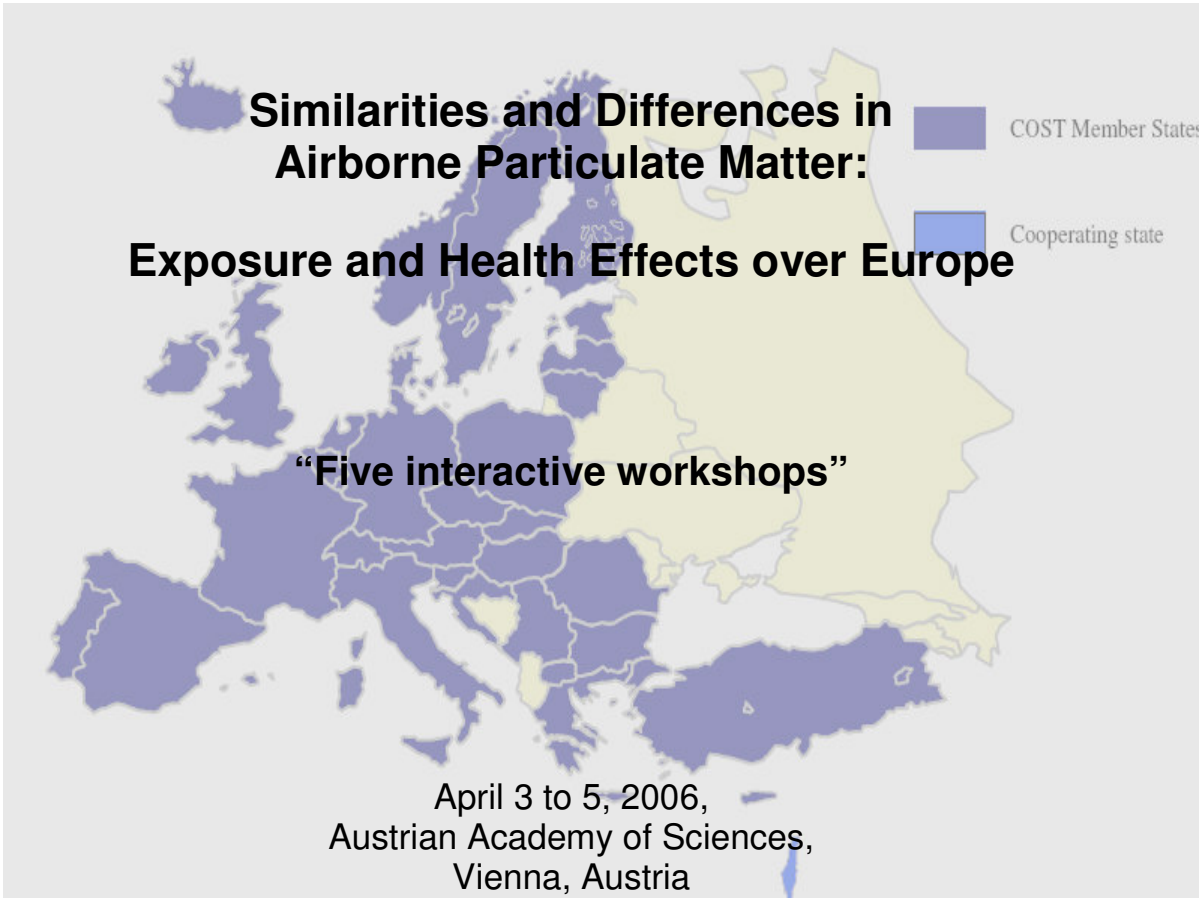
COST ACTION 633
Particulate Matter:
Properties Related to Health Effects

Proceedings of the international conference



Similarities and Differences in Airborne Particulate Matter:

Exposure and Health Effects over Europe

"Five interactive workshops"

April 3 to 5, 2006,
Austrian Academy of Sciences,
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Abstract

The impact of airborne particles on human health is currently seen as the most important environmental issue in Europe. Scientists with the diverse scientific background coming from all over Europe discussed the issue of “Particulate matter and health” in 5 interactive workshops.

Two main conclusions:

- There has been a tremendous increase in knowledge related to airborne particles and their effects on human health over the last decade: the complexity of PM is recognised and requires both new metrics and better understanding of source contributions for effective policy measures.
- Still, major knowledge gaps remain and it is seen that integrated approaches combining the different scientific areas covering environmental, socio-economic and medical research in selected regions in Europe are a prerequisite to effectively tackle the uncertainties European wide.

Specific needs were identified:

- Extension of the current monitoring network with additional particle parameters in urbanized areas.
- Improvement of PM mass measurement accuracy.
- Standardisation of analytical methods for aerosol measurements
- Better integration of epidemiology and toxicology, using for instance same health indicators (biomarkers of effect) with emphasize on oxidative stress.

Policy relevance was explicitly seen in the guidance on additional measures and abatement strategies from specific sources and in the explanation / increased confidence on biological plausibility and causal relationship by toxicology.

The need of collaboration and interdisciplinary approaches is obvious. Several urgent (definite) research needs were identified. A clear recommendation to conduct well organized concerted research studies in several regions in Europe comprising monitoring and research of air quality, exposure, health status, exposure-response functions, source specific toxicological studies as well as evaluation of abatement actions, was given by all members of COST 633 and participants of the workshop.

Preface

COST Action 633 was initiated by Othmar Preining and Helger Hauck with the support of the Clean Air Commission of the Austrian Academy of Sciences because of the clear need to address the topic "Particulate Matter and Health" through a multidisciplinary approach. Extensive knowledge and expertise was available in various relevant areas at the time, but interactions were often limited to experts within each of the fields of particle measurements, dynamics and transformation of atmospheric aerosols, epidemiology, toxicology and modelling of aerosol sources, atmospheric processing, exposure and health effects. Action 633 brings experts of these and other fields together and provides a truly interdisciplinary platform to formulate questions, discuss possible answers and identify research that needs to be performed in the near future and beyond.

The COST 633 conference provided an interdisciplinary discussion forum also to scientists and stakeholders who are not members of the MC or one of the working groups of the action. The results of our discussions form the main part of this scientific report.

The conference would not have been possible without the help and support of several institutions and persons. We are grateful to the Austrian Academy of Sciences for hosting the conference and for administrative support, the City of Vienna for the evening reception at the Vienna City Hall, and the University of Vienna for administrative support. The conference would not have been possible without the financial support given by the COST Office.

Gudrun Breschar of the Clean Air Commission of the Academy of Sciences was a great help with the administrative part of the conference at the Academy. Vera Meyer of the University of Vienna dealt with registrations, abstracts and other administrative issues. Peter Reisinger, Gerhard Steiner and Anna Wonaschütz helped with other preparations and provided on-site technical and administrative support at the conference. We are very grateful to them all – without their help and their dedication the conference would not have been the success it was.

As chair of the MC of COST 633 it is my great pleasure to express our gratitude to the topic leaders, rapporteurs and breakout group leaders for the efforts they put into the discussions and the report. Our heartfelt thanks go to the co-chairs of the program committee Flemming Cassee and Thomas Kuhlbusch for the hard work they did for the conference and the conference report – the Action thanks you for the success of the conference!

Regina Hitzenberger
Chair, Management Committee, COST 633

Executive Summary

COST Workshop 633 – Particulate Matter and Health

Similarities and differences in airborne particulate matter,

exposure and health effects over EuropeParticulate Matter and Health

The impact of airborne particles on human health is currently seen as the most important environmental issue in Europe. Recent assessments showed an expected loss in life expectancy of about 9 months in the year 2000 (EU-25, central CAFE baseline estimate[1]) due to exposure to (ambient) PM2.5 mass. The revisions of the Air Quality Directive and its daughter directives were discussed at the same time in Brussels which all form the background of this conference.

Various scientific areas covering a range of sciences from physics over chemistry, meteorology, engineering, toxicology, to epidemiology are necessary when tackling the still wide open issues in the research on particulate matter. Scientists with the diverse scientific background coming from all over Europe discussed the issue of "Particulate matter and health" in 5 interactive workshops. Each of which approached the conference topic "Similarities and differences in airborne particulate matter: Exposure and health effects over Europe" from a different perspective and resulted in answers to pressing (policy) questions.

The topics of the five workshops were:

1: Particle characterisation and characteristics
2: Sources of particulate matter
3: Modelling and (personal) exposure
4: Health effects - Epidemiology
5: Health effects - Toxicology

Figure 1 shows a flow chart on information and research areas necessary to assess the health impact of airborne particulate matter. It also illustrates how the topics fit into this overall scheme.

Two major issues were clearly stated by all participants of the workshop:

- There has been a tremendous increase in knowledge related to airborne particles and their effects on human health over the last decade: the complexity of PM is recognised and requires both new metrics and better understanding of source contributions for effective policy measures.
- Still, major knowledge gaps remain and it is seen that integrated approaches combining the different scientific areas covering environmental, socio-economic and medical

[1] Baseline Scenarios for the Clean Air for Europe (CAFE) Programme , Markus Amann, Imrich Bertok, Janusz Cofala, Frantisek Gyarfas, Chris Heyes, Zbigniew Klimont, Wolfgang Schöpp, Wilfried Winiwarter, Final Report to DG ENV, Feb. 2005

research in selected regions in Europe are a prerequisite to effectively tackle the uncertainties European wide.

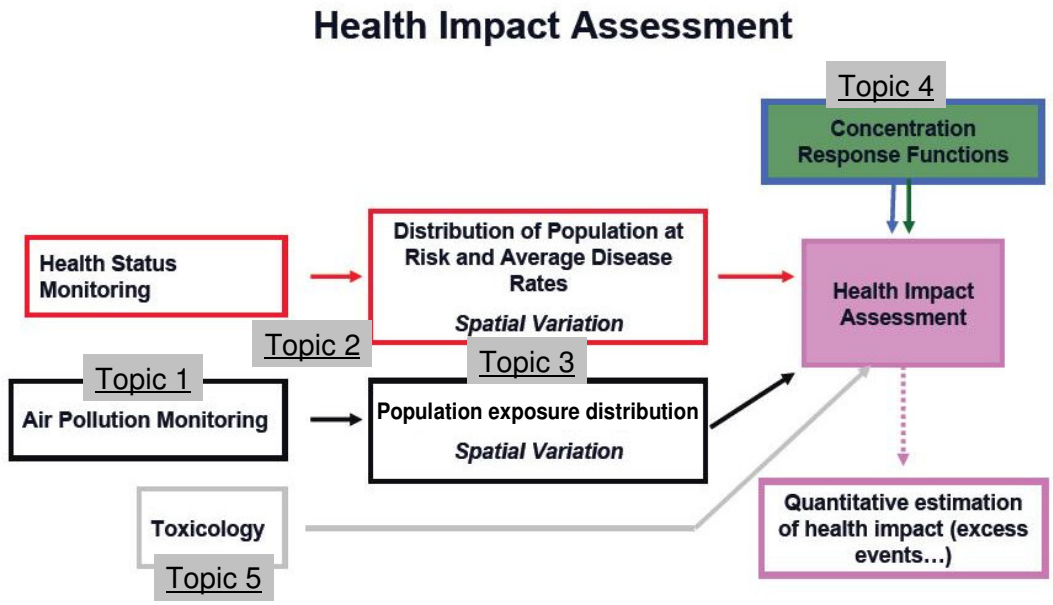

Figure 1: Scheme for health impact assessment [2]

Several major issues came up during the discussions in **workshop 1** on particle characteristics and characterisation. One block of recommendations is linked to monitoring. Specific needs were identified:

- Extension of the current monitoring network. Additional particle parameters should be measured and this with preference in urbanized areas.
- Improvement of PM mass measurement accuracy. Notably, the reference method EN12341 suffers from sampling artefacts and analytical bias.
- Standardisation of analytical methods for aerosol measurements that cannot be validated because standards do not exist (e.g. EC, particle number concentration).
- Better integration of epidemiology and toxicology, using for instance same health indicators (biomarkers of effect) with emphasize on oxidative stress.

The points listed above would be best addressed by setting up at least 3 aerosol (super) sites in urban areas located in different regions of Europe. These (super) sites would achieve a complete characterisation of the urban aerosol in relation with their health effect, and serve as platforms for instrument calibrations and intercomparisons.

[2] Analysis and design of local air quality measurements: Towards European Air Quality Health Effect Monitoring, T. Kuhlbusch, A. John, A. Hugo, A. Peters, S. Klot, J. Cyrys, H.-E. Wichmann, U. Quass, P. Bruckmann, Report to DG ENV, http://www.iuta.de/Verfahrenstechnik/Luftreinhaltung/euraqhem_final_report.pdf, April 2006.

Further important recommendations were

- Development of novel analytical capabilities related to aerosol-and-health e.g. PM oxidative stress potential, reactivity or the surface area of the particles' insoluble core.

Specific relevance to policy developments:

- Assessment of how pollutant emission abatement strategies affect PM characteristics.
- Guidance on the selection of new parameters to be measured for monitoring the health effects of PM.

The above recommendations are all linked to ambient air quality monitoring and its assessment while in **workshop 2** the main focus was on the sources of airborne particulates and their assessment. The importance of source apportionment in view of health effects and planning of abatement strategies was clearly stated. The following issues were identified to be of high importance for the future directions of source apportionment.

- A need for the development of a common methodology for certain questions/tasks is clearly seen, which shall be validated by comparison with secondary information and/or other methods.
- A possible new focus could be the combination of emission inventories, chemical transport models and source apportionment methods into an integrated approach. While each tool separately is not capable of answering all questions, in combination they could provide a more detailed insight to issues such as regional variability of contributions by traffic, wood burning, etc.
- The quantification of wood burning as a PM source is a concern for emission inventories. Source apportionment studies could help verifying or rejecting the current statistics, in order to determine whether the large differences reported across the EU are a fact or whether wood burning is simply not reported for some regions.
- One of the biggest challenges for source apportionment studies are secondary organic aerosols (SOA). Current knowledge on their formation processes and on the influence of natural or anthropogenic precursors is limited. Smog chamber experiments, modelling studies or the study of their polymerisation processes would provide an insight to this issue.

Linking source apportionment and health effect studies was identified to be of specific importance which should include the following points:

- Separate focus on the coarse and fine grain-size fractions, given that the health effects associated with these two fractions need to be differentiated (respiratory vs. cardiovascular).
- Extension to particle number concentrations, namely ultra fine particles.
- Short- and long-term health effect studies should be linked to source apportionment studies, thereby facilitating the identification of possibly harmful sources and particle properties.

Following recommendation was giving with specific regards to policy issues.

- Source apportionment studies shall be conducted for verification of the effects of the various European and local abatement efforts.

While ambient air and particle source apportionment studies are important tools linking particles and health a further major focus discussed in **workshop 3** was the linkage of measurements of (personal) exposure and how modelling can facilitate this linkage. The intense discussion of this topic enabled the identification of the following recommendations:

- There is a need to assess the uncertainty of existing models rather than to develop new models.
- Long-term exposure estimates need to be improved and developed, especially taking the indoor situation into account.
- Outdoor-indoor penetration of particles and their life time as well as indoor sources and their association with health effects require further investigations.
- Air quality models should be used to complement monitoring data allowing a better spatial distribution characterisation and hence enable improved exposure assessments.
- Exposure studies in Europe should take into account the different characteristics of climate zones, the specific behaviour of social groups, and regional habits.

Again, specific recommendations with regards to policy developments were identified to be:

- Assessment of transboundary transport of PM with advanced air quality models
- Increase number of PM parameters in air quality model outputs to include more health relevant parameters e.g. trace constituents, source contributions, ultra fines.
- Guideline values based on exposure rather than ambient concentrations are needed to improve public health.

The last two workshops were complementary to the first three. The first three were focussed mainly on the different aspects of exposure (air quality monitoring, source apportionment, indoor/outdoor, air quality modelling, personal exposure) while the focus of the last two was on health effects related to particle exposure. Epidemiology, its possibilities and limitations were discussed in **workshop 4**. The outcomes of this discussion are summarized in the statements below identifying future needs and possible directions.

- Physicochemical differences of particles need to be better defined and included into health effect models that include genetic and socio-economic differences.
- Development of high resolution spatial exposure models for the estimation of chronic, long-term particle exposure; studies in selected regions in Europe on long-term effects of air pollution with standardized procedures in both health and exposure assessment are needed. To appropriately investigate chronic effects, such studies must focus on early pathophysiological or functional markers of chronic diseases rather than on terminal outcomes.

- Inclusion of socio-economic and genetic differences in studies on exposure-response relationships between air pollution and pulmonary, cardiovascular or neurodegenerative diseases. The interrelation between socio-economic factors and the biologically relevant co-factors are poorly understood in different regions of Europe and need to be integrated in future air pollution research.
- Development of dosimetry models that can be used to refine the exposure-response function and for studying effects in secondary organs.
- Investigation of the consistency of concentration-dose-effect estimates for different sources, constituents, and European regions.

Major issues related to policy developments needs are seen to be:

- Abatement strategies need to be evaluated by integrated health effect studies concurrently with the time line of their integration.
- Integrated short-term health effect studies linking health effects and sources are needed to identify their potential hazards. This will allow for initiation of new effective measures.

The last **workshop 5** dealt with health effects mainly from the toxicological point of view. Clear concepts on how particle interact with human health were presented while major gaps were identified at the same time. Further needs of developments are seen for in the following aereas:

- Better integration of epidemiology and toxicology, using for instance same health indicators (biomarkers of effect).
- Conduct source related toxicological studies preferably using real world mixed samples of different regions of Europe.
- Long term exposure studies (that can also be used as toxicology-time series studies). Better animal models with the challenge for developing and using transgenic mouse models.
- Development of a test battery for oxidative stress that can ultimately be used to monitor the biological reactivity of air pollution in different regions of Europe.
- Development of tests to evaluate the effectiveness of control strategies e.g. for vehicle and wood combustion emissions.
- The role of the surface area of (the insoluble core of) PM has to be identified.
- The role of so-called non-toxic components (often also referred to as “natural”) in the total mixture of PM in view of health effects is still insufficiently studied. Can such particles interact to become cariers for toxic or allergic substances?
- Integration of air sampling in toxicological studies: Usage of PM sampling techniques that reduces sampling artefacts to a minimum such that it approaches the real-world situation of PM in the human airways.

Policy relevance was explicitly seen in the guidance on additional measures and abatement strategies from specific sources and in the explanation / increased confidence on biological plausibility and causal relationship by toxicology.

The need of collaboration and interdisciplinary approaches became obvious during the final discussion. Several urgent research needs were identified in specific research areas and a clear recommendation to conduct well organized concerted research studies in several regions in Europe comprising monitoring and research of air quality, exposure, health status, exposure-response functions, source specific toxicological studies as well as evaluation of abatement actions, was given by all members of COST 633 and participants of the workshop.

Conference Participants

COST 633 and the Conference

Background

COST Actions are to co-ordinate and promote research on specific topics in Europe. Othmar Preining and Helger Hauck, both of the Austrian Academy of Sciences, started to organize a new action on airborne particulate matter and health in 1999. Finally, this Action "COST Action 633 – Particulate Matter and Health" was approved by COST and started in 2002. The European wide interest in the topic can be seen in the participation of 20 EU countries in this action and more specifically in the topic.

Atmospheric particles or dust (particulate matter, PM) have been always considered a major component of air pollution. Epidemiological studies in recent years gave a strong hint on increased morbidity and mortality even at relatively low PM burdens.

The understanding of the corresponding causal chains of various parameters describing PM exposure and health effects is still incomplete. Many research projects on PM exposure and health effects due to PM have been initiated in the last years, mainly in the United States but also in Europe both on the European and the National level and other areas of the world. Consequently, ambient air quality standards for particulate matter are being established or revised in many countries. In fact the World Health Organization (WHO) just published its new Air Quality Guidelines and recommended AQG values for PM and other air pollutants[3]. Several workshops on research priorities within this field (EU, USEPA, HEI) indicated extensive needs for additional information.

In many European as well as other countries monitoring programs focus on PM and special parameters like carbon, acidity, semivolatile components, ultrafine particles and so on. The research goals of these studies are not always the same in detail, but generally particle properties are addressed with respect to effects on the environment and in particular on human health. Furthermore, epidemiological studies focused on different health endpoints and high risk groups were and are conducted. Since most of these studies are being done independently at present, an intensive exchange of information and experience between the groups working individually on either exposure or health effects would be of great benefit. A harmonisation of the available results and concerted planning of future activities is highly desirable.

[3] WHO Air Quality Guidelines Global Update 2005, http://www.euro.who.int/Document/E87950.pdf

Therefore, the COST 633 Action is to co-ordinate and promote research and activities on particulate matter and health effects in Europe.

The objectives of COST 633 Action are:

- to increase information on the particulate matter (PM) characteristics throughout Europe, describing the PM-system with respect to geographical and meteorological conditions, particle formation processes and their transport with special regard to the European aerosol situation (compared e. g. to the US).
- to increase the information on health effects of PM throughout Europe with special regard to geographical, seasonal and source related aspects.
- to improve the scientific basis for setting environmental standards in Europe and for defining cost-effective abatement measures to reduce particle and particle precursor emissions.

Three working groups are operating within the Action covering “Air quality measurements and instrumentation”, “Health related issues” and “Modelling – Source apportionment, dispersion and integrated assessment modelling”.

The conference

The conference summarized in this report is one major step of COST 633 to reach its objectives. The conference brought together scientists working in diverse fields (atmospheric PM system and measurements, epidemiology, toxicology and modelling) to gather in a multidisciplinary expert group.

Five interactive workshops were conducted summarizing the status quo on currently available information related to five topics (particle characterization; sources of particulate matter; modelling {personal} exposure; health effects - epidemiology and – toxicology) and whether this information already allows statements related to European similarities and differences on health effects caused by particulate matter.

The summaries and analyses of existing airborne particle datasets and health effect related information in view of similarities and differences within Europe were discussed extensively. This workshop facilitated a pan-European transdisciplinary approach

- combining information of particle composition, size, and morphology, (personal) exposure, epidemiology and toxicological effects of particles
- identifying short- and long-term future needs within the above research areas

The following chapters present the outcomes of the discussions along with recommendations on future needs to tackle the most urgent issues related to particulate matter and health.

Reports of the Break-out-Groups

Topic 1: Particle characterisation and characteristics

Jean-Phillipe Putaud, Axel Berner, Harry ten Brink, Roy Harrison

Questions

- **How important is it to know data uncertainties? What precision and/or accuracy is needed for health studies and model inputs and/or validation?**
- **What parameters (speciation, spatial resolution, time resolution, time span, size fractionation) are needed for health effect assessments and aerosol modelling?**
- **Is it possible to cluster regions showing strong similarities related to particle characteristics? Which properties of aerosol exhibit clear differences for different parts of Europe?**
- **What are the effects of meteorology on particle characteristics?**
- **Are there indications / evidence for differences in dose-response (effects) in the different regions in Europe, in time and space, due to heterogeneity of PM / differences in sources?**
- **What parameters (speciation, spatial resolution, time resolution, time span, size fractionation) are needed for health effect assessments and aerosol modelling? Does measurement equipment exist to provide these parameters?**
- **How important is it to know data uncertainties? What precision and/or accuracy is needed for health studies and model inputs and/or validation?**

Discussions and answers

How important is it to know data uncertainties? What precision and/or accuracy is needed for health studies and model inputs and/or validation?

The value of an experiment very much depends on the quality of the measurements. It was however impossible to reach a consensus on the level of uncertainties needed for aerosol health effect studies, perhaps because the level of accuracy required may vary according to the parameter studied. Should we be aiming for a maximum uncertainty of ± 10% or ±0.5 µg/m³ for PM mass and components, whereas guidelines for monitoring networks advise that uncertainty in PM measurements should be better than 25% only?. Let us not forget though that uncertainties in PM concentration measurements are generally far smaller than the difference between fixed site and personal exposure concentrations. The same probably applies to modeling: site representativeness is the most critical factor when comparing measured and modeled data, rather than measurement accuracy itself.
Sampling artefacts may affect PM mass concentration, chemical composition and microphysics. These artefacts are site dependent, and should be locally addressed, e.g. by looking at consistency among instruments based on different principles: chemical mass closure involving filters and TEOM, number size distribution-derived particle volume compared to gravimetric or

TEOM based mass concentration measurements, etc... At stations where just PM mass is measured, data quality may be difficult to assess.
For comparing aerosol characteristics at various locations, accuracy becomes essential. PM mass concentration measurements with the reference method are comparable up to a certain point (semi-volatile species may be lost, aerosol-bound water occurs at 50% RH). The use of correction factors to make on-line PM measurements comparable with the reference method includes further uncertainties. Quality assurance is inevitably incomplete when reference standards are unavailable. This is the case for e.g. number size distribution measurements and OC+ EC analyses. Analysing EC is more complicated than previously thought, because of matrix effects (e.g. inorganics can catalyse the EC oxidation in inert carrier gas). In some cases, the use of specifically defined procedures makes it possible to compare various sites, even if this procedure does not produce "true" values. Aerosol standards deposited on quartz fibre filter recently appeared on the market (NIST reference material RM8785). A TC reference value is assigned to this reference material, but no reference value could be given for OC and EC because the concentration values differed across the 2 best established thermo-optical analytical methods by 1.66 ± 0.23 (Klouda et al., 2005).

Generally speaking, there is a real need for method standardisation. Setting up at least 3 (super) sites in urban areas in 3 different regions of Europe for fully documenting the aerosol, and also to offer a platform for intercomparisons would be a solution to improve aerosol data quality in Europe.

This would not overlap with other on-going projects in Europe (e.g. EMEP, EUSAAR) which network non-urban sites and focus on climate change and impact on ecosystems rather than on aerosol health effects.

What parameters (speciation, spatial resolution, time resolution, time span, size fractionation) are needed for health effect assessments and aerosol modelling?

Epidemiologists consider that on top of a "pollutant dispersion factor" which is well reflected by PM mass concentrations, data on particle sources are more important than data on PM characteristics. Source apportionment may indeed differ in various countries because natural sources, vehicle fleets, emission by road erosion, road sanding material, and the relative importance of sources with incomplete combustion, may be different. For policy making, it may be useful to distinguish between sources that can be controlled, and sources that cannot be controlled (e,g, natural sources, long-range transport of pollutants.) Assessing the contribution of emissions occurring at the global, regional, down to street canyon level is important too. Source apportionment studies should also split the aerosol coarse and fine fractions. However, epidemiological studies have also provided insight into the significance of exposure to secondary particles, such as sulfate, compared with primary emissions, such as elemental and organic carbon (Schlesinger et al., 2006). PM chemical characterization can therefore be useful not only for source apportionment studies but also for health effect assessment.

Toxicology studies need much more detailed atmospheric particle characterisation (which may go beyond what is currently feasible) with a good time resolution to understand short term acute effects. The inhaled material biological reactivity seems to control the series of end points, but

due to the very small amounts of PM we inhale daily (10ths to a few mg), the health effect of PM can only be indirect. The parameters which could lead to PM acute health effects include the concentration of reactive organics producing O and N radicals (e.g. oxidised PAHs and NO_3-PAHs), the concentration of bio-available transition metals, and the red-ox potential of particles. Potentially important substances also include endotoxins (lipopolysaccharides coming from bacteria membrane fractionation). Endotoxins occur in both PM10 and PM2.5, and have a very strong inflammatory character. They are found more in coarse than in fine particles, and more during summer than winter. Resuspended dust may contain this bacteriological debris and thus have much stronger inflammatory effects than fresh mineral dust. The effect of small particles should not be forgotten either: there are microbiological components in the aerosol fine fraction too.

More than the bulk aerosol chemistry, characteristics of individual particles may be important, such as their surface area, once the liquid shell has been removed. Finally, not only the dose inhaled, but also where in the lungs this dose is deposited matters, which is linked to the particle size in the atmosphere and inside the respiratory tract (hygroscopic growth).

Modellers need data they can validate their calculations against: SO_4^{2-}, NO_3^-, and sea salt for instance are unambiguous well defined aerosol constituents. OC and EC are more problematic because they are instrumentally defined "species", of which sources, properties (hygroscopicity, density, …), formation pathways and kinetics (for secondary OC) are poorly known. Finally, modellers like measurements that are representative of the scale they look at. Remote sensing tools (LiDAR, sunphotometers) are nowadays available to retrieve vertical profiles of various aerosol characteristics and may be useful for validating certain models.

Regarding spatial resolution, it was stressed that personal exposure is the parameter that matters. Correlations between indoor and outdoor PM10 concentration have been observed and there are a few studies which combine exposure and ambient PM data (www.pamchar.org, www.ktl.fi/expolis/). Sulfate and nitrate show a good spatial co-variance: the differences between cities like Vienna, Munich, and Zurich for these secondary aerosols are not very important. Regarding traffic aerosols, one would certainly need a much better spatial resolution. The spatial covariance for trace elements is probably not good either.

The time resolution needed for aerosol measurements depends on the type of studies (e.g. epidemiology vs. personal exposure). From the practical point of view, daily averages for PM10 and PM2.5 are sufficient. Hourly concentrations may be relevant for very specific studies, e.g. when dealing with particle number concentrations and size distributions in relation with cardiovascular diseases.

The cut point at 2.5 µm aerodynamic diameter does not really split coarse and fine particles. There are always some coarse particles below the 2.5 µm cut point. Anyway, the coarse and accumulation modes of the atmospheric aerosol overlap to some degree, and cannot be rigorously separated by physical means

There is consensus to move forward from measuring just PM10 and PM2.5 mass concentrations. Indeed, the aerosol health effect community can currently only provide a list of possible and most likely particle characteristics to be of health relevance. This list should be elaborated and than used by the the aerosol monitoring community to collect aerosol data to confront health data with.

Is it possible to cluster regions showing strong similarities related to particle characteristics? Which properties of aerosol exhibit clear differences for different parts of Europe?

There are currently not enough data available to definitively answer these questions. However, PM mass concentration and PM components' mass concentrations are much more variable with time and space than PM chemical composition. From a previous work based on a limited number of sites (e.g. Putaud et al., 2004), it can be inferred that SO_4^{2-} accounts for 10 to 20% of PM10 mass for sites ranging from rural to street canyons. The contribution of SO_4^{2-} at remote sites might be up to 30%. NO_3^- accounts on average for 5 to 20% to PM10 mass, with a maximum contribution observed at near city and urban background sites. Its contribution is much more variable in time than that of SO_4^{2-}, partially due to the influence of temperature on NH_4NO_3 gas/particulate phase partitioning (see below). The recent 1 yr long EMEP OC/EC campaign (Yttri et al., in prep.) showed that the yearly mean contribution of OC to PM usually ranges from 15 to 3O%. Lowest contributions (<20%) were observed at sites located in NW Europe, i.e. Ireland, GB, NL and Belgium. EC accounted for 2-4 % of PM10, except at the marine site in Ireland (1%) or at an urban site in Belgium (5%). Larger contributions of EC have been observed at urban sites in other studies. These results are in line with the data compiled in the European aerosol phenomenology (Putaud et al., 2004), which also showed that the contribution of EC to PM10 may be larger than 10 % at kerbside sites. The most variable components are of course mineral dust and sea-spray, which are mainly present in the coarse aerosol fraction and therefore occur only at sites impacted by their specific sources.
Annual mean particle number concentrations range from 5000 to 50000 cm^{-3} from rural to street canyon sites at the locations included to the European aerosol phenomenology (Van Dingenen et al., 2004). Particle number concentration is very variable with time (even over 24 hrs) and space. The relationship between particle number and PM mass concentration is site-dependent and the correlation generally weak.

What are the effects of meteorology on particle characteristics?

As already mentioned, meteorology is the main factor controlling the aerosol concentration at a given site. Pollutants' dispersion indeed depends strongly on the height of the mixed boundary layer (vertical) and wind speed (horizontal). Precipitations also lead to a decrease in particle concentration by rainout or washout.
Temperature and relative humidity influence particle size and the gas - to - particulate phase partitioning of semi-volatile species such as NH_4NO_3 and part of the organic matter. The synoptic meteorological situation also controls the long-range transport of pollutants, which can have a large impact on measured PM concentration and composition at a given site.

Are there indications / evidence for differences in dose-response (effects) in the different regions in Europe, in time and space, due to heterogeneity of PM and / or differences in sources?

Evidence that the effects of a given concentration of PM10 are different in various places in Europe has been highlighted (e.g. APHEA project), but the reasons for this are not completely understood and might be multiple. Too little is still known about population exposure.

Regarding long-term effects, the only parameters epidemiologists can compare health data (mortality, morbidity, etc…) with are often just PM10 mass concentrations. This is clearly not sufficient.
Regarding short term effects, perhaps the aerosol characteristics we are currently able to measure are not relevant. Indeed, many minor compounds are not measured and we don't even know if they occur in atmospheric particles. Furthermore, many pollutants covary (see the role of meteorology), which make it difficult to highlight the impact of individual species.

What are the research needs & recommendations?

- Measuring more than just PM mass concentrations in monitoring networks.
- Improve PM mass measurement accuracy. Also the reference method EN12341 suffers from sampling artefacts and analytical bias.
- Standardise analytical methods for aerosol measurements that cannot be validated because standards do not exist (e.g. EC, particle number concentration)
- The points listed above would best be addressed by setting up at least 3 aerosol (super) sites in urban areas located in different regions of Europe. These (super) sites would achieve a complete characterisation of the urban aerosol in relation with their health effect, and serve as platforms for instrument calibrations and intercomparisons.
- Develop novel analytical capabilities to fulfil the requests of the aerosol-and-health community regarding e.g. PM oxidative stress, the surface area of the particles' insoluble core, etc…

Policy relevance

- Assessment of the effect of pollutant emission abatement strategies on PM characteristics
- Guidance on the selection of new parameters to be measured for monitoring the health effects of PM

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Topic 2: Sources of particulate matter

M. Viana, T. Kuhlbusch, M. Amann, T. Pakkanen

Questions

- **Can PM sources be clearly differentiated?**
- **What kind of information shall source apportionment deliver, in order to be useful for measurement design, atmospheric modelling or health studies?**
- **How can accuracy and representativeness of source apportionment results be tested?**
- **What kind of source apportionment methods are in use in the EU?**
- **To what extent can we extrapolate source apportionment results to other regions?**
- **Are there regional specific sources?**
- **How are natural sources currently reflected in source apportionment studies?**
- **Do similar sources of PM have the same health effects (endpoints, exposure-response functions) over Europe?**
- **Where should the focus of modelling and source apportionment studies related to health be (street/urban/regional scale; in/outdoor, hot-spot/background, industry/traffic)?**

Discussions and answers

Can PM sources be clearly differentiated?

Two main methods of source apportionment were differentiated: One based on multivariate statistics using internal covariance in data sets to obtain information on group of variables/ input data determining changes in the concentration; the other based on information of the sources such as source profiles for Chemical Mass Balance (CMB) and dispersion modelling based on emission inventories.

The differentiation of sources by the latter method is mainly dependent on the quality and completeness of the input data which are source profiles, emission inventories (including possible time activity profiles), and the quality/completeness in the determination of ambient compound specific PM concentrations. One of the major drawbacks of this approach is seen in its limitation to only see what you “put inside”. It can only attribute primary particle emission.

This is certainly different for the prior methods based on multivariate statistics. These methods only need time series of ambient compound specific PM concentrations. Factors are resolved from these data sets by using the internal correlation in the variance of the compounds. Ideally, factors resemble certain sources or source groups.

The major draw back of this method is seen in the clear differentiation into source groups. Several factors, such as meteorology and air transport, may influence this analysis significantly. It may be that the same source is found in two different factors (e.g. sea salt unaltered/altered by substitution of chloride by nitrate) or that the source is not singled out/separated (e.g. sea salt which is always transported some way inland and hence is statistically always linked to some contributions from sources on land).

Some especially problematic sources which currently are very difficult to identify on larger scale were named to be corrosion, abrasion, and resuspension. It is seen that these sources may contribute significantly at certain locations and/or during certain periods. These as well as some other diffuse sources are not yet well characterised.
It is concluded that there are difficulties in clearly separating sources and their contributions and that therefore currently at least two different methods should be applied to derive information on the uncertainty of the source apportionment results.

The development of a common methodology for certain questions/tasks is recommended, maybe based on a combination of methods (e.g. receptor and dispersion modelling)!
Verification of the results by comparison with other methods/information is strongly encouraged!

What kind of information shall source apportionment deliver, in order to be useful for measurement design, atmospheric modelling or health studies?

The main purpose of source apportionment on a regular basis was seen for health studies, independent of short- or long-term studies. The demands with regard to temporal and spatial resolution can be set more specific for certain kind of studies.
Regarding health studies linking sources to short term health effects the minimum requirements were set to a time series of about 3 years for 24 hours resolution at least resolving soil, salt, traffic, metals from anthropogenic sources and secondary inorganic ions. This should be done by measurements at least one site and coupled with spatial information by e.g. modelling.
The time resolution demand can be set much lower for chronic studies. The latter “only” needs average source contributions for a period of 1-2 years, but if possible with a very good spatial resolution down to 200 m.
For some short-term health effect studies with specific health endpoint it even may be advantageous to have access to source apportionment results on an hourly time resolution.
In all cases a linkage of sources to health effects was seen to be of paramount importance in order to identify the health relevant PM parameters as well as to set up effective abatement strategies. No clear recommendation on sources of specific health relevance could be given at this point beside those already discussed (combustion, metals, PAH).

It seems necessary to develop basic needs linking short-term time series studies and source apportionment studies. The need of an extension to long-term studies is also clearly seen.

How can accuracy and representativeness of source apportionment results be tested?

Accuracy is a problematic issue regarding all types of models. The main advantage of receptor modelling is related to the fact that it retrieves source contributions from measured ambient concentrations, while other types of models (e.g. transport or dispersion) proceed forward and results do not always match the measured ambient concentrations.
The calculation of uncertainties in source apportionment studies is seen as a pressing need by epidemiologists, as the absence of uncertainty data may induce a significant bias in the study of PM-derived health effects. It is thus suggested that source apportionment studies should provide uncertainty bars for their results. Detailed knowledge of the study area is considered a

useful tool to reduce uncertainty, given that it allows for the discrimination of potentially different sources with similar tracer elements. A first "feeling" on the certainty of source apportionment methods can be derived by conducting comparison studies between different methods on artificial and practical data sets. Anyhow, no golden standard is seen yet. Finally, the importance of obtaining independent datasets for validation is also highlighted as a means to minimise uncertainty and maximise representativeness of source apportionment results.

What kind of source apportionment methods are in use in the EU?

The most commonly methods in use in Europe are Principal Component Analysis (PCA), back-trajectory analysis, Lenschow approach, Positive Matrix Factorisation (PMF), Chemical Mass Balance (CMB), cluster analysis, isotopic mass balance, Constrained Physical Receptor Model (COPREM), Multi-linear Engine (ME) and UNMIX.
Methods discussed during the session were Chemical Mass Balance (CMB) and Constrained Physical Receptor Model (COPREM). CMB is based on the statistical comparison of the chemical profile of known sources with that of ambient particulate matter. The main disadvantage of this method is that chemical source profiles need to be determined at each study site in order to increase the accuracy of source identification. COPREM, on the other hand, requires a priori knowledge of the number of relevant sources and their approximate composition, and allows specification of some elements of the source profile matrix while leaving others unspecified. As regards input data for receptor models, PM chemical composition is generally used although alternatives such as particle size distribution data have also been used in the US and in Europe, with satisfactory results.
Given the large variety of models in use in EU, it seems evident that the major challenge for source apportionment studies is to ensure comparability between the results. A possible way to achieve comparability is by performing round-robin exercises, such as those which have been carried out in the US (Hopke et al., 2005) and are currently underway in EU.

To what extent can we extrapolate source apportionment results to other regions?

The output of receptor models is generally site-dependent, given that it is based on the variations of the levels of chemical species measured at a specific study site. Any integrative exercise for the entire EU would need to be extremely sophisticated, and current knowledge lacks enough data on factors such as regional variability of components or the effects of city-specific factors. Thus, it would currently be not advisable to try to obtain single EU-wide contributions for specific sources (e.g., traffic contributes with X% of PM10 in EU).

Are there regionally specific sources?

Chemical profiles and contributions of PM sources show a considerable regional variability across the EU. For instance, mineral matter in Mediterranean countries is originated from African dust contributions and soil dust re-suspension, and enhanced by low precipitation rates. Coal combustion for household heating is a source of PM in Poland, while it is mostly linked to industrial activities in the rest of the EU. Traffic fleets (both passenger cars and trucks) vary from country to country, and consequently traffic emissions are expected to show different chemical profiles. Likewise, the composition of road dust depends on the materials used in the pavement,

which present a large regional variability in Scandinavia. The use of different materials for road construction not only affects particle composition but also size distribution, depending on the hardness of the material. One other example of regionally specific sources is related to biogenic emissions, which vary widely in composition as a function of local vegetation (e.g. biogenic emissions originated in the tundra or in a Mediterranean forest).

How are natural sources currently reflected in source apportionment studies?

Natural sources are discriminated by most of the source apportionment methods applied in Europe. Emission inventories, on the other hand, do not generally include natural sources because countries are not obliged to report on this issue. However, an effort is currently being made to include natural sources in emission inventories from some of the EU countries. Thus, in the future emission inventories of natural sources will also be available. As stated above, these sources show a large regional variability.

Future research directions

A number of recommendations were obtained regarding future research directions:

- Source apportionment studies should focus separately on the coarse and fine grain-size fractions, given that the effects on health of these two fractions are clearly differentiated (respiratory vs. cardiovascular).
- Source apportionment should be extended to particle number concentrations
- A need for the development of a common methodology for certain questions/tasks is clearly seen, which shall be validated by comparison with secondary information and/or other methods.
- One of the biggest challenges for source apportionment studies are secondary organic aerosols (SOA). Current knowledge on their formation processes, or on the influence of natural or anthropogenically emitted precursors, is limited. Smog chamber experiments, modelling studies or the study of their polymerisation processes would provide an insight to this issue.
- The quantification of wood burning as a PM source is a concern for emission inventories. Source apportionment studies could help verify or reject the current statistics, in order to determine whether the large differences reported across EU are a fact or whether wood burning in some regions is simply not reported.
- A possible new focus would be the use of emission inventories and source apportionment methods in combination. Whereas both tools are not capable of answering all the questions separately, in combination they could provide a more detailed insight to issues such as regional variability of traffic fleets or wood burning.
- Short- and long-term health studies should be linked to source apportionment studies, thereby facilitating the identification of possibly harmful sources and particle properties.

- Source apportionment studies shall be conducted for verification of the effects of the various European abatement efforts.
- Similar studies should also be conducted for the evaluation of local measures of PM reduction.

References

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Topic 3: Modelling and (personal) exposure

Ana Isabel Miranda, Wilfried Winiwarter, Carlos Borrego, Matthias Ketzel

Questions

- **Can spatial and temporal variance of elements and source groups be modelled?**
- **How can (personal) exposure be assessed?**
- **Does ambient air quality monitoring reflect the exposure of the population?**
- **How do differences in personal activities and habits influence the exposure?**

Discussions and answers

General comments

Both key speakers gave an overview of the type and characteristics of models, which can be consulted in the enclosed summaries. Answers given below might depend on the type of model and the type of application.

There are the following main applications in exposure modelling:

1. For epidemiological and time series studies it is often desired to model the exposure at individual or address level and in high time resolution (e.g. days or even hours).
2. For risk assessment and risk management applications the exposure in a more aggregated way using averaged population exposure has to be assessed. Here local and short term variations are less crucial to be modelled exactly.

It is important to discuss in the modelling and health communities together what models are needed and have therefore to be developed or improved at the European level.
The models can have different level of sophistication and aggregation with respect to the following dimensions:

- time ==> averaging time
- space ==> modelled µ-environments
- person ==> groups size and population classes
- component and sources (PM10, PM1, ultrafine particles, gas tracers etc.)

Can spatial and temporal variance of elements and source groups be modelled?

There is a need to assess the uncertainty of such models. Studying uncertainty is not a trivial task, and also results derived are not readily scientifically appreciated as specta-cular progress. It is more often considered a tedious duty, as no ready-made concepts are available to directly use its results. Such concepts need to be developed, however. Models will need to be evaluated against their respective purpose, using their respective uncertainties. The key aim is to assess the need of the users, or in other words to test whether a model is fit for its purpose. As the questions directed towards models may differ strongly depending on model type and application; therefore no simple answer can be given once and for all.

Thus no new models need to be developed (models predicting transport and transformation of compounds in the atmosphere are available). Instead, methods need to be devised which are able to forward input and model uncertainty to the modelled outputs. While some approaches have been tested, a scheme is needed that allows to under-stand whether model results are "fit" for a respective user. Such a scheme needs an adaptation of error propagation through a variety of coupled/integrated models. Uncertainty of model input, as well as implicit model simplifications as a result of modelling assumptions need to be assessed and their consequences to the outputs tested or assessed during model validation. The respective results made available for a certain use allows understanding if answers to specific user questions can or can not be supplied reliably.

The participants agree that highly sophisticated modelling approaches are available, which allow to assess air quality at high spatial and temporal resolution also for several source groups or even individual sources. It is the quality of the results that need to be assessed. Even street canyon models are available and deliver plausible patterns.

How can (personal) exposure be assessed?

While assessing individual exposure by monitoring still may be cumbersome (personal monitors with active sampling devices are heavy and difficult to carry around) it can be fast at this level of technology development. Small personal sampling devices are conceivable, even if the number of such devices will not easily become sufficiently large to account for a representative sample of the population. Moreover an exposure-monitoring programme is normally quite an expensive and labour-intensive process. Developments are needed to facilitate this kind of personal monitoring.

Exposure modelling may be an even more promising approach to exposure assessment, using ambient air concentration modelling (or interpolation of measurements) and the respective spatial and temporal distribution, combined with population statistics, to get a statistical assessment. Still information may become more personalized, using traffic models' information on optimized daily trips (work, school, shopping) or even tracking individuals' mobile phone positions. Technology for exposure assessment is available; it is a matter of costs, data handling, and data privacy regulations to access such information. Anyway, a statistical assessment may prove sufficient for many purposes.

Still any exposure assessment is limited to a short-term activity. Long-term exposure estimates suffer from a variety of difficulties: exposure model results or ambient measurement interpolations are available only for short term; differentiation between indoor, outdoor and street level (e.g., in-car) exposure becomes highly problematic; and a predisposition to smoking exposure (either active smoking or passive exposure) is highly inaccurate to estimate. Questionnaires to assess smoking history on an individual basis have proved to be highly unreliable.

Issues to discuss/clarify:

Ultrafine particles exposure is clearly related to traffic emissions, however there is a need to better understand ultrafine particles toxicity and health effects. Open questions are:

1. Dependency on the vehicle type (e.g. diesel vs gasoline ultrafine emissions are different and have different effects regarding exposure)
2. Relation between noise and ultrafine particles

3. Ultrafine particles inside a vehicle (concentration can be very different from outdoors values, depending on the vehicle type, traffic situation and even weather condition, and this is not measured by a monitoring network).

Does ambient air quality monitoring reflect the exposure of the population?

Systems to monitor air quality do not always adequately assess the population exposure to air pollution. The spatial variation in the pollution concentration and the differences between the areas covered by the monitoring and the areas where the population is located create problems in using air quality data generated by routine monitoring networks. Air quality models are used to complement existing monitoring networks.
Furthermore, according to the PM sources the answer can vary a lot:

- PM2.5 and secondary particles present a good relation between air quality values and exposure since the spatial gradients within an urban area are moderate
- For area sources as ambient combustion particles from heating the relation might be acceptable
- For emissions from very local sources as ultrafine particles and road dust resuspension the location of the monitoring station is crucial and several stations are needed to reflect the spatial variation of the exposure.

Indoor environments are (in the absence of strong indoor sources) assumed to be much less exposed to PM than outdoor air, due to the filtering effect of the building. Still data show that ambient monitoring reflects also the temporal variation that is seen indoors, at least for PM10. This supports the use of ambient data as a proxy for the overall exposure.
Very little information is however available on the long-term validity of such an assumption. Moreover, it is clear that the fraction of time people spend indoors changes between seasons, between places (especially northern vs. southern Europe) and also depends on work habits that may change with time (mechanisation of outdoor labour, but also leisure activities). Finally, influence of the outdoor environment to indoor air quality, or the indoor particle formation will strongly depend on age and national habits, partly again triggered by environmental conditions (e.g. need for heat insulation).
Together with the need to look into other metrics than PM10 alone (PM1, number concentrations, non-soluble material etc.), this indicates the importance of better understanding the extent people are exposed to particulate matter – with exposure as the potential to inhale and retain a certain dose of material. For assessing the influence on long-term health, despite of all the problems involved, there will be no way to circumvent specific exposure assessment.

Issues to discuss/clarify:

- PM outdoor-indoor penetration and the life time indoor is very dependant on particles size, composition and physical properties (e.g. volatility, hygroscopicity).
- Up to date, only some initial “baby-steps” have been done in separating indoor and outdoor particles generation.
- How can the exposure to indoor sources (cooking, cleaning, candle lights, smoking) be assessed and what are the health implications of those sources.

Experimental and modelling approaches should be used and we should take profit of their capabilities

How do differences in personal activities and habits influence the exposure?

They influence a lot. Indoor and outdoor activities will be responsible for quite different exposure levels, according to the ambient air concentration and to the indoor characteristics.
Different age groups have different behaviours and spend most of their time in different indoor environments.
Exposure along Europe can be quite different according to population habits. People from a north European city or from a south European city are differently exposed to ambient air concentrations.

What are the research needs & recommendation?

Several tasks were identified of which the following are seen as the most important once:

- There is a need to assess the uncertainty of existing models rather than development of new models.
- Long-term exposure estimates still need to be improved and developed.
- PM outdoor-indoor penetration and the life time indoors and indoor sources including health effects need further investigation.
- Air quality models should be used to complement monitoring data allowing a better spatial distribution characterisation and hence enable improved exposure assessments.
- Exposure studies along Europe should take into account the different characteristics of climate zones, the specific behaviour of social groups, and national habits.

Policy relevance

- Assessment of transboundary transport of PM with advanced air quality models.
- Increase number of PM parameters in air quality model outputs also to include more health relevant particle parameters e.g trace constituents, source contributions, ultrafines.
- Guideline values based on exposure instead of ambient concentrations are needed to improve public health.

Topic 4: Health effects – Epidemiology

Michael Riediker, Raimo Salonen, Nino Künzli, Joseph Cyrys,

Questions

Short-term health effects

- **Do particle characteristics explain heterogeneities of health effects in European populations?**
- **Are dosimetry-based population exposure models useful to assess the health impact to various risk groups?**
- **Can we demonstrate the effectiveness of policies to reduce PM concentrations?**
- **What are urgent research needs for the study of short-term health outcomes?**

Long-term health effects

- **Do particle characteristics explain heterogeneities of health effects in European populations?**
- **What is the role of population-characteristics (e.g. genetic differences and socio-economic factors)?**
- **Are dosimetry-based population exposure models useful to assess the health impact to various risk groups?**
- **Are there "less harmful" components?**
- **What are the research needs & recommendation?**

Discussions and answers

Short-term health effects

Do particle characteristics explain heterogeneities of health effects in European populations?

Epidemiological time-series studies on the association of daily variations in ambient air mass concentration of thoracic particles (PM10), fine particles (PM2.5) or black smoke with cardiovascular or respiratory mortality and morbidity have shown regional heteroge-neities as well as source-related heterogeneities in concentration-response relationships. Particulate matter originating from local combustion sources (e.g., automotive traffic, resi-dential heating with coal and wood, heavy metal industry) has had more consistent and stronger relationships with both respiratory and cardiovascular outcomes than particulate matter from other sources. However, there is currently not enough scientific evidence to declare any source or chemical composition as "non-toxic", because even sea salt and soil-derived particles from desert may be involved in adverse health effects in ubanized areas via interaction with local anthropogenic particles.
It is possible that the so far poorly defined physicochemical differences in particulate mixture (including those related to atmospheric photochemical activity) contribute to the observed

regional heterogeneities (South vs. North and East vs. West) in PM10-associated daily mortality and hospital admissions in Europe, but there may well be other reasons for these heterogeneities such as ambient temperature (despite statistical approaches to control for confounding) and differences in personal exposure patterns due to different building ventilation practices and different time activities of the populations. Moreover, in study periods preceding EU harmonization the siting of monitoring stations may have systematic differences between cities / countries. The role of population characteristics (e.g. genetic and socio-economic differences) in regional heterogeneities of short-term exposure-response relationships is not known, but some other factors like dietary intake of antioxidants or use of such supplements could well have an impact. However, it is difficult to collect this kind of individual information due to ethical and practical reasons, because it is much more detailed than what is available from routine population-based registers on mortality and hospital admissions. In general, the exposure-response relationships for the common particulate indices and a whole variety of health outcomes in short-term epidemiological studies have been linear with no obvious threshold even at concentration ranges extending to very low ambient levels.

Source-related heterogeneities can be observed for cardiovascular or respiratory mortality and morbidity. However, there is currently not enough scientific evidence to declare any source or chemical composition as “non-toxic”.

Are dosimetry-based population exposure models useful to assess the health im-pact to various risk groups?

Current exposure models do not seem very helpful for improving the exposure assessment in short-term epidemiological studies on ambient air particulate matter, but future models validated with ambient air monitoring data may prove more useful. However, it might be interesting to perform an exercise, in which a previously conducted epidemiological study with actually measured ambient particulate concentrations and health outcomes would be compared with modelled higher resolution exposures and modelled health outcomes for the same study population and period. Lung dosimetry and modelling of particulate clearance from the lower respiratory tract may help in explaining some of the differences in ex-posure-response relationships between e.g. children / aged cardiorespiratory patients and healthy adult subjects, but their use does not increase the validity of the analysis, because each subject is her / his own control in time-series studies.

Dosimetry is **currently** of limited use for short-term effects studies.

Can we demonstrate the effectiveness of policies to reduce PM concentrations?

There are three well-described studies, in which a local policy or other event has simultaneously caused a dramatic decrease in particulate levels and adverse health impacts:

- a ban of coal sales for domestic heating in Dublin was followed by a profound decrease in ambient air black smoke concentration and respiratory and cardiovascular mortality (Clancy et al. Lancet 360:1210-1214, 2002 [PMID: 12401247])

- an SO2 reduction policy in Hongkong lead to a decrease in PM?-associated mortality (Hedley et al. Lancet 360:1646-52, 2002 [PMID: 12457788])
- a one-year strike in a poorly controlled steel mill in Utah Valley was accompanied by a profound decrease in ambient air PM10 concentration and respiratory hospital admissions of children (Pope et al. Arch Environ Health 47:211-7, 1992 [PMID: 1596104])

The effectiveness of an intervention can be best shown in epidemiological time-series studies, if it causes an abrupt improvement in ambient air quality.

Intervention studies demonstrate the effectiveness of policies to reduce particles.

What are urgent research needs for the study of short-term health outcomes?

The most urgent research need with regard to acute health outcomes concerns exposure-response relationships of source- and chemical composition-specific particulate exposures among susceptible population groups. This kind of new evidence should be produced in highly integrated multicentre studies using harmonized methodologies for: (1) characterization of the sources and chemical composition of the complex particulate mixture, (2) measurement of the source-specific personal particulate exposures and epidemiological health outcomes, and (3) testing the toxic activities of collected particulate samples in cell and animal studies. Only this kind of elaborate study setups can produce strong evidence on the causality of certain particulate sources and chemical compositions in adverse health outcomes as well as on the biological plausibility of the epidemiological findings. The regulators need a reliable answer to the question, which sources produce the most harmful and harmless ambient air particles in order to be able to prioritize their resources and to develop the best management strategies for their control. Moreover, the engineers and technological industry need to know which constituents must be reduced with future technologies in the particulate emission sources.

Long-term health effects

Do particle characteristics explain heterogeneities of health effects in European populations?

Few European studies investigated so far long-term health effects of exposure to ambient particles. The existing data allows the conclusion that chronic, long-term exposure to particles causes health effects that go beyond those expected for repeated short-term exposures. However, too few studies and data exist to draw conclusions about heterogeneities of long-term health effects in European populations and the relation of such heterogeneities to sources or particle characteristics. This gap of knowledge exists on a European as well as on a world-wide scale.

Studies require sufficient exposure gradients to detect effects. Central monitoring sites are a useful tool to examine acute health effects in time-series studies. This is due to the fact that the temporal sequences of air pollutants at different sites in a given region are usually well-correlated even if the absolute levels of local exposures are different. This allows the study of acute health effects based on relative exposure gradients even if the “true” levels of personal exposure remain undefined. In contrast, long-term effect studies face a major challenge: They have to estimate the level of personal cumulative long-term exposure. Comparing cities with

different background exposure levels is not a satisfying solution since city background levels ignore the often substantial spatial differences in pollution concentrations within cities. The development of high-resolution regional and local expo-sure scenarios that can be combined with subject information such as home and work-place addresses was proposed as the most promising strategy. Such exposure models need to be developed and validated for different regions in Europe so that differences related to climate, co-pollutants and population characteristics can be examined. Ideally, these exposure models will include source contributions and particle characteristics.

Particles cause long term effects. However, we do not have the necessary information or sufficient studies to make a statement about heterogeneities.

What is the role of population-characteristics (e.g. genetic differences and socio-economic factors)?

Socioeconomic factors are proposed to affect the dose-response relationship between air pollutants and health by two different ways: They can affect the exposure, and they can affect the susceptibility of the individuals to the pollutants. For example low socioeconomic household have a higher exposure to traffic pollutants since they may be more likely to reside near large and busy streets. Lifestyle factors are suspected modifiers of susceptibility such as smoking, sports or dieting. Moreover, underlying diseases such as diabetes and other states of chronic inflammation may modify the effects of air pollution. Lifestyle factors and co-morbidities are influenced by the socioeconomic status.
Some polymorphisms that modify the response to oxidative stress and/or systemic inflammation are established modifiers in acute effect studies. However, we do not sufficiently understand how this translates into the chronic risks, and other genes and pathways are expected to play a complementary role in the development of chronic diseases. Susceptibility (ageing, diabetes, obesity) as well as socio-economic differences are increasing across Europe. These differences are expected to influence both exposure and response-functions. There are many questions surrounding socioeconomic variables and the under-lying co-determinants of health in Europe. Thus, it is not known how susceptibility factors affect the association between air pollution and chronic health effects in Europe.

Socio-economic factors influence exposure and susceptibility to air pollutants in complex ways.

Are dosimetry-based population exposure models useful to assess the health impact to various risk groups?

For establishing the association between sources or components and long-term health effects, more details about the long term exposure are necessary. Dosimetry can improve these models by taking into account that deposition in the lungs. This deposition depends not only on particle size but also on subject characteristics (age, height, exercise etc.). Improved understanding of dosimetry of various co-pollutants may enhance knowledge about the relevant toxic components and sources. This may also be helpful for the planning of policies, since this information can be used to compare different possibilities / policies (especially with regard to components and sources).

Research in this field has just started. For example, how much do we need to know about mechanisms to know what we need to know about dosimetry? How should we combine dosimetry with information about size, chemical composition and shape for particles originating from different sources?
One possible way to include dosimetry could be to calculate dosimetry based on energy consumption of the individuals can be helpful - but this information is difficult to obtain. Generalizations for deposition across population groups (e.g. young people) may be difficult and can lead to misinterpretations. Even less is known about dosimetry for the study of chronic effects in secondary organs, for which the relevance of ultrafine particles may be higher. However, risk assessors may borrow very rich and exhaustive dosimetry information from other fields of research such as radiation and health where particle dosimetry plays an important role.

As in case of acute effect studies, dosimetry studies per se cannot inform about the long-term effects of air pollution on chronic diseases. This requires epidemiological approaches. However, dosimetry models can improve the adjustment of epidemiologically derived exposure-response functions for subgroups with different dosimetric characteristics.

Are there “less harmful” components?

(This addresses the lower-key question: “Pan-European differences: If there is toxicological evidence that some parts of sea salt water and/or crustal material are far less harmful than other parts of PM and this can be determined in air quality networks, can epidemiology proved new air concentration-response relationships be used to set a new standard that does not include these fractions?”)
Several research studies investigated whether specific sources and components of particles were more harmful than others. Traffic particles, for example, are proposed to be more harmful. This raises the question, whether there were also “harmless” sources or components. Toxicological studies propose that pure crustal materials and salts may be less harmful. However there are very few epidemiological studies on the impact of crustal material on mortality: from 8 studies, 7 were valid and only 3 did not find effect. This directs to the biological possibility that the “non-toxic” components facilitate, permit, or interact with the toxic effect mechanisms. Furthermore, these particles may have other, dangerous particles attached to them.
There is an additional problem to the idea to subtract salt and crustal material: The relative risk used for all the models include concentration - response relationships that had been calculated using a particle mass number that included these components. The mortality and morbidity effects of the remaining particles would thus be higher. At the end, it is likely that little would change for the total risk and the policies that are based on these total risk estimates. A final question that was addressed was the idea to subtract “natural components” without taking into account their toxicity. This was considered a very suspicious approach for several reasons. E.g. Secondary organic aerosols are in part a product from biological VOC and anthropogenic pollutants. Biological components are also known to boost the response to air pollutants. A proposition to subtract “natural components” would be in disagreement with current scientific knowledge.

There is insufficient data to draw any conclusion regarding “less harmful” particle components for long-term effects due to long-term exposure and it is too early to propose regulations. There is no scientific evidence to allow regulators to subtract any portion from the ambient PM concentrations to determine compliance with standards.

What are the research needs & recommendation?

- Physicochemical differences of particles need to be better defined and included into health effect models that include genetic and socio-economic differences.
- Development of high resolution spatial exposure models for the estimation of the chronic, long-term particle exposure; and Europe-wide studies on the long-term effects of air pollution with standardized procedures in both health and exposure assessments are needed. To appropriately investigate chronic effects, such studies must focus on early anatomical or functional markers of chronic diseases rather than on terminal outcomes.
- Future studies need to include socioeconomic and genetic differences when they study the exposure-response relationship between air pollution and pulmonary, cardiovascular or neurodegenerative diseases. The interrelation between socio-economic factors and the biologically relevant co-factors are poorly understood in Europe and need to be integrated in future air pollution research.
- Develop dosimetry models that can be used to refine the exposure-response function and for studying effects in secondary organs.
- Investigate the consistency of concentration-dose-effect estimates over time and for different European regions for different sources and constituents.

Policy relevance

- Integrated short term health studies linking health effects and sources are needed to identify their potential harzards. This will allow for initiation of new effective measures.
- Abatement strategies need to be evaluated by integrated health effect studies concurrently with the time line of their implementation.

Topic 5: Health effects - Toxicology

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Questions

- **Do toxicological studies provide plausibility and causal proof of epidemiology-based associations between health effects and PM?**
- **Which particle characteristics are important in the toxicological view of health effects?**
    A. *Which parameters shall be monitored in toxicological studies?*
    B. *Is there source specific toxicity?*
    C. *What is needed to relate sources to health effects?*
    D. *What do we know about the contribution of natural sources to the observed health effects?*
- **Can toxicology provide insight in alternative indicators for air quality measurements?**
- **What are the mechanisms and modes of actions of adverse health outcomes that can be directly related to outcomes observed in epidemiological studies?**
- **Can results from toxicological studies induce new epidemiological studies?**
- **Why do toxicological studies show much larger potency differences than epidemiological studies?**
- **Why don't humans develop tolerance to PM exposure?**
- **How can toxicological data be used for risk assessment of PM and for regulation purposes?**

Discussions and answers

Do toxicological studies provide plausibility and causal proof of epidemiology-based associations between health effects and PM?

The wealth of recent toxicological studies provide sufficient plausibility to explain the modes of action and underlying mechanism of health effects associated with ambient PM demonstrated by the huge base of epidemiologic studies. However, there was little or at least conflicting evidence that the toxic effects also occur at lower, closer to ambient levels of PM. In addition, toxicology has shown that at on equal mass basis, PM from different places or sampled at different seasons have different toxic properties. Toxicologists have still the obligations to work more on biological plausibility. Even though there still are admitted gaps of understanding the general principle of induction of the viscous circle of oxidative stress and inflammation provides a reasonable causality of almost all respiratory and cardiovascular diseases associated with PM exposure. In this respect PM effects have not yet been compared with effects with the most potent oxidant in ambient air, ozone. Toxicology provides only very little information on the adverse health effects of the total air pollution mixture which included gases such as ozone and nitrogen dioxide. The use of particles in experiments does not easily mimic human exposure due to its complexity and whatever toxicologist do, it remains virtually impossible to mimic reality.

There is a need for more integrated and focused research, trying to get real ambient PM into animals and come to the point that you really get an effect, eventually starting with high PM concentrations.

Which particle characteristics are important in the toxicological view of health effects?

Current scientific knowledge is sufficient to determine mass-concentration-based parameters like PM10 and PM2.5; however, toxicology has shown that this metric does not correlate well with toxic effects of PM at various locations or in various seasons of the year. From the toxicological point of view the chemical composition is an important denominator and three groups appear to be important:

- Transition metals because of their redox capacity and their ability to form oxygen and/or nitrogen radical species altering the oxidant / antioxidant balance, protease / antiprotease balance, and through their metabolization by xenobiotic metabolized enzymes(XME).
- Reactive organic compounds because of their redox capacity and their ability to form oxygen and/or nitrogen radical species altering the oxidant / antioxidant balance, protease / antiprotease balance etc.
- Particle surface area of the insoluble core of particles because this surface provides the substrate for the interactions of particles with biological fluids, membranes and cells.

In addition, particle surface area and number concentration will affect the toxicity of the total PM mixture. Apart from a focus on the chemical and physical particle characteristics, other measures independent of these particle properties are relevant. In this respect, it has been demonstrated that the biological reactivity is essential for understanding very important aspects of particle toxicity such as the interaction of the particles with complementary sets of biological

systems like biomolecules, nucleic acids, proteins, membranous receptors, cells etc. redox reactions, adduct formation and the disturbance of homeostatic balance.
Furthermore, characteristics representing ageing processes are considered to be important. Such processes are based on the chemistry of the containing compounds within the particle coagulating with other particles and reacting with surrounding gases under the thermodynamic conditions of the ambient aerosol.

a. Which parameters shall be monitored in toxicological studies?

- Continuations of mass concentrations PM10 and PM2.5
- size fractions and distribution depending on the various moments of the diameter of the particles (moment 0 = number; moment 1 = length; moment 2 = surface area; moment 3 = volume, mass)
- Transition metals and reactive organic compounds
- Particle surface area of the insoluble core of particles; based on recent studies the active surface area (Fuchs surface area) seems to be of less importance for toxicological interactions with biological systems.
- Solid versus liquid particle fractions (water and lipid)
- Bioavailable compounds versus biopersistent compounds in the remaining core of particles
- Biological reactivity of particles, i.e. the interaction of the particles with complementary sets of biological systems like biomolecules, nucleic acids, proteins, membranous receptors, cells etc. leading to redox reactions, and disturbance of the homeostatic balance
- Parameters determining particle penetration into cells involving active processes like endocytic incorporations by vesicles (phagosome, caveolae, clathrin coated pits, etc.) versus seemingly passive processes like diffusion.

b. Is there source specific toxicity?

Yes, there is evidence for source specific particle toxicity. The currently dominated discussion relates to combustion type particles originating from various sources:

- Traffic (gasoline and diesel engine exhaust, lubrication oil, tire and brake wear dust)
- Fossile fuel burning (power plants, domestic heating)
- Industrial (welding fumes)
- Wood burning

Besides that there is evidence that mineral dusts re-dispersed from e.g. road surfaces express different toxic potentials.
Indoor PM might have to be included, but it is recognised that we need to do more source oriented toxicological studies. Indoor is difficult due to limited amounts.

c. What is needed to relate sources to health effects?

Study designs need to focus on different regions with contrast in sources of emissions, with good chemical and physical characterization of PM and other components. In particular inhalation studies at locations dominated by one source will contribute to the understanding of source related health effects. Laboratory studies using source specific PM, such as diesel engine exhaust or wood smoke will be useful to rule out contributions of other sources in the PM mixture and at the same time to include the gaseous components.

d. What do we know about the contribution of natural sources to the observed health effects?

We know that sodium chloride as major component of sea salt is relatively harmless at ambient levels. However, we currently do not know to what extend sea salt or any potential harmless constituents contribute to the total PM effect. All other components in PM, whether from anthropogenic or natural origin, are likely to contribute to some extend to health effects. Natural origin does not equal harmless. In fact there are cases were combinations of natural and combustion particles interact to produce stronger effects, a striking example is the adjuvant effect of inorganic urban dust and allergenic pollens. It seems logical that there is a focus to reduce PM from antropogenic origin.

Can toxicology provide insight in alternative indicators for air quality measurements?

Yes, most biomarkers such as cytokines and chemokines in bronchoalveolar lavage C-Reactive Protein and other acute phase proteins and/or molecules (e.g. CC16) being used in epidemiological studies result from clinical and / or toxicological studies. Markers for oxidative potential, free radical activity and particle number can serve as additional indicators. In the end, we still need to know what sources contribute to these effects.

What are the mechanisms and modes of actions of adverse health outcomes that can be directly related to outcomes observed in epidemiological studies?

There are a few examples where rodents exposed to dirty air in a metropolitan area expressed more toxic effects than animals housed in clean environments. Technologies are available to do inhalation studies in rodents or humans at the same time and locations as epidemiological studies, but they may have major logistic problems.
The general principle of induction of the viscous circle of oxidative stress and inflammation provides a reasonable causality of almost all respiratory and cardiovascular diseases associated with PM exposure. In addition, PM exposure is involved in exacerbation of allergic reactions. Furthermore, there is toxicological evidence that PM exposure can induce artheromatous plaque rupture which is the initial step of thrombogenesis stroke and myocardial infarction. There is a lack of good robust animal models that mimic the human population at risk.

Can results from toxicological studies induce new epidemiological studies?

Yes, certain measures in animal toxicological or human clinical studies can be transferred to an epidemiological study, for example lung function, heart rate (variability) and blood pressure, clotting parameters. There are intensive efforts under way to identify genetic markers which then are used to search polymorphisms and groups of susceptible individuals suffering from asthma COPD etc.

Why do toxicological studies show much larger potency differences than epidemiological studies?

Toxicological studies are mostly short-term studies applying rather high doses which are usually less common in environmental settings. In fact, very frequently no effects are found at low doses because the biological test systems provide no long-term history of exposure resulting in pre-developed lesions and susceptibilities. However, toxicology uses biological systems which are as much controlled as possible; hence, reactions with single biomolecules, nucleic acids, proteins, membranous receptors, cells or even multiple cell systems or genetically equal animal models or knock-out mouse models can be tested to disentangle specific reactions without the multitude of promoting or counteracting processes found in the delicately balanced homeostatic system of the entire organism. Toxicology can use more sensitive measures which are predictive for the health outcomes in epidemiological studies such as increased severity of a disease. Toxicology can also perform repeated studies, even within the same subjects, and do these kinds of studies under well-controlled conditions.

Why don't humans develop tolerance to PM exposure?

There is confusing evidence. From epidemiologic studies after short-term exposure it is concluded that there is some kind of adaptation to a given level of exposure which may vary greatly from location to location; health effects occur in response to increases beyond this level. This phenomenon is even more pronounced in smokers which inhale very high doses of tobacco smoke and yet they seem to be sensitive to increments of PM10. On the other side epidemiological studies based on long-term exposures clearly demonstrate the risks of mortality and morbidity increases with increased levels of exposure to ambient PM.

How can toxicological data be used for risk assessment of PM and for regulation purposes?

There is a need to differentiate between dosimetry and toxicological modes of actions and underlying mechanisms. Dosimetric aspects like (a) the deposition probability of inhaled particles to the various regions of the respiratory tract, (b) particle retention in the various regions of the respiratory tract, and (c) particle accumulation in secondary target organs are clearly important pre-requisites of risk assessment. However, toxicological modes of actions and underlying mechanisms can only serve creating advanced knowledge of the pathogenic development of different diseases. Therefore, it cannot directly serve as quantifying tools for risk assessment.

What are the research needs & recommendation?

- Need for better integration of epidemiology and toxicology, using for instance same health indicators (biomarkers of effect).
- Source related toxicological studies preferably using real world mixtures.
- Long term exposure studies (that can also be used as toxicology-time series studies). Better animal models with the challenge for transgenic mice.
- Development of a test battery for oxidative stress that can ultimately be used to monitor the biological reactivity of air pollution.
- Effectiveness of control strategies of vehicle emissions from the toxicological point of view.
- The role of surface area of (the insoluble core of) PM.
- The role of so-called non-toxic components (often also referred to as “natural”) in the total mixture of PM. Can such particles interact to become for instance a carrier for a toxic or allergic substance (not even a particle).
- Integration of air sampling in toxicological studies: Usage of PM sampling techniques that reduces sampling artefacts to a minimum such that it really resembles PM in air.
- Improved insight in biological mechanisms.

Policy relevance

- Guidance on the selection of new or additional measures to control health effects due to PM.
- Guidance of abatement strategies to reduce PM from specific sources.
- Increased confidence on biological plausibility and causal relationships.

Presentations to the topics

Topic 1: Particle characterisation and characteristics

Spatial and temporal variances in aerosol characteristics in the EU

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Introduction

Satellite-borne measurements of the aerosol optical thickness (AOT) obviously provide an excellent spatial coverage of the column–integrated concentration of particles, if data are averaged on a long enough time period though (no measurement possible in presence of clouds). Nevertheless, satellite measurements are not always relevant for assessing the health effect of particulate matter. Indeed, it has been reported at several sites that PM10 mass concentration at ground level (where people breath) and AOT seasonal variations are anti-correlated. This is due to the fact that PM10 at ground levels depends strongly on the mixed boundary layer height, whereas AOT does not. However, a huge amount of data regarding PM10 mass concentrations does exist. For instance, the EuroAirnet database maintained by the European Environment Agency (EEA) contains data from more than 30 European countries collected in more than 6000 monitoring stations located at urban sites (EEA Topic Report 26/1996 – Air pollution monitoring in Europe - Problems and trends). Working out such a dataset, it has been possible to highlight that PM10 is on average >3 times as large in Budapest as in Helsinki, and >twice as large is Milan compared to Paris. It can also be seen that PM10 concentrations have not been significantly reduced in many European cities over the 5 past years.

Several epidemiological studies (e.g. the 6 City Study, Dockery et al., 1993) indicated a good correlation between PM mass concentrations and health effects (mortality or morbidity). But more recently, it has been shown that the relative risk for mortality per 10 μg/m3 increase in PM10 levels is very much city-dependent in Europe (Katsouyanni et al., 1997). To understand whether these differences in PM10 effects may be due to differences in PM characteristics, data regarding e.g. the aerosol chemical composition, size distribution and main sources should be looked at.

Aerosol Chemical Composition over Europe

PM10 mass concentrations have also been measured at some of the EMEP (Co-operative program for monitoring and evaluation of the long-range transmissions of air pollutants in Europe) stations from about 10 years ago. On the top of PM10, a considerable amount of particulate SO_4^{2-}, NO_3^-, and NH_4^+ concentration data in aerosol (http://www.nilu.no/projects/ccc/emepdata.html) is available in the EMEP database. Long-term records indicate that SO_4^{2-} concentrations have significantly decreased (> a factor of 2) over the past decades, whereas such a clear trend is not visible for NO_3^-. At sites like IT04 in northern Italy, most of the PM mass concentration reduction observed over the past 20 years (20%) was due to the decrease in NO_3NH_4 and $(NH_4)_2SO_4$, which might well not be the PM component contributing most to the PM adverse health effects.

Potentially more dangerous are heavy metals (HM). HM concentrations in aerosol have also been measured at more than 35 EMEP sites, in some cases for close to 20 years. EMEP reports (Heavy metals and POP measurements, 2003) show large differences (factor 3-5) among sites, with the highest values usually observed in central Europe. Long-term time series show e.g. that Cd and Ni concentrations decreased in Slovakia (SK06) over the last 10 years, but not that much in UK (GB14) or Austria (AT02). In contrast, Pb concentrations significantly dropped down in UK (GB14) over the last decade, but did not significantly change at SK06 or AT02. As concentration even increased in Spitsberg (NO42).To normalize for dilution effects, one can look at HM/PM10 concentration ratio. It still appears that atmospheric particles contain more Cd in e.g. Germany (DE09) and Austria (AT02) than in Norway (NO99) and Spain (ES09), but about the same fraction of As, Ni, and Pb (except at NO99 for Pb).

Also carbonaceous aerosol may be responsible for adverse health effect. These compounds have been measured at a limited number of EMEP stations, and the lack of standardized methods for sampling and analyses make it difficult to compare organic carbon (OC) and elemental carbon (EC) concentrations from various sites. This is why EMEP organized a “carbonaceous aerosol” campaign over Jul. 2002 – Jul 2003, during which samples were collected with similar sampling trains at 13 sites over 12 countries in Europe, and all analyzed at NILU with the same method. This exercise showed that PM10 contains on average twice as much OC in Stara Lesna (SK04) and Virolahti (FI17) compared to Ghent (BE02) in summer, or in Ispra (IT04) and Illmitz (AT02) compared to Kollumerwaard (NL09) in winter. The EC/PM10 ratio also ranges from 2 to 4%, excluding remote and urban sites.

Aerosol chemical and physical characteristics collected at stations spread all over the world can also be found in the WMO-GAW-World Data Centre for Aerosol (http://wdca.jrc.it/). However, the number of stations reporting also PM mass concentrations is limited (see Table 1).
Aware of the fact that it would be worth making more aerosol data collected in the framework of research projects available to a large community, scientists recently started to work in this direction. The FP5/GMES project CREATE (Construction, use and delivery of a European aerosol database) aimed at contributing to the compilation of European aerosol data. The database it has established contains a quite comprehensive set of data collected at 10 research sites in Europe.

This brief overview shows that a lot of data are available regarding PM mass concentration at urban sites on the one hand, and that a more limited but significant amount of data regarding detailed aerosol characterizations at rural to remote sites exist on the other hand.

Table 1: Aerosol characteristics reported to WDCA for stations including PM mass concentration.

Station	PM mass	ions	OC +EC	elements	
Beresinski (BR)	X			X	X (OPC)
Iskrba (SI)	X	X			
Hohenpeissenberg (DE)	X	X	X		X
Kosetice (CZ)	X			X	
Neuglobsow (DE)	X				
Mace Head (IE)	X	X	X		X
Schauinsland (DE)	X			X	X
Jungfraujoch (CH)	X	X			X

Data collected at stations of any type (remote to kerbside site) where PM mass concentration **and** aerosol chemistry **or** number size distributions were available were compiled in a single document entitled “A European aerosol phenomenology”, to emphasize the fact that this was a picture of the aerosol over Europe, as it appeared to us at that time. Some robust conclusions were drawn out of this work, such as:

- Sulfate and organic matter are the 2 main contributors to PM, except at kerbside sites where mineral dust (incl. trace elements) and black carbon predominate.

- No useful correlation ca be established between PM mass and particle number concentrations.

  It also stimulated positive comments from e.g. modelers who found there a source of data for testing their calculations against, and constructive critics such as:
- Lots of relevant data sets are not included in this compilation
- The lack of harmonisation in sampling and analytical techniques makes comparisons between sites difficult.

The COST633 activity 1 (Aerosol characteristics and characterization) actually proposed to work on these 2 topics:

- compiling more data obtained in the frame of research programs, at sites ranging from rural to street canyons
- reviewing and assessing the sampling and analytical artefacts which may render results obtained at various sites with different techniques not comparable, and if possible, propose solutions

Thanks to COST633 working group 1 members, more than 250 datasets from 15 countries have already been identified (where, when, what, how, by whom). Some data from Denmark, Finland, Italy, Spain, and The Netherlands have been uploaded to a specially created temporary databank with password restricted access, which would make it possible to average and plot for comparing these data. However, a lot of data are still missing before it makes sense to work out these data, which would allow us to produce a unique compilation going **much beyond what is currently existing**. Eventually, the “COST633” data might be on request transferred to the WDCA to ensure a safe archiving and worldwide availability.

COST633 provides the opportunity of achieving this terrific task, which will be realizable only with an additional little effort by each of us (COST633 WG1 members) though.

Use of Macro Tracers for PM2.5/PM10 Source Analysis

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To reduce ambient particle concentrations, knowledge about the magnitudes of individual source contributions is necessary. Emission inventories may help to understand the relative contributions of primary emissions. However, large contributions from fugitive sources as well as from secondary formed aerosol are generally not accounted for in emission inventories. Therefore, for the analysis of source contributions methods, based on the analysis of the ambient PM2.5 and PM10 aerosol combined with special data evaluation techniques have been emerged. A six component macro tracer concept has been introduced to derive source attributions of PM2.5 and PM10 ambient aerosol. The model consists of three sources based on organic tracers, two sources, based on inorganic tracers, and the diesel exhaust source. Although only six sources contribute to the organic carbon in our model (Diesel emissions, coal combustion, wood combustion, secondary organic aerosol represented by humic like substances and dicarboxylic acids, and plant debris), the agreement between observed organic carbon and organic carbon explained by the six sources is remarkable. The winter OC is explained nearly quantitatively by the six sources; summer OC is explained to more than 2/3. The largest fraction of wintry OC at the Vienna AKH site is from wood smoke, followed by OC from Diesel exhaust and “humic like substances”. In the summer case Diesel exhaust is the dominant fraction in OC, followed by coal combustion and the presumably secondary components “humic like substances”.
Concerning the PM2.5 particulate mass at Vienna site the foremost source contribution is secondary inorganic aerosol (ammonium, sulfate and nitrate) in the winter (48%), as well as in the summer half year (44%). Diesel off road and road emissions account for around 15% in both seasons. Based on a off road-road emission split, the contribution of road traffic exhaust emissions to PM2.5 is around 10% in Vienna.

Topic 2: Sources of particulate matter

Results of Source Apportionment Studies in Spain and comparison with other European Regions

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Results on the measurements of levels, speciation and source apportionment analysis of PM_{10} and $PM_{2.5}$ at 25 monitoring sites of Spain for the period 1999-2005 are presented. Measurements were performed with data coverage of at least one year at each site and included regional and urban background sites, traffic hotspots and urban background sites with high industrial influence. The data were obtained using manual gravimetric PM_{10} and $PM_{2.5}$ high volume captors and quartz micro-fibre filters. Following sampling, PM_{10} and $PM_{2.5}$ filters were analysed for mass and major and trace elements and compounds, with a total of 57 determinations per sample. The components analysed included: a) crustal component (Al_2O_3, SiO_2, CO_3^{2-}, Ca, Fe, K, Mg, Mn, Ti and P); b) marine components (Cl^-, Na^+ and indirectly calculated marine sulphate); c) organic matter plus elemental carbon (OM+EC, value obtained after applying a 1.2 factor to the OC+EC concentration); d) secondary inorganic aerosols (SO_4^{2-}, NO_3^- and NH_4^+); and e) to support the source apportionment analysis a number of metals and trace elements were also analysed for each sample. Source apportionment analyses were performed for most of the data series obtained, by applying the widely used method based on a first Principal Component Analysis followed by a Multilinear Regression Analysis (PCA-MLRA).

Average ranges of PM_{10} and $PM_{2.5}$ concentrations and chemical composition in Spain show significant variations across the country, with current PM10 levels at several industrial and traffic

hotspots exceeding recommended pollution limits[4]. Such variations and exceedences are linked to patterns of anthropogenic and natural PM emissions, climate, and reactivity/stability of particulate species. PM_{10} and $PM_{2.5}$ concentrations reach 14-22 $\mu gPM_{10}/m^3$ and 8-12$\mu gPM_{2.5}/m^3$ in most rural/regional background sites, 25-30 PM_{10}/m^3 and 15-20$\mu gPM_{2.5}/m^3$ in suburban sites, 30-46 $\mu gPM_{10}/m^3$ and 20-30$\mu gPM_{2.5}/m^3$ in urban background and industrial sites, and 46-50$\mu gPM_{10}/m^3$ and 30-35$\mu gPM_{2.5}/m^3$ in heavy traffic hotpots. Spatial distributions show sulphate and carbon particle levels reach maxima in industrialized areas and large cities (where traffic emissions are higher), and nitrate levels increase from the Atlantic to the Mediterranean (independently of the regional NO_x emissions, and probably linked to higher eastern ammonia emissions). African dust outbreaks have an influence on the number of exceedances of the daily limit value, but the additional African particulate load on the mean annual PM_{10} levels is only highly significant in Southern Iberia and Canary and Balearic islands. The marine aerosol contribution is near one order of magnitude higher in the Canaries compared to the other regions. Important temporal influences include PM intrusion events from Africa (more abundant in February-March and spring-summer), regional scale pollution episodes, and weekday *vs.* weekend activity. Higher summer insolation enhances $(NH_4)_2SO_4$ but depletes particulate NO_3^- (as a consequence of the thermal instability of ammonium nitrate in summer) and Cl^- (due to HCl volatilisation resulting from the interaction of gaseous HNO_3 with the marine NaCl), as well as generally increasing dry dust resuspension under a semi-arid climate.

The results of the source apportionment analysis of PM_{10} are presented as an example of the outputs of the studies performed. These may be summarised as follows:

1. Direct road traffic contributions reach from 30-48% of the PM_{10} annual load in urban sites, not including secondary aerosols which are formed from gaseous precursors from traffic.
2. Industrial contribution reaches, in most industrial areas, loads close to 30% of the PM_{10} annual values.
3. There is an important and constantly identified source of crustal composition which is in fact not a single source but includes a mixture of natural and anthropogenic mineral matter (road dust, demolition and construction dust among others) deposited in urban areas and constantly being re-suspended by traffic and wind. This load represents 25 to 35% of the PM_{10} mass in urban areas, but up to 50% around industrial areas with primary particulate emissions. It is important to note the marked North to South

[4] Number of maximum allowed exceedances of the 50 µg/m³ PM10 daily value are exceeded in many hotspot sites, but also the annual 40 µg/m³ PM10 average is exceeded in a number of these sites.

increasing gradient found when looking at mineral matter levels in PM_{10} as aridity (caused by low rainfall rates) and proximity to Africa increases.

4. The marine contribution is limited down to 3-5% in continental sites, 5-10% in coastal sites, but reaches 30-35% in urban sites of the Canary Islands on an mean annual basis. These high percentages are due to a relatively low PM_{10} concentrations and high Atlantic wind speeds.
5. There is a large contribution of unaccounted mass for model outputs obtained for regional background sites. This is attributed to the relatively high proportion of water contained in PM_{10} which is still not extracted from the sample under 25 or 50% humidity chambers after 48 h of equilibrium. This artefact is inherent to the filter sampling strategy and is more important in remote sites due to the lower ambient concentrations of actual PM components compared with absorbed water with respect to the polluted sites.
6. Another limitation we found from our source apportionment studies is the frequent identification of a secondary inorganic aerosol (SIA) regional contribution source resulting in lower loadings of other components. This may account for up to 30% of the PM_{10} levels and is in fact a regional or external source stemming from a mixture of contributions, not a single source. The relative load of this mixed sources component is lower in polluted sites because part of its contribution is assigned by modelling to local emission sources that are partially associated with SIA and with defined chemical tracers.
7. There is an increasing gradient from the lower levels of the Atlantic side to relatively higher levels in the Mediterranean side of Spain concerning the regional-SIA load. This is attributed to lower dispersion of pollutants, lower rainfall, and probably to higher NH_3 levels in the Eastern part of the country.

The OC/(OC+EC) ratios in urban sites of Spain are very high (0.6-0.8). If diesel vehicles are a major road traffic emission source and the OC/EC ratio of the PM emissions of these vehicles is usually low, a mismatch between the theoretical impact of these emissions on PM levels and the PM speciation may arise.

8. Some low stability (such as NO_3^-) and photochemically dependent (such as SO_4^{2-}) PM species may show opposite seasonal concentration patterns, even if these are arising from a single source. This may give rise to erroneous source differentiations by receptor modeling.

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Topic 3: Modelling and (personal) exposure

Determination and Modeling of Exposure to PM from Different Sources

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Rationale

Attribution of PM exposures to sources as well as modeling of exposures from different sources are crucial elements to answer key questions in PM *risk assessment* (Which sources contribute how much to the exposure and risk?) and for *risk management* (What is the impact of a given source reduction on exposure? How big reduction on which source is required to achieve a wanted reduction of exposure and risk?).

The dominant sources of PM exposure for urban individuals - excluding ETS and other indoor sources - are typically secondary inorganic PM formation from gaseous precursors, SO_2, NOx and NH_3, followed by primary long range transported PM (which together form the *extraurban background*). Superimposed on this background is the *urban background* PM, consisting to a small degree from fresh secondary PM and sometimes salt PM from street deicing, and to greater and highly varying degrees from local street and soil dust and primary combustion particles from traffic and stationary sources. Further superimposed are the localized *peak levels* of traffic generated PM on and in the vicinity of the major streets, combustion particles from residential heating and small industrial sources, and occasional burst of wind blown dust.

Exposure & source attribution modeling principles

The following cross table (WHO/IPCS, 2005) presents the four modeling principles.

	Mechanistic	***Empirical***
Deterministic	Mathematical constructs of physical/chemical processes that predict an exposure for a set of inputs.	Statistical models based on measured input and output values (e.g. regression models that relate ambient pollutant concentrations with person exposures).
Stochastic	Mathematical constructs of physical/chemical processes that predict the probability density distribution of population exposure using full ranges of input data	Regression-based models, where model variables and coefficients are probability distributions, which represent variability and/or uncertainty in the model inputs and parameters.

These principles apply to source to exposure as well as source attribution of exposure modeling.

Exposure modeling

Sources: Exposure modeling changes from relatively straightforward and accurate towards complex and inaccurate as one proceeds from modeling of exposure to the compounds (and source emissions) of extraurban background PM towards modeling exposure to traffic PM including the exposure while in transit.

Averaging time and population: Similarly, along these dimensions, modeling the probability density distributions of PM exposures for populations of entire conurbations over one year is straightforward and accurate compared to modeling the PM exposures for given individuals at given locations & times, which can be extremely data hungry, computing intensive and still produce poor correlations between the predicted and true (measured) exposures. The former is needed e.g. for long term cohort studies or the evaluation of policy options, the latter e.g. for case-control studies, or gene-environment interaction studies.

Attribution of exposure to sources

Source attribution methods fall into two main categories, empirical (statistical) and mechanistic. The former include the variants of factor analysis, which require little or no *a priori* knowledge of the sources or their emission profiles, and can therefore point out unexpected sources, but require a number of identically collected and analysed samples.

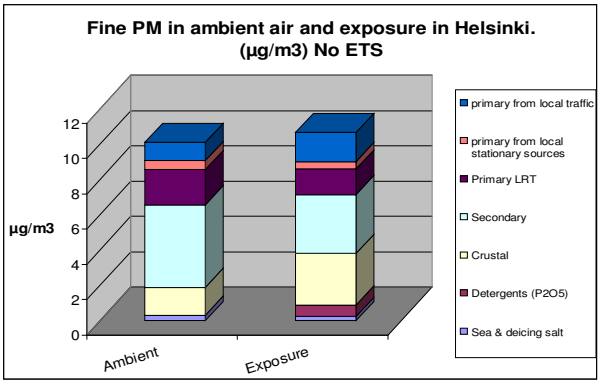
The results of statistical source attributions apply for the whole data set, but attribution of individual samples to sources is possible by subsequent regression analysis, although some sample variation is missed in the process. Because the method is data driven, statistical attributions of multiple data sets do not necessarily result in identically defined sources. The methods include variants of *target transformation factor analysis*, *principal components analysis*, *structural equation modeling* and most recently *positive matrix factorisation*, which is superior to the earlier methods in its ability to deal with missing data, differently weighed data and data uncertainties.

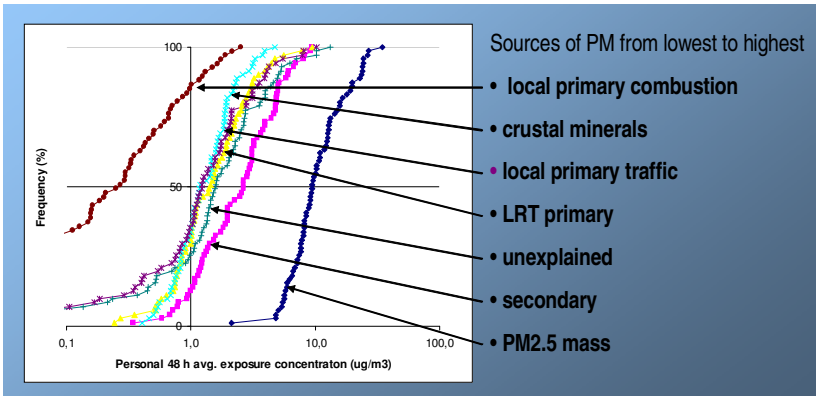
Mechanistic methods at their simplest are based on marker elements such as S for secondary PM or Pb for traffic (up to 1980's), include the more advanced *chemical mass balance* method, which requires knowledge of the relative chemical compositions for the emissions of competing sources and reconstructs the source proportions which most closely matches the sample composition, and *source reconstruction*, in which the total PM mass from a specific source is computed from the elemental composition of the sample (e.g. crustal source component = 2.20xAl + 2.49xSi + 1.63xCa + 1.58xFe + 1.94xTi + 1.41xK). Mechanistic methods require *a priori* knowledge of the sources, attribute individual samples to sources, and are capable of producing fully comparable source attributions for different data sets.

Experience has shown that multiple source attribution techniques should be applied to the same data and the results should be compared and assessed before drawing far reaching conclusions.

Exposure to PM from different sources

There are many studies where ambient PM has been attributed to sources. Similar studies on exposures are quite rare in comparison. The following upper figure shows a comparison between average ambient air and personal $PM_{2.5}$ concentrations for non-smoking individuals in Helsinki, and the contributions of various sources to both. Compared to the ambient PM, personal exposure has less extraurban PM and more traffic and crustal PM. The lower figure shows the full probability density distributions - rather than just average values - of 48h $PM_{2.5}$ exposures of the same population. The dominant role of long range transported extraurban background (primary and secondary) PM is still obvious. Traffic exhaust PM and street dust are also significant, but in Helsinki the role of primary PM from local stationary sources is, due to district heating, quite small.


Fine PM in ambient air and exposure in Helsinki.
(µg/m3) No ETS
µg/m3
0
2
4
6
8
10
12
Ambient
Exposure
primary from local traffic
primary from local stationary sources
Primary LRT
Secondary
Crustal
Detergents (P2O5)
Sea & deicing salt


Frequency (%)
100
50
0
0,1
1,0
10,0
100,0
Personal 48 h avg. exposure concentraton (ug/m3)
Sources of PM from lowest to highest
• local primary combustion
• crustal minerals
• local primary traffic
• LRT primary
• unexplained
• secondary
• PM2.5 mass

Modelling of urban population exposure to PM

Carlos Borrego, Ana Isabel Miranda and Joana Ferreira

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1. Introduction

Air pollution is a major environmental health problem causing approximately three million deaths per year in the world, as result of exposure to particulate matter (PM) (EEA, 2003).
For population exposure assessment, a combination of the spatial distribution of both air quality and population density is required. Numerical models are useful tools for the mapping of air pollutants, once the monitoring networks are able to assess the air quality in the single stations of the monitoring network, and not the whole area of interest. Dispersion and photochemical models have been applied, at different scales, to evaluate air pollution due to particulate matter concentrations and its effects on human exposure and health.
Health effects of air pollution are the result of a chain of events, going from the release of pollutants leading to an ambient atmospheric concentration, over the personal exposure, uptake, and resulting internal dose to the subsequent health effect (Figure 1). The conditions for these events vary considerably and have to be accounted for, in order to ensure a proper assessment (Hertel *et al.*, 2001).

Figure 1: Source to dose assessment

This paper will focus on Ambient Concentrations as an input for exposure estimation. Atmospheric pollutants concentrations are generally obtained by direct measurements in air quality stations, equipped with analysers in continuum. Air quality national networks include different types of stations in order to characterize different types of environments like traffic, kerbside, industrial, background, urban, suburban and rural. Even having a wide covering of the area of interest, air quality concentrations are obtained at discrete sites. Depending on the location and dimensions of the region to be studied, monitoring data could not be sufficient to characterize the air quality of the area or to perform related human exposure estimations.

Therefore, numerical modelling can be seen as a complementary tool, once it is able to reproduce a 2D or 3D air pollutants concentration field for a given area of interest.

2 Air Quality Modelling

Air quality models use mathematical and numerical techniques to simulate the physical and chemical processes that affect air pollutants as they disperse and react in the atmosphere. Based on inputs of meteorological data and source information like emission rates and stack height, these models are designed to characterize primary pollutants that are emitted directly into the atmosphere and, in some cases, secondary pollutants that are formed as a result of complex chemical reactions within the atmosphere. These models are important in air quality management systems because they are widely used by agencies in charge of controlling air pollution providing the identification of source contributions to air quality problems and assisting in the design of effective strategies to reduce harmful air pollutants. For example, air quality models can be used during the permitting process to verify that a new source will not exceed ambient air quality standards or, if necessary, determine appropriate additional control requirements. In addition, air quality models can also be used to predict future pollutant concentrations from multiple sources after the implementation of a new regulatory program, in order to estimate the effectiveness of the program in reducing harmful exposures to humans and the environment.

3 Types of Models

The most commonly used air quality models include the following:

- Dispersion Modelling - used to estimate the concentration of pollutants at specified ground-level receptors surrounding an emissions source, considering only the dispersion and not the chemical transformation processes.
- Chemical Modelling - used in regulatory or policy assessments to simulate the impacts from all sources by estimating pollutant concentrations and deposition of both inert and chemically reactive pollutants over large spatial scales.
- Receptor Modelling - observational techniques which use the chemical and physical characteristics of gases and particles measured at source and receptor to both identify the presence of and to quantify source contributions to receptor concentrations.

Chemical air quality models have become widely recognized and routinely utilized tools for regulatory analysis and attainment demonstrations by assessing the effectiveness of control strategies. These models simulate the changes of pollutant concentrations in the atmosphere

using a set of mathematical equations characterizing the chemical and physical processes in the atmosphere.

There are two types of air quality models commonly used in air quality assessments: the Lagrangian trajectory model that employs a moving frame of reference, and the Eulerian grid model that uses a fixed coordinate system with respect to the ground.

Lagrangian models consider air parcels, which travel with the wind (i.e., they are advected). These models are often also referred to as trajectory models, since the air parcel under consideration follows a trajectory defined by the winds.

Eulerian models consider a mathematical framework anchored to the surface of the earth, as shown in Figure 2. Eulerian models are often also referred to as grid models, since the framework is a three dimensional grid, with pollutants being emitted into the grid at the appropriate points. Pollutants travel through the grid, under the influence of the local winds, undergoing chemical and physical transformations as they go.

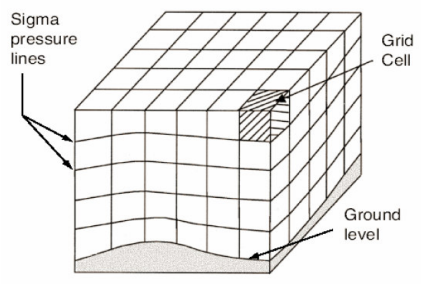


Figure 2: Eulerian modelling framework

Most of the current operational chemical air quality models have adopted the three-dimensional Eulerian grid modelling mainly because of its ability to better and more fully characterize physical processes in the atmosphere and predict the species concentrations throughout the entire model domain (Reid *et al.*, 2003).

Regarding the scale of application, models can be classified into global, regional, mesoscale, urban, local and microscale.

As the name implies, global models consider the transport of pollutants throughout the atmosphere, with no artificial restriction of the domain. The large spatial extent of these models dictates that the spatial resolution (grid spacing) must be relatively coarse to keep the computational demands within reasonable bounds. To date, most global modelling has been

confined to carbon dioxide and the climate change issue, which also means that chemical transformation is not treated. However, expansion to other pollutants has recently taken place. Mesoscale to regional scale models consider spatial scales ranging from a few hundred to a few thousand kilometres, over which many of the most pressing air pollution concerns are important (NIWAR, 2004). Models on these scales are generally the most important for policy makers. Examples of current use Lagrangian models include Calpuff and Hysplit, while Eulerian models include Models-3/CMAQ, UAM-V, CAMx, CHIMERE and TAPM.
Urban scale modelling (including local scale) is typically used to assess the impact of single sources, or small groups of sources, over distances ranging up to tens of kilometres. Examples of Gaussian type models in current use include ISC, AERMOD, AUSPLUME. VADIS and FLUENT are examples of Computational Fluid Dynamics (CFD) local models.

4 Choice of model

The choice of an appropriate model is heavily dependent on the intended application. Even though a model may be freely available, it does not follow that it is necessarily the most appropriate for the situation under investigation. In particular, the science of the model must match the pollutant(s) of concern. For example, if the pollutant of concern is fine particulate matter, the model chemistry must be able to handle reactions of NO_X, SO_2, volatile organic compounds (VOC), ammonia, etc. Reactions in both the gas and aqueous phases must be included, and preferably also heterogeneous reactions taking place on the surfaces of particles. Apart from correct treatment of transport and diffusion, the formation and growth of particles must be included, and the model must be able to track the evolution of particle mass as a function of size. The ability to treat deposition of pollutants to the surface of the earth by both wet and dry processes is also required.

5 Input data

Modern models require a considerable volume of data. The specific needs reflect the science incorporated in the model, but will typically include the following:
Emissions - For all sources treated by the model (for each grid square of an Eulerian model) the rate of emission is required for each of the chemical species followed by the model, specifically including each of the VOC species or categories used in the model chemistry, i.e., both anthropogenic and biogenic.
These emissions should relate to the specific time period being studied. For the modelling of fine particulate matter formation and transport, emissions data are also required for primary

particle emissions as a function of particle size, as well as emissions of ammonia. Neither of these is particularly well categorised yet.

Geophysical data - Information is required on a range of surface parameters, including topography, land use category and vegetation type.

Meteorology - Meteorological information is typically required to drive the transport in the model. This information is needed at several vertical levels in the atmosphere, and must also be for the period to be modelled.

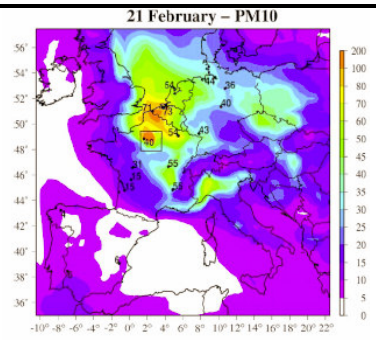
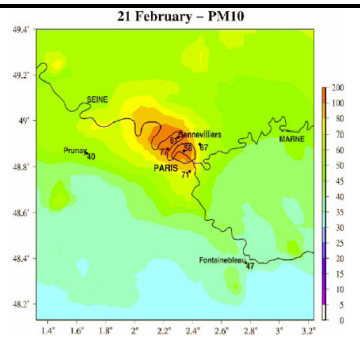
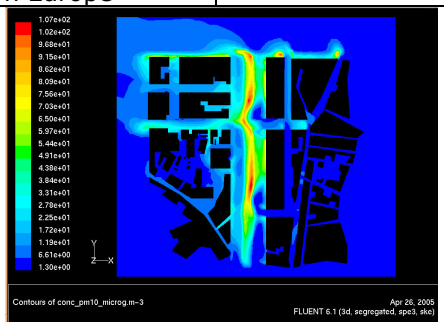
Initial and boundary conditions - It is usual to specify the initial chemical concentrations for a number of the major species in the model. These will be taken from typical or average values measured, or previously modelled, for the region of interest. It is also necessary to specify concentrations at the boundaries of the model, except for global models. It is relatively simple to estimate initial and boundary conditions at the surface based on measurements, these values are also required at higher levels in the atmosphere, where measurements are very much sparser. Current practice, which addresses the specification of initial and boundary conditions, is to nest the model. In nesting an initial run is carried out for a large domain at relatively coarse resolution. This is followed by a run at finer resolution for a smaller subdomain, using predictions from the first run to provide initial and boundary conditions (Reid *et al*., 2003).

6 Modelling applications for human exposure

Air quality models are widely applied in Europe for various purposes. As diagnostic tools, they are used to improve knowledge of the chemical and physical behaviour of the atmosphere, to help decision makers on the development of policies and air quality management systems for protection of ecosystems and human health. Models are also important instruments for prognostic, applied to a future situation, in order to evaluate the efficiency of a policy or a taken measure to reduce a certain type of emissions, or for air quality forecast, and consequently human exposure and health effects prevention.

Some examples of air quality modelling application are given in the table below, for different goals and thus, covering various scales. Air pollutants concentration fields estimated by the models allow to better understand outdoor microenvironments, than monitoring data, and, combined with gridded population and microenvironments information, should be used for exposure modelling and estimation of doses and health effects, concluding the source to dose assessment (Figure 1).

Table 1 – Examples of air quality modelling applications

Type of Model	Example of results
CHIMERE Mesoscale/Regional Eulerian model (Bessagnet *et al.*, 2005)	 Western Europe  Paris region
FLUENT CFD local model (Martins *et al.*, 2005)	 Liberdade Avenue, in Lisboa city centre

A number of exposure modelling studies based on air quality modelling results have already been performed, namely in the scope of the European Project FUMAPEX (URL1). As an example, Figure 3 presents the results of the estimation of children (< 15 years) exposure in Turin city area. A mesoscale model was used to obtain PM_{10} ambient concentrations in a 1*1km horizontal resolution grid, that were than used to estimate the indoor concentrations necessary for exposure calculations (Hänninen *et al.*, 2005).

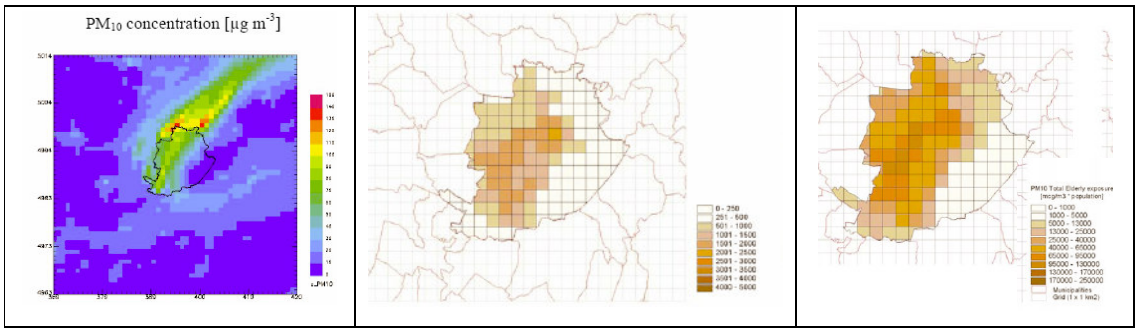


Figure 3 – PM_{10} simulated concentration field, children population living in each grid cell and spatial distribution of 24-hours total PM_{10} exposure.

7 Final remarks

Numerical models are useful tools for the mapping of air pollutants, once the monitoring networks are able to assess the air quality in the single stations of the monitoring network, and not a whole area of interest.

All models are useful. The choice of a model depends on the type and dimension of the area, the pollutants to simulate, and the final goal of the study: air quality management, exposure and health estimations, etc.

Knowing the physical state and composition of atmospheric aerosols is of great significance, especially when anthropogenic perturbations are examined, because of their role in atmospheric processes and climate forcing. Mathematical models which simulate the evolution of both gaseous and aerosol species have only recently started to appear, due to the complexity and variability of the processes in which particulate matter is involved (Nenes, 1999).

Air quality models are already widely applied for exposure and health related issues, but an effort is still needed to take more advantage of the third generation models (Eulerian models including aerosol chemistry) on epidemiological studies.

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URL1 – FUMAPEX Project Home Page http://fumapex.dmi.dk/

Topic 4: Health effects – Epidemiology

Acute Health Effects Perspectives

Gerard Hoek
Utrecht University, The Netherlands

The presentation focused on evidence from epidemiological studies for answering the following four questions:

- What are the relevant health effect end points to be assessed?
- Which parameter shall be monitored for epidemiological studies?
- What is the current status related to exposure- response functions?
- Do current information allow to assess pan European similarities and differences?

A large variety of endpoints have been studied in epidemiological studies, that differ widely in their degree of "adverseness". Cause-specific mortality, Hospital / Emergency room admissions for respiratory and cardiovascular disease, Primary care consults, School / work absenteeism, Medication use, Asthma attacks, symptoms, Lung function, are among the classical health endpoints studied. Increasingly, epidemiology also assess more physiological markers, that provide hints for potential mechanisms of air pollution. Examples include Heart rate variability, Blood markers of systemic inflammation, Blood viscosity, -segment changes, CC16 in blood / urine and Inflammation markers in nasal lavage.

Concentration response functions for PM10, NO_2 and ozone have recently been reviewed by WHO. The USEPA has also derived summary estimates for PM10, PM2.5 and ozone recently. The EU-funded project APHEIS also provides useful information.
Heterogeneity of response was the main topic. The issue is of interest because Sensitive subgroups may show a larger response than the population average and Simple indicators such as PM10 may not have the same relationship with health everywhere. Recent epidemiological studies such as the APHEA_2 study have provided evidence that there is indeed significant variation of the effect of air pollution on mortality within Europe. There was some evidence that this effect might be due to the composition of particles or the climate or the region within Europe, but it is difficult to disentangle these factors. A few other studies have suggested that particles from combustion sources such as traffic had a stronger association with more mortality and cardiovascular morbidity than crustal particles and sea salt. A study is in the US suggested that

the effect estimate for PM10 and hospital admissions was larger in cities with less air conditioning (consistent with more penetration of particles in indoor air of homes). Several studies have documented that air pollution effects on mortality and morbidity depend on pre-existing disease status (e.g. COPD, diabetes), nutrition (e.g. anti-oxidant intake). In conclusion, health effects of air pollution are likely different in different locations and subjects, but the explanations include a complex of particle composition, sources, indoor/outdoor relationship, co-exposure to other pollutants and risk factors and subject characteristics.

Long-term Effects of Air Pollution on Health

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The organizers have asked to discuss mainly four questions:

- What are the relevant health end points to be assessed?
- What shall be monitored for epidemiological studies?
- What is the current status related to exposure-response functions?
- Does current information allow to assess pan European similarities and differences?

The first paragraph clarifies terminologies prior to address the four questions. Needs and open questions will be emphasized at the end.

1. Definitions / Terminology

Long-term effects are the consequence of the cumulated every-day 'chronic' exposure to ambient pollution. Most typically, the ultimate result of such effects is a chronic condition. In contrast to purely acute consequences of air pollution, these effects do not disappear if we have a few days of clear skies. A useful model of long-term effects is smoking. Long-term effects may not simply be the sum of all acute effects. The underlying mechanisms leading to acute versus chronic effects may well be different and independent or complementary mechanism may be important. It is thus a necessity to specifically design studies to investigate long-term effects. As a further complication, one has to be aware that the same (epidemiologically measured) health outcome may be the consequence of either acute or long-term or both effects [1]. An obvious example is "death due to lung cancer". Patients with lung cancer are more likely to die on days with high air pollution [2] and it is plausible that terminally ill patients to be particularly susceptible to oxidiatve stress, air ways infection and pneumonia which are all enhanced on days with higher levels of pollutants. However, associations between residential concentrations of ambient pollutants 20-30 years ago and lung cancer death [3 4] cannot be explained with acute effects but may reflect the result of the carcinogenic effects of constituents of air pollution including chronic oxidative stress with adverse effects on DNA and other pathways[5].

2. What are the relevant health end points to be assessed?

Health endpoints in chronic effect investigations are those that reflect the result of long-term underlying pathophysiologic processes (e.g. carcinogenesis, remodeling of air ways, thickening of artery walls due to calcifications). Time to death (life expectancy), and the onset of chronic diseases (such as lung cancer, asthma, COPD, atopy) are useful outcomes. Of particular relevance in long-term effect investigations are early markers of chronic health states or anatomic structures (lung function, thickness of the artery wall, chronic inflammation etc.). In contrast, the use of acute outcomes with a chronic underlying pathophysiology, such as myocardial infarction lead to far more ambiguous conclusions with regard to the time period and duration of toxic action of pollution. It is inherently difficult to disentangle the acute contribution of air pollution to triggering the infarction from the hypothesized long-term effect of air pollution on atherogenesis as the primary underlying pathology. Reconstruction of long-term exposure does not necessarily resolve the problem given that a location with high long-term average pollution is also more likely to be more polluted on any day [6].

3. What exposure shall be monitored for epidemiological studies?

As in all outdoor air pollution studies, it is most important to characterize exposure to pollution from outdoor origin rather than 'total exposure'. This need has to determine the strategies of exposure assessment. For example, if NO_2 was used as the marker for outdoor air pollution, measurements taken *outdoor* will be more relevant and more appropriate than measurements taken on person or indoors as indoor sources such as smoking or gas cooking would in both cases contribute to the personal NO_2. This would jeopardize its use as a marker of *outdoor pollution*. Of course, if investigations of the effects of NO_2 per se are of interest, personal exposure assessment would be considered the gold standard. Future studies will need rigorous attempts to characterize source specific exposure as this will be of direct relevance to policy makers.

Probably the biggest challenge in exposure assessment in chronic effect investigations – and the most important difference to studies of acute effects – originates from the need to maximize and characterize *spatial differences* (in contrast to purely temporal contrasts) of exposure. This is a demanding task given that some pollutants vary very little on the local level, thus subjects must be recruited from different areas to get sufficient contrasts in exposure. This strategy often comes with difficult to control area-specific confounding. Moreover, some pollutants vary drastically within short distance, within communities, thus residential location and time-activity patterns can be important determinants of exposure and a source of large non-systematic errors

if one cannot assess exposure on this spatial scale. Studies like the Dutch cancer cohort developed elegant solutions to characterize long-term exposure on various spatial scales [7] showing rather strong association between living close to busy traffic arteries and mortality. An increasing number of studies points in the same direction [8 9]. The European Community Respiratory Health Survey with 21 European centers participating in the air pollution module will give further evidence for the necessity to characterize exposure differences *within cities* rather than just across cities [10 11]. Contrasts of pollution concentrations – in particular of primary pollutants from traffic – can be as large within cities as they are across a wide range of European cities.

4. What is the current status related to exposure-response functions?

Exposure-Response functions are not available for chronic effect studies if exposure is correctly defined as the contact between pollution and a subject. So far, personal exposure measurements are the domain of acute effect studies, if at all. However, *concentration-response functions* are available from an increasing number of studies and outcomes.

For early markers of chronic diseases the most important and recent evidence originates from studies on the association between lung function growth in children and adolescents (e.g. [12]) and lung function level among adults (e.g. [13]). However, these studies were conducted in very few areas such as South California, Switzerland, Germany, or Austria and generalizations across Europe remain uncertain. Moreover, air pollution and the decline in lung function among adults has not yet been published, and studies on COPD (using the GOLD criteria) are extremely rare but needed for an assessment of the evidence. Only one Los Angeles based study investigated the association of air pollution with early markers of atherosclerosis, namely the artery wall thickness [14]. The positive findings among these participants of clinical trials need to be confirmed in light of an emerging literature reporting atherogenic effects of ambient particles in animal models [15].

The first cohort studies addressing long-term pollution and mortality were published in the U.S. (e.g. [16]. Meanwhile, several European countries published qualitatively comparable results based on Dutch [7], French [8], German [9], and Scandinavian [3 4] studies as well as among a subgroup of the European EPIC study [17]. However, approaches and definitions of exposures are diverse thus a formal assessment of concentration-response functions rather difficult at this point in time.

Studies on the incidence of chronic diseases are rare. Several studies published associations between air pollution and lung cancer, but, again, exposure definitions and methods are divers.

Studies on the incidence of COPD are needed to confirm the limited evidence of cross-sectional associations with air pollution [13]. More but not many studies investigated new onset of asthma and air pollution with results being in line with the increasing number of studies showing associations between asthma prevalence and proximity to traffic, but definitions of traffic exposure are as divers as the number of publications, thus a quantitative synthesis would be a challenge. Onset of asthma among adults has not yet been investigated in air pollution research and remains to be addressed. Long-term studies on air pollution and the onset of atopy are also needed given the experimental evidence of interactions between, allergens and diesel particles[18]. An integration of measures of exposure to primary pollutants from traffic will be needed in these studies as well.

5. Does current information allow to assess pan European similarities and differences?

Investigations of long-term effects of air pollution are limited to a few mostly national studies employing different approaches to characterize exposure. All are based on some markers of the complex and potentially divers mixture. A formal comparison of effects across Europe is not possible at all. It would require standardized European-wide projects. ECRHS would be a model for such an undertaken among adults but, as discussed, characterization of within-community contrasts in exposures will be a necessity.

6. Future Needs and open questions

The conclusion from all the above is that long-term effects of ambient air pollution need to be further investigated across Europe. The focus on objective early markers of chronic pathophysiologies that underlie chronic diseases will be of particular importance to understand the chronic contribution of pollution as opposed to the triggering of acute events. Studies need to employ hypothesis-driven designs for both the outcomes and the tools to characterize exposure. Methods to capture contrasts in exposure to local pollutants that are heterogeneously distributed in space need to be developed, standardized, and employed in chronic effect studies. Given the inability to measure exposure on persons over long periods of time, combinations of measurements and modeling will play an important role in future research. Attempts to characterize long-term exposures to pollution from specific sources need to be made. Studies that try to disentangle effects of specific PM constituents and of gaseous co-pollutants will be of relevance for scientists and policy makers alike.

Investigations of chronic effects of air pollution targeting at the same time the respiratory and the cardiovascular systems could make major contributions to understanding common pathways of

chronic pathologies, its environmental determinants, and the underlying susceptibility factors (see Figure 1).

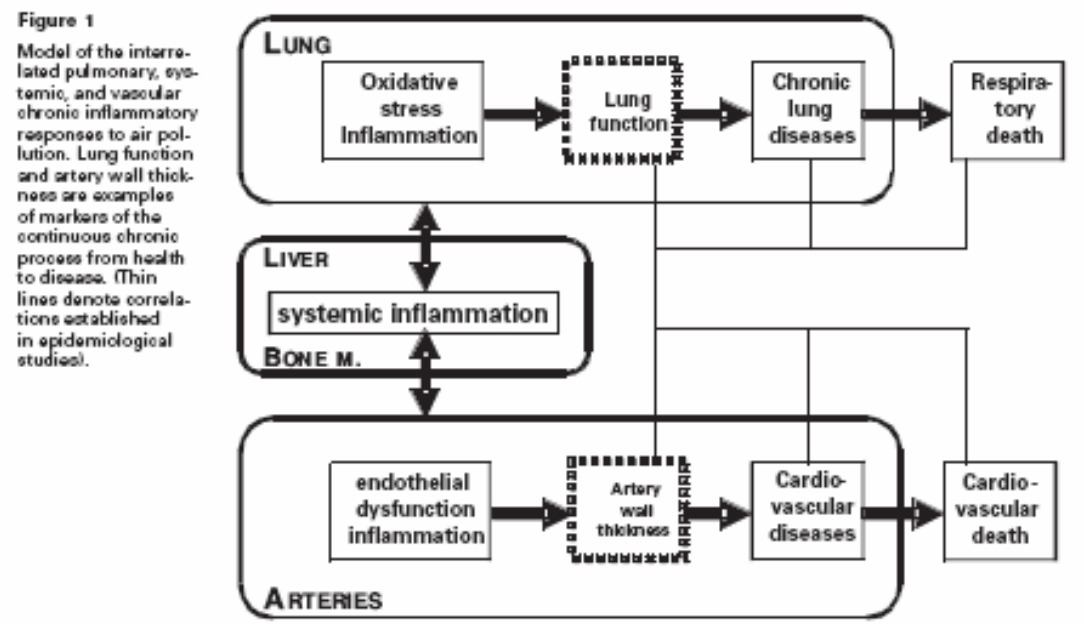

Figure 1 from Künzli and Tager, Swiss Medical Weekly 2005; 135: 697-702. (with permission)

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Topic 5: Health effects - Toxicology

Question-What particle characteristics should we measure? Answer- Ability to cause oxidative stress

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Summary: Recognizing that the mass collected by the PM10 or even PM2.5 convention are not the optimum measure for risk assessment, the question was –What particle characteristics should we measure? To answer this question, first of all the diseases associated with PM exposure were identified along with our best understanding of the mechanisms underlying these diseases. The diseases associated with PM increases are inflammatory lung diseases, cardiovascular disease and cancer. All of these conditions have oxidative stress and inflammation as part of their mechanism. Numerous studies have identified oxidative stress as a key activity of PM and several components frequently found in PM, such as surfaces, metals and organics, are known to generate free radicals in biological systems. A range of different assays are available for measuring the potential of a PM sample to deliver oxidative stress and it is by no means certain which of these would provide the best basis for the metric. It is hoped that the recognition that oxidative stress could be a metric that would approach the biologically effective dose, will act as a catalyst activating research into new technologies for rapid measurement of the free radical generating activity of particle samples. Whilst a measure of the oxidative activity of PM would not replace PM10 or PM2.5 mass at the moment, this is an attractive metric that has real health relevance for risk management.

Are there regional differences in toxicological effects of particulate matter?

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Epidemiological multi-centre studies of the health effects of air pollution have shown some heterogeneity of risk estimates of PM effects in different cities. This variation in risk estimates could be due to different composition of PM, but other factors may also play an important role. Experimental studies have examined how PM may induce acute health effects in voluntary individuals in exposure chambers, acute and chronic effects in animal studies as well as biological responses in cultured cells.

How may the toxicological end points be linked to disease? Inflammation appears to be a major determinant in PM-induced health effects. The inflammatory responses involve the production and release of a range of signaling molecules, which operate in a complex network between epithelial cells, alveolar macrophages and other immune cells, including neutrophils, eosinophils and T cells. The recruited immune cells may release cytokines, ROS, lipid mediators and toxic proteases, which may contribute to epithelial damage. This may in turn lead to an increased release of cytokines and chemokines from the epithelium, subsequently increasing and/or prolonging the inflammatory reaction, overwhelming the defense mechanisms and possibly progressing to chronic inflammation.

The differential attraction of immune cells is associated with different diseases. In allergic asthma the attraction of eosinophils involves different mediators than those that attract neutrophils in non-allergic inflammation-dependent diseases. Another difference is the production of specific IgE antibody in the response in allergic diseases exacerbated by particles. Cell death may also be involved in inflammation and development of both acute health effects and chronic lung diseases. These processes may occur in parallel to a remodeling of the lung tissue, and include proliferation of fibroblasts and myofibroblasts, which are involved in the development of disease. Changes in cell death and proliferation, induction of inflammation and the induction of DNA damage are linked to the development of lung cancer, but the relative importance of these processes has not been clarified.

Cardiovascular end points comprise very different effects ranging from arrhythmia through altered nervous regulation in response to particles, to vascular changes and cardiac stress

responses to particles, soluble components and mediators from the lung. The vascular changes may involve increased tendency of coagulation and mediators of a systemic inflammatory response. The cardiac stress responses may involve mediator production and activation of cellular stress proteins. These effects may contribute to outcomes such as heart failure and infarction.

What were the results of investigating effects of PM from different sites in Europe? Three EU-funded projects RAIAP, HEPMEAP and PAMCHAR, using these types of experimental systems, have attempted to dissect out which particle characteristics play a major role for the elicitation of effects. Studies using cell culture models indicate that there are differences between the effects of PM from different sites in Europe. The production of pro-inflammatory mediators is increased in both primary lung cells and cell lines from human and rat lung. In RAIAP, particles from Lodz and Rome were more potent than particles from Amsterdam and Oslo. Winter samples showed less pro-inflammatory activity than summer or spring samples. Coarse particles exhibited greater potential to induce mediators than fine particles at similar mass. The mediator results from the corresponding animal studies correlated with the cell culture data. Studies of particle effects on systemic allergic responses indicated that the sampled PM enhanced the allergic response, and that fine particles were more potent than coarse particles. A respiratory allergy model did not reveal any difference between fine and coarse particles, except that histopathology indicated a stronger effect of coarse than fine particles. The particle component concentration of metals and endotoxins showed increased levels in coarse fractions, which corresponds with the data for increased mediator levels. In an overall assessment of a link between groups of components with specific sources organic components in PM were more associated with respiratory allergy end points than other end points, whereas metals and endotoxins, linked to crustal material were more associated with inflammatory end points in cells and animals. The greater effect of coarse versus fine particles was, however, in this study not adjusted for differential deposition efficiencies, as inhalation studies were not included.

What are the results of comparisons of particles from different sources? Only a few studies have compared the effects of PM from different sources. Examples of such sources are wood combustion, road dust and industrial activity. Road dust consists of different stone particles that are made up of different types of minerals. Mineral composition seems to determine the potential of the stone particles to elicit pro-inflammatory responses in cells and inflammation in rat lung. Studies with cells indicate that the higher the content of plagioclase is, the lower is the pro-inflammatory potential. Though these particle samples were laboratory made, a sample from a tunnel in which the asphalt consisted exclusively one stone type that

was also included in the analysis, confirmed the potential of this stone type to induce pro-inflammatory effects in cells.

Wood smoke differs considerably in composition from traffic derived PM in that metals dominate the traffic-derived PM, whereas PAH dominate the wood smoke PM. Inflammatory effects in animals have been reported. In human monocytes wood smoke induces the cytokine IL-8 to approximately the same extent as traffic-derived PM, but a different mechanism may be involved.

Cardiovascular effects may be elicited in several different ways, through effects on the nervous system that may lead arrhythmias; through endothelial dysfunction and increased coagulation; through direct effects of soluble components or particles on heart cells leading to cardiac stress responses and increased risk of heart failure. Effects on fibrinogen in rat blood have been demonstrated with Ottawa dust and particles sampled in The Netherlands. No significant differences were demonstrated (F. Cassee et al., 2005). Ultrafine carbon particles were found to induce stress responses in isolated heart cells, both monocultures of myocytes and heart fibroblasts and in co-cultures. Cardiovascular effects have also been demonstrated in studies using exposure to CAPS, and the effects were associated with metals such as zinc, vanadium or iron (See AIRNET, Toxicology report). Zinc and copper were found to induce cardiovascular effects in rats (Bagate et al., 2006). In similar experiments as with the ultrafine carbon particles, metals were shown to induce stress responses in isolated rat heart cells and co-cultures of myocytes and fibroblasts. However, no synergism of metal effects could be detected.

How are responses initiated in the cells? The inflammatory responses involve the release of pro-inflammatory cytokines from macrophages and epithelial cells. This response seems to involve the formation of reactive oxygen (ROS) and/or nitrogen species (RNS) and the activation of receptors in the cell membrane. ROS formation may be initiated by the particles themselves, even outside the cells in the epithelial lining fluid, or at the cell membrane, or are elicited by the uptake of particles into the cell. A cascade of signaling events may follow the particle binding, uptake, and ROS formation. Certain proteins are involved in regulating the activity in the nucleus that leads to the induction of cytokine release or initiation of the process of cell death.

Soluble compounds may contribute in different ways. PAHs may induce DNA damage that may lead to cell death, but some PAH stimulate more the survival pathway leading to the survival of cells with DNA damage. This may enhance carcinogenesis. Some metals may not necessarily act through ROS formation to stimulate cytokine production pathways, but instead inhibit proteins that reduce the activity of such pathways. Thus there may be a dual effect or a possibility for synergism.

Concluding remarks. Particles sampled in different locations exert effects in various biological systems – rat and human lung cells, rats, mice and humans. The potential to induce effects various with the location the PM was sampled and with the season PM was sampled in. For non-allergic inflammatory reactions the coarse sample had a greater potential than the fine fraction to induce effects at equal mass. However, none of the measured particle components could alone explain the differences observed, though the non-allergic inflammation seemed to be more associated with metals and endotoxin and the allergic reaction more with organic components. The intrinsical potential of mineral particles to induce inflammation seemed to depend on the mineral composition. However, apparently non-toxic components may become more toxic by binding other components that occur in the air. Thus, composition is an important factor in addition to size and surface area

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Poster presentations - abstracts

Spatial variation in PM_{10} mass, particle number and chemical composition at UK sites

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Keywords: particle number concentration, PM_{10}, chemical components, regional transport, local influences.

Hourly measurements of PM_{10} have been made at an increasing number of sites in the UK since 1992. More recently these measurements have been enhanced by addition of co-located measurements of $PM_{2.5}$ (at 4 sites since 1998), particle number (at 8 sites since 2000), and by chemical composition data - organic and elemental carbon, and chloride, nitrate and sulphate at some of the sites since 2002. These measurements, along with some shorter term series of chemical composition measurements, allow the inter-relationships of various particle metrics to be compared across the UK. The sites at which particle metrics other than PM_{10} are measured are shown in Figure 1.

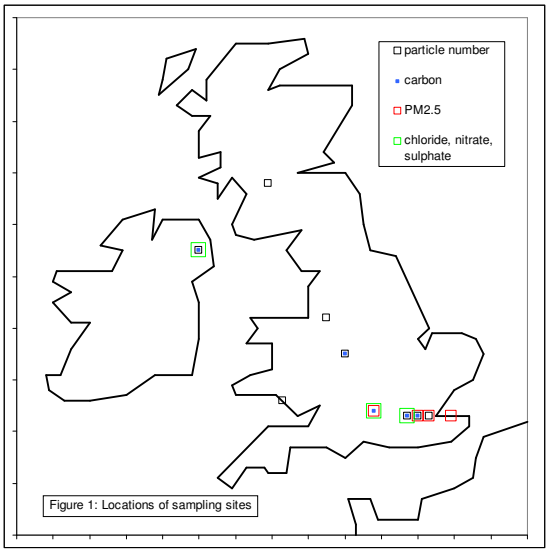

Figure 1: Locations of air sampling sites.

Comparisons of the magnitudes of the various particulate metrics, and the relationships between the metrics, enables the degree of spatial variability of particulate matter and its composition across the UK to be determined. Distinctions are drawn between measures of chemical components such as nitrate or sulphate which show a regional pattern (Abdalmogith and Harrison, 2005, 2006), and particle number concentrations which appear to be determined by more local sources (Harrison and Jones, 2005).

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SO2 remains the major determinant of air pollution-related mortality in Upper Silesian Industrial Zone, Poland

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Keywords: air pollution, mortality, time series study, epidemiology.

Impact of air pollution on human health is subject to extensive research, involving various health outcomes. Most evidence concerns daily mortality and hospital admissions in various ambient air pollution scenarios. The expert opinion from the last meeting of COST 633 action in Brussels suggests that the largest effect on air-pollution specific mortality in Europe is due to exposure to airborne $PM_{2,5}$. Ambient air pollution composition in urban areas in Poland is distinct by elevated levels of SO_2, compared to the Western EU regions, even if air quality in Poland has improved over the recent decades.

Changing air pollution profile justifies investigation into comparison of its impact on morality, over a longer period. This assumption triggered a study undertaken to compare current and earlier estimates of air pollution mortality risk (2001-2002 vs 1994-1995) in Upper Silesian Industrial Zone (USIZ). The densely inhabited area (ca 3.5 million inhabitants, ca 35000 deaths/year) is located in Katowice Voivodship, Poland. Analysis of association of daily mortality with daily concentrations ('on line' measurements) of air pollutants involved adjustment for relevant meteorological variables (atmospheric pressure, humidity and temperature). The table below shows the results of multiple-regression analysis (regression coefficients and their statistical significance for each pollutant and meteorological variable) in USIZ, in 2001-2002.

Table 1: Association of daily mortality with air pollution in USIZ, 2001-2002 (regression coefficients and their statistical significance, * - p<0.05)

Daily mortality	Regression coefficients				
	Pollution		Temperature	Humidity	Pressure
All causes	NOx	0.073	-0.28*	-0.09*	-0.07
	PM10	0.084*	-0.27*	-0.09*	-0.08*
	SO2	0.154*	-0.20*	-0.09	-0.08
Cardiovascular Diseases	NOx	0.017	-0.34*	-0.06	-0.08
	PM10	0.038	-0.34*	-0.05	-0.09
	SO2	0.129*	-0.26*	-0.05	-0.10*
Respiratory Diseases	NOx	0.062	-0.17*	-0.05	-0.03
	PM10	0.075	-0.17*	-0.05	-0.04
	SO2	0.129*	-0.11*	-0.05	-0.03

Current results confirm a predominant effect of SO_2, also seen in 1994-1995. Moreover, the risk estimates (risk of additional death ascribed to an increase in air pollutant concentration by 10 $\mu g/m^3$) appeared similar in the compared periods. Table 2 shows the results of comparisons for cardiovascular death among the elderly.

Table 2: Risk of additional death from cardiovascular diseases per day in elderly (65+) related to an increase in pollution by 10 $\mu g/m^3$ in USIZ, in 1994-1995 and 2001-2002

Pollutant	1994/1995	2001/2002
PM10	1,01	1,06
SO2	1,21	1,23
Nox	1,01	1,04

The results suggest that in USIZ ambient air concentration of SO_2 is the most important air pollutant in terms of public health (air pollution-related health disorders). The predominant effect of SO_2 should be carefully controlled in studies aiming at effects of other contaminants, such as PM_{10}, $PM_{2.5}$.

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Overview of source apportionment methods in use in European COST633 Action member countries

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Keywords: receptor modelling, ambient concentrations, exposure

The application of effective abatement strategies to reduce PM levels is only possible if emission sources are uniquely identified and characterised. Source apportionment studies attempt to identify and quantify the contribution of PM sources based on the measured ambient concentrations of different PM fractions and components at a given study site. A number of techniques are available for this purpose in Europe and the U.S.

The main objective of this work is to provide an overview of the source apportionment methods in use throughout Europe and their applications, in order to identify gaps and possible future research directions for the modelling community, as well as to provide an input for health-related studies. To this end a questionnaire was designed within Working Group 3 (Sources, Emissions, Modelling, Economic Aspects) of COST Action 633 and distributed amongst the Action's participant countries. The questionnaire was aimed to compile data on the source apportionment methodologies used in the different countries, including data on the PM fraction targeted, sampling methodology and study area. Despite the fact that replies to the questionnaires were not obtained from all the member countries, the return may be seen as a useful overview on source apportionment activities in Europe. It should be noted, however, that this overview does not provide a full set of all source apportionment studies performed in the European COST633 Action member countries.

The replies to the questionnaires reported a total of 46 publications on source apportionment in 6 countries (Austria, Belgium, Finland, Germany, Portugal, Spain, Switzerland), based on 10 different methodologies: Principal Component Analysis (PCA, Thurston & Spengler, 1985), CMB (Chemical Mass Balance, US-EPA, 1987), Positive Matrix Factorisation (PMF, Paatero & Tapper, 1994), UNMIX, ME (Multi-Linear Engine, Paatero, 1999), Lenschow approach (Lenschow et al., 2001), back-trajectory analysis, cluster analysis, COPREM (Constrained Physical Receptor Model, Wåhlin, 2003) and isotopic mass balance using C-14 (Szidat et al., 2004). The most frequently used method was PCA (30% of the studies), followed by back-trajectory analysis and the Lenschow approach (15%), and PMF (10%). The remaining methodologies were used in at most 2 studies each. Regarding the number of research groups applying the different techniques, PCA appeared to be most widely used (in 5 different countries), whereas PMF and the Lenschow approach were applied in 4 and 3 countries, respectively.

The most common target metrics in the different studies were PM10 (55% of the studies) and PM2.5 (33%), even though models were also occasionally applied to PM1, PM2.5-10, TSP and fine particles (<1 µm). Taking into consideration the increasing evidence on health effects related to the finer grain-sized fractions, these results highlight the need for further research on PM2.5 sources and source contributions. Receptor sites at which models were applied were described as urban background (51%), kerbside (18%), rural (18%, including remote and agricultural sites) and industrial (13%), indicating that current source contribution analyses are focused on urban environments (mainly residential, suburban). This raises the question on whether future research should continue in this direction or be directed towards highly polluted environments (industrial, kerbside), taking into account not only ambient concentrations but also exposure.

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Source-specific air pollution exposure characterization for a large population-based Swiss Cohort (SAPALDIA)

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Keywords: traffic exposure, dispersion modelling, GISSeveral epidemiologic studies have provided evidence of long-term effects of particulate air pollution, with evidence pointed toward the smaller particles, $PM_{2.5}$, as a more likely culprit than PM_{10}. Recently, a plethora of studies started looking into health effects related to source-specific air pollution. In particular, traffic exhaust air pollution has been linked to exacerbation of respiratory symptoms among asthmatic children and sudden cardiac arrests. While evidence on acute health effects related to traffic exhaust is accumulating, there is paucity in long-term effects of source-specific air pollution in the general population, specifically among the European populations. The goal of this paper is to evaluate whether dispersion model estimates supplemented with detailed site characteristics can provide accurate source-specific exposure estimates for ambient air pollutants.

The SAPALDIA study recruited 9,651 subjects from 2 large cities (Basel, Geneva), 2 medium-sized cities (Aarau, Lugano), 2 rural areas (Payerne, Wald), and 2 rural alpine areas (Davos, Montana). The first health examination was conducted in 1990, with a second follow-up occurred in 2002. Each area was monitored at up to 3 central monitoring sites for criteria air pollutants including PM, NO_2, NO_x, and other gaseous pollutants. In 1999-2000, a sampling campaign was conducted for PM_{10}, $PM_{2.5}$, and BS (a marker for traffic exhaust) at 16 sites including 9 background and 7 traffic sites across Switzerland. In 2002-2003, passive NO_2 measurements were collected strategically over the year outside and inside ~60 homes per area. Annual average concentrations of $PM_{2.5}$, PM_{10}, NO_2, and NO_x (source-specific and total) were estimated using a Gaussian dispersion model with GIS to match individual residences of the SAPALDIA subjects. We conducted analyses to address three major questions: 1) How dose the source-specific dispersion model perform as compared with the actual measurements? 2) How did PM_{10} and NO_2 sources vary by area and over time (1990-2000)? 3) What are the implications of these findings to long-term air pollution epidemiologic studies? The dispersion modeling results for PM_{10}, $PM_{2.5}$, NO_x, and NO_2 were evaluated against the measured values at all Swiss monitoring sites including traffic sites in 1990 and 2000, respectively, using paired t-tests and regressions. Traffic-specific model predictions were compared with measured black smoke and NO_x. Spatial variations of traffic-specific and regional transport modeling results for individual residences were examined. Because the modeling results were spatially interpolated based on 200x200 m grids, we smoothed the individual residences to certain distance from the fixed monitors to examine the actual representative range of area. In addition, we estimated annual averages between 1990 and 2000 for each individual based on the measured and modeled values. We examined the emission profiles, the source-specific pollution levels, and ambient measurements across cities and over time. Error propagation was performed to estimate the total error associated with individual exposure estimates based on the dispersion modeling approach as compared with the central-site measurements alone. This paper makes advances in evaluating predictions of exposures to source-specific air pollution, including $PM_{2.5}$, PM_{10}, NO_2, and NO_x, in a population-based cohort in Switzerland. These source-specific exposure estimates are being examined against a series of respiratory and cardiovascular health effects in other papers. The poster will present results of these analyses.

Frequency occurrence situation with increased concentration of PM10 and PM2.5 in related to assessment health risk inhabitants in Southern Poland

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Keywords: ambient air quality, human exposure, epidemiology, meteorological conditions, data mining

Particulate matter pollution is one of the most serious environmental hazards of the beginning of XXI century (CAFE, 2005, EPA, 1999; WHO, 2004) Intensive development of transport together with still insufficiently reduced industrial dust emission cause that quite restrictive admissible values of particulate matter in air are permanently being exceeded in most European countries. At the same time current epidemiological data confirm the association between PM concentration and morbidity or mortality rates-general and specific due to respiratory system and circulatory system diseases. Similar association exists for daily hospital admission rate, also in case of short term episodes of increased PM concentrations or at the levels assumed so far to be safe for humans. Developments in statistical methods and in measurement techniques, including the establishment of automatic measurement systems makes it possible to monitor the association between air concentration of different PM fraction and meteorological parameters. Main target of this work is the identification of factors determining concentration levels of PM as well as its profile (chemical composition) together with the assessment of exposure of industrialized urban population under conditions of reindustrialisation and recultivation. The work complies with COST 633 Action, and in particular the results have used for modelling aerosols particles and related to health effects.

Since 2002 year the parallel investigations of PM10 and PM2.5 fraction are presently performed at a sampling site located in Zabrze, which has representative for dense populated urban region.

The aim of this research is the classification of meteorological and air quality (AQ) condition, determination of PM fractions pattern in a given meteorological situation. As a result, there were PM concentrations that is to say groups of meteorological factors effecting on health risk. The frequency occurrence of situation responsible for cause-specific mortality and morbidity was made done by variability of contamination factors AQI (Air Quality Index) (EPA, 1999, Osrodka et al., 2005). For this purpose the selected data mining methods will be used.

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Surface characteristics of fine particles (PM2.5) in Zabrze, Upper Silesia, Poland

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Keywords: aerosol, PM2.5, reflection of filters, XPS (X-ray photoelectron spectroscopy) In the last decades the focus of studies on associations between particle mass and adverse health effects shifted from total suspended particulatese (TSP) and PM10 to finer particles such as those with a 50% cut-off aerodynamic diameter of 2.5 µm (PM2.5). The rapidly increasing number of papers on the environmental and health aspects of PM2.5 can be observed. However, especially important properties of these fine particles seems to be related with their surface. It is the surface that is directly accessible to biological fluids after inhalation or ingestion (Hutton and Willimas, 2000; Kendall et al, 2001). Besides, chemical transformations of particles in the atmosphere occur on the particle surface. However, during the last thirty years only few studies have addressed the surface chemistry of urban air particulates (Novakov et al., 1974; Craig et al., 1974; Hutton and Williams, 2000; Kendall et al., 2001; Zhu et al., 2001; Pastuszka et al., 2003; Wawros et al., 2003).

The aim of this work was to identify the surface elemental composition of the aerosol fine particles collected in Zabrze, Upper Silesia, in the years 2001-2003. Additionally, the optical properties of these PM2.5 particles have been investigated.

The information on the chemical structure of the particles surface has been obtained by X-ray photoelectron spectroscopy (XPS). The PHI 5700/660 Photoelectron Spectrometer (Physical Electronics, USA) has been used to obtain the XPS spectra of aerosol samples. The optical properties has been studied on the basis of the reflectance of the PM2.5 filter deposit, determined using a digital smoke stain reflectometer (Model 43D, Diffusion Systems Ltd., London, UK).

The analysis of XPS spectra indicates that carbon- and oxygen-containing species dominated the particulate surface with traces of N, S, Si, Cl, Na, Zn, Al, Cu, Ca, K, Mg, Pb, and P present.

Table 1 gives the monthly data of the reflectance coefficient obtained for the fine particles collected in Zabrze during 2002. These data agree well with the surface concentrations of elemental carbon. The obtained results indicate also the important role of oxides and sulfates in promoting the reflectance of light.

Table 1: The reflectance coefficient of airborne fine particles in Zabrze

Month	Jan.	Feb.	March	April	May	June	July	August	Sept.	Oct.	Nov.	Dec.
Reflectance coefficient [%]	9.9	12.1	11.8	33.2	51.8	43.7	42.5	37.9	41.2	20.8	9.9	3.7

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Seasonal variability level of PM frictions and their effect on human morbidity and mortality in Upper Silesian Agglomeration, Poland

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Keywords: aerosols particles, respiratory and cardiovascular disease, meteorological conditions, frequencies analysis

Recent studies suggested that high concentration PM2.5 in agglomeration areas was associated with significantly increased risk for many adverse respiratory outcomes like: cough without infection, bronchitis, broncholitis, pneumonia and asthma etc. (CAFÉ, 2005; WHO, 2004). The proposed project is designed to test the hypothesis that daily number of death and emergency hospital admission in Upper Silesian Agglomeration (respiratory and cardiovascular diseases) are related to exposure to particulate matter, particularly PM2.5. The project complies with main task COST 633, and in particulate with the scope working group WG1 and WG2.

The purpose of the work was performing seasonal and daily changes in concentration particulate matter and their effect on health risk population have lived in urban-industrial region southern Poland. Air quality measuring stations in 1997-2004 collected the data related to 1 hour average concentration. The continuous measurements PM10 and PM2.5 have been since 2002 year, at one sampling site, located in chosen urban area, with different emission profiles. The Fourier analysis, statistics analysis and wavelet analysis was used to analyse data. The obtained results were comparable taking into account another urban region in Europe. The results obtained confirmed the hypothesis that have done correlation (significant statistic level) between cause-specific mortality and morbidity and seasonal increased level PM2.5.

CAFE Program (2005) *Baseline Scenarios for Clean Air for Europe*. Final report. Luxemburg.
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Characterisation of particulate matter for health effect assessment in Krakow, Poland

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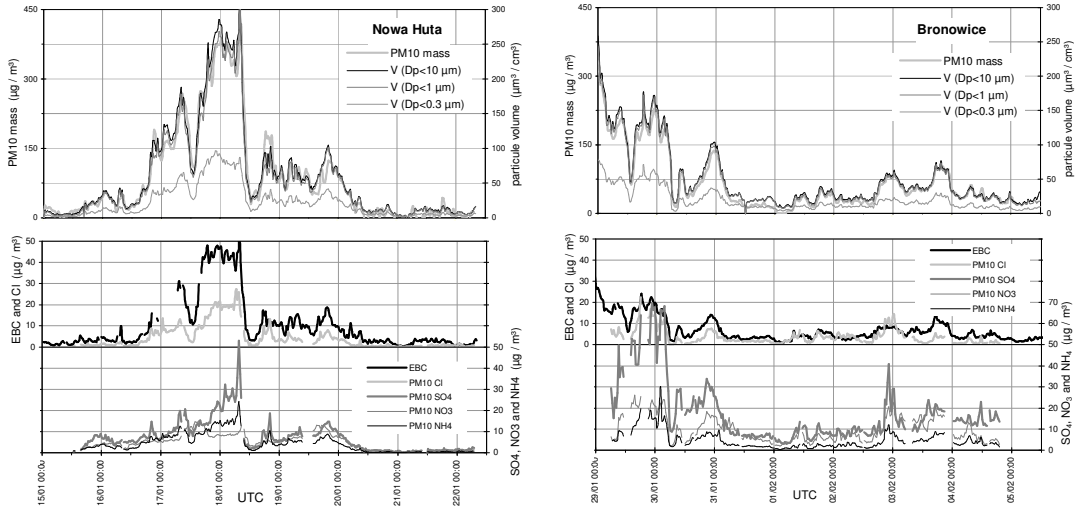
Keywords: atmospheric aerosol, PM10, on-line measurements, physical and chemical characterisation

The 24 hr - limit value for PM10 mass concentration (50 µg/m³) is frequently exceeded in large parts of the EU-25, in particular during winter time. Regulatory monitoring of the air quality revealed significant correlations between mortality, morbidity and PM10 concentrations at various locations around the world. However, the factors responsible for their harmfulness are still poorly understood. PM10 mass measurements cannot provide any hint for understanding what type of particle, size fraction or aerosol species is responsible for the health effect. For designing abatement strategies which would efficiently reduce the health effect of the aerosol, a much better knowledge of its characteristics, in relation with its sources and effects, is needed. The aim of the COST633 action is to highlight similarities and uncertainties in aerosol characteristics, sources, and effects across Europe. A way of contributing to this goal is to carry out detailed characterisations of the aerosol, which go well beyond simple PM10 mass measurements. Maximum improvement in our understanding of the health effect of aerosol may be expected when such characterisation is performed in the frame of an integrated project comprising the development of emission inventories, atmospheric chemistry and physics modelling, and health studies. This was the case of the Krakow 2005 project.

This paper presents a fraction of the ambient aerosol characterization work from a physical and chemical viewpoint. PM_{10} mass concentration and major inorganic components (Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, etc…), particle size distribution (10 nm – 10 µm), volatility, hygroscopicity, and absorption coefficient (shown as Equivalent Black Carbon concentration, EBC) were measured on-line by the EC-JRC mobile laboratory for aerosol characterisation at two sites: Nowa Huta (industrial site) from Jan 15th to 22nd, and Bronowice (near-city background site) from Jan 28th to Feb. 5th. The time resolution for these measurements was 30 min or shorter. Some of the results we obtained will be presented.



Major PM10 pollution events were captured at both sites. The first striking observation was the very large contribution of smaller particles to PM10 mass concentrations. Particle volume size distributions indeed suggest that particles < 1 µm generally accounted for > 90% of PM10 at a both sites. A second observation was the very high correlation between PM10 mass and EBC concentrations at both sites ($R^2 = 0.93$). Actually, the chemical composition of PM10 did not vary during each leg of the campaign. PM10 concentration appeared to be mainly controlled by meteorological conditions (mixed boundary layer height, precipitations). Large amounts of C and Cl^- in PM10 suggest coal burning as a major source of aerosol during this period.

Particle number was dominated by ultra-fine particles (Dp < 100 nm), and was generally not correlated with PM10 mass concentration. Non-volatile particle size distribution and particle hygroscopicity growth factor, which are key parameters for assessing the aerosol climate effect, also provide important information on the nature of ultra-fine particles, which are so important for health effects.

Acknowledgements: Local support of the VIEP staff in Krakow is acknowledged. This work is a contribution to COST Action 633.

Mass Size Distributions of Atmospheric Aerosol on Two Urban Background Sites in Prague: Meteorology Influence

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Keywords: urban aerosol, mass size distribution, influence of meteorology

The project "Comprehensive size resolved characterization of atmospheric particulate matter in Prague" was based on parallel sampling at two urban background sites in Prague. The first site was located at northwest suburbs of Prague in the campus of the Institute of Chemical Process Fundamentals (ICPF) 285 m ASL, the second one is located downtown of Prague city on the roof of the 4 storey building of Faculty of Natural Sciences of Charles University in Benátská street, 225m ASL. The distance between both sites is about 7 km. Both sites were equipped with following sampling devices and on-line instruments: scanning mobility particle sizer (SMPS) with CPC model 3022, 10 stage Berner low pressure cascade impactor (Berner and Lürzer, 1980)), PM1, PM2.5 and PM10 filter samplers and Gent SFU sampler. On-line gas analyzers have provided data of NO, NO_2, NO_x, O_3, CH_4, and non-methanic hydrocarbons. Meteorological data like temperature, humidity, wind velocity and wind direction and total sun radiation were provided by a meteo-station. The samples will be analyzed by IC, PIXE, INAA, AAS and OC/EC. Here we report the results on the influence of meteorological parameters on measured mass size distributions obtained by cascade impactors on both sites during three campaigns in winter (2 weeks 2004, 2 weeks 2005), summer (4 weeks), autumn (2 weeks) and spring (2 weeks).

The results showed very similar results for both sites despite their location. The typical influences of several meteorological parameters were found. The dilution effect of wind velocity was clear, the higher velocity the lower total aerosol concentration were found. The influence of relative humidity cause the shift in accumulation mode position, with maximum at about 0.3µm for daily average humidity smaller than 60%RH, and about 0.7 µm for RH>75%. The most interesting was the influence of wind direction (Fig. 1). We divided wind directions into three categories related to the position of suburban site. South-eastern winds were coming to the site over the city, while north-western winds were coming to the city, over the area with relatively low traffic. The other winds were unresolved. The average size distribution was calculated for each wind direction and two periods determined by average daily temperature (T<5°C, T>5°C). The contrasting picture was obtained for colder and warmer period. While during warmer period NW wind were clearly the cleanest as it was expected, during the colder period those winds were the most polluted. The similar picture was found also for downtown site. Therefore it seems, that during colder period, the local heating using coal and wood and even some waste on the border and outside the city can increase the pollution level inside the city.

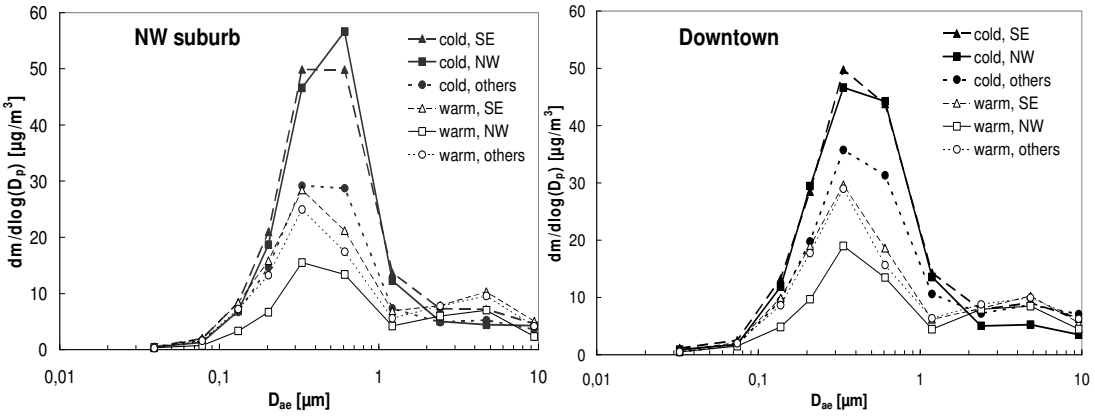

Fig 1: The influence of wind direction for average MSD of aerosols on NW suburb and downtown urban background sites of Prague for cold (T<5°C) and warm (T>5°C) period.

Acknowledgements: The support of grant GA ČR No. 205/03/1560 is greatly acknowledged.
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PM10 and PM2.5-bound PAHs in an industrial area in Northern Greece

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Keywords: PM10, PM2.5, PAHs

16 EPA Polycyclic aromatic compounds (PAHs) associated to ambient PM10 and PM2.5 were determined in an industrial area in Thessaloniki, Greece, for one week on April 2005. Particulate matter and PAHs are among the pollutants of interest within the frame of COST633.
The mean PM10 concentration was 90.84μg/m^3 exceeding the EC annual limit of 40μg/m^3 , while the mean PM2.5 concentration was 36.45μg/m^3 a value below the mean daily limit of 65μg/m^3 established by the EPA. PM2.5 standard deviation was about 4 times higher than the one calculated for PM10, indicating that the fine fraction may have a local origin (Bourotte et al, 2005).
Fig. 1 shows the mean PAH concentrations measured on PM10 and PM2.5. No exceedance of the EC annual limit of 1ng/m^3 for B[a]Py was observed. PAH concentrations on PM10 were significantly lower than those reported from Mantis et al. (2005) for the city of Athens. The 48,28% of ΣPAHs were present on the PM10 fraction while the 51.72% were on the PM2.5. Fig. 2 shows the mean PAH profiles for PM10 and PM2.5. In the PM10 fraction, the dominant compounds were DbTh (15.7%), NaP(9.54%), BghiPe(8.3%), BkFa (8.14%), and BeP(8.22%). The dominant PAHs on PM2.5 were DbTh (18.55%), NaP (7.48%) and BkFa (7.48%). PAHs with 4 (Fa, Py, B[a]A and Chr) and 6 rings (IcdP and BghiPe) were dominant on the PM10 fraction representing the 20.57% and 16.29%, respectively, of the total PAHs concentrations. These compounds were also dominant on the PM2.5 fraction (15.42% for the four- and 13.37% for the six-ring PAHs). The fine particles are emitted from the source and retained in the atmosphere for a long time. PAHs associated with particles of this size are easily transported through the upper respiratory tract into the bronchiole and alveoli of the lungs where they pose a direct adverse health impact (Zhou et al., 2005). 5-ring PAHs (BaP, BahA, BkF) exhibited the same contribution on PM10 (12.73%) and PM2.5 (12.74%).

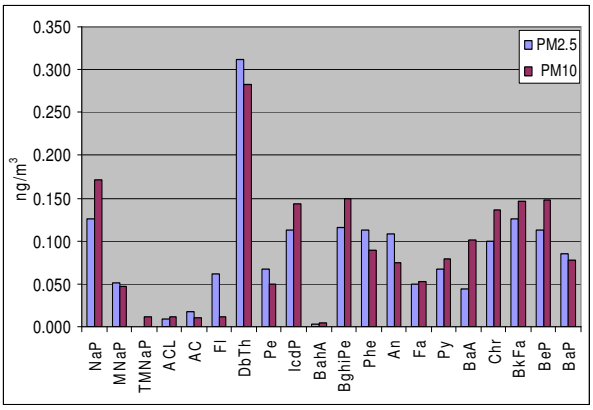

Figure 1: Mean PAH concentrations on ambient PM10 and PM2.5.

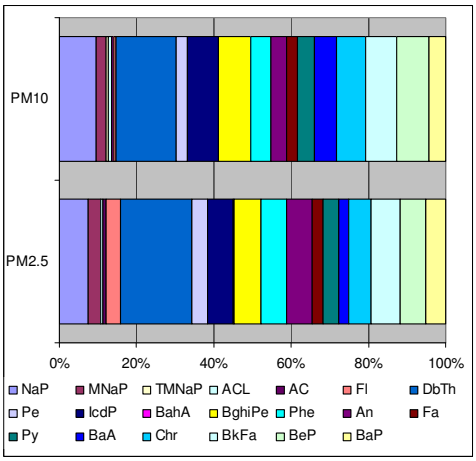

Figure 2: Mean profiles of ambient PM10 and PM2.5.

Acknowledgements: The financial support of the Municipality of New Eykarpia, in Thessaloniki is gratefully acknowlegdged.

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Size-segregated aerosol chemical composition from different sources and in different environments

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Worldwide epidemiological studies have shown a consistent increase in cardiac and respiratory diseases due to exposure to particulate matter (PM), see e.g. Samet *et al.* (2000). Air quality legislations presently regulate air concentrations of PM (PM_{10} and/or $PM_{2.5}$), but heath effects of PM also depend on particle chemical composition in the different size ranges. In fact, depending on emission source characteristics, particles may have very different chemical composition in different parts of the world and in different areas. Information is especially lacking on the amount and composition of carbonaceous particles, which are thought to be particularly harmful for human health. In order to improve our knowledge on this issue and to provide input for future regulatory legislation some efforts are underway, see e.g. the European-wide studies by Putaud *et. al.* (2004) and van Dingenen *et al.* (2004).

This study presents an extensive data-set on size-segregated organic and inorganic chemical composition of aerosol samples collected in different environments: an urban area of Europe, a high-elevation free-tropospheric site, a Saharan dust outbreak, a continental industrialized area in Asia, biomass burning aerosol in the Amazon basin, a boreal forest site, a remote marine site. Sampling was carried out by multi-stage cascade impactors and the substrates have been analyzed using the same protocol, aimed at mass closure. In particular, the following measurements have bee performed for all aerosol types in different size intervals:

- total mass;
- water-soluble inorganic ions (WS_INORG);
- total carbon and elemental carbon;
- water-soluble organic carbon (WSOC)
- WSOC speciation by function group analysis based on liquid chromatography and nuclear magnetic resonance (HNMR).

This integrated sampling/analytical approach allows a simplified representation of the aerosol chemical composition in which the size-segregated PM can be subdivided into: water soluble inorganic ions (WS_INORG), water soluble organics (WSOC), and insoluble mass (INS), calculated as the difference between total measured mass and total analysed soluble mass. In the sub-micron size range, INS mainly comprises insoluble organic species, while in the coarse size-fractions INS is mainly composed of inorganic crustal components. In particular, the technique for characterization of WSOC and inorganic ions allowed a simplied description of PM water-soluble chemical composition by a limited number of model compounds, which will be particularly useful for modeling purposes.

The high degree of variability of the relative chemical composition of organic and inorganic water-soluble components, insoluble fraction and total aerosol mass is clearly evident for the various aerosol types in the different size intervals. Despite the large differences observed, all aerosol types exhibit a high content of organic matter in the sub-micron fraction (in this size range organic matter comprises WSOC and almost the entire INS fraction).

Acknowledgements: The financial support of the European Commission (Projects SMOCC, QUEST, ACCENT) and of the Italian Ministry of Environment (Italy-USA Cooperation on Science and Technology of Climate Change) is gratefully acknowledged.

Putaud J.-P., F. Raes, R. Van Dingenen, J.P.U. Baltensperger, E. Brüggemann, M.C. Facchini, S. Decesari, S. Fuzzi, R. Gehrig, H.C. Hansson, C. Hüglin, P. Laj, G. Lorbeer, W. Maenhaut, N. Mihalopoulos, K. Müller, X. Querol, S. Rodriguez, J. Schneider, G. Spindler, H. ten Brink, K. Tørseth, B. Wehner and A. Wiedensohler (2004) *Atmos. Environ.*, **38**, 2579 - 2595.

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Physico-chemical characteristics of particulate matter in Slovenia in 2004 and 2005

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Keywords: PM10, mass size distribution, chemical composition of water soluble fraction

Besides the elevated concentrations of O_3 in summer time, the most problematic area regarding air quality in Slovenia is pollution with particulate matter. Aerosol particles are involved in many processes in atmosphere. In addition, numerous recent studies showed their negative impact on human health. The concentration and chemical composition of atmospheric particles vary with their size and depend on time and location of sampling. The fine particles (smaller than 1 µm) originate mainly from chemical processes in the atmosphere, while the coarse particles are caused by wind erosion of the soil and sea spray. Particles emitted from different combustion processes can be present both in fine and coarse size fraction.

In this contribution we will present results of the monitoring of particulate matter in Slovenia in 2004 and 2005. Additionally, during this period short term sampling campaigns were performed, focused on mass size distributions and chemical analysis of water soluble fraction.

Monitoring of particulate matter in Slovenia is performed at eight different locations by Environmental Agency of the Republic of Slovenia. Continuous measurements of PM10 are done by TEOM (Tampered Element Oscillating Microbalance), while according to European standard (EN12341) reference samplers (Leckel) are used. Size segregated sampling was performed by Berner low-pressure cascade impactors with 10 collection stages in the size range from 15 nm to 16 µm. For chemical analysis combined samples were prepared, approximately matching sizes of the ultrafine particles, accumulation particles and coarse particles. Deposits on foils were extracted with Milli-Q water in an ultrasonic bath. Chemical analysis was focused on major ions and selected trace elements (mainly toxic metals).

In 2004, the highest concentrations of PM10 particles were measured at urban-traffic locations (Maribor, Trbovlje), followed by urban background locations (Ljubljana). The lowest concentrations were measured at a rural background location (Murska Sobota). Average monthly concentrations at five different locations are presented in figure 1. At all measuring sites, the concentrations of PM10 were higher in wintertime because of the additional emissions due to heating. Moreover, meteorological conditions with frequent temperature inversions in basins (e.g. Ljubljana, Trbovlje) were also unfavorable during the cold part of the year.

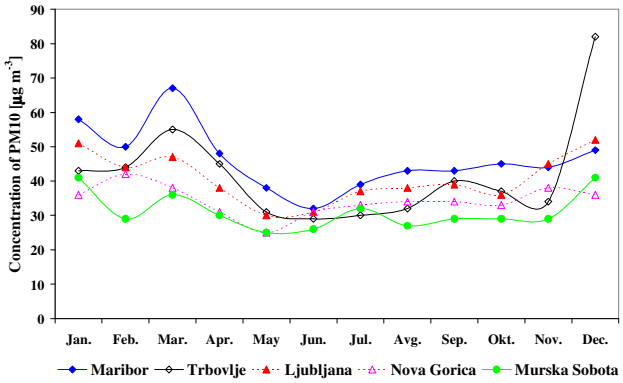

Figure 1: Average monthly PM10 concentrations at five different locations in Slovenia in 2004.

Mass size distributions showed that at all locations the accumulation mode is the most pronounced in winter time. However, the fraction of the coarse particles in cold part of the year is higher at urban locations. In summer, the mass fraction of particles in the coarse mode is higher at all locations and the difference between urban and background sites regarding the fraction of coarse particles is not so pronounced.

In winter time, the highest fraction in the accumulation fraction is attributed to NO_3^-, SO_4^{2-} and NH_4^+. In summer, the fraction of NO_3^- and NH_4^+ was significantly lower and can be attributed to losses of NH_4NO_3 during sampling. In the coarse particles the content of ions varied depending on location and season.

Acknowledgements: The financial support of Ministry for Higher Education and Science and of the Ministry for Environment and Spatial Planning of the Republic of Slovenia is gratefully acknowledged.

Characteristics of aerosol particles properties related to the human health in Vilnius

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Keywords: aerosol particles, PM_{10}, air mass backward trajectories.

The epidemiological studies have shown, that common morbidity and mortality cases related to small aerosol particles (Pope, 2000; Kreyling et al., 2004). Transport (particularly heavy duty) is the main source of aerosol particles in settled territories. Other particles formed due to nucleation, coagulation and condensation processes. The large particles are mechanically generating due to turbulence and wind erosion.

Total aerosol concentrations and size distributions were analyzed in Vilnius. During the experiment the highest concentration of particles was observed in range of small submicron particles (Dg < 0.1 μm), which are exhaust direct from cars engines. However, high concentration of pollutants in the air can trigger chemical reactions on the surface of aerosol particles and stimulate growth of these particles. From the diurnal data of gas precursors and PM_{10} a few important episodes can be separated, when very high concentrations of pollutants were observed (maximum concentration of NO_x exceed 200 μg/m^3, PM_{10} – 130 μg/m^3). For the other days the concentrations of gas precursors and PM_{10} were much lower.

It was shown, that both source of pollution (local cars transport and long-range transport) are important to air quality of Vilnius. When the air mass backward trajectories were from North over Scandinavia and Baltic Sea very low concentrations of sulphur and nitrogen compounds were observed. When the air mass backward trajectories were from South or West Europe the concentrations of mentioned compounds greatly increased.

Physical parameters, which determine formation and growth of particles, were calculated (Table 1). There can be concluded, that small particles exist in shorter time interval, because they coagulate with larger particles, which are in atmosphere. Sudden growth of particles depends on rate of condensable vapor source (*Q*), which at these days exceeded average values. The lifetime of nucleation mode particles depends on the new particles formation rate (*J*). Condensational sink (*CS*) depends on the amount of airborne condensable vapor and shows how condensation influences the growth of particles (*GR*).

Table 1: The trajectory direction and parameters influencing the formation and growth of aerosol particles

Date	*GR*, nm/h	*J*, 1/(cm^{3}s)	*CS*, 1/s	*Q*, 1/(cm^{3}s)	Trajectory direction
03.28	3.9	0.76	3.40E-03	1.82E+05	SE
03.29	2.9	1.14	3.60E-03	1.43E+05	SE
03.31	7.4	0.20	4.30E-03	4.36E+05	S
04.01	3.7	0.14	2.90E-03	1.47E+05	SW
04.02	3.7	0.50	6.90E-03	3.50E+05	W
04.03	3.9	0.64	9.80E-03	5.24E+05	E
04.04	4.4	0.98	6.40E-03	3.86E+05	SE
04.05	5.5	0.37	6.50E-03	4.90E+05	S
04.06	3.2	0.25	2.90E-03	1.27E+05	N
04.07	2.8	0.22	1.90E-03	7.29E+04	N
04.08	3.0	0.18	2.80E-03	1.15E+05	N
04.09	3.1	0.11	3.00E-03	1.27E+05	NE
04.10	4.4	0.49	5.10E-03	3.07E+05	SW
04.11	4.3	0.64	5.00E-03	2.95E+05	S
04.12	9.0	0.91	5.30E-03	6.53E+05	S

Acknowledgements: The financial support of Lithuanian State Science and Studies Foundation and FP6 project ACCENT is gratefully acknowledged.

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Modelling Particulate Matter in European COST633 Action Member States

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Keywords: numerical modelling, aerosols, ambient concentrations, exposure

Nowadays, particulate matter (PM) is considered as one of the critical air pollution problems in Europe due to the high levels of concentration reported by the majority of the member countries, mainly in urban agglomerations, where exposure and human health effects are also higher (EEA, 2005). Air quality models can provide important information allowing understanding how PM emissions and ambient air concentrations are related through the simulation of their transport and transformation in the atmosphere. Therefore, modelled PM concentration fields are a fundamental tool when exposure and health effects are under assessment and/or air quality management strategies are under definition.

The main objective of this work is to provide an overview of the application of PM models in European COST633 Action member countries, within Working Group 3 (Sources, Emissions, Modelling, Economic Aspects) activities. The purpose was mainly case-study oriented trying to summarise PM modelling application in each country.

Ten member countries replied the questionnaire reporting a total of 30 case studies applications. Primary aerosols were simulated in all the case studies, whereas secondary aerosols were included in 50% of the cases, estimating not only PM_{10}, but also $PM_{2.5}$ and SOA (secondary organic aerosol). Several models were applied, in a total of 20, with different scopes and covering different modeling scales, from the local (5 models) to the regional scale. At local scale applied models only simulate primary PM. Validation work is associated to each reported case study.

This overview work is limited to the COST633 member countries that replied to the questionnaire and does not intend, for now, to include other information sources as CITY-DELTA (Thunis and Cuvelier, 2004), or EUROTRAC-2 (Noone et al., 2002). It allowed verifying that, in general, member countries are already simulating PM and using models as useful tools to air quality management, namely in what concerns the Air Quality Framework Directive.

Acknowledgements: The authors would like to thank all the researchers who provided data for the questionnaires. This work was carried out under COST Action 633. The financial support of Calouste Gulbenkian Foundation for the grant of Joana Ferreira is gratefully acknowledged.

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A field campaign to assess air pollution effects on school children health

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Keywords: air quality, particulate matter, children health, field campaign.

Air pollution is a major environmental health problem causing approximately three million deaths per year in the world, as result of exposure to particulate matter (PM) (WHO, 2001).

Sources of exposure to particulate air pollution are many, covering not only outdoor but also indoor. Since people spend most of their time indoors, it is essential to evaluate the PM concentrations not only in open air, but also in different indoor locations, called microenvironments. Indoor sources include cooking, heating appliances and pets and, also, outdoor PM penetrating indoors, usually the most significant source in the absence of smoking (WHO, EC, 2002).

Many epidemiological studies have shown adverse health effects from exposures to airborne PM. Asthmatic children are a subpopulation of particular interest given their increased susceptibility to adverse respiratory health effects from airborne exposures due to airway inflammation and hyperresponsivenes. However, most studies examining the relationship between acute asthma in children and PM relied on measurements collected at a centrally located stationary monitoring site (Wu *et al.*, 2005).

In this context, the Portuguese national research project "Health and the Air we breathe" aims to assess and evaluate the relationship between ambient levels of atmospheric pollutants and school children's health. Viseu, an urban area located in the interior of the country and, apparently, with no air pollution problems, but which future development that could lead to an increase of atmospheric pollutants concentrations, was selected as the study area. To evaluate the influence of air pollution on 6 to 10 years old asthmatic children, a field campaign was designed covering environment and medical monitoring to this children group. Children to be monitored were selected based on the obtained answers to a school health evaluation questionnaire validated and widely applied by the ISAAC project (International Study of Asthma and Allergies in childhood - URL1). Apart from the children follow, the air quality of the study area, directly related to health effects, will be characterized. Therefore, measurements of indoor and outdoor air pollutants concentrations will be taken.

Outdoor concentrations will be measured by 3 luggage-vans, placed respectively at an urban school, a suburban school and a traffic area of the town, and equipped with a meteorological master and air quality analysers in continuum for O_3, NO, NO_2, CO, SO_2, BTX (benzene, toluene, xilene) and PM_{10}. A grid of 20 points was defined to place diffusive tubes, to be changed once a week, for O_3 and NO_2 sampling, allowing obtaining weekly average concentrations. Indoor concentrations of O_3, NO_2, BTX and formaldehyde will be taken by passive samplers left inside schools and children homes. Due to the high importance of ambient air particulate matter effects on health, additional measurements will be made, using high volume and low volume samplers for PM_{10} and $PM_{2.5}$ outdoor concentrations and indoor concentrations. During the same period, medical tests to assess children respiratory system responses will be performed at Viseu's hospital, by specialized doctors.

The results of the field campaign will be analysed aiming to combine information from children's health and ambient concentrations measured in the places where a child of 6 to 10 years old spends most of the day: school, home and outdoors. Based on the obtained relations children human exposure will be estimated.

Acknowledgements: The financial support of Calouste Gulbenkian Foundation is gratefully acknowledged.

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URL1 - http://isaac.auckland.ac.nz/

Characterization of surface functional groups present on aerosols

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Keywords: Particulate matter, aerosols.

The increase of exposure to PM_{10} and $PM_{2.5}$ (particulate matter with aerodynamic diameter smaller than 10 □m and 2.5 □m, respectively) has been found to be associated with a range of adverse health effects, including cancer, pulmonary and cardiovascular diseases. Surface characteristics (chemical reactivity, surface area) are considered of prime importance to understand the mechanisms which lead to harmful effects. A hypothetical mechanism to explain these adverse effects of particulate matter is the ability of some components (organics, metal ions) adsorbed on these particles to cause oxidative stress in biological systems (Donaldson, 2003).

In the framework of the present research project that focuses on occupational exposure to Diesel particulate, we have used a novel and promising method to characterize the surface functional groups present on aerosols that have been collected on suitably passivated filters (Demirdjian et al., 2005). This method makes use of a heterogeneous chemical reaction between the aerosol condensed phase and a gas-phase probe molecule. For each type of functional group present on the aerosol surface (such as carbonyl, acidic, basic and oxidizable groups), the interaction of an appropriate gaseous molecule specifically reacting with a single functional group is studied in a Knudsen flow reactor. The type and number of probe molecules taken up by a deposited aerosol sample, whose surface area has been previously measured, reveals the type and number of functional groups present on the aerosol surface. Furthermore, heavy metals (total iron, copper and manganese) adsorbed on aerosols have been analyzed by the Atomic Adsorption method.

Preliminary results from a field measurement campaign, where the aerosols have been collected on High-Volume Sampler for subsequent interrogation by gas-phase probe molecules, will be reported. Number and size distribution of the sampled aerosols will also be discussed.

Acknowledgements: This research is supported by the Swiss State Secretariat for Education and Research (OFES) within the framework of the COST 633 project “Particulate Matter – Properties related to Health Effects”.

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Characterisation of aerosols in Danish air

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Keywords: Size distribution, ultrafine, $PM_{10}/PM_{2.5}$, chemical composition, traffic, wood.

The overall objective of the Danish particle programme was to provide new and broader knowledge on the adverse health effects of atmospheric particles with the aim to develop strategies to reduce the adverse health effects of man-made sources. This presentation focuses on characterisation of particles and their sources, i.e. emissions, size distributions, chemical composition, transformation and dispersion of particles, which all are key issues in COST 633.

Approach. The experimental research is mainly taking outset in the Danish air quality monitoring programme, with long time series of the main pollutants. The particle studies included measurement campaigns supported by data from the monitoring programme. The combination of long time series of traditional pollutants and the special particle campaigns permits establishment of relationships between sources and the properties of the particles.

The studies. The experimental studies of PM_{10}, $PM_{2.5}$ and ultrafine particles were performed in busy street canyons, urban background and rural locations, and included chemical composition, e.g. the content of elements, elemental carbon, PAHs and other organic compounds and size distributions (10-700 nm). Emission factors of particles from traffic were estimated under actual driving conditions on the basis of the air quality measurements. Application of air quality models supported the data analysis (Ketzel et al, 2003).

Main findings. The road traffic (Wåhlin et al. 2003) and wood stoves (Glasius et al. 2005) are the particle sources, which cause the highest outdoor human exposure due to high emissions at low release heights and in urban areas that is where the population lives. Diesel vehicles are the dominating source of nano-particles and ultrafine particles. The highest emissions of particles take place from traditional diesel vehicles without filters or catalysts. The particle filters are generally very efficient (>95%) for all particles including nano-particles and ultrafine particles. In addition to the tailpipe, the non-exhaust emissions from wear of road surfaces, tires, brakes etc contributes significantly to the PM_{10} pollution from diesel as well as from petrol vehicles (Wåhlin et al. 2006). Long-range transported particles, i.e. primary inorganic particles, organic particles, and secondary particles formed by oxidation of SO_2 and NO_x emitted at the European continent, are dominating in urban background and comparable with the traffic contribution in busy streets (mass concentration, $PM_{10}/PM_{2.5}$).

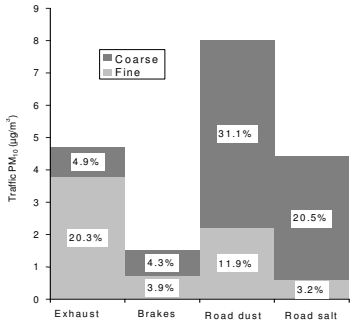

Figure 1. Road traffic contributions to fine and coarse particles in a street in Copenhagen (67,000 veh./day).

Preliminary results are published (Palmgren et al. 2003). The presentation will focus on selected examples of results about PM from road traffic and wood burning.

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Chemical variation in inhalable particulate matter from Spanish population centres: illustrating the complexity of the inorganic aerosol cocktail

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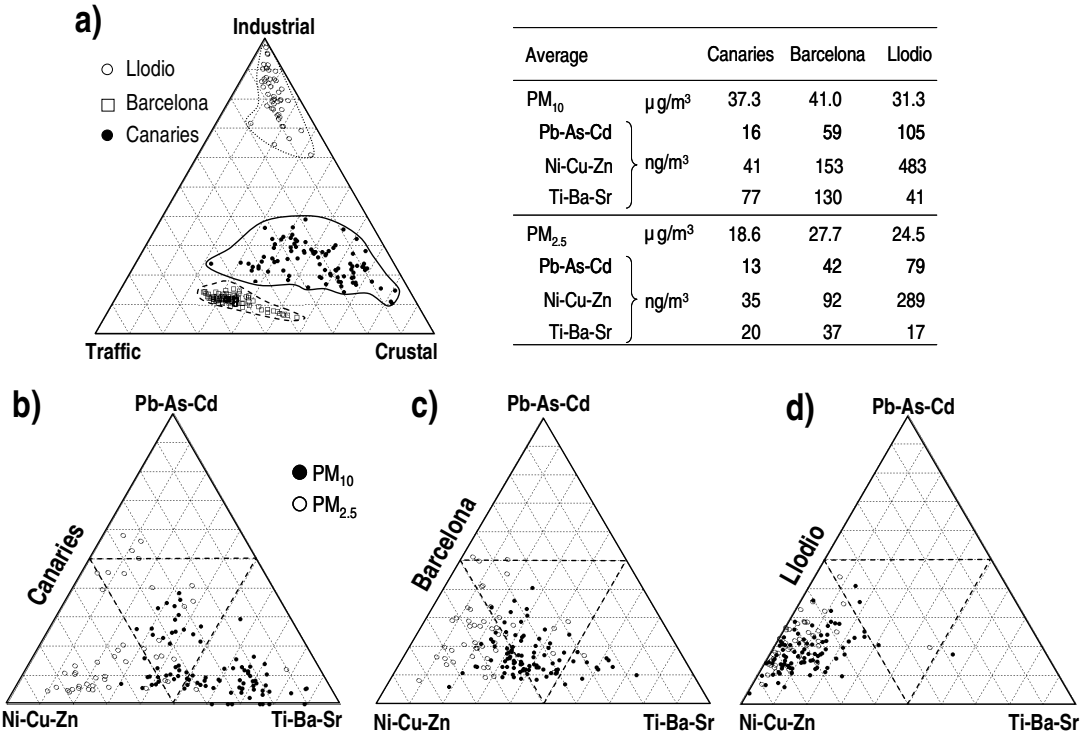
Keywords: PM chemistry, trace elements, source apportionment, health effects.

The majority of the Spanish urban population breaths air containing inhalable ambient airborne particles at average concentrations of 30-40 µg/m^3 (PM_{10}) and 20-28 µg/m^3 ($PM_{2.5}$). Source apportionment analysis demonstrates that these particles originate mainly from traffic, industry and natural materials such as crustal rock forming minerals. There are, however, large variations in the relative contributions of different PM sources between different population centres, as illustrated by the TIC triangle below (Fig. 1a) which takes three of our collection sites and compares a Mediterranean big city (Barcelona), a heavy industry town (Llodio), and the Canary Island town of Las Palmas de Gran Canaria which is frequently visited by African dust outbreaks. Such differences in PM sources inevitably result in great chemical variation: as an example Figures 1b-d illustrate differences in selected trace element contents of PM_{10} and $PM_{2.5}$. Whereas the Canaries site shows many PM_{10} analyses dominated by elements of low toxicity, such as Ti and Ba, Barcelona, and especially Llodio, are much richer in more toxic metals. Furthermore, as figures 1b and 1c also illustrate, $PM_{2.5}$ samples collected from urban sites across Spain typically show preferential enrichment in the more toxic trace metals as compared to PM_{10} from the same sites. Many such metalliferous particles emanate from primary technogenic emissions and are extremely small, with a high potential for in vivo mobility and bioreactivity. Even though the average weight of particles inhaled may be similar, marked variations in PM cocktail chemistry between towns predict that health effects will be different.



Average		Canaries	Barcelona	Llodio
PM_{10}	µg/m^3	37.3	41.0	31.3
Pb-As-Cd	ng/m^3	16	59	105
Ni-Cu-Zn	ng/m^3	41	153	483
Ti-Ba-Sr	ng/m^3	77	130	41
$PM_{2.5}$	µg/m^3	18.6	27.7	24.5
Pb-As-Cd	ng/m^3	13	42	79
Ni-Cu-Zn	ng/m^3	35	92	289
Ti-Ba-Sr	ng/m^3	20	37	17

Fig. 1: a) Source apportionment TIC triangle for PM_{10} trace elements; b-d) comparison of PM_{10} and $PM_{2.5}$ concentrations of selected trace elements with varying toxicities (high: Pb-As-Cd, medium: Ni-Cu-Zn, low: Ti-Ba-Sr).

Acknowledgements: This study was supported by the Spanish Ministries of Environment (*D.G. de Calidad y Evaluación Ambiental)* and Education and Science (project CGL2004-05984_C07-02/CLI), and research contracts supported by the Autonomous governments of Catalunya, Euskadi and the Canary Islands.

Effects of diesel exhaust particle extracts and nitrated polycyclic aromatic hydrocarbons (PAHs) on apoptotic related cell signalling that may have implications for their carcinogenic effects

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Keywords: diesel exhaust particle extracts, polycyclic aromatic hydrocarbons, apoptosis, cancer

PAHs in ambient air are often bound to particulate matter (PM) and considered to be contributing to the aetiology of human lung cancer. Nitro-PAHs are an important subgroup of the PAHs, found on diesel exhaust particles (DEP) that have been suggested to be related to the development of lung cancer. Studies indicate that several of the nitro-PAHs are found to cause mutations and tumors in animal models. Some of the PAHs are converted to reactive electrophilic metabolites that may covalently bind to macromolecules including proteins and DNA, thereby causing damage to cells. The cells will detect the damage, try to repair or trigger death (apoptosis) if too much damage is found.

In previous studies on Hepa1c1c7 cells, we have shown that some of the PAHs and cyclopenta-PAHs may trigger cell survival and inhibit pro-apoptotic signals, which suggested that the ensuing result may be that more cells may survive the DNA damage, but with an increased probability of having mutations and chromosomal aberrations[1-3]. Such findings may explain why various types of DNA damage have different mutagenic potential and contribute to the scientific basis on which a refinement of current classification of carcinogens and cancer risk assessments can be undertaken.

In the present study, we have examined the toxic effects of the nitro-PAHs 1-nitropyrene (1-NP), 1,3-dinitropyrene (1,3-DNP) and 1,8-dinitropyrene (1,8-DNP) with that of diesel exhaust particle extracts (DEPE). The potencies of the various substances including DEPE were different, but all introduced changes in gene expression and enzyme activities, as well as triggering of apoptotic and necrotic cell death. These findings suggest that the nitro-PAHs contribute to the effects of DEPE. Furthermore, considering the potency and amount of the different nitro-PAHs, the contribution of 1-NP seems to be substantial.

The cell death induced seemed to be triggered by DNA-damage resulting in phosphorylation and nuclear translocation of the tumour suppressor gene p53. These effects lead to further activation and cleavage of enzymes central for receptor- and mitochondria-related apoptosis. Furthermore, we observed an increased phosphorylation of the stress related MAPKs p38b and JNK which have been suggested to modulate the apoptotic response. The compounds also increased the level of certain cell survival signals including Akt.

Of special importance was the finding that the most mutagenic and carcinogenic compound tested, 1,8-DNP, induced less, if any, cell death, despite the fact that this compound seemed to give most DNA damage judged by increased phosphorylation of p53 and accumulation of cells in S-phase. Further immunocytochemical studies revealed that 1,8-DNP did not result in a translocation of the p53 protein into the nucleus. It is hypothesized that some genotoxic, carcinogenic chemicals induce DNA damage that "normally" should have resulted in cell death, but that they at the same time inhibit the triggering of the apoptotic process. One possible implication may be that more cells survive the DNA damage, but with an increased probability of having mutations and chromosomal aberrations. Such properties may contribute to the explanation to why certain DNA damaging chemicals are particular mutagenic and carcinogenic.

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Acknowledgements: The financial support of Aurora is gratefully acknowledged.

Interpolation methods for European scale air quality mapping and their application to population exposure estimates for PM10

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Keywords: particulate matter, interpolation, population exposure, European scale

The ETC/ACC is currently carrying out a research task for the EEA (in support of the Structural Indicator work of EEA to DG ENV) that reviews and further develops interpolation methods for use in European wide air quality mapping. In the work carried out so far emphasis is placed on the development of interpolation methodologies for both ozone and PM_{10}, with the subsequent preparation of high resolution maps that resolve urban agglomerations. For PM_{10} both the annual mean and 36'th highest daily average indicators are interpolated. The ultimate aim of the task is to produce maps that show population at risk and population weighted indicators on a European wide basis by combining the pollutant maps with population density maps.

The basic observational data used for the interpolation are taken from the AirBase and EMEP databases which provide data for the study years (2000-2003). 205 rural background and 724 urban/suburban PM_{10} stations have been used. These data are spatially interpolated using a variety of techniques to a resolution of 10x10 km.

A number of interpolation methods are tested and applied including ordinary and log-normal kriging, ordinary and log-normal cokriging, regression techniques and various combinations of these. Supplementary data used in cokriging and regression analysis include altitude, geographical position and climatic parameters as well as concentration fields calculated with the EMEP unified model. Interpolation is carried out for both the rural and urban stations separately and then combined using population weighting into a complete European map. Interpolation accuracy is assessed by examination of the root mean square error (RMSE) using cross-validation.

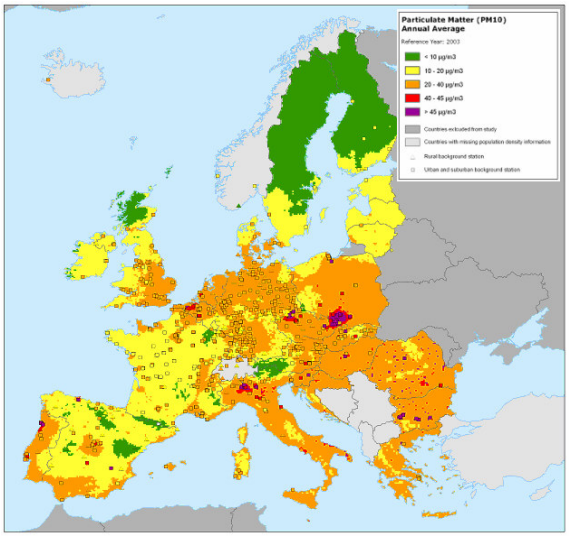

Figure 1: Example of an interpolated map showing the annual mean concentration of PM_{10} ($\mu g*m^{-3}$) for 2003. Also included are the monitoring sites used and their values.

The interpolation method chosen for PM_{10} makes use of regression relations that take into account EMEP model fields, elevation and sunshine duration. The residual, after adjustment for these regression relations, is then interpolated using ordinary kriging. The resulting fields give the lowest RMSE of all the methods tested. An example of the concentration field produced for PM_{10} is shown in Figure 1.

In this poster the techniques developed and applied for producing the maps will be briefly described and an assessment of the uncertainty of the maps given. The maps constructed will show PM_{10} concentration and indicator fields along with preliminary estimates and maps showing population weighted indicators and population at risk for PM_{10}.

Acknowledgements: The project is fully financed by the European Environmental agency (EEA)

On-line deposition of organic aerosols onto lung epithelial cell cultures – morphological analyses and cytokine responses

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Keywords: organic aerosol, on-line particle deposition, lung, epithelium, macrophages

Comprehensive epidemiological studies have shown consistent associations between the exposure to particulate air pollution (PM_{10} and $PM_{2.5}$) and acute increases in morbidity and mortality rates, especially for persons with obstructive lung and cardiovascular diseases. The interaction of ultrafine particles with the inner surface of the lungs is still poorly understood.

It is the aim of this project to investigate the interaction of ultrafine organic particles with cellular and acellular lung components ultrastructurally and biochemically. One of the main open questions is, which particle surface properties are responsible for the biological effects.

Organic particles are generated in a flow tube reactor with different degrees of oxidation serving as surrogates for freshly emitted and aged atmospheric particles. In a later stage of the project, organic particles produced from volatile precursors in a large photochemical reactor (Secondary Organic Aerosols) and diesel exhaust particles from a car test bench will also be used. As shown in Figure 1 aerosol particles of different sizes between 50-500 nm are transferred in on-line experiments to a specially designed cell-culture system enclosed in an exposure chamber. The cell culture will be representative for the inner surface of conducting airways and contain a differentiated epithelium and macrophages on the apical side. In addition, the aqueous lung lining layer will be supplemented by a surfactant film at the air-liquid interface. Cellular responses to particle exposure will be assessed by microscopic and biochemical analyses.

At first we constructed a special aerosol chamber for efficient particle deposition on cells cultured on filter inserts in multiwell plates. We tested particle deposition characteristics in the exposure chamber using fluoresecent polystyrene particles of 50-200 nm in diameter. Aerosols were transferred to the cells for 2 h. At various time points before and after aerosol exposure, the cell cultures were either chemically fixed for ultrastructural analysis or tested for survival, antioxidant depletion and changes in cytokine expression profiles.

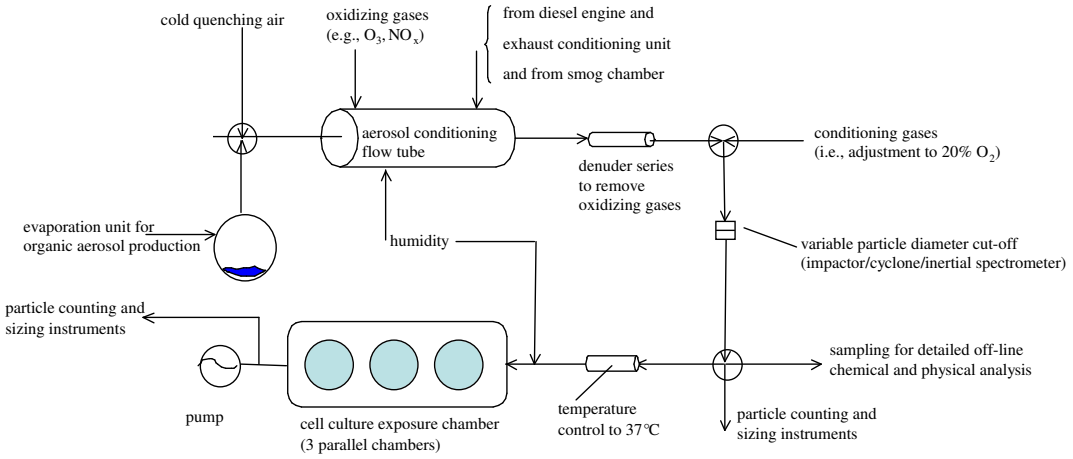

Figure 1: Experimental set-up of the on-line particle production and lung cell exposure chamber.

Acknowledgements: The technical support by Mrs. U. Gerber and the financial support of the State Secretariat for Education and Research, COST 633 grant CO3.0052, are gratefully acknowledged.

Exposure of children to particulate matter in urban areas – model calculations

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Keywords: outdoor sources, inhaled dose, particulate matter

Calculations of individual aggregated respiratory dose of particulate matter related to outdoor sources have been made for a family of four living in Oslo, the largest city in Norway (~500 000 inhabitants). Their daily routine and activity level decide the quantitative contribution to the dose in the various microenvironments. The main emission sources for particulate matter in Oslo are traffic, domestic wood burning and resuspension.

The method
A routinely used air quality management tool, AirQUIS, has been extended to provide exposure estimates based on most common microenvironments, and an estimate of aggregated respiratory dose of particles based on activity level and physiological parameters. The outdoor concentrations are calculated using an Eulerian dispersion model (e.g., http://www.airquis.no/models_dispersion.htm) and the indoor concentrations are calculated on the basis of outdoor concentrations taking into account house age and ambient wind speed. Based on defined daily routes, the hourly concentration of particulate matter is calculated for various microenvironments. The respiratory deposition for various particle sizes is calculated on the basis of the microenvironmental concentrations, activity level, gender and age, using a standard respiratory deposition model. The aggregated daily dose is calculated from the hourly values. The inhalation dosimetry module is based on a mechanistic description of particle dynamics in the human respiratory tract, using Weibel's lungs scheme and the commonly used ICRP66 respiratory tract model.

Description of calculations
22 March 2003 represents a typical winter situation in Oslo. Calculations have been made for a family of four, living in a residential area established in the 1950s in the eastern part of Oslo. The **youngest child**, aged 5, attends a kindergarten situated in the middle of the residential area exposed to both traffic and domestic wood burning. The **oldest child**, aged 10, goes to school five minutes walk from home. The school is situated next to the woods with a moderately trafficated road passing by. The **father** works at home. In the morning and the afternoon he walks with his youngest child to and from the kindergarten, taking a circular route that also passes by the school of the oldest child. The **mother** works in an office in the centre of town, and commutes by car. Her route takes her along the main roads from the residential area into town.

Results and conclusions
The calculated outdoor concentrations are comparable to the measured values, and for the 24 hour period, are between 15 and 50 microg/m^3 hourly averages (compared to between 20 and 100 for an urban background site in a more traffic-exposed part of the city). The PM10 estimated concentration for the indoor home environment is between 35 and 55% of the outdoor estimated concentration when outdoor air is the only indoor source.
The two children receive the highest respiratory doses. The main contribution to the total aggregated dose come from the time they spend outside playing. The youngest child receives the highest total respiratory dose due to his time spent outside the kindergarten playing.
The two children and their mother spend the same amount of time at home and the differences in respiratory dose at home are mostly due to lung capacity.
The results reflect the differences in concentrations in the various microenvironments, the time spent there, the activity level and the variations in respiratory capacity due to age and gender.

Acknowledgements: The work has been done with contribution from the European Communities, contract nr. EVK4-CT-2002-00090, Urban Exposure, from the Research Council of Norway, and from NILU.

Ultrafine insoluble particles bind specifically to lung-lining fluid proteins: a potential way to pass the air-blood barrier of the lung.

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Keywords: ultrafine particles, protein-particle-complex, particle surface properties

Currently there is increased interest about translocation of inhaled ultrafine insoluble particles (UFP) into systemic circulation and distribution to extrapulmonary organs. There are conflicting reports about the fractions of UFP translocating across the alveolar-capillary barrier into te blood circulation. Reported varying amounts of translocated UFP may be due to different UFP-specific chemistry and surface properties. Little is known about translocation mechanisms through the air-blood barrier of the lungs. Complexes formed between UFP and specific proteins play a role in translocation mechanisms; such complexes determine the fate of UFP following deposition in the respiratory tract. We have studied binding of lung-lining fluid proteins obtained from broncho-alveolar lavage fluids (BALF) of rats (WKY/Kyo- and SHR-rats) in an in-vitro system with different UFPs (TiO_2, Printex-90, SiO_2). In the same in-vitro system we also studied binding of human and rat blood serum proteins with UFPs. Proteins forming particle-protein complexes were separated from unbound proteins and size-analyzed by gel electrophoresis. We found clearly differing protein patterns between the blood serum of humans and rats and different UFPs. A number of specific particle-protein complexes were observed for each type of UFP after 1-hour incubation, but also unspecific proteins binding to each UFP such as albumin. The intensity of the unspecific protein bands in blood serum decreased from SiO_2 to Printex90 and to TiO_2 when the same mass concentration of UFPs was incubated indicating different protein affinities of different UFPs.
Protein-bands found for both BALF protein and serum proteins were at 95 kDa for SiO_2, at 44 and 49 kDa for Printex-90 and at 16, 24, 65, 74, 80 kDa for both UFPs. First proteins were identified using MALDI-TOF MS and verified by Western-blot technique, see table 1.
Table 1: Molecular weights (MW) of serum proteins binding to UFPs; proteins were separated by gel electrophoresis, identified with MALDI-TOF MS and verified with westernblot technique

Protein MW (kDa)	Protein (MALDI-TOF MS)	Verified by Western-blot
30.0	Apo-lipoprotein A-I	X
35.7	Apo-lipoprotein E	X
44.4	Apo-lipoprotein A-IV	
68.7	Serum Albumin	X
76.3	Serotransferrin	X

The ability to bind proteins varies widely between different particles and seems to be dependent on their surface ligands. Under physiological conditions hydrophobic particles show the highest affinity to albumin and transferrin.
Conclusion: UFP binding to proteins occurs both specifically and unspecifically; therefore, UFP translocation across the air-blood barrier is likely to depend on the formation of UFP-protein complexes. We hypothesize such protein-binding can either promote or inhibit UFP translocation.

Similarities and differences in mass and chemical composition of atmospheric aerosol at selected sites in Hungary

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Keywords: rural aerosol, urban aerosol, mass closure

Atmospheric aerosol research in Hungary dates back to 1962; regular sampling and measurements have been performed since 1982. The main locations where the studies were organized repeatedly or on a regular basis include a rural site at K-puszta and an urban site at Debrecen (both located on the Great Hungarian Plain), several urban and urban background sites in Budapest, and a rural site at the Lake Balaton (Tihany). The locations are indicated in Fig. 1. As a coordinated and joint activity of the Hungarian research teams, aerosol samples were collected in parallel at the sites indicated above, and some mobile measurements were also performed from 21 July through 2 August 2003. The samplers deployed at the fixed sites included stacked filter units, PCI impactors and Dekati impactors. The samples were investigated by gravimetry, proton-induced X-ray emission analysis, thermal carbon analysis, and ion chromatography according to identical procedures. Evaluation of the analytical data and modelling calculations are in progress. The study contributes to the European Science Fund COST Action 633 project as well.

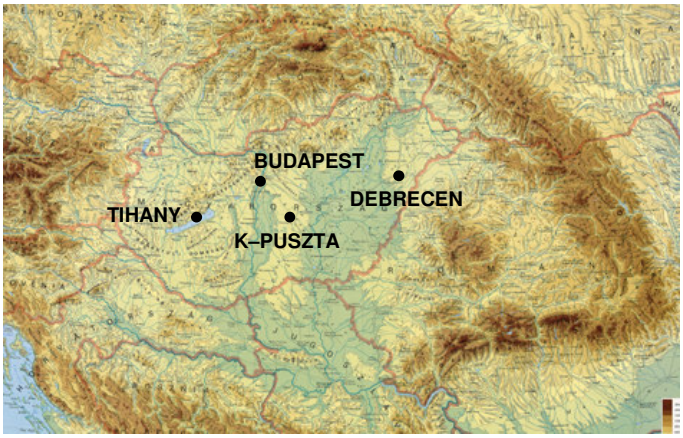

Figure 1: Location of aerosol research sites in Hungary

Some typical aerosol characteristics are summarized in Table 1. It is seen that the fine aerosol mass derived for the different sites is comparable, and the source contributions (except for elemental carbon) only show modest differences. The coarse size fraction mainly consists of crustal matter, and reflects the local source types and processes, but includes some site-specific constituents or specialties as well.

Table 1. Typical particulate matter mass concentration (in µg/m^3) in the fine size fraction, and contribution of major aerosol components to the fine mass (in %) at different research sites in Hungary

Sites	Budapest	Debrecen	Tihany	K-puszta
Particulate matter mass	23	17	12	20
Sulphate	13	24	25	17
Organic matter	43	–	38	49
Elemental carbon	21	16	–	–
Crustal matter	14	6*	–	–
Nitrate	6	–	4	6
Ammonium	6	–	5	8
Others/unaccounted	–	54	28	20

*Sum of Al, Si, P, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Br, Ba, Pb.

Seasonal and spatial variations of outdoor particle concentrations in Greater Munich

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Keywords: outdoor, particle, seasonal, spatial, variation

Considering health aspects [1] and the new EU-Guideline it is of main interest to know more about variance of particles. Within the PIA-Study (real-time parallel indoor and outdoor particle measurement) we measured particle concentrations at 10 locations in Greater Munich for one week each season. We analyzed seasonal and spatial variations like Gehrig et al. [2] and Monn et al. [3].

In Figure 1 we present the means and 95% CI's of around 2,000 measurements (5 min) from five different locations (#2, 5, 12: urban, #8: urban with heavy traffic and #9: rural). Data are evaluated at each season (**sp**ring, **su**mmer, **au**tumn, **w**inter) for the size fraction 2,0 – 3,0 µm. Only at location 5 we don't get a significant difference between spring and summer. At all other locations and seasons the seasonal variations were significant.

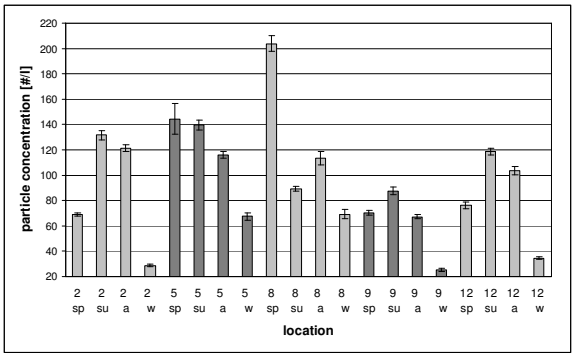

Figure 1: Mean (95% CI) particle number conc. (0.2 – 0.3 µm)

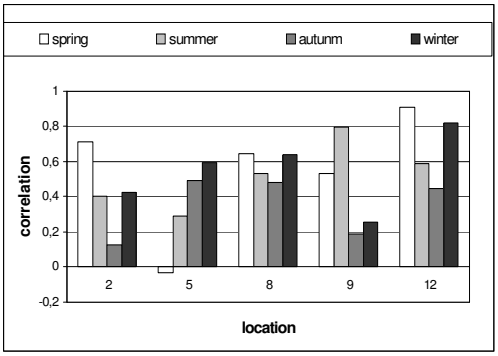

Figure 2: Correlation with State Site (3h-means).

To look for spatial variations we correlated 3hours means of size fraction 2,0 – 3,0 µm with PM10-means of a central located State Monitoring Site (Figure2). Only in some cases (2 sp, 8 sp, 8 w, 9 su, 12 sp and w) we found good correlations compared to local data. Interestingly, we got a good correlation with the rural location (# 9) on summer. At location 12, which was located at a distance of 15 m around a corner, correlations were reasonable.

ANOVA statistic showed significant seasonal variations for all size fractions from 0.3 - 0.4 µm until 5.0 - 7.5 µm, but only indifferent spatial variations.

Acknowledgements: The Bavarian State Ministry for Environment, Health and Consumer Care supported the PIA study.

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2. Gehrig, R. and B. Buchmann, *Characterising seasonal variations and spatial distribution of ambient PM10 and PM2.5 concentrations based on long-term Swiss monitoring data.* Atmospheric Environment, 2003. **37**(19): p. 2571-2580.
3. Monn, C., *Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended particulate matter, nitrogen dioxide and ozone.* Atmospheric Environment, 2001. **35**(1): p. 1-32.

A Selective Particle Size Sampler for Biological Exposure Studies of Diesel Particulate

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Keywords: Diesel exhaust, biological exposure, inertial impactor, diffusion battery.

Diesel exhaust from modern engines has a typical particle size distribution between 10 nm and 400 nm, and a mean mobility diameter of approximately 100 nm. Moreover, it is known that the particle deposition fraction on the lungs is highly dependent on particle (Oberdörster, 1994). For the study of the above dependence and the assessment of the environmentally related health issues of airborne pollutants – such as diesel exhaust - a new in vitro screening tool has been designed. This tool is a Selective Particle Size (SPS) sampler that can be used for biological exposure studies in conjunction with appropriate sample dilution and conditioning.

The selection of particles according to their size can be effected combining two different mechanisms: diffusional deposition in a multi-channel Diffusion Battery (DB) (Knutson, 1999) and inertial separation under low pressure conditions in an impactor (I). In order to adapt the two techniques for the design of the SPS sampler according to required specifications of collaborating biologists regarding desired particle sizes, concentrations and sample amount, the effect of several operational parameters was studied.

For the DB a sensitivity analysis was performed in terms of the operational flowrate, length, temperature, and aerosol concentration. The final design of the DB is based on an array of square channels. For the selected aerosol flowrate of 0.3 lpm particles are deposited on the channel walls due to their random Brownian motion and this shifts the mean mobility diameter of the aerosol size distribution at the exit from 90 nm to 130 nm. Additionally, the resulting particle size distribution is narrower than the original one, since the geometric standard deviation of the latter is σ_g=1.75 and the DB delivers a size distribution with a σ_g=1.5.

In order to choose the design and sequence of the impactor plates (Kwon et al., 2002), a detailed study of the impactor's geometric characteristics, such as the number of nozzles and the nozzle diameter, was performed. The final design consisted of a three-stage system, based on the combination of a 19-nozzle impaction plate with nozzle diameter of 0.3 mm, followed by a 21-nozzle impaction plate, with nozzle diameter of 0.3 mm and finally by a 27-nozzle impaction plate, with nozzle diameter of 0.3 mm. All the plates were constructed of stainless steel. This system shifts the original mean diameter of the size distribution from 90 nm to 50 nm while the geometric standard deviation decreases to σ_g=1.6.

Apart from the two main separation devices the SPS sampler contains a number of auxiliary aerosol conditioning components. In order to dilute the initial aerosol sample, a two-stage dilution system was developed. Mass flow and temperature controllers control the different flows to achieve good and repetitive separation performance.

The prototype SPS sampler developed combines the two separation techniques resulting in a continuously operating device connected to the exhaust line of a diesel engine and delivering on demand either a particle distribution of "large" or "small" size. The selected "large" particles are provided by tuning the DB and MI operating conditions.

By applying the SPS sampler to diesel exhaust it is possible to obtain two widely separated size distributions for size-related biological exposure studies. The SPS is currently employed for size-depended toxicological and DNA alternation studies by the group of Prof. J. P. Morin at INSERM (Rouen, France)

The development of the SPS sampler is supported in part by the European Commission Quality of Life Project: "Multidisciplinary Approach to Airborne Pollutant Health Related Issues: Modelization with Combustion Engine exhausts" (MAAPHRI).

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Kwon S. B., Kim M. C. and Lee K. W., (2002). *Effects of Jet Configuration on the Performance of Multi-Nozzle Impactors*. Journal of Aerosol Science, 33 (6), 859-869.

Oberdörster, G., J. Ferin, B.E. Lehnert, (1994). *Correlation between particle size, in-vivo particle persistence, and lung injury*. Environ. Health Perspect., 102 (S5), 173-179.

Monthly and Annual Averages of Diurnal Pattern of PM(10-2.5) at Urban and Rural Sites in Austria

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Keywords: $PM_{(10-2.5)}$, diurnal variations, contribution by local traffic

Recently a comprehensive aerosol study – the Austrian AUPHEP project – has been finished. First results, among them diurnal pattern of PM10, PM2.5, and PM1 for one-year periods, have been presented (Gomišček et al. 2004). The PM signals were sampled by TEOM instruments operated side by side at a site; so the $PM_{(10-2.5)}$ can be derived from the PM_{10} and $PM_{2.5}$ by subtraction.

The $PM_{(10-2.5)}$ fraction of the atmospheric aerosol is important because its relation to automobile traffic. As has been remarked earlier the aerosols emitted by traffic possess a strong coarse mode with modal sizes at about 4 µm and geometric standard deviations around 2.8 (Berner et al. 2004). The $PM_{(10-2.5)}$ fraction covers this mode almost entirely.

As demonstrated by figure 1, the annual and monthly averages of the diurnal $PM_{(10-2.5)}$ pattern exhibit rather low concentration after mid night until the early morning hours, a mode around 9:00 a.m. and an other mode around 6:00 p.m. This pattern is typical for traffic density in a city.

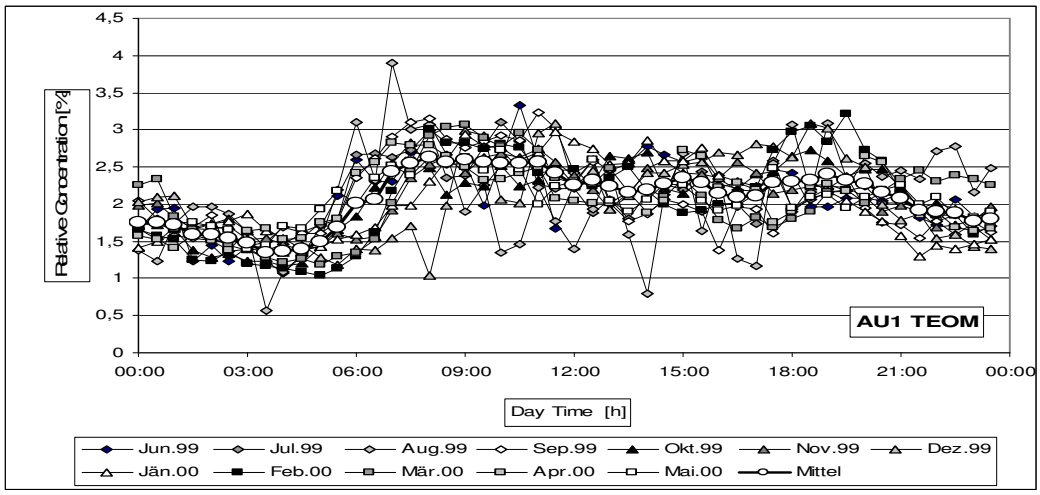

Figure 1.
Monthly and yearly averages of $PM_{(10-2.5)}$ concentrations at the urban site in Vienna.
The concentrations are given in percentage of the daily total concentration, the sum of 48 1/2h $PM_{(10-2.5)}$ values.

Conclusion: When the low concentrations around 3:00 a.m. represent the $PM_{(10-2.5)}$ fraction of a back ground aerosol which would not exhibit a diurnal pattern, this aerosol would contribute about 65% to the daily $PM_{(10-2.5)}$, on the annual average. Local traffic would contribute another 35%. During the day the $PM_{(10-2.5)}$ concentrations are about 1,7 times higher than the concentrations of the $PM_{(10-2.5)}$ back ground.

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Berner, A., Galambosm, Z., Ctyroky, P., Frühauf, P., Hitzenberger, R., Gomišček, B., Hauck, H., Preining, O., Puxbaum, H. (2004). *Atmospheric Environment 38:3959-3970.*

Source apportionment of carbonaceous aerosols with 14C

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Keywords: source apportionment; environmental radiocarbon; organic aerosol

Carbonaceous particles are a major component of the fine aerosol. They originate from anthropogenic (mainly from fossil fuel combustion and biomass burning) and biogenic emissions. For the identification and quantification of these sources, many elemental and organic molecular tracers have been employed, but their reliability often suffers from limited atmospheric lifetimes due to their chemical reactivity and highly variable emission factors (Fuzzi et al., 2005). Thus, there is a large uncertainty about the importance of anthropogenic emissions for the total carbonaceous aerosol burden of the atmosphere. In contrast to these tracers, radiocarbon (^{14}C) determinations enable a direct distinction of contemporary and fossil carbon in ambient aerosols, because ^{14}C has decayed in the latter material. Therefore, this isotopic method offers a unique possibility for unambiguous source apportionment of carbonaceous aerosol particles with the potential to point out similarities and differences of particle composition over Europe within COST 633.

The carbonaceous aerosol (total carbon, TC) was differentiated into elemental carbon (EC) and organic carbon (OC) according to Szidat et al. (2004a) and ^{14}C was measured with accelerator mass spectrometry (AMS) (Szidat et al., 2004b). EC is introduced to the atmosphere either from fossil fuel combustion or biomass burning. Consequently, ^{14}C measurements allow a direct apportionment of both sources for EC. Unfortunately, this simple two-source model cannot be applied to OC: it neglects the fact that biomass burning and biogenic emissions jointly contribute to the contemporary carbon fraction. In order to distinguish between these sources, the biomass burning fraction of OC was estimated from the respective fraction within EC using an average EC/OC emission ratio. Excess contemporary OC is defined as biogenic OC, comprising primary and secondary components.

This advanced model was applied to ^{14}C measurements of carbonaceous particles from different sites in Switzerland and Sweden. Urban background aerosols from Zurich showed that EC originated nearly exclusively from fossil fuel usage during summer, whereas biomass burning emissions from residential heating became substantial during winter with ~25%, even though this process contributes only marginally to the local energy production; for OC, biogenic sources were dominant in summer and less pronounced in winter. First results from urban particulate matter from Gothenburg suggest smaller importance of biomass burning during winter compared to Zurich. On the other hand, organic emissions from residential wood heating were identified as the dominant source for two rural sites in southern Switzerland during winter, contributing up to 90 % to the total ambient particulate matter.

Fuzzi S. et al. (2005) *Atmos. Chem. Phys. Discuss.*, **5**, 11729-11780.
Szidat S. et al. (2004a) *Radiocarbon*, **46**, 475-484.
Szidat S. et al. (2004b) *Nucl. Instr. Meth. Phys. Res.*, **B 223-224**, 829-836.

Biological responses to particulate matter in compromised rats: the role of transition metal and polycyclic aromatic hydrocarbon contents

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Keywords: particulate matter, cardiorespiratory effects, susceptibility

Statistically significant associations have been shown for ambient particulate matter (PM) and morbidity and/or mortality with higher risks for diseased and elderly. The heterogeneity in effects found in epidemiological studies might be explained by the composition of the air pollution mixture and especially by specific components like polycyclic aromatic hydrocarbons (PAHs) or metals.

The objectives of this study were (a) to investigate *in vivo* (cyto)toxicity, inflammatory activity and systemic toxicity in compromised animals exposed to coarse ($PM_{10-2.5}$) and fine ($PM_{2.5-0.1}$) PM and (b) to relate the outcomes to the chemical composition in particular polycyclic aromatic hydrocarbon (PAH) and transition metal content.

PM samples were collected using a high-volume cascade impactor in six European cities representing typical contrasting situations in urban PM pollution. The selection of PM samples was based on results of preceding *in vitro* studies and *in vivo* experiments in healthy animals. Spontaneously hypertensive rats were exposed to a single PM dose (7 mg/kg of body weight) by intra-tracheal instillation and biological effects were investigated at 24 hours after exposure.

Exposure to a PM sample with high metal or PAH content resulted in elevated lactate dehydrogenase, and protein levels as well as increased neutrophil numbers in bronchoalveolar lavage fluid compared to exposure to a PM sample with respectively low metal or PAH content. In addition, blood coagulation was positively related to metal or PAH content but there was no result on other health effect markers in the blood. In general, samples with PAHs bound to particles induce stronger effects than particle free extracts. This study showed that PM samples with higher metal or PAH contents induce stronger cellular toxicity, inflammatory activity, and blood coagulation. The outcomes of the present study support the hypothesis that on an equal mass basis, particle toxicity differs due to composition.

Acknowledgements: This study formed part of the EU 5th-framework-project PAMCHAR (QLK4-CT-2001-00423) and the financial support of the European Commission is gratefully acknowledged.

Preparatory Work for Optimised European Air Quality and Health Effect Monitoring (EURAQHEM)

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J. Cyrys[3)4)], S. v. Klot[4)], H.E. Wichmann[4)5)], A. Peters[4)]

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Keywords: Europe, Air Pollution, Exposure Assessment, Health Effects

The European Commission project "Analysis and Design of Local Air Quality Measurements" has the objective to provide recommendations for new or modified legislation so that air pollution assessment will be more health relevant and that health effects related to air pollution can be assessed in an adequate way. The project includes

a) analysis on the health relevance of the current air pollution assessments based on monitoring and modelling as conducted in air pollution networks, monitoring strategy and assessment methodology,
b) analysis on the current assessments of air pollution related health effects,
c) proposal of a design of a network and methodology for the assessment of health effects by air pollution and
d) proposal of a design for the systematic assessment of health impacts by air pollution.

Within this project a scheme (Figure 1) was developed on how a European wide improved monitoring network, air pollution data and health impact assessment could be achieved. This concept will be presented and discussed. In addition, several areas of future research and development tasks related to such an improved European wide Health Impact Assessment (figure 1) have been identified, e.g.

- need of accurate information on spatial representativeness of measurement sites and air pollution data for exposure assessments
- need of information on personal exposure from different origins (outdoor versus indoor) and different sources,
- use of updated concentration-response function derived from studies outside Europe for Health Impact Assessment to assess the impact of current efforts to improve ambient air,
- need of identification of the most relevant PM characteristics and their concentration-response functions

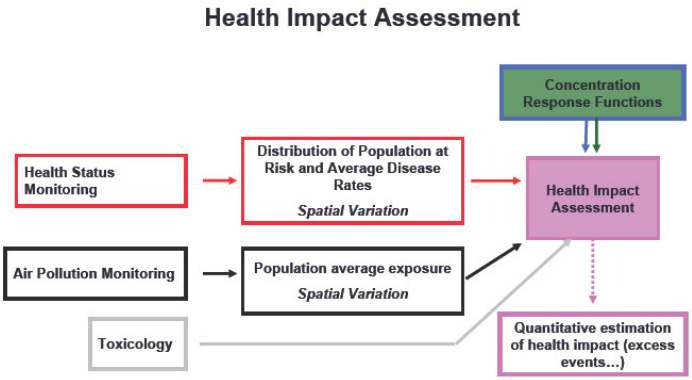

Figure 1: Information needed for health impact assessment

Analysis and design of local air quality measurements, Towards European Air Quality Health Effect Monitoring, T. Kuhlbusch, A. John, A. Hugo, A. Peters, S. von Klot, J. Cyrys, H.-E. Wichmann, U. Quass, P. Bruckmann, Report to DG ENV, http://www.iuta.de/Verfahrenstechnik/Luftreinhaltung/euraqhem_final_report.pdf, April 2006.

The chemical composition of particulate matter in Cork city centre

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Numerous studies worldwide have highlighted the detrimental effects of airborne particulate matter (PM) on human health. The toxicity of airborne PM is dependant on two factors; size and chemical composition. Generally the smaller the particle the greater the toxicity, leading to the classification of PM_{10} (particulate matter with a diameter less than 10 µm) and $PM_{2.5}$ (less than 2.5 µm). PM_{10} passes the initial clearance mechanisms in the nose and throat and enters the lungs, so is termed the 'inhalable' fraction. $PM_{2.5}$ reaches the sensitive gaseous exchange regions of the alveoli, so is termed the 'respirable' fraction. The exact mechanism by which airborne PM exerts its toxic effect remains unclear. However, increasing the concentration of metals has been found to increase lung injury and acidity may play a role in increasing the bioavailability of these toxic components.

Seasonal sampling of $PM_{10\text{-}2.5}$ and $PM_{2.5\text{-}0.1}$ was undertaken using a high volume cascade impactor (HVCI) in Cork city centre, a background urban site and a background rural site. Microwave digestion coupled with inductively coupled plasma atomic emission spectroscopy (ICP-AES) was used to analyse 15 crustal and anthropogenic trace metals (Ca, Fe, Ni, Zn, Mg, Pb, Mn, Cr, V, Cd, Cu, Si, As, Ti, Al) and an aqueous extract was also analysed to quantify the solubility (bioavailability) of the different metal components. Ultrasonic water extraction followed by ion chromatography (IC) was utilised to quantify seven anions (fluoride, chloride, bromide, nitrite, nitrate, sulphate and phosphate) and six cations (lithium, sodium, ammonium, potassium, magnesium and calcium) and the relative acidity was measured using the sulphate to ammonium ratio. This information on chemical composition will be vital in an upcoming source and distribution modelling study due to commence mid 2006.

Spatial and temporal variability of PAH concentration in Cork urban air.

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According to the World Health Organization, 4–8% of deaths occurring annually in the world are related to air pollution. The main pollution sources are associated with anthropogenic activities and the emission of gases and particulate matter. The particles are generally classified by their size rather than composition. Thus "coarse" particles have aerodynamic diameters larger than 2.5 μm and "fine" particles possess diameters lower than 2.5μm. Particulate matter can contain a wide range of chemical species ranging from elements (*e.g.* carbon, trace metals, silicon) to inorganic ions (e.g. nitrate, sulphate) and organic compounds such as polycyclic aromatic hydrocarbons (PAHs). PAHs have started to be widely studied and a special concern is being paid to environmental subjects due to their carcinogenic and mutagenic properties.

In this framework, the purpose of this study is to assess the spatial and temporal variability of PAH concentration in Cork urban air by measurements obtained from two different sampling campaigns in 2001 and 2005. The samples were collected using two types of samplers, a high volume cascade impactor (HVCI) and a dichotomous Partisol sampler. Both samplers separate the collected PM_{10} into the coarse ($PM_{10\text{-}2.5}$) and fine ($PM_{2.5\text{-}0.1}$) fractions. Three locations were chosen for the sampling programme including the city centre, a "background urban" site and a "background rural" site. Soxhlet extraction followed by sequential elution through a silica gel solid phase extraction column and gas chromatography-mass spectrometry (GC-MS) has been used to analyse a range of organic compounds including Polycyclic Aromatic Hydrocarbons (PAHs).

Particulate matter characterization to different altitude.

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Keywords: particulate matter, chemical-physical characterization, vertical dispersion

Air pollution interest is high, because this phenomenon influences public health. Air is the world breath, but many people inhales a new air because evolution of healthy-unhealthy air. The present study is turned to sampling and chemical physical characterization of particulate matter (PM), a dangerous polluting for the cardiac (Gonzalez-Flecha,2004) and lung disease (Englert,2004).

Today the legislation doesn't consider the movement of particles in atmosphere and their chemical composition [3]. The aim of this study is to estimate the particulate matter concentration at the different altitude of the ground. We have collocated two samples: the first sample at 3 meters and the second at 12 meters from the ground, with an air flow of 20 lit/min and a sampling time of 11h. The particulate concentration has been determinate by gravimetric method. The physical characterization (size and morphology) has been determinate by microscopy electronic and the chemical composition has been determinate by atomic adsorption. We have observed that the particulate size is in function of the different height. Figure1 shows a spatial particulate dispersion with a greater concentration at 3 meters. Figure 2 shows the adsorption atomic results. We observe heavy elements at 3 meters.

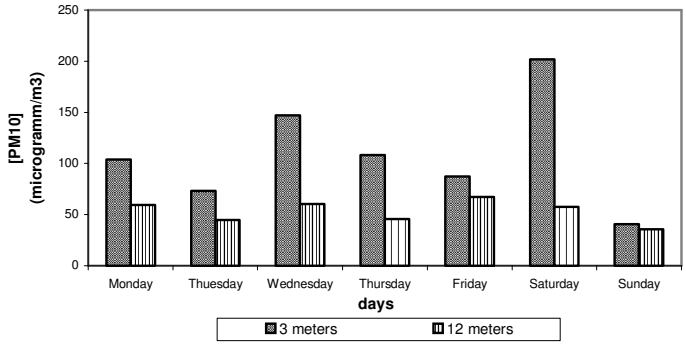

Figure 1 - PM Concentration

In both cases the analysis with electron microscopy shows that the particulate has an aerodynamic diameter less than 10 micrometer (PM10), with a different particulate size at different altitude. Infact at 3 meters we have observed particles with a diameter greater than 2.5 micrometer, while a 12 meter we have observed particles with a diameter less than 2.5 micrometer. In particular at 12 meters the prevailing particulate is PM1 (particles with an aerodynamic diameter less than 1 micrometer). PM1 are most dangerous for public health, because they penetrate deeply in lungs. Figure2 shows particulate matter sampled at 3 meters. Figure3 shows particulate sampled at 12 meters. The analyses of this phenomenon suggest producing a legislation that considers the particles dispersion.

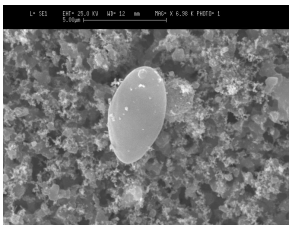

Figure3 –PM example at 3 meters

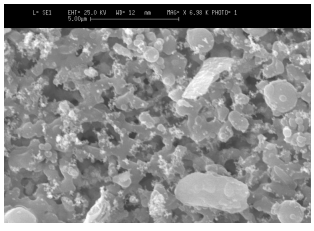

Figure2- PM example at 12 meters.

[1] Gonzalez-Flecha B., (2004), *Molecular Aspects of Medicine*, **25,** 169-182
[2] Englert N., (2004) Toxicology Letters,**149**, 235-242.
[3] Council Directive 96/62/EC

Source Apportionment of Fine particulate matter in the Netherlands

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Adverse health effects have been associated consistently and coherently with particulate matter. To reduce health risks European directives have recently been implemented in the legislation of the member states. As in many countries limit values are exceeded abatement strategies are being designed on a local, national and European scale both directed on technology as well as the volume of emissions. The central issue in developing an abatement measure and the assessment of its impact is the contribution of the source to the total concentration. On the other hand, the ongoing statistical and epidemiological studies are using concentration data of air pollution parameters, such as PM10. Knowledge on the substantial contributions of sources would open the identification of harmful emission and their association with health effects.
Source apportionment methodology offers ways to assess the contribution of sources to particulate matter. In recent years source apportionment methods are developed and evaluated that enable the identification of source profiles without laborious and therefore costly emission profiling, an important drawback of methods such as Chemical Mass Balance. A method that will provide profiles with direct measurement is Positive Matrix Factorization (PMF).
A study, designed to acquire information on the composition of particulate matter in the Netherlands, both the fine and the coarse fraction, produced data that is used for source apportionment. This paper describes the results using PMF on PM2.5 composition data. PM2.5 was either sampled over one day each sixth day or its concentration continuously monitored over a period of one year (9-1998 – 9- 1999) at 6 sites in the Netherlands, characterized as marine/rural, agricultural, street, city-background and industrial. PM samples were analyzed for mass, for the elemental composition (XRF), the ionic content (IC) and carbon (EC/OC). A selection of the samples was analyzed with scanning electroscopy – XRF. Seven source profiles could be identified, including traffic, two distinct profiles for long range transport (consisting mainly of inorganic secondary aerosol), sea spray, crude oil combustion, and one unidentified. The traffic and sea-spray profiles contained some crustal material indicating that both traffic and wind force creating sea spray also resuspended dust from the soil and road surface. Assuming a constant correlation between the crustal material in the fine and the coarse fraction the profiles and their contribution are separated further.
Both the long- range-transport profiles together with the traffic profiles could explain 60% of the average observed PM2.5 mass.
The paper describes the monitoring campaign, the imputation of the data set and its processing using PMF to arrive at the source contributions for the different sites and over the entire study period. Wind roses are calculated to give more credence to the obtained results.
The results of this study show the feasibility of source apportionment methodology and its potential support for policy decisions.

A Web based Data Analysis and Decision Support System for Particulate Matters

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Keywords: Decision Support System, Data Analysis, Forecast.

Introduction

E2SP (Environmental Enterprise Service Provider) online reporting and forecasting platform, funded by the European Commission in eTEN programme, supports environmental scientists and agencies in fulfilling their reporting and data analysis activities, by providing a cost effective, Internet based data processing platform in ASP (Application Service Provider) mode. E2SP supplies a wide range of environmental management tasks (integrating data, data validation, data analysis, reporting, modelling, etc.), with a specific focus on PM_{10} and $PM_{2,5}$. Among E2SP partners, the Municipality of Bari, the University of Bari (Italy), IPIS-PAN (Poland, COST633 partner) and Upper Silesia Regional Inspectorate for Environmental Protection (WIOS, Poland).

On line data analysis, reporting and forecast for Particulate Matters

Air quality monitoring networks and laboratories generate huge amounts of data about pollutants and PM. Collected data are aggregated to build indicators, made available through reports, whose aims are to fulfil legislative requirements, to build an historical view of air quality and to disseminate accurate environmental information. These activities, joined with accurate forecasts, are fundamental in the environmental decision processes. E2SP is a Web based pilot service covering PM reporting, data analysis and forecasting. Environmental analysts can (figure 1):

- validate and investigate data according to "free" and personalized analytical paths (multidimensional OLAP),
- create new combinations of environmental indicators and indices, according to various space-time aggregation criteria,
- analyse PM dispersion in atmosphere through mathematical models.

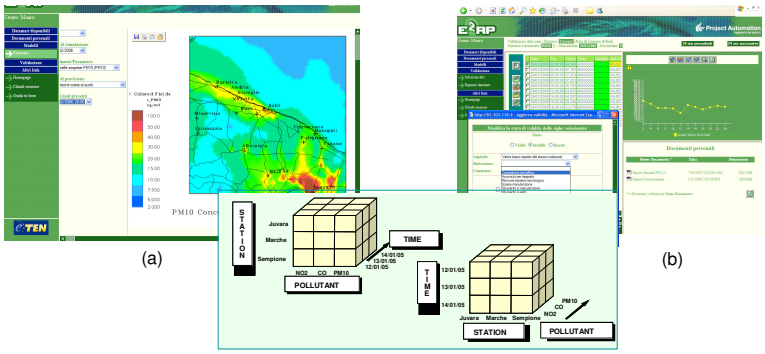

Figure 1: Examples of (a) PM_{10} forecast and (b) PM_{10} and $PM_{2,5}$ data validation and (c) multidimensional PM_{10} and $PM_{2,5}$ data analysis

E2SP forecast system produces daily forecasts of particulate matters, primary and secondary pollutants. It uses MM5 meteorological model to drive different dispersion models: the multi-scale chemistry-transport models CHIMERE and FARM gridded Eulerian model, and the Lagrangian Particle model SPRAY. PM_{10} forecasts, jointly with O_3, NO_2, CO wind fields and temperatures, are supplied on daily basis.

Acknowledgements
The financial support of the eTEN programme of the European Commission is gratefully acknowledged.

M. Cislaghi, K.Klejnowski, A.Bucko-Serafin et al., (2005) in EnviroInfo2005 proceedings ISBN80-210-3780-6, **899** : *E^2SP : a new environmental reporting platform in ASP mode.*
G. De Gennaro, (2006), TAU International conference, Milano, *E^2SP: uno strumento web di supporto alle decisioni per la gestione della qualità dell'aria.*
E2SP project website (2005), URL *e2sp.esaprojekt.pl.*

High Resolution Measurements of Particle Fluxes of Particulate at Street & City Scales

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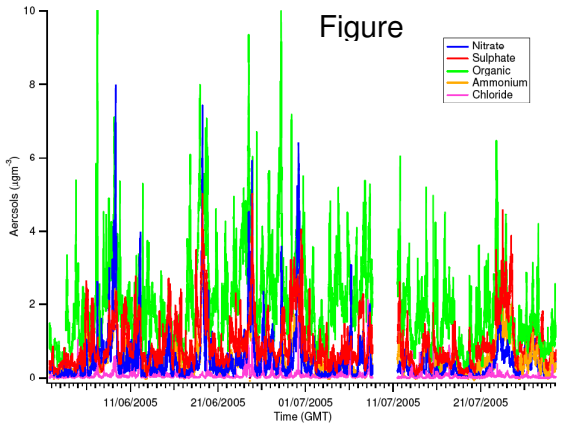
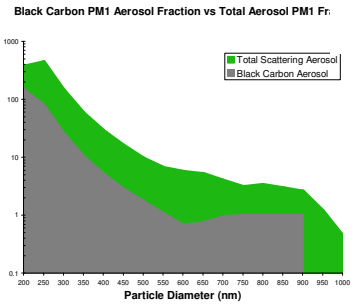
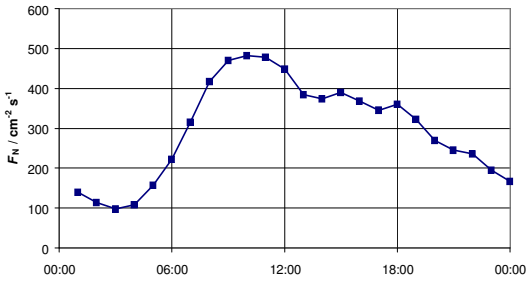
Keywords: Particulate, Fluxes, Emission, Organic, Micrometeorological, Exposure

In the urban environment gases and aerosols are emitted from a melange of sources (vehicles both fleet and personal, residential and institutional combustion, small plant and construction activities). Many of these chemical compounds can be highly reactive and are usually emitted in hot exhaust plumes where chemical processing starts immediately as the plume cools, mixes and dilutes and material is lost, converted or transported away from the source. Many issues directly relevant to epidemiological exposure type, level and frequency arise as a result of these processes with relevance for personal air quality within the city and atmospheric composition change as the particulates are transported out of the city.

The CityFlux project is currently engaged in addressing some of these issues, specifically to investigate the following questions: What are the emissions in the urban area and what are the emission factors of individual activities found in the urban area? What fraction of the gases and aerosols in the city is primary and what is secondary? How does the gas/particle partitioning change as the pollutants age after emission in the street canyons and above the city? What are the time-scales involved? A range of high resolution, fast response instrumentation, many as components of micrometeorological eddy correlation flux techniques are being applied to assess the variability of size and chemically segregated particle fluxes and their subsequent chemical transformation and variability both at the personal exposure level by investigating vertical and horizontal fluxes that occur within individual street canyons and on a citywide scale using large tower based measurement sites under real atmospheric conditions. CityFlux aims to derive emission factors of a range of air pollutants by two independent techniques and provide updated emission factors for UK conditions. CityFlux in upcoming experiments will also investigate; the chemical processing and transport of urban emissions within the urban environment, starting at the emission point, with respect to reaction time-scales, the subsequent evolution of aerosol size-spectra, gas/particle equilibria and oxidation state using field deployable aerosol mass spectrometers; the pollutant transport within and out of street canyons and quantify chemical processing and relative venting rates of aerosols, and; to compile a dataset to inform model development and for validation of future model simulations of urban air chemistry. Figure 1 shows high resolution (1-10 minute) measurements of aerosol mass (□g m^{-3}) as a function of chemical composition (ammonium, nitrate, sulphate, organic and chloride) over 55 days in a Manchester street. Figure 2 shows the vertical aerosol flux (particles cm^{-2} s^{-1}) and PM1 size segregated carbon aerosol number fraction measured within the canyon. These data will be combined within CityFlux to quantify emission rates and exposure levels of near-primary and secondary aerosols.







Acknowledgements: This work was funded by the UK Natural Environment Research Council.

On regional correlation of atmospheric PM signals

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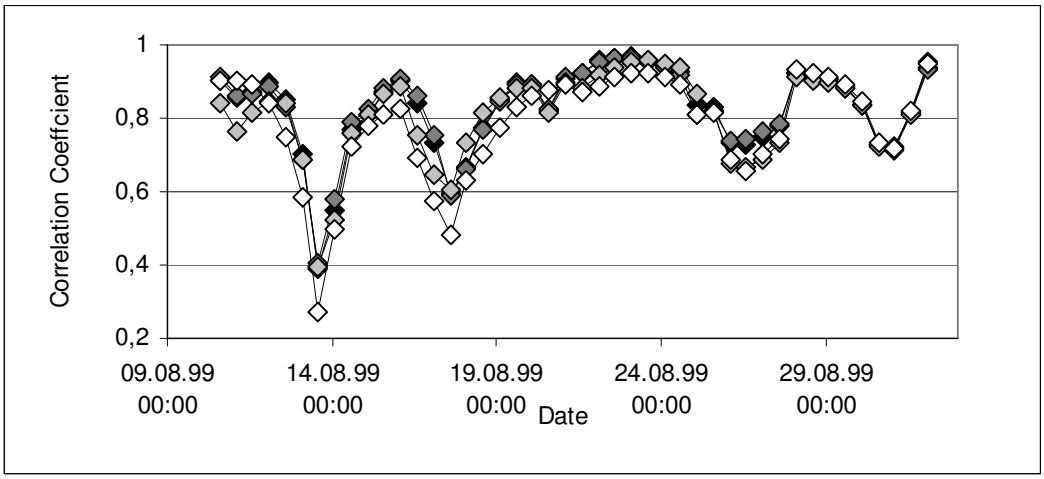
Keywords: regional correlation, episodic structure, meteorologic relations

Recently, regional correlation of atmospheric PM signals has been addressed in the literature (1). A follow-up investigation of atmospheric PM (2) indicated that time series of partial correlation coefficients of regional PM signals exhibit typical structures. Periods of rather high correlation coefficients are interrupted by short periods of rather low coefficients.

In our work the atmospheric PM signals (time series of PM2.5 and PM10) are based on rather accurate impactor data (2), that is, mass size distributions for 12h sampling periods over four weeks in summer 1999 and winter 2000. Partial correlation coefficients are calculated for a few subsequent samples (5 samples, 7 samples, 9 samples). An example is shown in the figure, showing the typical structure indicated above.



As shown by the analysis of the back trajectories for the two sampling sites, the minima observed in the partial correlation coefficients coincide with extreme maxima of trajectory distances. They are related to an abrupt change of the weather conditions.

According to the few first results this rule of correspondence does not hold when local emissions dominate the PM concentration at either one or both sites. Hypothetically the partial correlation coefficients are a supplementary indicator whether sampling sites are dominated by a regional back ground aerosol or by local emissions.

References:
(1) A. Berner, Z. Galambos, P. Ctyroky, P. Frühauf, R. Hitzenberger, B. Gomiscek, H. Hauck, O. Preining, H. Puxbaum (2004). On the correlation of agtmospheric aerosol components of mass size distributions in the larger region of a central European city. Atmospheric Environment 38 (2004) 3959-3970.

(2) M.E. Hazenkamp-von Arx, T.G. Fellmann, L. Oglesby, U. Ackermann-Liebrich, T. Gislason, J. Heinrich, D. Jarvis, C. Luczynska, A.J. Manzanera, L. Modig, D. Norbäck, A. Pfeiffer, A. Poli, M. Ponzio, A. Soon, P. Vermeire, N. Künzli (2003): PM2.5 assessment in 21 European study centers of ECRHS II: method and first winter results. Journal of the Air and Waste Management Association 53 (2003) 617-628.

Heterogeneity in chemical, toxicological, and source charactristics of urban air coarse, fine, and ultrafine particles from six sampling campaigns in Europe (PAMCHAR)

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Urban air fine particles ($PM_{2.5}$; diameter < 2.5 μm) have been associated with extensive premature mortality and morbidity among subjects with chronic cardio-respiratory disease. Also coarse particles ($PM_{10-2.5}$) have been found harmful at least to respiratory health in epidemiological studies. The responsible sources and constituents as well as toxicity mechanisms for the particulate effects are not well known. Seven-week sampling campaigns were conducted in six European cities during selected seasons of public health interest: Duisburg (autumn), Prague (winter), Amsterdam (winter), Helsinki (spring), Barcelona (spring) and Athens (summer). Urban air $PM_{10-2.5}$, $PM_{2.5-0.2}$ and ultrafine particulate ($PM_{0.2}$) samples were collected with a modified Harvard high-volume cascade impactor (HVCI) for toxicological studies, while $PM_{10-2.5}$ and $PM_{2.5}$ samples were collected in parallel with three virtual impactors for in-depth chemical and source characterisation. Analyses of the HVCI and virtual impactor samples revealed large variations between the sampling campaigns in chemical composition (e.g., organic matter, PAH, heavy metals, crustal material) of the size-segregated particulate samples. Chemical mass closure as well as size-segregated mass, ionic and specific source tracer (e.g., levoglucosan) concentration data proved useful for estimation of the contributions of various local and distant particulate sources to these variations. The differences in chemical composition and sources of the complex urban air particulate mixture were consistently reflected in the inflammatory activity of the particulate samples in the mouse RAW264.7 macrophages and the mouse lung. The inflammatory activity of the $PM_{10-2.5}$ samples was higher than that of the $PM_{2.5-0.2}$ samples, but the latter exhibited larger differences between the sampling campaigns. The $PM_{0.2}$ samples had negligible inflammatory activity, but some of them were highly cytotoxic. In addition to emission sources, atmospheric photochemical activity is suggested to be a major factor determining the toxicity of urban air particulate matter. Finally, the substantial variations in chemical composition and toxic activities of size-segregated urban air particles with geographical region and season are likely to have public health implications in Europe.

Acknowledgements: Funding by the EU-FP5 (QLK4-CT-2001-00423), Academy of Finland (Contract 53307) and Tekes (Contract 40715/01), and field campaigns by the teams of Thomas Kuhlbusch (Institut für Energie- und Umwelttechnik, Duisburg, DE), Martin Branis (Charles University in Prague, CZ), Bert Brunekreef (University of Utrecht, NL), Jordi Sunyer (Institut Municipal d'Investigació Mèdica, Barcelona, ES), and Klea Katsouyanni and Ilias Kavouras (National and Kapodistrian University of Athens and National Observatory of Athens, EL).

Role of Particle Composition and Size in the Effects of Urban Particulate Matter in Vitro

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[1]Laboratoire de Cytophysiologie et de Toxicologie Cellulaire, Université Paris 7, and [2]LEPI, France

Epidemiological investigations have associated exposure to ambient particulate matter (PM) with an increase in cardiorespiratory disorders. PM is a complex mixture of particles which composition varies according to both location and season sampling but that in urban areas is characterized by the presence of nanoparticles produced by combustion. Using *in vitro* models of airway epithelial cells we have previously demonstrated that PM2.5 and DEP (Diesel exhaust particles) induce a proinflammatory response characterized by the release of the GM-CSF cytokine (*Baulig et al. TIV, 2003)* as well as an overexpression of amphiregulin, an EGFR ligand *(Blanchet et al., AJRCMB, 2004*) and that the expression of these 2 biomarkers of the effects of PM2.5 exposure involves reactive oxygen species (ROS).
The aim of the present study was to investigate which PM2.5 component is involved in the induction of these 2 biomarkers and whether their induction differs according to particle size.
For this purpose we used (1) PM2.5 sampled in two sites of Paris (kerbside and urban background). 50% of the particles have a diameter ≤ 260nm. Their content in metals and polycyclic aromatic hydrocarbons (PAH) has been previously characterized and shown that there are twice more PAH in kerbside than in background site PM2.5 (*Baulig et al, EST, 2004*). The contribution of the different components was analysed comparing native particles to their respective organic and aqueous extracts. (2) size-fractionated PM sampled in an urban background site of Paris with a 13 stages low pressure impactor (10µm to 0.03µm). Three size classes (PM1-2.5, PM0.1-1, PM0.03-0.1) were tested for their biological reactivity towards airway epithelial cells.
PM2.5 induced amphiregulin (AR) expression (assessed by northern blots and RT-PCR after a 18h exposure) and a dose-dependent AR secretion (from 1 to 30 µg/cm² of particles, after a 24h exposure) in both human bronchial (16HBE cell line) and nasal (primary cultures) epithelial cells. GM-CSF secretion was also dose-dependently induced by PM2.5.
AR and GM-CSF releases were higher when cells were exposed to organic extracts and PM devoided of hydrosoluble compounds than to aqueous extracts and carbon black particles (Ø 95 nm), suggesting that neither soluble metals nor carbonaceous core are involved in these biological responses. In addition the PM2.5 organic fraction also mainly contributes to the intracellular ROS production assessed by a specific fluorescent probe. Despite their difference in metal and PAH composition, PM2.5 (10 µg/cm²) from kerbside or background sites induced similar levels of AR and GM-CSF secretion as well as ROS production. It could be due to differences in organic compounds bioavailability. This hypothesis results from the observation that both the expression (assessed by quantitative RT-PCR) and activity (measured by the ethoxy-resorufin assay) of CYP1A1, the P450 cytochrome induced by PAH and involved in PAH metabolization were highly induced by background site PM2.5, but less by kerbside site PM2.5.
Finally, preliminary experiments performed with size-fractionated PM revealed that GM-CSF secretion was principally induced by the smallest PM whereas amphiregulin secretion was mainly induced by PM1-2.5.
In conclusion, we have shown that the biological reactivity of airway epithelial cells induced by PM2.5 is mainly dependent on PM2.5 organic component and its bioavailability. In addition according to PM size, the induction of the two studied biomarkers differs and should be analyzed in light of their physico-chemical characteristics. (*This work was supported by ADEME, PRIMEQUAL, INSERM and Renault)*

PRIMEQUAL : The french program for the impact of atmospheric pollution on health: The PM studies

Francelyne Marano [(1)], Pascale Ebner [(2)], Alain Weill [(3)]

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[(2)] MEDD, ministry of environment and sustainable development, in charge of PRIMEQUAL program, 20 avenue de Ségur,75302, Paris SP07(France),
[(3)] CETP, 10-12, Avenue de l'Europe, 78140 Vélizy (France) and scientific council of PRIMEQUAL

PRIMEQUAL is a scientific program initiated by the French ministry of Environment since 1995 and involved in air quality study. One of its objectives is to develop high level scientific applied research to get insight into air pollution to help policy and decision makers.

At the beginning of the program, analytic disciplinary studies were conducted. These studies were concerned by pollutant sources, metrology, chemical reactivity, physicochemical modelling, epidemiologic and toxicological researches. Several researches carried out on human factors were also undertaken.

Since 2001, due to a better analytical knowledge, more systemic approach became necessary and a more transversal and interdisciplinary research has been incited. Therefore, three scientific programs with more interactions between disciplines were launched.

1. One about different aspects of air pollution which was orientated toward air pollution risks in 2001
2. One about anthropogenic particles and more particularly on thin particles following a seminar with a state of art about particles scientific knowledge deficiency.
3. More recently a program about pollution evaluation and perception of pollution has been initiated in a framework where technical and human factors were supposed to be taken into account.

Campaigns of measurement as POVA devoted to alpine valleys pollution, ESCOMPTE concerning summer pollution in a complex terrain Mediterranean city of Marseille and Biopollat'm related to relationships between pollutants and agricultural ecosystems are also described. Results from the very large multicentric epidemiological study of GENOTOXER are then expressed showing in three French cities genotoxicity of pollutant particles and effectiveness of pollution due to road transportation.

Research actions from program 2 more particularly related to small atmospheric particles and associated health impacts have been selected. Particularly, a project on complex aerosols impacts on heart, kidney and reproductive impacts was selected, as well as one on biologic effects of PM2.5, PM1 and PM0.1 of the urban aerosol pollution background.

For the purpose to complement small particles health impacts knowledge, and to better determine physicochemical particles properties, different projects have also been selected as:

- Metrology of ultra-thin particles
- Methods to discriminate sources of particles
- Ageing and degradation of different types of particles
- Analysis of soot related to aircrafts
- Indoor particles
- Particles impact on building

In conclusion we point out that the objective of this program 2 lies within the framework of PRIMEQUAL strategy and complement already obtained epidemiological and toxicological results concerning diesel particles and health. As a perspective, envisaged PRIMEQUAL actions about proximity pollution, climate change and air quality and prospective scenario on air quality for year 2050 must leave a very large place on research devoted to anthropogenic particles and health impacts.

Programm of Conference

Monday 3rd April:

08.00 - 09.00	Registration
09.00	**Official opening** - Regina Hitzenberger (University of Vienna)
09.15	Scope of the workshop and outline of what to come! - Thomas Kuhlbusch (IUTA Duisburg)
09.45	**Topic 1: Particle characterisation and characteristics** Chair: Hans Puxbaum (University of Vienna)
09.45	Chemical characterization of PM from various Sources - Hans Puxbaum (University of Vienna)
10.15	Spatial and temporal variances in particle characteristics in the EU - Jean-Phillipe Putaud (JRC-Ispra)
10.40	Introduction into the topic question - Harry ten Brink (ECN-Petten)
10.45	**Coffee / tea break**
11.00	Workshop 1 (2 parallel sessions) Group leaders: Harry ten Brink (ECN Petten) Roy Harrison (University Birmingham) Rapporteurs: Jean-Phillipe Putaud (JRC-Ispra) Axel Berner (University of Vienna)
12.00	Plenary: session reports and general discussion
12.30 - 14.00	**Lunch break**
14.00	**Topic 2: Sources of particulate matter** Chair: Rainer Friedrich (University of Stuttgart)
14.00	PM Emission inventories - Rainer Friedrich (University of Stuttgart)
14.30	Results of source apportionment studies in Spain and comparison with other European regions - Xavier Querol (CSIC Barcelona)
14.55	Introduction into the topic question - Markus Amann (IIASA Laxenburg)
15.00	**Coffee / tea break**

15.15 Workshop 2 (2 parallel sessions)
Group leader:
Tuomo Pakkanen (FMI Helsinki)
Markus Amann (IIASA Laxenburg)
Rapporteurs:
Thomas Kuhlbusch (IUTA Duisburg)
Maria del Mar Viana (CSIC Barcelona)

16.00 Plenary: session reports and general discussion

16.30 - 18.00 Poster session (incl. buffet).

Tuesday 4th April:

09.00 **Topic 3: Modelling and (Personal) Exposure**
Chair: Matti Jantunnen (KTL-Kuopio)

09.05 Determination and modelling exposure to PM from different sources
- Matti Jantunnen (KTLKuopio)

09.30 Modelling of urban population exposure to PM
- Carlos Borrego (University of Aveiro)

09.55 Introduction into the topic question
- Matti Jantunnen (KTL-Kuopio)

10.00 **Coffee / tea break**

10.15 Workshop 3 (2 groups in parallel)
Group leader:
Carlos Borrego (University of Aveiro)
Matthias Ketzel (NERI Roskilde)
Rapporteurs:
Wilfried Winniwater, (ARC - Seibersdorf)
Ana Isabel Miranda (University of Aveiro)

11.00 Plenary: session reports and general discussion

11.30 **Topic 4: Health effects: epidemiology**
Chair: Gerard Hoek (IRAS- Utrecht)

11.35 Acute Health Effects perspective
- Gerard Hoek (IRAS- Utrecht)

12.00 Chronic Health Effects perspective
- Nino Kuenzli (IMIM- Barcelona)

12.25 Introduction into the topic question
- Gerard Hoek (IRAS- Utrecht)

12.30 **COST 633 MC meeting** (with buffet lunch provided)
Everyone else: lunch break.

14.30 Workshop 4 (2 groups in parallel)
Group leader:
Josyph Cyrys (GSF Munich)

Nino Kunzli (IMIM- Barcelona)
Rapporteurs:
Raimo Salonen (KTL Kuopio)
Michael Riediker (IOHS Lausanne)

15.15 Plenary: session reports and general discussion

15.45 **Coffee / tea break**

16.00 - 16.30 Integrated Assessment Modelling
- Markus Amann (IIASA Laxenburg)

19.30 Reception at the city hall

Wednesday 5th April:

09.00 **Topic 5: Heath effects - toxicology**
Chair: Ken Donaldson (University of Edinburgh)

09.05 Which particle characteristics are important in view of health effects?
- Ken Donaldson (University of Edinburgh)

09.30 Are there regional differences in toxicological effects of particulate matter?
- Per Schwarze (Folkehelsa Oslo)

09.55 Introduction into the topic question
- Franceline Marano (Université Paris)

10.00 **Break (plus press conference and poster viewing)**

10.45 Workshop 5 (2 groups in parallel)
Group leader:
Franceline Marano (Université Paris)
Per Schwarze (Folkehelsa Oslo)
Rapporteurs:
Wolfgang Kreyling (GSF Munich)
Flemming Cassee (RIVM Bilthoven)

11.45 Plenary: session reports and general discussion

12.15 - 13.30 **Lunch break**
(parallel meeting of rapporteurs and break out group leaders to sum up the reports and prepare the last session)

13.30 **Synthesis: Session with stakeholders and policy makers**
Presentation of all the topic group discussions and reports, discussion on future work; Chair: Thomas Kuhlbusch (IUTA)

15.00 - 15.15 Summary and closing remarks
Regina Hitzenberger, COST633 Chair
Flemming Cassee, Scientific committee

Conference Committees

Organizing Committee

Chair: Regina Hitzenberger (COST633 Chair), University of Vienna, Austria

Gudrun Breschar (Kommission Reinhaltung der Luft, Akademie der Wissenschaften, Austria), Agnes Molnar (Hungarian Academy of Sciences, Hungary), Maria del Mar Viana (CSIC, Spain), Vera Meyer (University of Vienna, Austria), Peter Reisinger (University of Vienna, Austria), Gerhard Steiner (University of Vienna, Austria), Anna Wonaschütz (University of Vienna, Austria)

Scientific committee

Chairs: Thomas Kuhlbusch (Chair), IUTA e.V., Germany

Flemming Cassee (Co-Chair), RIVM, The Netherlands

Axel Berner (University of Vienna, Austria), Harry ten Brink (ECN, The Netherlands), Carlos Borrego (University of Aveiro, Portugal), Christian Dye (NILU, Norway), Wolfgang Kreyling (GSF, Germany), Franceline Marano (Université Paris 7, France), Michael Riediker (Institute for Occupational Health Sciences, Switzerland), Imre Salma (Eotvos University, Hungary), Maria del Mar Viana (CSIC, Spain), Wilfried Winiwarter (ARC systems research GmbH, Austria)

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List of attendees: COST633 conference, Vienna, 3-5 April 2006

(in alphabetical order; (at) is used instead of @ to minimize spam).

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