

Workshop measurements and modelling of PM_{2.5} in Europe Overview and Proceedings

This is a publication of the Netherlands Research Program on Particulate Matter


Netherlands Environmental Assessment Agency



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BOP report

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Overview and Proceedings

Jan Matthijsen, PBL; Peter Bultjes, TNO



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Report 500099017

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Contact: karin.vandoremalen@pbl.nl

ISSN: 1875-2322 (print) ISSN: 1875-2314 (on line)

This is a publication in the series: BOP reports

Project assistant: Karin van Doremalen

English editing: Annemieke Righart

Figure editing: PBL editing and production team

Layout and design: RIVM editing and production team

Cover design: Ed Buijsman (photographer: Sandsun)

ECN Energy research Centre of the Netherlands

PBL Netherlands Environmental Assessment Agency

TNO Built Environment and Geosciences

RIVM National Institute for Public Health and the Environment

This study has been conducted under the auspices of the Netherlands Research Program on Particulate Matter (BOP), a national program on PM₁₀ and PM_{2.5} funded by the Dutch Ministry of Housing, Spatial Planning and the Environment (VROM).

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The complete publication, can be downloaded from the website www.pbl.nl

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Workshop measurements and modelling of PM_{2.5} in Europe

Summary

In 2008, the European Directive on air quality went into force (2008/50/EC). The new directive combined four existing EU directives, and established air quality standards for fine particulate matter (PM_{2.5}). The knowledge on particulate matter, specifically on PM₁₀, has increased greatly, both in the scientific and the applied sense. The PM dossier still carries many uncertainties and, generally, only little experience has been gained in the measuring and modelling of PM_{2.5} in aid of policy support. During the workshop different aspects of PM_{2.5} have been addressed in thirteen presentations. Countries have started to include measurements of PM_{2.5} and its components in their national air quality monitoring networks. Harmonisation of the national air quality networks is on its way but takes a considerable amount of effort. As a consequence it is difficult to make a consistent view of PM_{2.5} Europe wide. Measurement artefacts due to semi-volatility of some particulate matter components also limit consistency between model results, emission inventories and measurements. Policy assessments which aim at finding adequate conditions for compliance to the PM_{2.5} standards are therefore still very uncertain. In addition there is growing evidence that, like PM₁₀, also PM_{2.5} is a rather crude indicator for the health effects associated to particulate matter. Combustion emission sources like traffic appear to have a larger health impact than others. So it is necessary to design smart policies which aim at the health improvement and take at the same time the current uncertainties into account about the efficiency of the current PM_{2.5} air quality standards for health improvement. Other, additional, PM indicators may be helpful to plan such policies.

Introduction

The aim of this workshop was to discuss with a number of European experts the current state-of-knowledge concerning all aspects of PM_{2.5}. This means observations, emissions, modelling, source apportionment and health effects. In view of the European air quality standards for PM_{2.5}, it is essential to exchange information, and to try to come to a coherent assessment with respect to PM_{2.5}. An important example is the introduction of the average exposure index, which is an indicator for the average urban background concentration in a country. This indicator is new and countries have so far little experience in dealing with it and consequently many

questions arise on how to measure and manage reductions which are necessary to meet the standards for the average exposure indicator. Also the other gaps in knowledge were discussed, hopefully leading to a common research agenda, and – funded – projects.

The workshop was attended by about 40 participants, from 9 European countries. In the following paragraphs an overview of the presented papers is given, and some conclusions are drawn. The workshop programme, abstracts of the different presentations, a list of participants and the most important hyperlinks used or presented in the workshop are given in annexes.

PM_{2.5} in the European Directive for Air Quality

For the improvement of air quality in Europe the following article from the 6th Environmental Action Plan is presented as a leading term: ‘achieving levels of air quality that do not give rise to significant negative impacts on and risks to human health and the environment’.

The European Air Quality directive includes five objectives for PM_{2.5}: Target and limit value of 25 µg/m³ for average annual PM_{2.5} concentrations to apply everywhere. It is a target value by 2010 and a limit value by 2015. An indicative limit value of 20 µg/m³ has been set for 2020 and is to be confirmed at the review of the Directive in 2013. The other two objectives aim at limiting and reducing exposure to PM_{2.5} in urban areas, based on the so-called national average exposure indicator (AEI): the Exposure Concentration Obligation of 20 µg/m³ by 2015 and the Exposure Reduction Target to reduce national average measured urban background concentration with a value between 0 and 20% between 2010 and 2020.

The national value of the exposure reduction target depends on the AEI in 2010. The exposure reduction target is subject to the review. The AEI is the 3 years moving average annual PM_{2.5} concentration at urban background sites. The AEI is one number per Member State as it is an average across cities. The Directive gives guidelines on how many urban background stations a Member State should have to calculate the AEI.

In 2013 the European Commission will review the Air Quality Directive 2008/50/EC. This review includes with regard to PM_{2.5} the following aspects:

- Legally binding exposure reduction target, review exposure concentration obligation
- More ambitious limit value
- Confirm/modify indicative limit value which is currently 20 µg/m³
- Monitoring

Observations

Observations are essential to the assessment concerning the current situation of PM_{2.5}. Throughout Europe, the knowledge on current PM_{2.5} concentration levels is still rather limited. Many Member States have only recently started to include PM_{2.5} measurements in their national air quality monitoring networks. Routine measurements of PM_{2.5} using automated samplers are thought to be more uncertain than measurements of PM₁₀ because the semi-volatile PM fraction, which is a main source of measurement uncertainty, resides predominantly in the fine fraction. Consequently, the climatology of PM_{2.5} throughout Europe is not yet well understood. Several presentations gave an overview of local efforts to measure PM_{2.5} and its components. The Joint Research Centre (JRC) showed results of a measurement campaign to support harmonisation with regard to observations of particulate matter performed according to the EU guidelines.

Urban background stations measuring PM_{2.5} are required with respect to the EU- Air Quality Directive starting from 1 January 2009. Per 1 million inhabitants a minimum of one urban background station is required summed over agglomerations and additional urban areas in excess of 100.000 inhabitants. In rural areas the chemical speciation of PM_{2.5} should be measured to support modelling and source attribution. The stations should be in operation now, and the observations will have to be reported by the beginning of 2010.

Between 2006 and 2009 JRC and the AQUILA network carried out a number of investigations and measuring campaigns of parallel measurements of PM₁₀, PM_{2.5} and PM₁. AQUILA is a formally established network - open to all of the National Reference Laboratories across Europe - that verifies and supports the correct implementation of air quality directives in Europe. The parallel measurements show that correlation with JRC PM data is in general high, but differences of the averages up to 30-50 % are found. Comparison between JRC data and those of national reference laboratories is good for gravimetric methods, but appear problematic at very high or very low concentrations.

It appears from the parallel measurements that yearly averaged concentrations of PM_{2.5} have an average uncertainty of about 25 % (2-sigma). Day-to-day differences can easily attain about 50 %. European regulations allow a maximum uncertainty of 25% (2-sigma) in the averaged annual measured data. However, possible values for the national exposure reduction target will be 20% or less. Since the allowed uncertainty in the measurements is larger than the exposure reduction target it appears that a reduction of this

magnitude will not be easily measurable. The significance of the average exposure indicator reduction, based on reference measurements, is a topic currently being addressed by AQUILA.

Observations of averaged annual PM_{2.5} concentrations in the Netherlands were obtained for 2008 by using the reference method. Imputation techniques were successfully applied to reduce the uncertainty in the average annual data due to limited data coverage. The rural-to-urban gradient and the urban-to-street gradient were both about 1 µg/m³. At 9 rural background stations the averaged annual concentration was 16 µg/m³ with a standard deviation of 2 µg/m³. At 9 urban stations the averaged annual concentration was 17 µg/m³ (stdev. 2 µg/m³). At 9 traffic stations the averaged annual concentration was 18 µg/m³ (stdev. 1 µg/m³).

In the United Kingdom (UK) averaged annual PM_{2.5} concentrations were measured in 2007/2008 and varied between 10 µg/m³ at rural background locations and about 12 µg/m³ at urban locations. To establish the spatial differentiation of PM_{2.5} concentrations in the UK PM_{2.5} maps for 2005, 2006 and 2007 were made by combining model results and measurements. From these maps average annual PM_{2.5} concentrations were calculated to be usually well below 15 µg/m³, also in most urban areas. However, during years with unfavourable meteorological circumstances, like 2006, averaged annual PM_{2.5} concentrations were on average several up to about 5 µg/m³ higher. Consequently in some urban areas average annual PM_{2.5} concentrations might have exceeded 20 µg/m³.

The 'three-site study' for a rural, urban traffic site in Birmingham showed a gradient from roadside to urban background to rural sites, most particularly in the content of elemental carbon (EC), organic carbon (OC) and iron. Secondary OC is important and shows similarities in seasonal behaviour to nitrate. For PM₁₀ similarly a gradient was found of mean roadside to urban background and from urban background to rural concentrations. Gradients for PM_{2.5} were found with roadside greater than urban background greater than rural in one study and urban background greater than rural in another. The gradients were, however, small compared to those of components like elemental carbon. Data for all seasons from the central urban background site show notably higher sulphate in summer and lower nitrate in summer.

Measurements of the chemical speciation of PM_{2.5} show the contributions of sulphate, nitrate, ammonium, carbonaceous aerosol, both EC and organic matter. Organic carbon in PM is present in many individual carbonaceous compounds, which not only contain carbon but also other elements, such as oxygen and hydrogen. These elements are usually not measured, but they do contribute to the mass. The additional mass of these elements can be accounted for by translating carbon into organic matter (OM) or also called Total Carbonaceous Mass (TCM). OM or TCM are derived from the total carbon mass (OC), usually by application of a multiplication factor (typically 1.3-1.4). First results have been obtained to discriminate recent and fossil carbon by the C-14 method, and separated in EC and OM. Further information

to distinguish several primary sources and secondary organic aerosol can be obtained by using the Aerodyne Aerosol Mass spectrometer (AMS). The AMS is a fast aerosol mass measurement for non-refractory fine particulate matter in the size range of 40 nm to about 1 µm.

The MARGA-sampler, based on continuous wet denuders and steam jet aerosol collectors to measure the chemical composition of PM_{2.5}, and PM₁₀, has been used in combination with the AMS during the EMEP Intensive measurements periods of June 2006, January 2007, October and November 2008 and February and March 2009. Both MARGA and AMS provide a good measure of ambient concentrations of volatile species like ammonium nitrate.

Low volume or high volume samplers – LVS/HVS - were found not to be suited for measurement of particulate nitrate due to measurement artefacts. Application of a denuder, which removes gaseous components from the sampled air, can reduce measurement artefacts. The reference method for PM₁₀ (EN12341;1998) and for PM_{2.5} (EN14907;2005) use either LVS or HVS. As a consequence, the contribution of particulate nitrate to the PM₁₀ and PM_{2.5} concentration, when measured according to the reference method, may be substantially different from ambient particulate nitrate concentrations. This constitutes a problem in measurement / model inter-comparisons. Measurement providers should be involved to consider what exactly the different instruments measure and how this might relate to the modeled compounds. Also for an adequate assessment of reduction measures it is necessary that models, used for this purpose, can take measurement artefacts into account.

Observations at Melpitz over a six year period 2003-2008 showed, next to detailed speciation measurements of PM_{2.5}, that the ratio PM_{2.5}/PM₁₀ varies between winter values of about 0.8 to summer values of about 0.65, and are also a function of wind direction, which means source areas. The Melpitz measurement site is a rural background station in the Eastern part of Germany. The average three year mean PM_{2.5} concentration from 2006 to 2008 was 17.6 µg/m³. The highest PM mass concentrations with high amounts of sulphate, nitrate and carbon were observed during air mass transport in winter from the East.

In general over Europe there is no significant trend over the last 5 years in the observed PM₁₀ concentrations, nor in the observed PM_{2.5} concentrations. The number of the PM_{2.5} observations and the length of the PM_{2.5} time series are still limited.

Emissions

Emissions play a crucial role in understanding ambient PM_{2.5} observations. The emissions are input for predictive models and the information on source-specific contributions can be translated in emission reduction strategies aiming to reduce the ambient concentrations. Emission data are available for primary emitted PM_{2.5} and for the precursor emissions of SO₂, NO_x, NH₃ and NMVOC. Estimates are available concerning the percentage of EC, OM and dust in the primary emissions of PM_{2.5}. A European wide emission data base for all these pollutants exists for the year 2005, on horizontal grids of

0.125° x 0.0625° latitude - longitude (ca. 6x6 km²), divided in area, line and point sources. For a number of countries, more refined emission information is available, for example in Germany on 1 min x 1 min (ca. 1,2 x 1.8 km²). Furthermore, as in the case of Germany, additional high resolution emission height information may be available for the national emission data base.

The 2005 emission database is consistent with national particulate matter inventories. About half of the total European primary PM_{2.5} emissions are carbonaceous aerosols (EC and OC). Diesel use in transport and fuel wood by households are dominant sources, responsible for about 60% of both EC and OC in PM_{2.5}. The high resolution inventories serve as input for atmospheric modellers. Comparison of measured with modelled concentrations help to verify the emission data bases and may lead to further improvements of these. This approach generates many ideas about further improvements, but it is difficult to put these ideas into funded work when they are not directly linked to reporting requirements or limit values.

Emissions of primary PM_{2.5}, and its speciation, are still rather uncertain, although in the long term they are believed to become more accurate than those of primary PM₁₀ because PM_{2.5} emissions are relatively more dominated by combustion processes. There are several aspects with regard to the emission uncertainties. For instance, primary emissions of PM_{2.5} are still often derived as a fraction of primary PM₁₀ emissions. In addition, accidental releases from regulated sources become more important for air quality, whereas these usually large emissions during a few hours or days do not appear in the official emission reports.

Modelling and source apportionment

Modelling with 3-D Chemical Transport Models has become a powerful tool, enabling to provide air pollutant concentrations fields and the determination of the contributing emission sources and the impact of future emission reductions strategies. Several model inter-comparison studies and evaluation studies have shown good and acceptable results for species like O₃ and NO₂, but less good performance for PM₁₀ and PM_{2.5}. In general, an underestimation is found, showing lower calculated concentrations than observations. The differences in calculated PM_{2.5} concentrations by different models are smaller than ±25%. Apart from not well described or even missing emissions like re-suspension by traffic, windblown dust and organic material, large uncertainties are related to the seasonal dependence of ammonia emissions and the formation of ammonium nitrate. Although the calculated inorganic fraction is in a reasonable agreement with observations, the calculated fine and coarse nitrate differs from the observed ratio.

Air quality models are currently being extended to be able to assess possible effects of climate change. Not only plays particulate matter an important role in affecting climate but also climatic changes can have considerable effects on air quality. Some sensitivity tests were presented, showing effects of increased temperatures on soil emissions and air quality concentrations.

Contribution of elemental carbon (EC) and organic carbon (OC) to $PM_{2.5}$ in the Netherlands is several tens of percentage points. Also in other European member states EC and OC constitute an important part of the mass of $PM_{2.5}$. Traffic and residential combustion are the important sources of these carbonaceous aerosols.

Source apportionment is used to analyse, from the observed speciation and knowing the speciation of the emissions and information concerning specific tracers (like Copper and Barium for break ware), the specific emission sources which cause the observed concentrations. Positive Matrix Factorisation – PMF - can be used to distinguish the type of organic aerosols from AMS organic aerosol mass spectra. It is also applied to determine the origin of inorganic PM components. PMF is a commonly used statistical technique for source apportionment based on PM component measurements. Using these techniques, in Zurich, six different sources of organic carbon were identified. The contribution of organic aerosol due to wood burning was with 20% to PM_1 much higher than expected. Secondary organic aerosol (SOA) is often dominating the organic fraction and is not only high in summer but also high in winter. The different sources of SOA cannot yet be distinguished.

In the United Kingdom measurements show that major $PM_{2.5}$ components are sulphates, nitrates and carbonaceous material. Nitrate is especially important in episodes of high PM_{10} . This behaviour has been observed in many other countries especially in NW Europe. Road traffic is normally the main contributor to primary carbonaceous particles, but the gasoline/diesel split is hard to determine from a chemical mass balance (CMB) model. The contribution of non-exhaust particles from traffic may also be significant and may also contain health relevant components.

Health effects

Epidemiological studies have provided evidence for an association between $PM_{2.5}$ and adverse health effects, albeit that there is far less information available compared to PM_{10} . However, it is not known yet which constituents or sources of emissions can be held responsible for the health effects. There is growing evidence that certain emission sources like traffic and other combustion processes have a larger health impact than other emissions such as inorganic aerosol.

The epidemiological evidence is expressed as shortening of life expectancy or impaired lung development and/or lung function of otherwise healthy people living near a busy road. Toxicological evidence shows that relative high levels of particulate air pollution can result in oxidative stress, inflammation of the lung, worsening of lung diseases and cardiovascular disorders and impairments. The toxicity may not only be caused by the particles themselves, but can also be caused by chemicals on the surface of particles and gases as well as influence by size. Particles of different sizes can cause different type of health effects.

Although the acute effects of particulate matter on health do not appear to be very large, its overall effect can be substantial since the whole population is exposed. In addition there are also effects below current PM_{10} standards

since, so far, epidemiological studies have not been able to demonstrate a threshold. Due to the linear relationship that epidemiological analysis has provided, each $10 \mu\text{g}/\text{m}^3$ increase results in the same increase in health effects, irrespective the levels. It needs to be mentioned that it is unrealistic to expect PM levels to be reduced to zero. PM is a complex mixture in which a part does not adversely affect our health. So, although there are many sources of PM_{10} or $PM_{2.5}$ they are likely not all equally potent. Abatement strategies should however be directed toward the most toxic part of PM in order to ensure health benefits of reducing PM levels. It also appears that PM_{10} and $PM_{2.5}$ are rather crude indicators for the health effects associated to the PM exposure. It is uncertain whether the health relevant PM fraction is reduced, although PM levels are gradually decreasing in mass. Therefore, new additional indicators such as measures for the oxidative capacity of PM, which are better for predicting health effects, are under study. At present it is not clear whether these can be used in a regulatory setting. Air pollution control measures are usually on a source basis whereas health effects are often related to component. Other uncertainties concern the effects of chronic low dose exposures. Next to the lungs also the cardiovascular system is affected due to PM exposure. Recently it was found that ultra-fine particles ($< 0.1 \mu\text{m}$) can likely reach the brain and may be associated with neurodegenerative diseases such as Parkinson's and Alzheimer's disease. So for the health effects of PM not only components are important but also size matters. In addition the air pollution settings and history can play a role in the effects since synergy of effects can take place when for instance a person is exposed to both ozone and PM.

Assessments

The EU-Directive includes standards for the average annual $PM_{2.5}$ concentrations $25 \mu\text{g}/\text{m}^3$ as a target value by 2010 and as limit value in 2015. Other more strict limits have been issued for average annual $PM_{2.5}$ concentrations: $15 \mu\text{g}/\text{m}^3$ in the United States, $12 \mu\text{g}/\text{m}^3$ in California and $10 \mu\text{g}/\text{m}^3$ has been set by the World Health Organisation (WHO) as air quality guideline level. Next to the EU target value, an average exposure indicator is defined, to ensure that public exposure to $PM_{2.5}$ is further reduced. First analyses indicate that the current limit value for averaged daily PM_{10} concentrations is stricter than the $PM_{2.5}$ limit value. The PM_{10} limit value for averaged daily concentrations allows no more than 35 exceedances of $50 \mu\text{g}/\text{m}^3$. This leads to the open question whether the attainment of the $PM_{2.5}$ air quality standards will lead to an improvement of the air quality compared to the situation that Member States comply with the PM_{10} limit values which came into force in 2005.

The available $PM_{2.5}$ measurements for Europe indicate that several EU countries will face more serious problems than the Netherlands in attaining the target and limit value of $25 \mu\text{g}/\text{m}^3$ on time. In some Member States, measured $PM_{2.5}$ concentration levels are well above $30 \mu\text{g}/\text{m}^3$. The European policies, which focus on reducing pollutant emissions from vehicle engines, will lead to lower $PM_{2.5}$ concentrations at all traffic locations, Europe wide. However, the traffic-related contribution to $PM_{2.5}$ from non-exhaust emissions and re-suspension remains, and these components vary in magnitude throughout Europe. The limited amount of

data on aspects such as local traffic prevents a Europe wide assessment on attainability in all Member States regarding the target and limit value of $25 \mu\text{g}/\text{m}^3$.

The assessment over Europe of the current population exposure to $\text{PM}_{2.5}$ can only be determined in a limited way, due to the currently small amount of $\text{PM}_{2.5}$ observations. Such an exposure study indicated that in a number of urban areas in Europe people are exposed to $\text{PM}_{2.5}$ concentrations higher than $25 \mu\text{g}/\text{m}^3$. Exceedances appear to take place in the Po-valley and eastern European countries.

Meeting the exposure concentration obligation by the 2015 deadline may be difficult for several Member States without measures that go beyond the European ambitions. Furthermore, it is unclear whether Member States will face problems meeting their national exposure reduction target value (ERT), for two reasons:

- The national ERT values of the individual countries are still unknown simply because they depend on future $\text{PM}_{2.5}$ concentration levels.
- The level of implementation of technical and non-technical reduction measures differs throughout Europe.

When all Member States would meet the exposure reduction target in time it would be three times more effective in reducing the years of life lost than when the $\text{PM}_{2.5}$ limit values would be attained on the European scale.

The variability of the $\text{PM}_{2.5}$ levels was estimated, Europe wide, which are inferred from PM_{10} measurements reported to the EEA air quality database (AirBase). This approach takes advantage of the abundance of PM_{10} measurements, the fact that PM_{10} includes the fine fraction and the spatial statistics on the $\text{PM}_{2.5}$ to PM_{10} ratio. For this purpose, $\text{PM}_{2.5}$ to PM_{10} ratios were derived from a selected set of collocated AirBase measurements. The study showed that the average exposure indicator (AEI) was in eleven Member States in 2005 well above the obligation for 2015 – irrespective of the calculation method. In three Member States, the AEI was, depending on the calculation method, just below or above the level of $20 \mu\text{g}/\text{m}^3$. In the other twelve Member States, the AEI was estimated to be well below the binding limit value of $20 \mu\text{g}/\text{m}^3$. The AEI estimate for the Netherlands was between 18 and $19 \mu\text{g}/\text{m}^3$, which was in line with the observed urban background concentrations.

Appendix 1 Workshop programme

■ Workshop

Measurements and Modelling of PM_{2.5} in Europe
Bilthoven, The Netherlands 23-24 April 2009
Address: Antonie van Leeuwenhoeklaan 9, Room T007

Organizers:

J. Matthijsen, Netherlands Environmental Assessment Agency (PBL)
P.J.H. Bultjes, TNO Institute for Applied and Scientific Research

Final Programme

Thursday 23rd April 2009

12:30 - 13:00 : Registration

13:00 – 13:15 : Opening: Peter Bultjes

Chairman: Jan Matthijsen

13:15- 13:55 : Population Exposure To PM_{2.5} At An European Level - Frank de Leeuw (EEA - European Topic Centre for Air Quality and Climate Change, The Netherlands Environmental Assessment Agency)

13:55 – 14:20 : The New Air Quality Directive 2008/50/EC Requirements Regarding Fine Particles PM_{2.5} – Andrej Kobe (European Commission, DG-Environment, Brussels)

14:20 - 15:00 : European Emissions of PM_{2.5} and its precursors – Hugo Denier van der Gon (TNO, The Netherlands)

15:00 – 15:30 Coffee/Tea Break (posters)

15:30 – 15:55 : Compilation of Spatially and Vertically Highly Resolved Emission Inventories For Germany - Jochen Thelocke (Universität Stuttgart, Germany)

15:55 – 16:35 : PM_{2.5} Measurements in Europe - Annette Borowiak (Joint Research Centre, Italy)

16:35 – 17:15 : PM_{2.5} Measurements and Source Apportionment - Roy Harrison (University of Birmingham, United Kingdom)

20:00 Workshop Diner

Friday 24th April 2009

09:00 - 09:15 Coffee/Tea

Chairman: Peter Bultjes

09:15 – 09:55 : Health effects of particulate matter: facts and uncertainties - Flemming Cassee (National Institute of Public Health and the Environment, The Netherlands)

09:55 – 10:35 : PM_{2.5} Speciation/ Source Apportionment – Urs Baltensperger (Paul Scherrer Institut, Switzerland)

10:35 – 11:00 : PM_{2.5} High-Volume Measurements in East Germany – a six year study at Melpitz site - Gerald Spindler (Leibniz-Institut für Troposphärenforschung, Germany)

11:00 – 11:25 : Real-time Measurements of PM_{2.5} and PM₁ Chemical Position: Experience and Results from the UK Supersites and EMEP Intensive Measurements Periods - Eiko Nemitz (Centre for Ecology and Hydrology, United Kingdom)

11:25 Coffee/Tea Break (posters)

11:55 – 12:35 : PM_{2.5} Modelling: “Research and Policy Challenges” - Laurence Rouil (INERIS, France)

12:35 – 13:00 : Modeling PM_{2.5} Concentrations for the UK and Projections to 2020 - Sally Cooke (AEA, United Kingdom)

13:00 - 13:25 : PM_{2.5} Measurement results with the reference method and modeling for 2008 in The Netherlands - Ronald Hoogerbrugge (National Institute of Public Health and the Environment, The Netherlands)

13:30 Lunch

14:30 End

Appendix 2 List of abstracts

* Speaker

Oral presentation

Population exposure to PM_{2.5} at an European level

Frank de Leeuw* (1), Jan Horálek (2), Bruce Denby (3), Peter de Smet (1)

(1) Netherlands Environmental Assessment Agency (PBL), P.O.Box 303, 3720 AH Bilthoven; (2) Czech Hydrometeorological Institute (CHMI), Praha, Czech Republic; (3) Norwegian Institute for Air Research (NILU), Kjeller, Norway.

The exposure to PM_{2.5} is known to have adverse health effects. Assessment of the health impact of PM_{2.5} at the European levels is hampered by a lack of (reliable) information. The scarcity of the PM_{2.5} monitoring data precludes the interpolation of the data to a European scale. Atmospheric transport models tend to underestimate the measured concentrations. Here European PM_{2.5} concentrations maps have been prepared on the basis of PM₁₀ interpolated maps. Firstly, the PM₁₀ maps have been prepared by Kriging interpolation methods by combining the observed data from the more than 1500 monitoring stations available from AirBase, the results of the EMEP model and supplementary meteorological data. In a second step, the PM_{2.5} maps have been prepared using a PM_{2.5}/PM₁₀ ratio inferred from the measurements. Different PM_{2.5}/PM₁₀ ratios have been used depending on location and on type of station (rural, urban or traffic). The estimated PM_{2.5} maps are input to a health impact assessment. Estimates of premature deaths have been made for all-cause, cardiopulmonary and lung cancer mortality.

The new Air Quality Directive 2008/50/EC Requirements regarding fine particles PM_{2.5}

A. Bertello⁽¹⁾, A. Kobe^{*(2)}

(1) Province of Turin (on secondment to DG Environment European Commission); (2) DG Environment European Commission

The new *Directive 2008/50/EC*¹ of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe (AQD) entered into force on 11 June 2008. It merges and streamlines existing legislation with the exception of Directive 2004/10/EC (the 4th daughter directive); clarifies treatment of contribution from natural sources; provides, under conditions, more time to comply with PM₁₀, NO₂ and benzene limit values, and most importantly sets new air quality objectives and associated assessment requirements for PM_{2.5} (fine particles). Objectives include an annual average target value of 25 µg/m³ in 2010, to be replaced in 2015 by the binding limit value of the same nominal value, and exposure related objectives – exposure concentration obligation and exposure reduction target. The later are set at the national level and are based on the *average exposure indicator (AEI)*. Their aim is to ensure that public exposure to PM_{2.5} and thus adverse health impact of this non-threshold pollutant is further reduced, in particular in the urban areas, even if compliance with the limit value is already achieved.

AEI is determined as a 3-years running annual mean PM_{2.5} concentration averaged over the selected monitoring stations in agglomerations and larger urban areas, set in urban background locations to best assess the PM_{2.5} exposure to the general population. Exposure concentration obligation with AEI below 20 µg/m³ has to be ensured by 2015, while during an 2010-2020 interval the national exposure reduction target sets an objective to reduce AEI by a specific percentage progressively depending on the AEI determined in 2010².

The new Directive also provides criteria for determining the minimum number of sampling points that shall be operated to assess compliance with the PM_{2.5} limit value and the exposure objectives, and to ensure information on PM_{2.5} and its chemical speciation in the rural background. The sampling points for AEI determination (at minimum one sampling point per million inhabitants summed over agglomerations and additional urban areas in excess of 100,000 inhabitants), should be already selected and operational since 1 January 2009. They should be set at urban background locations in agglomerations or other urban contexts, reflecting adequately the exposure of the general population. Commission expects that all stations are set within conurbation in excess of 100.000 inhabitants, unless special consideration has been required, and that larger number than minimally required will be used, in particular by the smaller Member States, to ensure representativeness and robustness of AEI.

To ensure adequate assessment, and where necessary monitoring of PM_{2.5} throughout the territory, minimum requirements are set for the combined number of PM_{2.5} and PM₁₀ sampling points, their ratio and type³.

Member States shall operate measurement sites at rural background locations away from significant sources of air pollution, for the purposes of providing, as a minimum, information on the total mass concentration and the chemical speciation concentrations of fine particulate matter (PM_{2.5})⁴. As these stations can be operated by cooperation between the neighbouring States, the Commission is preparing guidelines to support the selection and implementation of common background measuring stations for PM_{2.5} considering the number and representativeness of sites, synergies with the EMEP monitoring programme, and the responsibilities for data reporting.

The new air quality directives, once complemented by the Implementing provisions on reporting, will present a complete framework for the assessment and adequate information of the PM_{2.5} levels that should lead to and effective management of the pollutant and its precursors. The Commission supports exchange of the PM_{2.5} implementation issues within the established fora such as AQUILA and Air Quality Committee, and strongly encourages any scientific research or exchange at the technical or decision-making level that would further facilitate implementation and policy development.

¹ OJ L 152, 11.6.2008, p.1

² For instance if the AEI in 2010 (2008-2009-2010 average) is between 18 and 22 µg/m³ (e.g. 19 µg/m³) the reduction target that should be met in 2020 is 20%. The AEI in 2020 (2018-2019-2020 average) should be 20% lower than the one calculated in 2010 so lower than 15.2 µg/m³ (19-20%=15.2 µg/m³). The “exposure concentration obligation” is an absolute value; the value of the AEI calculated in 2015 on the basis of 2013, 2014 and 2015 data shall be below 20 µg/m³.

³ Annex V.

⁴ Article 6(5).

European Emissions of PM_{2.5} and its Precursors

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Emissions play a crucial role in understanding ambient PM_{2.5} concentrations and achieving desired reductions in these concentrations. Emissions are the pressure on the environment that causes certain concentrations to occur. If we want to model the concentrations more accurately, emission data quality needs to improve accordingly. If we conclude that concentrations need to be reduced, this calls for emission reductions. What are the emission sources than, and can they be controlled? If we want to be more spatially explicit in our exposure assessments, predicted concentrations (and therefore emission data) need to become more spatially explicit. So, even though emission numbers seem rather abstract (what is the relevance of 10 tons of PM_{2.5} emission?), knowledge of emissions is crucial.

In this paper emissions of PM_{2.5} and its precursors will be discussed. PM_{2.5} concentrations in the air are caused by direct (primary) PM_{2.5} emissions and formation of secondary particulate matter from gaseous components (precursors) in the atmosphere. The most important precursors for PM_{2.5} are nitrogen oxides (NO_x), sulfur dioxide (SO₂) and ammonia (NH₃). By coupling all emission process to source-specific spatial distribution proxy's we are able to make high resolution, spatially explicit emission maps suitable as model input. Total PM_{2.5} for UNECE Europe excluding international shipping amounts to 3400 kt and about half of this is carbonaceous aerosol (organic matter). Primary sources of *non*-carbonaceous particles are combustion in industries and residential combustion, comprising fly ash and suspended product particles. Defining components in the primary PM_{2.5} emissions may help in singling out the most health-relevant sources. Despite the capability to deliver detailed emission inventory data there is also a darker side to PM_{2.5} emissions. Most estimates are based on (assumed) fractions of PM_{2.5} in PM₁₀, direct PM_{2.5} emission factor measurements are limited and not all sources are represented. These shortcomings will be discussed. Further research and input from other disciplines is needed to make the necessary improvements.

Oral presentation

Emissions Inventories For PM_{2.5} and Precursors in Germany, Compilation of Spatially and Vertically Highly Resolved Emission Inventories for Germany

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According to projected and modelled ambient air concentrations, large scale exceedances of PM_{2.5} limit values in Germany are expected despite the implementation of current national and international legislation. Identification of the causes of these (current and projected) exceedances is crucial to develop successful mitigation options. Furthermore, it is necessary to examine in what way further national abatement measures are able to effectively ensure the expected PM_{2.5} limit values in the near future.

Thus a methodology is needed to determine the causes for exceedances of PM_{2.5} and to identify appropriate and cost efficient emission abatement measures to reduce the PM_{2.5} concentrations in Germany. For this, the contribution of transboundary transported particle precursors as well as the influence of biogenic and natural emission sources has to be considered. Thus, emission inventories in a high spatial and vertical resolution are necessary. In this contribution methodologies for preparing emission inventories in high spatial and vertical resolution are described. As result emission inventories for PM_{2.5}, PM₁₀, NO_x, SO₂, NH₃ and NMVOC developed for Germany in a high spatial and vertical resolution and vertical (1 min x 1 min) will be presented. The gridded emission data is based on geographical and statistical information on point sources as well as line sources and area sources that have been quality checked with national experts. The inventories have been compiled on the basis of the German central emission data base (ZSE) that has been extended by additional expert data bases, e.g. for resuspension of particles from road transport. A huge effort was made to develop new proxy data for the spatial distribution, e.g. for a better representation of emissions from wood fires in households.

For the development of mitigation strategies the compilation of reference emission data sets for 2010, 2015 and 2020 is crucial. Hence, for future years highly resolved emission inventories have also been compiled on the base of official emission data for Germany.

PM_{2.5} Measurements in Europe

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Measurements of particulate matter have been carried out in Europe since many years. During the 80's total suspended particulates had been measured by means of the "Black Smoke OECD" reference method (Directive 80/779/EC). Studies on health impact assessment of particles have led to the revision of European air quality policy during the 90's. The PM₁₀ fraction of PM, considered as inhalable, has been introduced with the publication of Directive 1999/30/EC. In line with the Clean Air for Europe strategy of the European Commission to minimize harmful effects of pollution on human health and the environment and to improve monitoring and assessment of air quality, the measurement of PM_{2.5} has been launched by the recently adopted Directive 2008/50/EC. Member States now have to measure as well PM_{2.5} concentrations on the basis of "common methods and criteria".

The CEN Technical Committee 'Air Quality' - Working Group 15 (PM₁₀ and PM_{2.5}) has been dealing with the set up of a standardized measurement method for PM_{2.5}. Extensive validation measurement campaigns had been carried out in order to describe the best suitable consensus method for PM_{2.5} measurements for the purpose of Directive 2008/50/EC. The European PM_{2.5} Standard method EN 14907 is under discussion since its publication and currently being revised by the Working Group. The results achieved by applying the standard method can vary due to influencing factors like choice of sampler, filter material, conditioning of filters, sampling duration, sampling time of the day, temperature, etc. Recent results of studies on filter material and the influence of filter conditioning will be presented.

For the purpose of harmonizing PM measurement methods the European Commission's Joint Research Centre and the AQUILA Network of National Air Quality Reference Laboratories have decided to organize a PM quality assurance campaign in Europe. From 2006 until 2009 18 measurement campaigns have been carried out in the European Member States by means of the JRC mobile laboratory carrying out parallel measurements to the Member States National Reference Laboratory and a routine monitoring station. The JRC mobile PM laboratory is equipped with reference samplers for PM₁₀, PM_{2.5}, PM₁, an optical particle counter, a continuous PM₁₀ instrument (TEOM FDMS) and a semi-continuous elemental and organic carbon analyzer. Results of the comparability of PM_{2.5} measurements will be presented.

PM_{2.5} Measurements and Source Apportionment

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Airborne particulate matter samples have been collected at urban roadside, urban background and rural locations. In addition to mass measurements, chemical characterisation has been conducted. Gradients in both mass concentrations and individual chemical constituents are explicable in terms of known sources, especially road traffic. Application of the University of Birmingham Pragmatic Mass Closure model has given good quantitative insights into the contributions of different broad source categories but lacks detail in terms of the organic matter fraction and the coarse traffic-related particles. In order to better understand the source apportionment of organic matter, in a recently completed project, organic molecular marker species have been analysed and the results used in a chemical mass balance model in order to estimate the contributions of individual sources such as coal combustion, natural gas combustion, wood smoke etc.. Preliminary results from this work will be presented. Additionally, results will be shown of measurements of non-exhaust traffic components.

Health Effects of Particulate Matter : Facts and Uncertainties

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Evidence accumulated over more than 50 years of epidemiologic and clinical research has established the adverse effects of air pollution on human health. The London smog of December 1952 caused > 4,000 excess deaths (Ministry of Health, 1954); in spite of the dramatic decreases in levels of air pollution that have been achieved since then, the association between air pollution and cardiorespiratory morbidity and mortality persists (Anderson et al. 2003; Dockery et al. 1993; Peters and Pope 2002). These associations are strongest for fine particulate matter (PM < 2.5 µm in aerodynamic diameter; PM_{2.5}), and the majority of excess deaths are due to cardiovascular events (Pope 2000). Despite the strength of the epidemiologic evidence and the emergence of promising hypotheses, the constituents and biological mechanisms responsible for the cardiovascular effects of air pollution are only beginning to emerge. However, there is scientific evidence that the mass-based exposure-response relationships of particulate matter may vary in different regions of Europe as well as locally due to larger than average exposures to particles from certain emission sources (e.g. traffic, domestic heating with solid fuels, resuspended dust, poorly controlled metal industry). Moreover, coarse thoracic particles (size 2.5-10 µm) and ultra fine particles (size < 0.1 µm) have shown health effects independent of PM_{2.5}.

In view of the emerging evidence implicating that fine particles are associated with health effects, it has been recommended by e.g. EU COST 633 (www.cost633.dk) that an additional and/or alternatives metric for such as ultrafine PM and/or source-related fractions should be developed. It is not possible for the exposure to PM to be set to zero for both technological and economic reasons, even though at PM concentrations well below current European Union standards, human reaction will still be present. In other words no threshold has been determined yet for the occurrence of adverse health effects to “at risk” populations. This presentation will provide reflections on the current state of knowledge on PM associated health effects.

Oral presentation

PM_{2.5} Speciation / Source Apportionment

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The major components of PM_{2.5} are usually inorganic constituents like sulphate, nitrate and ammonium and carbonaceous aerosol consisting of elemental carbon (EC, also called black carbon, BC) and organic matter (OM). While the sources of the inorganic constituents have been known reasonably well for quite a while, very little has been known until recently about the sources of the carbonaceous aerosol.

A powerful method to discriminate between 'recent' and 'fossil' carbon is the C-14 method: due to its age ¹⁴C has completely disintegrated in fossil substances, whereas modern plant material is on the contemporary radiocarbon level. This intrinsic isotopic information characterizes the sources of ambient PM independent of its history regarding emission conditions or atmospheric transport. Such C-14 measurements have been performed for quite some time, but only a discrimination of OC (organic carbon) and EC prior to the C-14 determination reveals the full power of this method: EC is often dominated by fossil fuel, while OC typically has a higher biogenic fraction. These features are not accessible in a C-14 analysis of the total carbon (Szidat et al., 2006).

Chemical mass balance (CMB) analysis has been used for a long time for source apportionment. This approach is based on tracer substances from emission measurements to perform source apportionment studies at a variety of locations. It is based on the assumption that the tracers are inert i.e., not susceptible to atmospheric degradation processes. In addition, only primary organic aerosol components can be identified, while secondary organic aerosol (SOA) is only indirectly accessible, by assuming that the ambient OC that is not apportioned by the CMB model belongs to SOA.

Another approach, which proved to be highly successful in recent years, is based on measurements with the Aerodyne aerosol mass spectrometer (AMS). This instrument provides on-line, quantitative measurements of the total mass and size distributed non-refractory chemical composition of the submicron ambient aerosol at a high temporal resolution. The full mass spectra of OM are then subjected to positive matrix factorization (PMF). As a result, several primary sources such as fossil fuel combustion, charbroiling or wood burning as well as secondary organic aerosol could be identified (Lanz et al., 2007). This approach, which was first applied for the aerosol in Zurich, was then rapidly applied also to other areas, and was found to be highly useful for such source apportionment studies. This is especially true when PMF of AMS data is combined with C-14 analysis.

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PM_{2.5} High-Volume Measurements in East Germany – a Six Year Study at Melpitz Site

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Long-time measurements of daily high volume (HV) quartz fibre filter samples PM_{2.5} were started in January 2003 (synchronous to PM₁₀ measurements remaining since 1993, Spindler et al. 2004) in the rural background of Eastern Germany at the IfT research site Melpitz (12°56'E, 51°32'N, 86 m a.s.l.) located downwind of Leipzig near the city of Torgau. The sampling device is the DIGITEL DHA-80 HV-sampler (Walter Riemer Messtechnik, Germany) with a flow rate of 30 m³/h (Gnauk et al., 2005). The PM samples were characterized for mass by weighing, the water soluble ions Cl⁻, SO₄²⁻, NO₃⁻, NH₄⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺ (WSI) were analyzed after extraction with demineralised water by standard ion chromatography and organic (OC) and elemental carbon (EC) analyzed by a two-step thermographic method (VDI 2465, Part 2, 1999).

The chemical species WSI, OC and EC shall be included in PM_{2.5} measurements in future (European Union Directive, 2008). The three year PM_{2.5} mass concentration average (2006 to 2008) for the rural Melpitz site is 17.63 µg/m³ (99.73 % data availability) and observes from the today's point of view the limit for the average exposure indicator of 20 µg/m³ for 2015, which would be based on the future mean concentration for 2008 to 2010. This value is < 18 µg/m³ and therefore the reduction target for 2020 would be 15 % (European Union Directive, 2008).

The monthly mean percentage of PM_{2.5} within PM₁₀ varies typically between winter (91.06 % January 2006) and summer (63.18 %, August 2003). The highest values were reached during cold and dry days in winter and the low percentage of PM_{2.5} within PM₁₀ in summer caused by re-emission of coarse particles in the surroundings (agricultural activities). The PM_{2.5} and PM₁₀ mass concentration and the content of WSI, OC and EC varies also strongly with the two main long range air mass transport pattern for the Melpitz site. These are more maritime and mostly dry continental air masses from West or East, respectively. Days with long range transport from these both sectors were selected using 96-hour backward trajectories (NOAA-Hysplit-Model). The highest particle mass concentration, with high parts of SO₄²⁻, NO₃⁻, OC and EC was reached in winter during the transport of air masses from East. Reasons are the long-range transport of anthropogenic emissions and a secondary particle mass formation. The main source regions for these air masses are located outside (Russia, Belarus and Ukraine) and inside (Poland, Czech Republic and Slovakia) of the European Union. Such situations can contribute to high PM concentrations in East German cities, when mixing volume and wind velocity are low.

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Real-time Measurements of PM_{2.5} and PM₁ Chemical Composition: Experience and Results from the UK Supersites and EMEP Intensive Measurement Periods

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Measurements of PM_{2.5} conventionally fall into two categories: mass measurements with manual or automated (real-time) techniques and composition measurements using (daily) filter measurements. However, new developments in measurement technology allow PM_{2.5} chemical composition to be monitored at hourly time resolution or better. At the Scottish EMEP Supersite Auchencorth PM_{2.5} (and PM₁₀) chemical composition has continuously been monitored since June 2006 using a sampler based on continuous wet denuders and steam jet aerosol collectors (MARGA) and a second instrument has now been installed at the English Supersite Harwell. We report the experience with the long-term operation of such equipment, inter-comparisons with other measurement techniques and demonstrate the value of hourly time-resolution in interpreting the sources and controls of PM_{2.5} concentrations.

An alternative measurement approach is based on the Aerodyne Aerosol Mass Spectrometer (AMS) which measures the non-refractory chemical components of sub-micron aerosol, although a new model is being developed as a monitoring tool for PM_{2.5} composition.

A combination of MARGA and AMS instruments was deployed across Europe during the EMEP Intensive Measurement Periods of June 2006, January 2007, October / November 2008 and February/March 2009. Results from these measurements will be presented, and the individual merits of the two real-time measurement techniques will be discussed.

PM_{2.5} Modelling: "Research and Policy Challenges"

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Particulate matter (PM) pollution control is one of the main challenge highlighted by the Thematic Strategy on Air Pollution (CAFE for Clean Air For Europe), adopted by the European Commission in October 2005, under its 6th Environmental Action Program. The CAFE strategy states that particulate matter (and especially fine particles with diameter smaller than 2.5 micrometers- PM_{2.5}) is responsible today for an average reduction of life expectancy of about 8 months in Europe. Other mortality indicators also charge atmospheric particulate concentrations. This raise in concern will be translated in both European Directives related to Ambient Air quality and to national emission ceilings, currently under revision. Consequently, special care will be accorded by policy makers to PM concentrations monitoring, trends analysis, and control.

These last decades, modelling has become a powerful tool, enable to provide air pollutant concentration fields and to assess the impact of future emission reduction strategies. Numerical models run in Europe for several years with operational and regulatory purposes (for instance in the context of integrated assessment modelling). Several intercomparison and evaluation studies have demonstrated that good and acceptable results could be expected from such models for ozone issues, while the question of PM modelling is much more opened. Good performances of PM models to predict atmospheric concentrations and deposition are less systematic and still highly varying, depending on the location and the period chosen for the evaluation.

Uncertainties in emission inventories can explain a part of the gaps between observations and model results. In particular, some sources like wood combustion remain not well qualified, and the spatial and temporal variability of some emission processes are not correctly taken into account in emission models. Emissions of some organic compounds which act as precursors of secondary organic aerosols are also unknown. Uncertainties in model parametrisations can also be high. They concern, for instance, the numerical approximation adopted for some complex processes involving the interaction between chemical compounds. That means that the air quality research community is currently very active and focused on the improvement of modelling tools for PM issues. But on the other hand, pressure from the general public and decision makers pushes development and use of operational integrated systems for PM forecasting and mapping.

This dichotomy between research and use for policy purposes will be basis of the proposed communication: Considering the state of art in the field of PM_{2.5} modelling, what can be

expected from the current models for operational issues? Which gaps should be uppermost filled in? How to combine modelling and measurement information to develop robust and reliable information systems? So many questions that we aim to cover with examples illustrating by concrete air pollution cases observed these last years. The PM_{2.5} episodes that occurred in spring 2007 in a large part of Western Europe as well as those observed at the beginning of this year will be analysed and commented, according to the modelling capacity developed in Europe.

Oral presentation

Modelling PM_{2.5} Concentrations for The UK and Projections To 2020

John Stedman, Susannah Grice, Sally Cooke* and Andrew Kent

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Modelling of PM_{2.5} concentrations across the UK at background and roadside locations has been carried out for the years 2005, 2006 and 2007. Projections have also been calculated for future concentrations of PM_{2.5} up to the year 2020 using 2005 as a base year. These projections are calculated on the basis of current national and European policies. The limited measurements for PM_{2.5} up to 2008 have been used to calibrate the models. The network of monitoring sites was greatly expanded in 2009, so from this year onwards there will be more measurement data available.

The Pollution Climate Mapping (PCM) Model has been used for the modelling work presented here. This model has also been used for the compliance assessments of PM₁₀ in previous years as part of the UK annual reporting required under the Framework Directive (Directive 96/62/EC). Maps of modelled PM_{2.5} concentrations are presented on a 1km x 1km grid for the UK for current and projected years.

The model described by Stedman et al (2007) has been further developed to calculate the components of PM_{2.5} to build up a complete picture of the concentrations of PM_{2.5}. The total concentration can be split into different sources such as: secondary inorganic aerosol, secondary organic aerosol, large point sources of primary particles, small point sources of primary particles, regional primary particles, area sources of primary particles, iron & calcium rich dusts, sea salt and a residual. We present maps of the spatial distribution of each of these contributions on a 1km x 1km grid. These source apportionments can also be summarised at a specific location (Stedman & Derwent 2008) or as population-weighted means. The population-weighted means are used to assess exposure.

The modelled results are compared to the PM_{2.5} European limit values, exposure concentration obligations and exposure reduction targets introduced in the new European Air Quality Directive (Directive 2008/50/EC).

The projected concentrations and source apportionment can be used to find the potential impact of current national and European policies on PM_{2.5} concentration levels in the future. These predicted future concentrations are also required to assess the likelihood of compliance with the exposure reduction targets. The future impact of alternative potential measures can also be modelled using the PCM model, but no alternative scenarios are presented here.

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PM_{2.5} Reference Measurements and Modelling for 2008 in The Netherlands

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One of the targets of the new air quality directive focuses on the reduction of PM_{2.5} levels in urban background situations. Depending on the level a reduction of 15 or 20 % must be shown over the period from 2009-2011 to 2018-2020. Although the reduction is quite considerable in the view of the various political options such a difference is still small in comparison with the measurement uncertainty. In order to enable measurements with sufficient accuracy, to demonstrate the reduction required, one should have a very stable measurement system. The National Dutch Air Quality Network of the RIVM was in the period 2007-2008 in the process of selection of the preferred automatic monitoring system for PM_{2.5}. In this selection process the decision was made that close corporation with other, local, Dutch monitoring networks was preferable in order to have optimal comparable results in the future. Therefore it was decided to start the PM_{2.5} monitoring with the reference method itself before the first of January 2008. To achieve such an obligation requires sufficient amounts of: reference samplers, suitable urban background locations, weighing facilities and well educated staff within a short time period. This obligation could only be achieved in close corporation with the local monitoring networks in the Amsterdam region and in the Rotterdam region.

The presentation will show the results for one year of measurement data with the European reference method. A particular problem with the reference method is the fact that the risk of missing measurement data is much larger than for automatic measurement systems. Methods for treating these missing data will be discussed. This first year of measurement data is used to calibrate a map of modeled PM_{2.5} concentrations for The Netherlands.

PM_{2.5} Measurements in The Netherlands; PM_{2.5} Performance Test

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In answer to the growing interest and towards implementation of the obligations applied in the Directive 2008/50/EC, the monitoring system of this finer fraction needs to be studied. To fulfill the requirements of the new directive on cleaner air for Europe and air quality a performance test is performed.

Consequently, within the Dutch monitoring systems there are strict criteria for the performance of continuous automatic PM_{2.5} measurement systems. To guarantee a univocal check of the particulate matter concentration, following the upcoming EU Directive on ambient air quality and cleaner air for Europe, the equivalence of the measurement systems to the PM_{2.5} reference method is of high relevance.

The performance test is performed in a framework of cooperation, which involves the institutes.

- DCMR Milieudienst Rijnmond,
- Public Health Service - GGD Amsterdam,
- Province Limburg,
- Province Noord-Brabant,
- Dutch National Institute for Public Health and the Environment (RIVM).

The random uncertainty is an uncertainty arising from a random effect ($\leq 2.5 \text{ g/m}^3$) assumed annual mean PM_{2.5} limit value of $20 \text{ }\mu\text{g/m}^3$.

The result of a PM_{2.5} continuous automatic monitoring system performance test is that there are two continuous automatic measurement systems available that are equivalent to the reference method. Only one system has hourly mean values available to inform the public continuously.

Results PM_{2.5} performance test

	Number	Random uncertainty (g/m ³)	Relationship between (REF) en measurement system (AMS)
I	119	3,2	AMS = 1,02 (0,05)*REF + 2,9 (1,0)
II	191	2,3	AMS = 1,04 (0,03)*REF + 0,1 (0,7)
III	209	2,9	AMS = 0,84 (0,03)*REF - 2,2 (0,8)
IV	202	3,4	AMS = 1,13 (0,04)*REF - 2,4 (0,9)
V	206	2,9	AMS = 0,92 (0,03)*REF - 1,7 (0,8)
VI	197	1,5	AMS = 0,89 (0,02)*REF - 1,2 (0,5)
VII	173	1,4	AMS = 0,92 (0,02)*REF - 1,2 (0,5)

* Results between () are extended uncertainties (95%) of coefficients.

Organic Carbon in Whatman-QMAN Field-Blanks

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Guideline EN-12341 prescribes the use of quartz fibre filters in the reference sampling of PM₁₀. It is well-known that quartz fibre filters exhibit artefacts: volatile OC is taken up during sampling and semi-volatile OC may be lost (Turpin et al. 2000). Adsorption is in general the most important artefact.

A large number of samples has been taken in EU counties with quartz fibre filters up to now and the OC-artefact must have lead to an appreciable artificial increase in the amount of PM₁₀. However, searching for information we could not find data on the importance of this artefact.

A minimum value for the adsorption artefact may be provided by the so-called field blanks. These are filters that are put in the sample holders without being loaded. The filters take up OC via diffusion. The networks in the US report field blanks that are large in comparison with the actual OC-data. Only some scattered data are available in Europe (e.g., Vecchi et al. 2009).

Filters as received from the manufacturer often contain OC and they are therefore cleaned by pre-firing in scientific studies. The commonly used Whatman-QMA filters are pre-fired in the factory according to information from the manufacturer.

In order to assess the current uncertainties around field blanks and lot-blanks in the PM-studies in the Netherlands we performed a dedicated study on the two types of blanks with Whatman-QMA filters as used in the national reference sampling for PM₁₀.

Field blanks

The study was part of a one-year investigation to assess the composition of PM in the Netherlands at regional and urban/kerb sites. Filter sampling was performed with automated reference samplers (KFG-Leckel) on 47 mm Whatman-QMA filters. There are two filter-magazines, one with the fresh filters from which every day a new filter is shifted into the filter holder. After sampling the filter is moved to a second carousel. Field blanks were filters that remained in the Leckel magazines, without sampling air.

150 field blanks were taken evenly distributed between the two carousels and over the stations. Analysis of the set of field blanks showed quite a variation in the OC-values. The average was 68 µg with an SD of 24 µg. The mentioned value for a total filter corresponds to a concentration of OC of 1.2 µg.m⁻³. This may translate into a value of Organic Matter (OM) of close to 2 µg.m⁻³.

There was no systematic difference in the blanks from the urban sites versus those from the three regional sites, which seems to indicate that the filters are saturated with OC. This could imply that the field blanks can serve as a proxy for the adsorption artefact during sampling.

Lot blanks

In addition to the field blank we made a study of the “lot”-blanks, i.e., Whatman-QMA as received from the factory. These “lot”-blanks were taken from two separate batches that were unsealed immediately before analysis.

It was consistently observed that filters from the top of a stack contained high OC values. These values were higher than the average field blank. Further down the stack of filters the values rapidly decreased to a value that was 40% of that of the average field blank. In one of the batches the values in the middle of the stack were at the detection limit (of 5 µg). The bottom filter of a stack had (again) a higher value.

Addendum: blanks for NO₃, SO₄ and NH₄

The average value in the (150) field blanks was resp. 1.1%, 2.1% and 0.7% of the average value in the actual samples. There was a single outlier for nitrate in the series of 150 field-blanks, still with a value that was 10% of the average of the loaded filters.

Acknowledgement

The investigation was performed in the framework of Netherlands Research Programme on Particulate Matter (BOP).

References

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Modelling PM_{2,5} Hot-Spot Concentrations in Practical Applications from a Consultant's View

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The amendment to the EU council directive on ambient air quality arises the following questions concerning PM_{2,5} for the municipal clean air planning:

- What are the current urban PM_{2,5} concentration levels?
- Where are the hotspots of PM_{2,5} concentrations?
- Are exceedances of the PM_{2,5} annual average limit value to be expected?
- How will PM_{2,5} annual average values at hotspots develop?

In Germany, external consultants like e. g. engineering companies are often commissioned to answer these questions. The poster will present two studies dealing with these issues:

1. "IMMISweb – First studies to include PM_{2,5}" (2008), for the North Rhine-Westphalian State Agency for Nature, Environment, and Consumer Protection. In this short study, PM_{2,5} emission factors for road traffic were assessed, implying a methodology to estimate non-exhaust emissions. The study has been conducted to prepare the introduction of PM_{2,5} into a web-service for a city-based air quality screening available to the local authorities of North Rhine-Westphalia.
2. "Updating of the calculations for the Berlin Clean Air Plan" (2008), for the Berlin Senat Department for Urban Development. In this study, the total concentrations of NO₂, PM₁₀ and PM_{2,5} were calculated for the Berlin major road network. A model chain was used for the calculations, combining large scale results of the model REM-CALGRID (carried out by FU Berlin), urban scale results of the regional model IMMIS^{net} and local scale results of the screening model IMMIS^{luft}. The large scale calculations included PM_{2,5} emission inventories and accounted for secondary particle formation. For the urban scale, PM_{2,5} emission inventories were derived from PM₁₀ emission inventories. For the local scale, assumptions according to a) were made.

Appendix 3 List of participants

List of participants

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Appendix 4 Hyperlinks used/ presented at the workshop

Programme; Workshop 'Measurements and Modelling of PM_{2.5} in Europe', 23-24 April 2009, Bilthoven, The Netherlands

<http://www.pbl.nl/en/dossiers/Transboundaryairpollution/content/Programme-Workshop-Measurements-and-Modelling-of-PM2-5-in-Europe.html>

Presentations; Workshop 'Measurements and Modelling of PM_{2.5} in Europe', 23-24 April 2009, Bilthoven, The Netherlands

<http://www.pbl.nl/en/dossiers/Transboundaryairpollution/content/Presentations-Workshop-Measurements-and-Modelling-of-PM2-5-in-Europe.html>

DIRECTIVE 2008/50/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 21 May 2008 on ambient air quality and cleaner air for Europe

<http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF>

Equivalence guideline

http://ec.europa.eu/environment/air/pdf/test_equivalencev31004.xls

AQUILA network (air quality reference laboratories)

<http://ies.jrc.ec.europa.eu/aquila-homepage.html>

European Commission, DG Environments site on air

<http://ec.europa.eu/environment/air/index.htm>

Netherlands research programme on PM₁₀ and PM_{2.5} (BOP)

<http://www.pbl.nl/en/dossiers/Transboundaryairpollution/content/Netherlands-Research-Program-on-Particulate-Matter.html>

Report on Attainability of PM_{2.5} air quality standards in the Netherlands in a European context

<http://www.pbl.nl/en/publications/2009/Attainability-of-PM2-5-air-quality-standards.html>

EMEP PM assessment report, 2007

<http://www.nilu.no/projects/ccc/reports.html>

Paul Sherrer Institut, Technology Assessment

<http://gabe.web.psi.ch>

Paul Sherrer Institut, Laboratory of Atmospheric Chemistry

<http://lac.web.psi.ch>

PAREST-Project (PAricle REduction STrategy for Germany)

<http://www.parest.de>

EEA European Topic Centre on Air and Climate Change

Assessment of the health impacts of exposure to PM_{2.5} at a European level ETC/ACC Technical Paper 2009/1

http://air-climate.eionet.europa.eu/reports/ETCACC_TP_2009_1_European_PM2.5_HIA

The report gives an overview and proceedings of the 2009 workshop on $PM_{2.5}$ organized by the Netherlands Environmental Assessment Agency (PBL) in cooperation with TNO. The aim of this workshop was, given the European standards for $PM_{2.5}$, to discuss with a number of European experts the current state-of-knowledge concerning all aspects of $PM_{2.5}$. This means observations, emissions, modelling, source apportionment and health effects. Countries have started to include measurements of $PM_{2.5}$ and its components in their national air quality monitoring networks. Harmonization of the national air quality networks is on its way but takes a considerable amount of effort. As a consequence it is difficult to make a consistent view of $PM_{2.5}$ Europe wide. Policy assessments which aim at finding adequate conditions for compliance to the $PM_{2.5}$ standards are therefore still very uncertain. In addition there is growing evidence that, like PM_{10} , also $PM_{2.5}$ is a rather crude indicator for the health effects associated to particulate matter. So it is necessary to design smart policies which aim at the health improvement and take at the same time the current uncertainties into account about the efficiency of the $PM_{2.5}$ air quality standards for health improvement.

The Netherlands Research Program on Particulate Matter (BOP) is a national program on PM_{10} and $PM_{2.5}$. It is a framework of cooperation involving the Energy research Centre of the Netherlands (ECN), the Netherlands Environmental Assessment Agency (PBL), the Environment and Safety Division of the National Institute for Public Health and the Environment (RIVM) and TNO Built Environment and Geosciences.

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