. Ideally, this is done initially through a risk evaluation. With ex situ remediation the volume removed from the site can be reduced on site e.g. through sifting and soil washing. In the next step, the transport distance and mode of transport will be crucial to make an environmental improvement.

Conclusions

The primary reason behind environmental impact, in the form of emissions into the air, caused by remediation of the Hexion site is the long, external movement of the contaminated soil to landfill. The external transport alternative for soil hazardous waste that generates most emissions is transport by boat (C) and the alternative that generates the lowest load is transport by rail (D).

To reduce the volume of emissions from external transport, the volume of soil that needs to be transported to landfill must be reduced. This can be achieved initially through a risk evaluation and by using various forms of soil pretreatment, e.g. sifting and/or soil washing. Improved on-site logistics, in combination with newer machinery adapted to stricter environmental stipulations, will reduce the total emission load generated in a remediation project.

When carrying out remediation, the environmental load is switched from the national environmental objectives “A non-toxic environment” and “A good built environment” to the environmental objectives “Reduced climate impact”, “Natural acidification only” and “Zero eutrophication”. By actively choosing the remediation alternative that generates as low total impact on the environment as possible, this contribution can be limited considerably.

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