

Second Position Paper on Particulate Matter

**CAFE Working Group on
Particulate Matter**

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Foreword

This Second Position Paper on Particulate Matter (PM) has been prepared by the CAFE Working Group on PM. The Working Group received additional support in drafting the document from a number of experts that have not been credited explicitly. The Working Group acknowledges this support, which is greatly appreciated. The Working Group further acknowledges support of the European Topic Centre on Air and Climate Change that provided Chapter 5 (Emissions).

In the final version comments given at the stakeholders' Workshop on PM in Stockholm, 20-21 October 2003, as well as from members of the CAFE Steering Group received to May 2004, have been taken into account.

The Working Group on PM has been established as integral part of the CAFE (Clean Air For Europe) programme, endorsed by the CAFE Steering Group to assist the European Commission in reviewing Directive 1999/30/EC, the First Daughter Directive on air quality. The European Commission will use the Position Paper as the basis for assessing particulate matter within the integrated assessment in the Clean Air For Europe (CAFE) programme.

The Working Group agreed the text for Chapter 11 (Recommendations concerning the PM metric and PM levels) and the executive summary at a meeting in December 2003. The conclusions of Chapter 11 were informed by the World Health Organization (WHO) report on "Health Aspects of Air pollution with Particulate Matter, Ozone and Nitrogen Dioxide" (January 2003). The WHO's answers to supplementary questions asked by the CAFE Steering Group in Spring 2003 were not available at the meeting in December 2003. They are however expected to be published around the same time as this Position Paper. The Working Group members were briefed at the December 2003 meeting about the WHO process for answering the supplementary questions. They agreed that the recommendations in the Position Paper about the particulate matter metric or the proposed range of concentrations for CAFE's integrated assessment process could be revisited at a later date if the WHO supplementary answers provided new scientific information that would likely lead the Working Group to different conclusions.

This document reflects the opinions of the members of the Working Group. The small number of cases where there was a majority view are noted in the text. This document should not be considered as an official statement of the position of the European Commission.

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Executive Summary

I. Introduction

1. What is the purpose and scope of this Position Paper? [Ch1]¹

This position paper was prepared by the EU Working Group on Particulate Matter in response to a request by the European Commission. It updates the 1997 Position Paper on Particulate Matter² that helped the European Commission prepare the First Daughter Directive on ambient air quality (1999/30/EC). Since 1997 there is considerably more information available on current and future ambient levels of particulate matter (PM) in Europe. There is also more knowledge on the health impacts of particulate matter. The paper assesses current and future ambient levels of PM in Europe in the context of the targets set in the First Daughter Directive. It assesses the attainability of current targets and also discusses the possible need and consequences of revising these targets for particulate matter.

This review is part of the policy development work within the Clean Air For Europe (CAFE) programme of the European Commission. It also supports the review of the First Daughter Directive under the Air Quality Framework Directive 96/62/EC.³

The Working Group on Particulate Matter comprised experts from EU Member States, Switzerland, Industry, NGOs, the World Health Organization (WHO) and the European Environment Agency. Annex 1 includes a list of members of the Working Group.

2. What is particulate matter? [Ch2]

The term PM is equivalent to the term atmospheric aerosol and defines a suspension of air-borne solid particles and/or droplets of various sizes. Size and chemical composition are regarded as the most important characteristics of such particles, whilst surface area and possibly particle number may also be important. A single particle usually contains a mixture of chemical and physical (solid, liquid) constituents. The PM₁₀ concentration is the mass per volume unit ($\mu\text{g}/\text{m}^3$) of particles with an aerodynamic diameter smaller than 10 micrometer (μm). The larger particles contained in the PM₁₀ size fraction reach the upper part of the lung. The smaller particles of this size fraction (in particular PM_{2.5} and PM_{1.0}, with diameters smaller than 2.5 and 1.0 μm) penetrate more deeply into the lung and reach the alveolar region. PM is often differentiated by chemical constituents (*e.g.* sulphates, heavy metals, organics), as well as by source-related constituents (*e.g.* diesel soot). Today, it has become common practice to denote the PM_{2.5} as the “fine fraction” and particles with diameters between 2.5 and 10 μm (PM_{2.5-10}) as the coarse fraction.

Size and composition of ambient PM not only depend on the emission process (see Section 3 below), but also, particularly for the finer fractions, on the atmospheric processes that the particles go through after emission. The particle mass is usually found in two size-related modes. The finer mode, up to around 1 μm , generally originates from high temperature processes and/or gas-to-particle formation processes in the atmosphere; these particles carry inorganic compounds (such as sulphates, nitrates and elemental carbon) and organic compounds, including semi-volatile components. Mechanical processes

¹ Refers to the relevant chapter in the Position Paper. The order of sections in the Executive Summary does not follow the order of chapters in the Position Paper.

² Ambient air pollution by particulate matter. Position paper. European Commission, Directorate-General XI, 1997. Available at http://www.europa.eu.int/comm/environment/air/pdf/pp_pm.pdf.

³ The important link between PM and climate change is not addressed in the draft Position Paper, but should be addressed in the CAFE process.

such as erosion, corrosion and material abrasion give rise to coarser particles, usually larger than 1 µm. These particles carry *e.g.* soil components and sea spray. Another fraction, the ultrafine particles, in size below 0.1 µm, is better characterised by the number concentration (number of particles per cm³), because despite their large number they contribute only little to the particulate mass.

PM measuring instruments do not collect particles of one single size, but rather within a certain size range. Hence size fractions such as PM₁₀ are to some extent defined by the sampling device. An important sampling artefact is that the semi-volatile fraction (such as ammonium nitrate) may not be measured, dependent on the method of sampling (see Section 9 below).

Large and very small particles have a limited atmospheric residence time due to deposition or coagulation. Particles in the size range between approximately 0.1 and a few µm remain much longer in the atmosphere (typically several days to a week) and can consequently be transported over long distances (1000 or more kilometres).

3. What are the sources of PM in Europe? [Ch5, 6]

PM is emitted directly from 'primary' sources (primary PM) and is also formed in the atmosphere by reaction of precursor gases (secondary PM). Other common distinctions are natural/anthropogenic sources and combustion/non-combustion sources. The emission estimates from non-combustion sources have a considerable degree of uncertainty.

In the current EU, primary PM₁₀ emissions are dominated by sectors including road transport, industry and fuel combustion in the residential sector. In the Acceding Countries, the largest sectoral contribution of PM₁₀ emissions comes from the energy industries sector, with a relatively small contribution from road transport.

The road transport sector contributes with both vehicle exhaust particles and resuspension of road dust. The main sources of precursor gases are fuel combustion in large installations (for SO₂), transport sector (for NO_x), solvent use and road transport (for NMVOC⁴) and agriculture (for NH₃).

A further contribution to total ambient particulate comes from a variety of natural sources, which vary widely from area to area and with time. These include sea salt (especially important in coastal regions), crustal material arising from natural erosion processes (especially important in dry regions) and biological material. Volcanic eruptions are an example of both a source for natural primary particle emissions and a source for precursors of secondary PM.

II. Health effects

4. What are the health effects of PM? [Ch10]

The WHO air quality guidelines⁵ formed the scientific basis of the PM limit values given in the First Daughter Directive. Since then, a large body of new scientific evidence has emerged that has strengthened the link between ambient PM exposure and health effects. New analyses have shown death being advanced by at least a few months on population average, at current PM concentrations in Europe, for causes such as cardiovascular and lung disease. Furthermore, there are robust associations between ambient PM and increases in lower respiratory symptoms and reduced lung function in children, and chronic obstructive pulmonary disease and reduced lung function in adults. Most epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on mortality and morbidity. It is likely that within any large

⁴ Non methane volatile organic compounds

⁵ WHO Air Quality Guidelines for Europe, Second edition. Copenhagen, WHO Regional Office for Europe, 2000 (WHO Regional Publications, European Series, No 91)

human population, there is a wide range in susceptibility so that some subjects are at risk even at the low end of current concentrations.

5. What is causing the health effects? [Ch10]

In its recent review⁶, WHO has concluded that there is a causal relationship between PM exposure and health effects. It has not however been possible to establish a causal relationship between PM-related health effects and one single PM component. This is in spite of intensive research roughly over the last decade. Nevertheless there is strong evidence to conclude that fine particles – usually measured as PM_{2.5} in health effects studies – are more hazardous than larger ones. This does not imply that the coarse fraction of PM₁₀ is innocuous. PM characteristics found to contribute to toxicity include: metal content; presence of polycyclic aromatic hydrocarbons and other organic components; endotoxin content, and small (less than 2.5 µm) and extremely small (less than 0.1 µm) size.

Epidemiological studies suggest that a number of emissions sources are associated with health effects, especially motor vehicles and also coal combustion. Toxicological studies show that particles originating from internal combustion engines, coal burning, residual oil combustion and wood burning have strong inflammatory potential.

6. Does the new scientific evidence on health effects justify reconsideration of the current WHO guidelines? [Ch10]

The present information shows that fine particles (commonly measured as PM_{2.5}) are strongly associated with mortality and other endpoints such as hospitalization for cardio-pulmonary disease, so that WHO recommended that air quality guidelines for PM_{2.5} be further developed. Based on these findings, WHO will update exposure-response relationships for the most severe health outcomes induced by particulate matter presented by air quality guidelines. In addition, WHO recommended to re-evaluate the value of black smoke as an indicator for traffic-related air pollution.

III. Current legislative provisions on ambient PM

7. What is current EU air quality legislation? [Ch1]

The First Daughter Directive to the Air Quality Framework Directive sets limit values for PM₁₀ to be met by 2005 and indicative limit values for 2010 and beyond (Table 1). The directives lay down requirements for Member States to assess air quality and report annually to the Commission.

Member States have to develop and implement measures to ensure compliance with the limit values in 2005. Currently, the First Daughter Directive is being reviewed. During the development of this directive, uncertainties regarding the fraction causing the association with health effects, PM sources and ambient concentrations were extensively discussed (see the first Position Paper on Particulate Matter). PM_{2.5}, then considered as a candidate metric for limit values, was deferred until the current review. The knowledge on ambient PM_{2.5} levels and its health effects was at that time considered insufficient for justifying a PM_{2.5} air quality threshold.

⁶ Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide. WHO, Bonn, Germany, 2003 (<http://www.euro.who.int/document/e79097.pdf>)

Table 1 Limit values for particulate matter (PM₁₀)

	Averaging period	Limit value	Date by which limit value is to be met
STAGE 1			
1. 24-hour limit value for the protection of human health	24 hours	50 µg/m ³ PM ₁₀ , not to be exceeded more than 35 times a calendar year	1 January 2005
2. Annual limit value for the protection of human health	Calendar year	40 µg/m ³ PM ₁₀	1 January 2005
STAGE 2 ⁽¹⁾			
1. 24-hour limit value for the protection of human health	24 hours	50 µg/m ³ PM ₁₀ , not to be exceeded more than 7 times a calendar year	1 January 2010
2. Annual limit value for the protection of human health	Calendar year	20 µg/m ³ PM ₁₀	1 January 2010

⁽¹⁾ Indicative limit values to be reviewed in the light of further information on health and environmental effects, technical feasibility and experience in the application of Stage 1 limit values in the Member States.

8. How are PM concentrations in ambient air currently assessed? [Ch1, 3, 4]

The First Daughter Directive requires all Member States to assess the ambient concentrations of PM₁₀ throughout their territory. The assessment has to be based on monitoring at a considerable number of sites and may be supplemented by modelling. Measurements of PM_{2.5} at a limited number of sites are also required. Many Member States and Acceding Countries have substantially changed their existing networks in order to comply with the new requirements. There are now over one thousand PM₁₀ stations in the EU plus the Acceding Countries, which measure concentrations in remote and rural areas, urban 'background' areas and at 'hot spots' (near road traffic and some industrial sources). The air quality directives also encourage assessment by modelling, especially for constructing maps of pollution levels.

PM measurements (mass and composition) are also part of the EMEP⁷ monitoring programme. In addition, many research activities related to PM are ongoing in Europe within individual Member States and also under the Fifth European Framework Programme for Research. Many of these projects aim at better understanding of the sources and atmospheric processes that give rise to PM in the atmosphere.

9. Are current assessment methods adequate and can they be improved? [Ch3]

- Measurement

Measurement techniques, even for the well-established PM₁₀ metric, are still intensely debated. The reference method (gravimetric: conditioning and weighing filters on a precision balance before and after sampling on a filter in the field) essentially defines what PM₁₀ is, by specifying the device that separates (with a finite cut-off range) particles up to 10 µm from larger particles. This method is labour-intensive and does not provide up-to-date information to the public. There are also other methods widely in use, in particular the beta-absorption and TEOM[®] methods. Discrepancies between these automatic methods and the reference method are significant (typically 10 to 30%, sometimes as much as 50%).

An EU working group has provided guidance⁸ on how to determine a correction factor to achieve the equivalence required by the First Daughter Directive. The equivalence problem may be larger for PM_{2.5}. However, the automatic methods are continuously being improved to become less susceptible

⁷ Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe under the UN-ECE Convention on Long-Range Transboundary Air Pollution

⁸ European Commission (2002), EC Working Group on Particulate Matter. Guidance to Member States on PM₁₀ monitoring and intercomparisons with the reference method.

to artefacts. A CEN report on procedures to establish the equivalency of measurement methods is being prepared and will be available in 2004.

The information from the improved monitoring networks, now reported routinely under the new directive, is dramatically improving our knowledge of PM₁₀ concentrations. The comparability between Member States and regions has increased, though it is still far from ideal; measuring methods have become increasingly harmonised; the First Daughter Directive specifies some data quality objectives and there is some guidance on correction factors. Station siting criteria have improved the information on the representativeness of station sites. However, station siting strategies are still not always similar between Member States, and the strategy on the location of the monitoring stations should be harmonised and information provision improved.

- Modelling

Computer models are another very important class of tools for assessing PM, particularly where it has not been measured. Models can be used to construct territory-covering pollution maps and overviews. Models are also indispensable for estimating future PM levels. Air quality modelling is by now a well-developed scientific field, but there are still considerable limitations, especially for PM. The limitations depend on the spatial scale and the PM metric addressed.

Many model calculations tend to underestimate concentrations, probably mainly due to missing sources or to low estimates in the emission data base. Formation of secondary components, as organic and nitrates, adds to the uncertainty on regional scale, while difficulties in describing resuspension of road dust and the dispersion of pollutants in a complex area play a role on a local scale. Conclusive validation of the models by measurements is very important. Concerning metrics others than mass, *e.g.* particle number concentration, the models are still in development and are presently tested in different research projects.

IV. Current PM Situation

10. What are current ambient PM levels in Europe? [Ch4, 6]

Information on PM₁₀ levels, and to a lesser extent on PM_{2.5} levels, has greatly improved following implementation of the Council Decision on Exchange of Information 97/101/EC and the First Daughter Directive. Information on several other metrics, notably PM_{1.0} and ultrafine particles, is still very scarce because they are not monitored on a regular basis. The general picture is that the mass fractions PM₁₀, PM_{2.5} and PM_{1.0} are distributed somewhat smoother in space than many other pollutants. This is due to elevated regional background levels in many parts of Europe.

Rural background levels of PM₁₀ are relatively high in some Acceding Countries and also in the Netherlands, Belgium and parts of northwest Germany. Urban PM₁₀ levels tend to be highest in Southern, Central and Eastern Europe (some Acceding Countries), and lowest in the Nordic countries; but the EU-wide pattern is rather complicated, spanning a concentration range of more than a factor of three. At hot spot locations (near busy traffic and certain industrial sources) PM₁₀ levels measured are typically 40% above the urban background level, with a considerable range (up to 100%). In 2001, the limit values for 2005 were exceeded at 34% of the 718 monitoring stations. The 24-hour limit value was much more frequently exceeded than the annual limit value. The indicative limit values for 2010 were exceeded more extensively: about half of the rural background stations measured exceedance.

Annual mean PM_{2.5} levels are roughly two-thirds those of PM₁₀, but substantial variations in space and time have been reported (ranging from 40% to 80% for individual stations). The spatial distribution of PM_{2.5} levels is not well known, but tends to be somewhat smoother than the PM₁₀ distribution. There is a tendency for lower PM_{2.5} levels in Scandinavian cities. At busy traffic sites, PM_{2.5} levels are typically about 40% higher than in the urban background; this is comparable to the local gradients for PM₁₀.

The majority of urban background sites met in 2001 an annual average PM_{2.5} threshold of 20 µg/m³, whereas this threshold was exceeded at traffic-exposed sites in 15 out of 23 cases.

The scarce data on PM_{1.0} indicate that the levels are typically about half of PM₁₀. There is evidence that the number concentration of ultrafine particles varies much stronger spatially, with a range of an order of magnitude going from rural to hot spot levels.

V. Historic and future trends of emissions and levels of ambient PM in Europe

11. What are the historic trends in emissions and concentrations of PM? [Ch5, 7]

Emissions of primary PM₁₀ have been reduced between 1990 and 2000 by 18% across Europe as a whole. Emissions of precursors of PM have decreased substantially.

Annual average PM₁₀ concentrations have generally decreased on the average by about 15-20% during 1997-2001, but there is considerable scatter in the time series due to variations in meteorological conditions and there are deviations from this general picture. There is evidence from several countries that PM₁₀ concentrations in 2002 and 2003 show an increase, mainly due to meteorological conditions.

The reasons for the overall downward trend are not well understood, but indications are that the trend is partially due to the success in reducing inorganic secondary particle precursor gas emissions (mainly SO₂ and NO_x). There are currently insufficient PM_{2.5} data to identify any trends, although a downward trend is to be expected because of reductions in secondary particle mass.

12. How will PM emissions and concentrations change in the future? [Ch7]

Projected PM₁₀ concentrations show a continued decrease in the coming decade. These modelled data are scarce and mainly relate to annual mean concentrations at background locations. Most of the calculated PM₁₀ reduction stems from secondary inorganic aerosol reductions, which models appear to compute fairly accurately. Regional scale models calculate only a small impact of primary PM related additional abatement measures on background concentrations, but the impact may be greater for urban and hot spot concentrations. In addition, the primary contribution may be underestimated because the observed trend in the past few years is faster downward than models indicate. The model predictions are uncertain because of incomplete data and understanding about source contributions from both natural and anthropogenic sources.

13. Will Member States meet the targets set by the First Daughter Directive? [Ch9, 4]

Without additional policies and measures there will be widespread non-attainment of the Stage 1 and Stage 2 limit values in the EU. The Stage 1 24-hour limit value is more stringent than the annual average limit value. With current policies, PM levels at many locations across the EU are likely to exceed the Stage 1 24-hour limit value in 2005; even with ambitious measures, these limit values seem unattainable in the most polluted areas by 2005.

The less stringent Stage 1 annual average limit value is likely to be attained by 2005 in most Member States. There are however likely to be exceptions at highly polluted locations and some urban background areas, mainly in some Mediterranean countries (Italy, Spain and Greece); this is also expected for central European countries such as Poland and Czech Republic. At this stage, there is not enough information to judge if additional measures will be sufficient to comply with this limit value in 2005.

One of the main barriers to attainability is the high regional background concentration of PM₁₀ caused by long-range air pollution. Attainability of the limit values, particularly the more stringent Stage 1 24-hour limit value and the Stage 2 indicative limit values, is largely outside the control of individual Member States. Europe-wide action on further reducing precursor emissions is needed if limit values are to be met.

Technical options are available to help Member States further reduce primary and PM precursor emissions. There is however comparatively little information *e.g.* on the most practical and cost-effective balance of local, regional, national and European scale measures to meet limit values.

Up to 50% reductions in PM₁₀ levels are needed in some locations by 2005. Measures that have an almost immediate effect on emissions would be required simultaneously at a local, regional and international level, but this is impractical.

Significant additional cost would be incurred by most Member States to meet the Stage 1 limit values and by all Member States to meet the Stage 2 limit values at all relevant locations. On the other hand, there would be considerable health benefits to the European population. These costs and benefits have however not been estimated, except in a few Member States.

VI. Possible further legislation and principal recommendations

14. What are possibilities for further reductions of PM levels? [Ch8]

Numerous control options are available for PM emissions from all major combustion sources. These include technical and non-technical measures. The size fraction affected depends on the source characteristics and the measure applied, but is mainly the fine fraction.

Numerous measures have also been described and tested for non-combustion sources, even though studies showing the quantified effects of such measures are scarce. Measures for non-combustion sources are more effective for reducing PM₁₀ than PM_{2.5} emissions.

IIASA⁹ has estimated the reduction potential of including additional measures in scenarios containing currently agreed abatement measures for 2010. In general, a large potential exists for reductions of precursors of secondary aerosols (SO₂, NO_x, NH₃ and NMVOC). The information on the potential for additional reductions of primary PM over current legislation scenarios is somewhat conflicting. In general larger reduction potentials for primary PM – additional to those arising from current legislation – can be expected for Acceding Countries compared to current Member States.

15. What are reasons for reconsidering the existing limit values?

- The review requirement in the First Daughter Directive [Ch1]

The mandatory review of the First Daughter Directive includes confirmation or revision of the indicative limit values for PM₁₀ in 2010. These limit values will be reviewed within the development of a thematic strategy for air pollution under the CAFE programme.

- Health effects [Ch10]

There is increasingly robust evidence of health effects associated with ambient PM. Severe health risks remain despite the current decreasing trend in PM levels. There are now stronger indications that particles smaller than 2.5 µm are the main cause of the health effects.

- Attainability [Ch9]

In many countries attainment of the 2005 limit values may not be possible everywhere. Attainment of the 2010 indicative values does not seem possible in most countries, even with maximum feasible emissions reductions.

⁹ International Institute for Applied Systems Analysis, Laxenburg, Austria

16. What are the consequences of revising PM limit values? [Ch11]

Key consequences of revising the PM limit values and/or metric include:

- Maintaining the PM₁₀ metric but changing the limit value or attainment year would affect the extent of attainment problems anticipated and the associated pace of health risks reduction in Europe.
- Replacing PM₁₀ with a metric for smaller particles such as PM_{2.5} would make reductions of mechanically generated particles, which are predominantly in the coarse size range, less useful for attaining the limit value.
- Choosing a metric for which ambient concentrations are markedly raised near emissions sources – such as particle number concentration –, rather than a limit value based on PM_{2.5}, would increase the effectiveness of local mitigation measures.
- Current measures to reduce PM₁₀ concentrations are also effective for reducing PM_{2.5} concentrations. The First Daughter Directive requires that measures taken to comply with the PM₁₀ limit value should also aim at reducing PM_{2.5}. Therefore it can be assumed that measures taken to comply with the current PM₁₀ limit value will also help comply with any possible new PM_{2.5} limit value.
- Replacing the PM₁₀ metric with a PM_{2.5} metric would require only minor and inexpensive changes to the current monitoring requirements. Changing PM₁₀ to PM_{2.5} monitoring should only require a change of the size pre-selector (sampling inlet) on existing instruments. Conversely, adding a PM_{2.5} threshold to the existing PM₁₀ limit value would require the setting up of new monitoring equipment.

17. What PM metric is recommended by the Working Group? [Ch11]

During the preparation of the 1997 PM Position Paper both PM₁₀ and PM_{2.5} were considered as a possible metric for limit values. It became apparent that there was not sufficient information to justify recommending PM_{2.5}. Since then, WHO has stated that "fine particles (commonly measured as PM_{2.5}) are strongly associated with mortality and other endpoints such as hospitalization for cardiopulmonary disease" and has recommended the development of air quality guidelines for PM_{2.5}. At the same time, it was stated "continuation of PM₁₀ measurement is indicated for public health protection" and that the coarse fraction of PM₁₀, i.e. PM_{2.5-10}, cannot be considered innocuous.

In the light of these health-related findings the PM Working Group recommends the use of PM_{2.5} rather than PM₁₀ as the principal metric for assessing exposure to PM.

It is further recommended by the majority of the Working Group that once PM_{2.5} limit values have come into force and have replaced the Stage 1 PM₁₀ limit values, the PM₁₀ indicative limit values currently set for Stage 2 in the First Daughter Directive should be reclassified as target values with the aim to help control the coarse fraction, PM_{2.5-10}. The numerical value of the PM₁₀ target value should be reconsidered in view of the results of the integrated assessment modelling. It would be sufficient to carry out PM₁₀ measurements at some 20% of the PM_{2.5} monitoring sites. Reasonable distribution of the PM₁₀ measurements should be ensured among the various types of sites (urban background, traffic, etc.), particularly where the coarse fraction may comprise a significant part of the mix.

The Working Group judges it too early to consider regulation of PM_{1.0} and ultrafine particles. However, Member States should be asked to carry out more research in the coming years to establish more valid information on these PM metrics with regard to both concentration levels and adverse health effects. The Commission should collect and review the results of this work within five years.

18. What PM concentration levels should be considered by the CAFE process? [Ch11]

In view of the serious health effects of PM (see Section 4 of the executive summary), the PM level should be as low as reasonably achievable for health protection. This is especially so for PM, where

there is a substantial natural component that is variable across Europe. While it is recognised that limit values have an important role to play in helping drive down exposure, the Working Group therefore recommends that the Commission consider the use of alternative approaches, such as gap closure or targets, to supplement (but not replace) the use of limit values.

If the limit value approach is to be followed, the Working Group found it reasonable to start considerations for PM_{2.5} levels from the advice developed by WHO. The results from studies of long-term exposures to PM_{2.5} suggest that an exposure-response relationship down to the lowest observed levels seems to be appropriate, with elevated risk even at (long-term) PM_{2.5} levels below 10 µg/m³. The majority of the Working Group concludes that a reduction of exposure to fine PM down to such levels would be desirable from the health point of view. However, the Working Group also noted the necessity to take other considerations into account like feasibility when establishing a legally binding limit value. Taking further into account the currently observed levels, the anticipated decreasing trends of fine PM, in particular the secondary inorganic PM, and that the attainment date for a new PM_{2.5} limit value would not be before 2010, the Working Group concluded that such a limit value should preferably not exceed 20 µg/m³. The limit value would apply where people are likely to be exposed for a period that is significant in relation to the averaging period of the limit value. Given that in setting a final level, the attainability of any such level would have to be taken into account, the Working Group concludes that no single PM level should be recommended at this time. Rather, values within the range 12 to 20 µg/m³ should be used as an input to the integrated assessment procedure. Consideration will also need to be given to the appropriate attainment date.

According to WHO both a short-term (24-hour) and a long-term value (annual average) should be established for PM_{2.5}. From the available information about the frequency distribution of 24-hour values – which is scarce –, the majority of the Working Group recommends that a 24-hour average limit value for PM_{2.5} around 35 µg/m³ (not to be exceeded more than 10% of the days of the year) seems reasonable as a starting point.

The value for the PM₁₀ target value should be reconsidered in view of the results of the integrated assessment modelling.

VII. Other Recommendations

1. There is a need to fill in the considerable gaps that remain in scientific knowledge and evidence on PM characteristics, to help inform policy makers about future strategies for reducing the health impacts of PM. Examples are the few data available for urban areas on PM_{2.5} and PM_{1.0} concentrations, particle composition and number concentration of particles. [Ch2]
2. The Reference Method for PM₁₀ (EN 12341, adopted in 1998) should be reviewed as soon as possible taking into account the forthcoming Reference Method for PM_{2.5}. This is well in line with the regular review cycle of CEN (5 years). [Ch3]
3. Current automatic monitoring methods for PM are, in many instances, not equivalent to the reference method. It is considered important that PM measurements in the EU are harmonised. To help ensure this, the Commission has prepared a 'Guidance on Equivalency' (to be published in 2004). In accordance with the First Daughter Directive, this allows for corrections to data to be applied before checking that the remaining uncertainty meets the data quality objectives. Member States should apply this guidance once it is available. [Ch3]
4. Member States are encouraged to intensify QA/QC exercises within their State but also between different Member States. [Ch3]
5. The European Reference Laboratory for Air Pollution, of the Joint Research Centre (JRC/ERLAP) is encouraged to intensify its efforts to support the harmonisation of European PM measurements.

AQUILA (European Air Quality Laboratories, the Network of National Reference Laboratories designated according to the Air Quality Framework Directive) should be active in this field. [Ch3]

6. In revising the First Daughter Directive it is important to address the current contradiction that the reference method cannot be used to meet the daily reporting requirements. [Ch3]
7. If new limit values are adopted for $PM_{2.5}$, it is recommended that careful consideration be given to establish appropriate PM monitoring and assessment strategies. [Ch3]
8. In view of the importance of station siting for the comparability of data it is recommended to review current practice in siting and to consider stricter siting criteria, particularly in the Exchange of Information Decision, which should be harmonized with the requirements of the First Daughter Directive. [Ch3]
9. To ensure better interpretation of results, it is essential to report a detailed description of monitoring sites used for compliance monitoring, including data on the exact locations, the surroundings and representativeness of the stations. Member States should clearly document and report any correction factors applied to the data. [Ch3]
10. Whilst considerable progress has been made in PM model development, there is still a need to improve, compare and validate models at all scales for a better understanding of PM and for provision of policy support. [Ch3]
11. As the uncertainty in model results is to a large extent determined by the uncertainty in anthropogenic and natural emissions of particulate matter, it is important to improve emission databases in Europe. For modelling there is a particular need for better emission data at the local authority level. [Ch3]
12. In view of the emphasis on $PM_{2.5}$ in the new WHO recommendations, $PM_{2.5}$ monitoring should be intensified as soon as possible to give representative data from Europe, covering all relevant area types. [Ch4]
13. There is limited information on other PM metrics or parameters related to PM, such as chemical composition including elemental carbon, $PM_{1.0}$, and the number concentration of ultrafine particles. It is recommended to set up some 20 monitoring stations throughout the EU where the different parameters are being monitored on a longer term. These stations should be set up and operated in a harmonized way to ensure comparability of data, and cover the relevant types of stations, e.g., rural, urban background and traffic sites. The data obtained from these monitoring sites should mainly serve to perform better source apportionment, validation models and trend analysis. They may also help to calculate correlations between the different parameters. It is recommended to aim this network also at facilitating better targeted future strategies for reducing the health impacts of PM and identifying the components and/or properties of PM which are particularly relevant for health effects. [Ch4]
14. Indoors, where citizens spend much of their time, PM levels are only partially dependent on outdoor levels. This is also true for passenger cabins of vehicles. More research is needed in order to identify the contributions of outdoor, indoor and other (personal activities and community) sources to personal exposure to PM. [Ch4]
15. Further measurements of particulate emissions are recommended to allow the development of more robust, country-specific emissions factors. Particular attention is also required to improve the emission factors for $PM_{2.5}$, to improve the quality of inventory reporting for this pollutant. [Ch5]
16. Countries are requested to improve emission reporting at European level, since these data are essential for the policy development process in Europe. [Ch5]

17. The UN/ECE Task Force on Emission Inventories and projection, in close collaboration with the EEA and national experts, is encouraged to work on improved inventories to get a harmonized European-wide PM emission inventory and projections. [Ch5]
18. Data on chemical composition of PM emissions is still scarce. Such data is needed for source apportionment and validation of PM emission inventories and models using monitoring data. Therefore, typical source profiles should be established for the main sources of primary PM. [Ch5]
19. Quantification of the resuspension source is still uncertain, and several models based upon different assumptions exist. Being an important source for near-road PM₁₀ occurrence and also relevant for PM_{2.5}, efforts should be put into better quantification of this source. [Ch5]
20. More information to quantify sources of both natural particulate emissions and natural secondary PM precursors is needed. [Ch5]
21. In rural areas, secondary inorganic species account for the largest contribution of PM₁₀ and PM_{2.5}, both about 35-55%. Second in mass is elemental and organic carbon, accounting for about 15-35% of PM₁₀ and 17-40% of PM_{2.5}. To reduce regional background concentrations, PM emissions abatement strategies should therefore focus on reducing primary carbonaceous compound emissions and gaseous precursors of secondary components of PM. [Ch6]
22. Road traffic is a major emissions source contributing to PM₁₀ and PM_{2.5} concentrations close to roads. Abatement strategies to reduce PM concentrations (especially concerning organic carbon, elemental carbon and mineral dust) close to roads should therefore focus on road traffic. [Ch6]
23. Natural PM sources can contribute significantly to PM_{2.5}, though less than to PM₁₀. It is recommended to include derogations for exceedances of a possible future PM_{2.5} limit value due to natural sources. [Ch6]
24. It is recommended to develop PM source apportionment further for differentiation of sources such as diesel, gasoline and wood combustion to help target better abatement strategies. [Ch6]
25. Member States should report PM₁₀ measurements for previous years to the full extent that data are available, to increase the basis for assessing historic trends and its comparison with historic emissions trends. Historic time series must be quality assured, particularly as concerns the correction factor used for measurements performed without using the reference method. [Ch7]
26. The historic time series should be analysed in terms of the influence of interannual meteorological conditions and other parameters, in order to separate the effect of emissions changes. [Ch7]
27. With some exceptions, there is a lack of PM concentration projections at the national and city scale. This hinders the ability to judge attainability of current and proposed PM targets and development of targeted abatement strategies. Member States are recommended to undertake their own national and city scale projections where they have not already done so. [Ch7]
28. Uncertainties in current projections should be addressed, particularly the underestimation of current PM₁₀ concentrations by some models as well as the possible underestimation of the impacts of abatement measures and consequent possible overestimation of future concentrations. [Ch7]
29. The First Daughter Directive (1999/30/EC) requires Member States to develop, implement and report air quality management plans to ensure that the limit values will be met in 2005. It can be expected that such plans will be developed for local hot spots, and also for urban regions. The

experiences gained in developing such plans should be exchanged and assessed in the coming years. [Ch8]

30. PM pollution has a strong regional component, but also local contributions. This poses problems for air quality managers, since they have to handle different scales concurrently in developing abatement strategies. Therefore, robust European scenarios on emissions and ambient (background) levels of PM should be made available to support air quality management at a local level. [Ch8]
31. Some of the measures (such as more stringent emission limits for mobile sources) are most effectively taken at a European scale. The Commission is therefore requested to investigate further measures on a Community level to support Member States in reaching the PM limit values and targets. [Ch8]
32. Several Member States will have major problems complying with the Stage 1 24-hour limit value, despite implementing many measures to reduce PM concentrations. Member States should document their attainment difficulties when preparing *plans and programmes* according the First Daughter Directive. [Ch9]
33. CAFE and the Commission should synthesise and assess information about attainability provided by Member States in their *plans and programmes* – and the information in this Position Paper – to inform a framework for Europe-wide additional actions and measures to help Member States meet the limit values in the First Daughter Directive.
34. Member States should also consult with each other to find a solution for significant pollution originating in other Member States. However, since levels of (secondary) PM are high throughout large parts of (Central) Europe, this consultation should not be bilateral, but should include all Member States, Acceding countries and the European Commission. [Ch9]
35. During this consultation (preferably within the CAFE framework), the effectiveness of measures at different scales should be investigated; in particular, measures on the Community level on precursor emissions and primary PM from the traffic and other sectors should be assessed. [Ch9]
36. The Commission should consider changes to the First Daughter Directive to ensure that targets are (1) challenging and achievable and (2) that Member States focus abatement strategies on achieving maximum health benefits from reductions in PM concentrations. One possible option to consider is to extend the date for compliance for the more stringent Stage 1 24-hour limit value, whilst maintaining the current date for the Stage 1 annual average limit value. In revising the current targets, the Working group recommends to take account of the recommendations formulated in Chapter 11. [Ch9]
37. Even though the evidence on the relationship between exposure to PM and health effects has increased dramatically over the past few years, there are still large uncertainties and considerable gaps in knowledge. These gaps can only be reduced by targeted scientific research. Areas where such research is urgently needed include: exposure to different PM components; dosimetry; toxicity of different components; mechanisms of injury; susceptible groups, etc. The development of methods for measuring particle size and type is still necessary as well as the development of atmospheric models and particle emissions inventories able to support PM policies and of any new standard. The European Community and national institutions are requested to make appropriate funding available to facilitate the corresponding studies. [Ch10]

1. Introduction

1.1 Scope

This paper is intended as an update to a previous Position Paper¹⁰ published in 1997 by an earlier Member State-led Working Group. That Working Group itself recommended that, because of uncertainties about the sources, concentrations and effects of particulate matter (PM) in ambient air, and because of the volume of research underway, the situation should be looked at again before too long. Its recommendation was carried forward into Article 10 of Directive 1999/30/EC. This Directive, amongst other things, sets limit values for PM, to be met in 2005.

This Position Paper looks at new data available from Member States owing to implementation of the Directive, results from recent research and the latest advice from the World Health Organization (WHO). It reports on progress, identifies problems that Member States are encountering, looks at whether they can be dealt with, and if so how, asks whether it is possible to identify certain types of particle as the main cause of health effects and makes recommendations for future action.

It is not however possible to make final decisions on what to do about PM in isolation from decisions on other air pollutants. This is because many of the sources of PM are also important sources of other pollutants, and because some types of particle are actually formed by interactions between gaseous pollutants such as sulphur dioxide, nitrogen dioxide, ammonia, and organic compounds. A further complication is that PM is in part a long-range transboundary air pollutant like ozone and acid rain. Concentrations of PM cannot therefore be controlled solely by local or even national action. It should also be noted that the important link between PM and climate change is not addressed in this draft Position Paper, but should be taken into account in the European Commission's Clean Air For Europe (CAFE) programme.

The Position paper is intended to help taking decision on PM in the context of the CAFE process (see <http://www.europa.eu.int/comm/environment/air/cafe/index.htm> for information on the CAFE programme).

1.2 Why is this paper needed?

1.2.1 Regulatory reasons

Council Directive 1999/30/EC established limit values and, as appropriate, alert thresholds for concentrations of sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air intended to avoid, prevent or reduce harmful affects on human health and the environment as a whole. The Directive also established criteria for Member States to obtain adequate information on concentrations and ensure this information is made available to the public.

The Directive requires that Member States take measures necessary to ensure that concentrations of particulate matter (as PM₁₀) in ambient air do not exceed the limit values laid down in Annex III of the Directive for specified dates. The limit values were developed starting from advice provided in 1997 by the Member State-led Working Group and published in the first Position Paper.

Drawing on studies in Europe and the USA, and considering the WHO summary of concentration-response relationships the majority of the Working Group then recommended a 24-hour limit value of 50 µg/m³ as a level at which public health effects were likely to be small. The group recommended this limit value be adopted as a 98th percentile of daily values over a calendar year. The majority of

¹⁰ Ambient Air Pollution by Particulate Matter. Position Paper, 8 April 1997

the group further recommended an annual average of 20 µg/m³. It did not say when limit values might be met, since a separate study on economic impacts had not yet reported on feasibility.

The Working Group at that time recognised that natural dust could play an important role in some Member States. they were not able to take this into full account and recommended that whatever decisions were taken on standards, they should be reviewed within 5 years after entry into force.

Following a separate economic evaluation, the Commission put forward a proposal that finally resulted in the adoption of Council Directive 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air. In Annex III, this Directive sets 24-hour and annual limit values for PM₁₀ which were less ambitious than the recommendations of the Working Group (see Table 1.1).

Table 1.1 Limit values for particulate matter (PM₁₀) as given in Annex III of Council Directive 1999/30/EC

	Averaging period	Limit value	Date by which limit value is to be met
STAGE 1			
1. 24-hour limit value for the protection of human health	24 hours	50 µg/m ³ PM ₁₀ , not to be exceeded more than 35 times a calendar year	1 January 2005
2. Annual limit value for the protection of human health	Calendar year	40 µg/m ³ PM ₁₀	1 January 2005
STAGE 2 ⁽¹⁾			
1. 24-hour limit value for the protection of human health	24 hours	50 µg/m ³ PM ₁₀ , not to be exceeded more than 7 times a calendar year	1 January 2010
2. Annual limit value for the protection of human health	Calendar year	20 µg/m ³ PM ₁₀	1 January 2010

⁽¹⁾ Indicative limit values to be reviewed in the light of further information on health and environmental effects, technical feasibility and experience in the application of Stage 1 limit values in the Member States.

Article 10 of the Directive requires that “no later than 31 December 2003 the Commission shall submit to the European Parliament and the Council a report based on the experience acquired in the application of this Directive and, in particular, on the results of the most recent scientific research concerning the effects on human health (...) of exposure to (...) different fractions of particulate matter (...) and on technological developments including progress achieved in methods of measuring and otherwise assessing concentrations of particulate matter in ambient air”. Article 10 also states that “the Commission will give particular attention to setting limit values for PM_{2.5} or different fractions of particulate matter, as appropriate”.

The present Working Group was set up to advise the Commission on advances in knowledge of PM since Directive 1999/30/EC came into force and what has been learned through Member States’ further experience in order to enable the Commission to decide whether or not to make further proposals.

The Terms of Reference of the Working Group are:

With the aim of supporting the European Commission’s review of the First Daughter Directive the group should:

- Assess the air quality situation with regard to the PM limit values set in the First Daughter Directive;
- Review the content of the Position Paper on PM published in 1997 with regard to information obtained since;
- Collect together information on predictive studies on the attainability of the limit values, considering at the same time contributions from long-range transport and local sources.

With the aim of supporting the production of the CAFE thematic strategy the group should:

- Consider the WHO work on health effects of PM with the aim of giving recommendations for targets for integrated assessment;
- Review the results of the integrated assessment modelling work on PM.

The Working Group on Particulate Matter comprised experts from EU Member States, Switzerland, Industry, NGOs, the World Health Organization (WHO) and the European Environment Agency.

1.2.2 Public Health reasons

In recent years scientific evidence has been strengthened by many epidemiological studies that there is an association between long and short-term exposure to ambient particulate matter and various serious health impacts. This relationship has been strengthened in recent years by a large body of new scientific evidence.

Current WHO air quality guidelines describe the relationship between ambient PM and various health end points. The available information indicates that there is no threshold below which effects would not be expected at the population level.

The Working Group was asked to make recommendations for standards despite the absence of thresholds.

In a report "Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide" prepared by WHO in support of the CAFE process (see Chapter 10), WHO states that in view of the large body of new scientific evidence that has emerged, "reconsideration of the current WHO Air quality guidelines and the underlying exposure-response relationships" is justified.

At the time of publishing this draft Second Position Paper, the WHO is in the process of revising the concentration-response relationships. The Working Group will review and if necessary revise its recommendations when the updated WHO guidelines are available.

1.3 What does this paper cover?

This Position Paper gives some general information on particle characteristics, and the problems related to measuring and modelling PM concentrations. It further provides information on current concentrations of different PM size fractions, namely PM₁₀, PM_{2.5}, PM_{1.0}, and of ultrafine particles (UFP, expressed as particle number concentrations). It addresses emissions and sources of PM, and discusses source apportionment and historic trends in emissions and concentrations. Also presented are the results of projections concerning PM concentrations for 2010 and various abatement options. The subsequent chapters are dealing with the attainability of set PM concentration levels, and present the currently available information on health effects as obtained from WHO. In the final chapter recommendations are made concerning PM metrics and PM targets for the future.

The Working Group itself did not look in detail at the health effects of PM, as the Commission had asked WHO to conduct a review of the most recent evidence on such health effects. WHO's review of the health effects of PM is included in Chapter 10 and the Working Group has taken it into account in reaching its conclusions.

PM and climate change

PM (otherwise known as aerosols) exerts radiative forcing and therefore an impact on climate change. There is significant uncertainty associated with the estimates of radiative forcing for aerosol species. Sulphate nitrate and organic carbon aerosols are estimated to have negative global annual mean radiative forcing, i.e. they could exert a cooling effect. On the other hand black carbon aerosols ('Black Smoke') are estimated to have a positive global annual mean radiative forcing, thereby contributing to global warming. It is not within the scope of this position paper to explore the interactions between PM and climate change. There are however important linkages between PM and climate change, for example concerning sources, emissions, abatement measures and atmospheric chemistry, that should be considered when developing air quality and climate change policies.

2 Characterisation of Particulate Matter

2.1 Scope

This chapter summarizes the currently available knowledge with regard to some essential features of particles in order to permit a better understanding of the following chapters. In particular, this chapter provides information on the size of particles¹¹, the residence time of particles in the atmosphere (“lifetime”), and the chemistry of particles. It also addresses the particular problem of long-range transport of particles, a phenomenon that has an important impact on ambient levels of particulate matter and pays particular attention to “African dust outbreaks”.

2.2 General

Atmospheric particles are constituted by solid and/or liquid particles that enter in the atmosphere from natural or anthropogenic sources. The term atmospheric aerosol is frequently used as a synonym of atmospheric particles.

Atmospheric particles are emitted by a wide variety of natural and anthropogenic sources. The nature of the source influences both the physical properties (e.g. mass, size, specific surface, density or number-density) and the chemical composition of the particles. Particles may be classified as primary or secondary depending to its formation mechanism. Primary particles are directly emitted into the atmosphere. Secondary particles are those formed in the air, generally by chemical reactions of gaseous precursors.

2.3 Particle size distribution

Particles associated with a specific source tend to have a specific size distribution and chemical composition. Their size may vary in the range from a few nanometers (nm) to several tens of micrometers (μm). Depending on the physical and chemical processes involved in the particle formation and growth, the particles are conventionally divided into different size fractions (generally called “mode”), the denomination of which mostly refers to how the particles are formed: nucleation, Aitken, accumulation, and coarse modes (Figure 2.1). It is important to know that instruments used to measure any integral parameter, e.g. the mass, of particles in the air do not collect particles of one single size or specific chemical composition, but rather particles that fall within a size range. Generally, the size distribution and chemical composition of PM emitted from a specific source are characteristic for that source.

The nucleation mode expands in the range less than 0.02 μm and usually presents the maximum number-density around 5-15 nm particle diameter. The only precursor gases shown to actually form new particles by homogenous nucleation in the ambient atmosphere are H₂SO₄, NH₃ and H₂O. However, the newly formed particles grow by condensation mostly by other condensing gases, organic and inorganic components. These particles have a lifetime of hours in the atmosphere as they rapidly coagulate with larger particles or grow into larger sizes by condensation. In background areas, growth in particle diameter of nucleation mode particles have been observed in the order of a less than one to several nm/hour (Kulmala et al., 2001). Classical nucleation theory shows that the nucleation highly depends on the concentrations of the gaseous precursors, relative humidity and temperature. In particular, the nucleation is favoured by decreases in the temperature and/or increases in the relative humidity (Eastern and Peter, 1994).

¹¹ Particles size is normally given in micrometers, μm (1 μm = 1/1000 mm). For convenience the size of very small particles (“ultrafine” particles) is given in nanometers, nm (1 nm = 1/1000 μm).

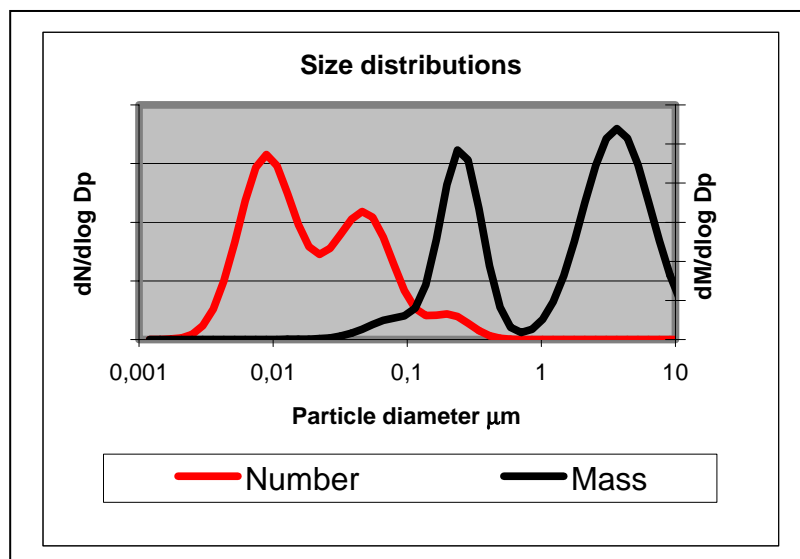


Figure 2.1 Particle number and mass distributions, during nucleation, are shown in a lin-log diagram. The nucleation and Aitken modes are clearly seen in the number distribution, while the accumulation and coarse modes show more in the mass distribution.

Aitken mode particles range from 0.02 to 0.1 μm and originate from either primary particles, natural and anthropogenic, or by growth of nucleation mode particles. As they still comprise a negligible amount of the total fine mode mass, little is known about their chemical composition. However, it is likely that secondary Aitken mode particles are formed by coagulation of nucleation mode particles, by condensation and by liquid phase reactions, as inorganic compounds are strongly water active and thus the particles are liquid even at low relative humidity. The actual influence of anthropogenic emissions on the number of Aitken mode particles is not clear at the moment. Soot can be seen as a major tracer of combustion emissions. Measurements of fresh emissions from different combustion sources, on fossil as well as bio fuel, show particles in sizes up to 150-200 nm with a major fraction below 100 nm. Soot is supposed to preferably be in particles larger than 30 nm (Matter et al., 1999). Considering the very large emissions of Aitken mode particles from combustion it is likely that the number of Aitken mode particles are affected at least on a regional scale.

Published measurement results show that Aitken mode particles are present at relatively stable concentrations, unless there is local disturbance, indicating a rather long residence time in the atmosphere. The number ranges from a few hundreds in remote locations over a few thousands in urban background to several tens of thousands per cubic metre in street level in city centre (Tunved et al., 2002 and Figure 2.2).

The expression ultrafine particles is used mostly in health related literature as notion for particles less than 0.1 μm , thus covering nucleation mode and most of Aitken mode particles. Sometimes the term ultrafine particles is also used for nucleation mode sized particles.

The accumulation mode covers the range between 0.1 and up to 1 μm . In the atmosphere Aitken mode particles grow to accumulation mode particles primarily by liquid phase reactions occurring in cloud droplets. The mass possible to transfer by condensation and/or nucleation/coagulation is not enough to cause any significant change in particles size compared with the observed growth (Hoppel et al, 1986, Bowers and Choularton, 1993). Due to the large water volume and rapid reaction rate in cloud droplets the mass transfer is strongly enhanced in spite of the short time a particle spends as a cloud droplet during its lifetime. About 90% of sulphate formed in the atmosphere is formed from liquid phase oxidation of sulphur dioxide occurring in clouds (Langner and Rodhe, 1992).

The coarse mode includes particles $>1\mu\text{m}$. However, by current convention particles between 2.5 and $10\mu\text{m}$ are called “coarse particles” whereas particles $<2.5\mu\text{m}$ are called “fine particles”. Most particles in the coarse mode range are formed by mechanical processes, such as the erosion of the surface of the Earth (mineral dust) or other materials, or the bursting bubbles on the ocean surface (sea spray). Sea spray is traditionally considered to add only to the coarse mode, but recently Mårtensson et al (2002) have shown that particles down to 20 nm are generated in higher number (about one order of magnitude) than coarse particles. However, the mass is totally dominated by coarse particles. The processes of formation and growing of particles in the range $<1\mu\text{m}$ does not tend to produce particles $>1\mu\text{m}$. On the other hand, mechanical processes forming primary particles cannot easily reduce the size of particles to diameters $<1\mu\text{m}$ owing to energetic limitations. Thus, the transfer of particles from different modes of the size spectrum presents a barrier around $1\mu\text{m}$.

The coarse mode is characterised by a low number concentration and a relatively high mass concentration. Although in the coarse mode the primary particles predominate, “secondary particles” may also be found. Such secondary particles are formed by chemical interaction of gases with primary particles ($>1\mu\text{m}$) of crustal or marine origin. For example, the reaction of the gaseous nitric acid with calcium carbonate or sea salt results in the formation of calcium nitrate or sodium nitrate (Harrison and Pio, 1983; Mamane and Mehler, 1987; Wall et al., 1988), respectively.

In heavy traffic situations particles in the nucleation mode range are usually observed. However, it is not clear at present whether these particles are primarily part of the engine exhaust or formed by nucleation processes when the exhaust gases are mixed with cold ambient air.

Nucleation mode particles occur with a very high variability in remote to urban background conditions. It can reach comparably high concentrations only in few hours under certain specific conditions. Finnish, German and Swedish background stations have registered about 50 nucleation episodes per year (Mäkelä et al. 1997, Birmilli and Wiedensohler, 2000, Tunved et al., 2003). Figure 2.2 gives an example of the difference of particle size distributions at two different types of site. The data refer to particle number, surface area, and mass concentration and were obtained during a 2-week measurement period in Stockholm in March 1999 at (a) one city centre kerbside site in a street canyon with roughly 40,000 vehicles passing per day and (b) one suburban background site in a residential area about 15 km south of the city centre.

Particles to be referred to as fairly fast growing nucleation mode particles or small primary Aitken mode particles dominate the total number of particles at the inner city site with almost $100\,000\text{ particles/cm}^3$. While particles around $0.1\mu\text{m}$ can be regarded as typical for primary diesel emissions, those below $0.1\mu\text{m}$ are most probably secondary particles, even though formed in the engine exhaust system. The surface area distribution shows on what particles gases condense and that collide with other finer particles (coagulation). These particles accumulate most mass and with the highest rate of condensing gases. This is especially interesting in the light of the relationship found between surface area and health. The surface area in traffic intense areas as shown in Figure 2.2 indicates particles around $0.1\mu\text{m}$ being the major sink for coagulating particles and condensing vapours. However, considerable surface area is also found in the coarse mode. Most striking is the high peak in the coarse mode mass distribution, which is due to high emissions of non-exhaust coarse particle, e.g. resuspension of road dust. In the fine particle size range, two smaller peaks are noticeable in the mass distribution. The one around $0.1\text{--}0.2\mu\text{m}$ is most likely originating from primary vehicle exhaust emissions, while the other one around $0.2\text{--}0.3\mu\text{m}$ is likely to originate from aged long-range transported particles.

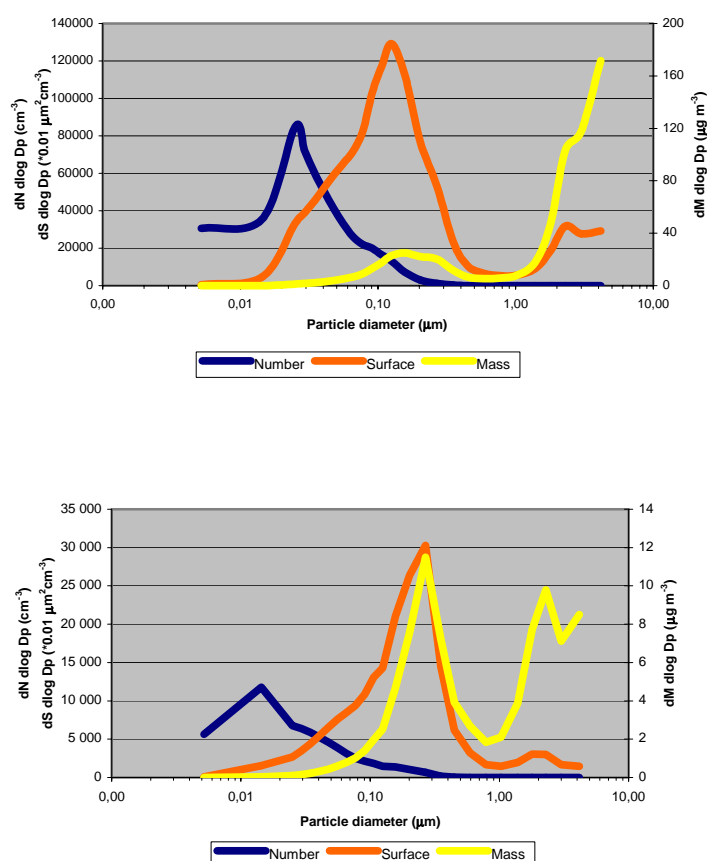


Figure 2.2 Particle number, surface and mass size distributions measured at a kerbside (top) and an urban background site (bottom) in Stockholm.

The picture changes for the urban background site (bottom of Figure 2.2). The nucleation mode particles are smaller due to lower concentrations and thus less coagulation and condensation giving lower growth rates. The maximum number is considerable lower than in the city centre although still totally dominated by the finest particles. Surface area distribution is shifted towards larger particles around 0.3 μm , i.e. a range dominated by the long-range transported particles. The mass is more equally distributed between fine and coarse particles. The fine mass is totally dominated by aged long-range transported particles, while the coarse mass is still mostly locally generated. Even though not close to major roads, $\text{PM}_{2.5}$ is still influenced by coarse mode particles from non-exhaust car emissions (pavement erosion, break and tyre abrasion) showing that dust is transported over at least several tens of kilometres.

The particle surface area has been found to be related to health effects (Maynard and Maynard, 2002; Moshhammer and Neuberger, 2003). As pointed out surface area is mostly dominated by particles in the size range 0.1-0.5 μm . This size range is mostly dominated by aged secondary-influenced aerosol, although primary particles can play the major role close to strong combustion sources.

These findings coincide with those of a more general study by Putaud et al. (2002) covering 33 different research sites in Europe. The authors conclude similarly that particle number concentrations (with aerodynamic diameter $D_p > 10 \text{ nm}$) increase more than particle mass in polluted areas. Number size distribution measurements explain this by a higher contribution of small ($D_p < 100 \text{ nm}$) particles when moving from clean to polluted sites. Particle size distribution measurements show that most (70-80%) particles have a diameter $D_p < 100 \text{ nm}$. At polluted sites those particles consist mainly of carbonaceous material (black carbon and organic matter). The large variation in number is attributed to local sources, mainly traffic in cities and natural nucleation events at background sites.

The relation between coarse and fine particle mass depends on the source strength of the fine and coarse particle sources that has been anticipated to be independent. However, Putaud et al. (2002) find that clear relationships do exist at individual sites, even though there are no “universal” (i.e. valid for all sites) relationships between $PM_{2.5}$ and PM_{10} mass concentrations. No clear relationship is found neither between $PM_{2.5}$ or PM_{10} on the one hand, and total particle number concentration or chemical composition on the other. Areskoug et al. (2003) found that vehicles emit 10 times more particulate mass of non-exhaust related particles (mainly coarse particles) than fine exhaust particles. Harrison et al. (2001) supports also that traffic is a strong coarse particle source, but in this case the emissions factors for non-exhaust PM_{10} reached values similar to those for exhaust emissions. These studies show that the traffic is both emitting fine and coarse particles in relation to number of vehicles, which cause a strong correlation between $PM_{2.5}$ and PM_{10} .

Secondary formation of components on particles takes time and is thus connected with long-range transport. High concentrations of PM_{10} are found downwind the major source areas, e.g. the Nordic countries, to depend on secondary particles. Putaud et al. (2002) found the ratio $PM_{2.5}/PM_{10}$ to increase with the PM_{10} concentration, except at kerbsides, where the opposite is observed, showing that even within a major source area as Europe secondary particle production and long range transport is important.

Sedimentation affects coarse particles, PM_{10} - $PM_{2.5}$ and larger, than the fine $PM_{2.5}$; consequently, mineral dust and marine aerosols (both with a prevalent coarse size) are preferentially deposited with respect to other fine PM components. Close to the emission source, both mineral dust and seas salt are dominated by $>10\ \mu m$ size particles, but during atmospheric transport the deposition of these very coarse particles shifts the coarse mode towards a size in the range 2.5 to $10\ \mu m$. On a regional scale, with long-range transport, deposition shifts the coarse mode towards even smaller sizes. It has been shown that special events such as African dust transport may induce high levels of $PM_{2.5}$ natural dust due to the special shape (clay flaks) and size of the particles.

2.4 Atmospheric lifetime of particles

Once the particles have been emitted into or formed in the atmosphere, they are subject to removal from the atmosphere by different processes, which together determine the atmospheric lifetime of the particles. Coarse particles are removed from the atmosphere by sedimentation and precipitation (the residence time for particles $>20\ \mu m$ is several hours while it is 2-4 days for 2-3 μm particles). Particles in the range 0.1-1 μm (accumulation mode and larger Aitken mode particles) exhibit the longest lifetime in the atmosphere, ranging from days to a few weeks. Primary particles from combustion and secondary aerosols are usually found in this size range. Consequently, these particles have a long residence time and as a result can be transported over long distances (\sim several 1,000 km).

The lifetime of a particle in the air determines to what extent it changes its properties, and its properties determine its lifetime. For example, a very small particle emitted from an automobile exhaust pipe will grow in size as it travels from the pipe toward the kerbside and further on. Since the exhaust particles are quite small and hydrophobic it will mainly be subject to dry deposition by diffusion, which is small. As the particles grow and accumulate inorganic components that usually are quite hygroscopic, they become subject to deposition by precipitation, which is the dominant deposition path for fine particles. The extent of such growth does not only depend on the distance travelled by the particle, but also on the atmospheric conditions. In addition, a particle's chemical composition changes with growing distance from the source depending on the nature of other pollutants present in the air. Natural particles, such as inorganic secondary species from volcanic emissions or African dust, may be transported at super-long-range scale ($>5000\ km$) due to: a) the large emission volumes, b) the release of PM into higher zones of the atmosphere where transport is favoured, and c) to the fine particle size caused by the photochemical origin (sulphate from volcanic SO_2 emissions) or by the size segregation caused by long-range transport (desert dust).

Particles in the range 0.1-1 μm often are efficient cloud condensation nuclei, i.e. easily forming cloud droplets, due to a sufficient water-soluble fraction. Thus, the time of residence of particles in the range 0.1-1 μm is considered similar to the time of residence of the water (around 10 days; Pósfai and Molnár, 2000). However, carbonaceous particles are usually difficult to wet and consequently, a period for surface oxidation is required before they can be wetted and removed from the atmosphere by wet deposition. This results in a longer lifetime of carbonaceous soot and organic particles (Gaffney et al., 2002) when compared to inorganic particles, e.g. sulphate.

On the other hand, the lifetime of several species depends also on their thermodynamic properties. Thus, the distribution of mass between the gas and aerosol phases in some chemical species experiences wide variations induced by the variations in the temperature and relative humidity typical of ambient air. This is especially important for species such as ammonium nitrate and some organic aerosols (Adams et al., 1999). Semi-volatile compounds tend to distribute their mass between the gas and aerosol phases to reach a thermodynamic equilibrium and this may be subject to important seasonal changes. Thus, in summertime a large proportion of particle-associated ammonium nitrate is not stable. Consequently, PM_{10} and $\text{PM}_{2.5}$ associated nitrate concentrations are usually characterized by low levels in summer and high levels in winter.

2.5 Chemistry of ambient particulate matter

As previously stated, atmospheric particles are a multi-component system constituted by a large number of natural and anthropogenic species. The major PM components are sulphate, nitrate, ammonium, sodium, chloride, carbon (organic and elemental), mineral dust and water. The predominance of these chemical components in PM_{10} and $\text{PM}_{2.5}$ and their size distribution are closely linked to the emitting source and the formation mechanisms of the particles.

2.5.1 Natural particles

The influence of the natural particle emissions on PM_{10} concentrations is especially important in Southern Europe. Specific climate and geographic patterns of Southern Europe may cause relatively high PM_{10} background levels. Thus at the 10 EMEP stations in Spain, annual PM_{10} levels ranged from 12-22 $\mu\text{g}/\text{m}^3$ in 2001. These high background levels are due to a high mineral dust input from frequent African dust events and local soil resuspension in dry areas. Mineral dust input contributes around 5 $\mu\text{g}/\text{m}^3$ to the annual PM_{10} average in regional background sites in Spain.

In the EU the natural mineral dust contributions to ambient PM_{10} may be caused by local resuspension of arid soils, or by long-range transport from distant large arid regions (Rodriguez et al., 2001, Viana et al., 2002). Although most of crustal PM occurs in the size range $>2.5 \mu\text{m}$, a considerable proportion of $\text{PM}_{2.5}$ is still present in ambient air during periods of atmospheric transport of African dust to Europe.

In specific locations at coastal areas of the EU, the marine contribution may also account for an important fraction of PM_{10} levels. Thus, at the Canary Islands and in specific coastal locations of the Netherlands the marine aerosol contribution accounts for 11.5 (annual averages of 2001 and 2002) and up to 7 $\mu\text{g}/\text{m}^3$ of the annual PM_{10} levels, respectively. Most of the mass of marine aerosols is in the fraction $>2.5 \mu\text{m}$.

Volcanic emissions are a source of primary mineral particles, but perhaps mainly secondary sulphur compounds (from SO_2 emissions). Although the contribution of such events to bulk particle mass levels in ambient air is generally limited, the particles can be ejected into very high altitudes, even the stratosphere (e.g. during the last Pinatubo eruption), and then stay suspended for a year and more, which can bring about a considerable change of the radiation balance.

The chemical composition and mineralogy of crustal particles may be subject to regional variation as a function of the geology of the source areas, but it is in general made up of various silicates, carbonates and minor amounts of some other constituents. In wind tunnel experiments (Alfaro et al., 1998) the suspended dust tended to show a size distribution characterised by three modes centred at 1.5, 7 and 14 μm .

The primary particle fraction of sea spray is mainly constituted by sodium chloride (NaCl) and some sulphates. The sea spray is mainly generated by bubble processes on the ocean surface and by the breaking of waves in the coastal areas. These processes are favoured by the surf processes activated by wind and produce sea spray aerosols mainly in the coarse size range (Warneck, 1987).

Biogenic sources of primary particles give rise to the fraction known as bio-aerosol or biological residues. This fraction is mainly constituted by vegetal debris, pollen, spores, and minor amounts of micro-organisms (e.g. viruses, bacteria, fungi, protozoae or algae). Particulate viruses and bacteria exhibit sizes in the range $<2 \mu\text{m}$, whereas plant debris and spores have sizes extending into the coarse range of PM (Pósfai and Molnár, 2000).

The natural secondary fraction of atmospheric particles is mainly made up of sulphate, nitrate and organic aerosol. Natural sulphate is formed from the oxidation of sulphur containing gases, such as SO_2 emitted by volcanoes, or dimethylsulphide of marine biogenic origin. Natural nitrate is the final product formed by oxidation of nitrogen oxides emitted by soil transpiration (Roelle et al., 2001) and lightning (Price et al., 1997a and b). Main sources of natural secondary organic aerosol precursors are found in the large forested areas (Harrison et al., 2001), from where important amounts of organic vapours, such as terpenes (Yu et al., 1999), are emitted. The oxidation of these organic vapours gives rise to low vapour pressure phases that support the formation of new particles (Christoffersen et al., 1998).

2.5.2 Anthropogenic particles

In urban environments, primary particles are emitted as a result of traffic-related combustion processes, which give rise to particles constituted of elemental carbon and various inorganic and organic compounds. Furthermore, primary particles result from erosion of the pavement by road traffic (road dust) and abrasion of brakes and tires. Particles mechanically generated by road traffic are coarse particles, whereas primary particles emitted by the vehicles exhaust are fine particles.

Particles in the sub-micrometer range are emitted by both diesel and spark ignition (gasoline) engines. The particles generated by diesels are generally larger in number and size than those from spark ignition engines, which results in a higher emission of mass per vehicle km. The difference in emissions between diesel and gasoline motor is largest at low velocities. The newly introduced particle traps for diesel engines have shown efficiencies of up to about 99%. This seems promising for achieving a lower mass emission of diesel engines (Matter et al., 1999).

Industrial activities such as building, mining, manufacturing of cement, ceramic and bricks, and smelters are typical sources of primary particles. In addition, combustion of coal should be mentioned. Although coal combustion has historically been an important source of primary particles (such as fly ash), the current use of emission abatement technologies and the progressive substitution of coal by other combustibles have reduced the importance of coal burning as a source of primary particles in most industrialised countries. Primary particles associated with smelter emissions are mostly fine particles, because most of these are formed by condensation of hot vapours. The chemical composition depends on the smelter type, but Ni, V, Mn and Cu are often emitted (Pacyna, 1998). The other industrial sources mostly emit coarse primary mineral particles. An important fraction of these emissions occurs as fugitive emissions due to the handling of dusty materials.

After emission of SO₂, oxidation gives rise to the formation of sulphuric acid which is to some extent neutralised by ammonia, and by reaction with calcium carbonate or sodium chloride to form the corresponding salts. Sulphuric acid and ammonium sulphate are found on fine particles (<1 µm), whereas the fractions neutralised by calcium carbonate and sodium chloride are more present on particles in the coarse mode (> 1µm) (Mildford and Davidson, 1987).

Nitrogen oxides are mainly emitted by traffic in urban environments, and by some industrial processes. By oxidation of NO_x nitric acid (HNO₃ gas) is formed, which may be neutralised and transformed into NH₄⁺ or Na⁺ nitrate (particle). The size distribution of nitrate depends on the neutralising agent. Thus, NH₄NO₃ is present in the fine mode (<1 µm), whereas Ca₂NO₃ (from reaction between HNO₃ and crustal CaCO₃) and NaNO₃ (from neutralisation by marine NaCl) are larger in size (Harrison and Kito, 1990; Wakamatsu et al., 1996). The ratio nitrate/PM mass is usually highest at rural sites and higher in urban background than at kerbsides (Putaud et al. 2002), because nitrate formation is slow.

Large agricultural areas and combustion of bio-mass and fossil combustibles are important sources of organic vapours, which are precursors of anthropogenic secondary organic aerosols. Aromatic and non-methane hydrocarbons stemming from gasoline usage are important precursors of organic aerosols (Odum et al., 1996, 1997a, 1997b). These vapours are mainly emitted by gasoline evaporation (fugitive emission) and in the combustion process (Watson et al., 2001). The organic aerosol is formed after the oxidation of the gaseous precursor, mostly likely by condensation to pre-existing aerosol resulting in organic matter on particles of sizes in the range <1 µm to 10 µm with a maximum in the fine mode particles.

Human activities may also give rise to anthropogenic bio-aerosols. Bacteria and fungi generated in solid waste recycling and composting plants represent a potential risk for human health (Marchand et al., 1995).

Brasseur et al. (1999) estimated for the global scale that the anthropogenic PM_{2.5} emissions are about 30-100% of the natural PM emissions. The anthropogenic coarse particle emissions (PM_{2.5}-PM_{1.0}) is only slightly larger than the PM_{1.0}, thus the anthropogenic PM_{1.0} emissions adds mostly to the PM_{1.0} budget.

Putaud et al. (2002) and APEG (1999) pointed out that in the EU sulphate and organic matter are the main contributors, about 20-30% each, to the annual average PM_{2.5} and PM₁₀ mass concentration, except at kerbsides where mineral dust is also contributing in high proportions to PM₁₀. Away from the kerbside sites in polluted conditions (PM₁₀ > 50 µg/m³), sulphate, nitrate and organic matter are the main contributors to PM₁₀ and PM_{2.5} which illustrates the importance of secondary formation and long-range transport. Black carbon contributes 5-10% to PM_{2.5} and somewhat less to PM₁₀ at all sites, including the clean background sites. Its contribution increases to 15-20% at some of the kerbside sites. Observations of PM_{2.5}/PM₁₀ ratios, particle size distributions and chemical composition indicate that traffic is responsible for the high PM₁₀ and PM_{2.5} concentrations, either through the contribution of nitrate and carbonaceous aerosols (at near-city and urban background sites) or due to the contribution of carbonaceous aerosols and re-suspended dust to the coarse fraction (at kerbsides). At polluted sites PM₁₀, PM_{2.5} levels and other aerosol parameters can be significantly higher during winter, although this is not always observed, e.g. the UK stations show very little seasonal variation. Chemical analysis explains this from condensation of semi-volatile species like nitrate and (unspecified) organics in the particle phase, a process that is favoured by cold temperatures. When selecting only polluted conditions (PM₁₀ > 50 µg/m³), organic matter and nitrate become the main contributors to PM₁₀ and PM_{2.5}.

2.6 African dust contributions

One of the most characteristic cause of natural particles affecting Southern Europe, and in particular the Mediterranean basin, is the dust transported from North Africa (Sahara and Sahel deserts). This source is far less significant in the Northern latitudes owing to the distance and meteorology of these areas.

At the Canary Islands 5 to 15 episodes per year with a mean duration from 2 to 22 days per event are usually registered each year. During these events 24-hour PM_{10} levels may reach extremely high values (24-hour mean values up to $1000 \mu g/m^3$ were recorded in 2001). Also in the Southern Iberia Peninsula the frequency of African dust outbreaks is relatively high (16 to 19 episodes per year with a mean duration of 1 to 10 days). Daily PM_{10} levels recorded for these events reach <150 and $<75 \mu g/m^3$ in winter and summer, respectively (Querol et al., 2003). African dust outbreaks over Northern Spain are less frequent compared with Southern areas. There, on average, 6 episodes per year are recorded with a mean duration of less than four days each and 24-hour PM_{10} levels below $100 \mu g/m^3$.

North African dust is injected into the atmosphere through resuspension processes in the source areas, and it is then transported at different altitudes (from sea level up to 4.5 km) over the Mediterranean and Southern Europe (Löye-Pilot et al., 1986; Bergametti et al., 1989; Dayan et al., 1991; Dulac et al., 1992; Chester et al., 1993; Molinaroli et al., 1993; Guerzoni et al., 1997; Moulin, 1997; Avila et al., 1997, 1998, 1999; Querol et al., 1998; Hamonou et al., 1999; Rodriguez et al., 2001), North Europe (Ryall et al., 2002), Canary Islands (Conde-Gaussen et al., 1989; Bergametti et al., 1989; Prospero et al., 1995), Cape Verde (Chiapello et al., 1995; Caquineau et al., 1998), Barbados (Prospero and Ness, 1986; Savoie et al., 1992; Arimoto et al., 1997), North America (Prospero et al., 1987; Perry et al., 1997; Prospero, 1999) or South America (Prospero et al., 1981; Swap et al., 1992). The dry climate and the scarcity of precipitation in the Mediterranean basin favour a long residence time of PM in the atmosphere with subsequent impact on the air quality.

The $PM_{2.5}/PM_{10}$ ratio measured in African dust outbreaks over Eastern Spain ranges from 0.4 to 0.8, but this rises to 0.7-0.9 in Northern Spain. Levels of Al and Fe are usually used as tracers for the African dust, deriving experimental factors to quantify mineral dust (e.g. [mineral dust] = $Al \times 12.2$ (Prospero et al., 1987) or mineral dust = $SiO_2 \times 3.03$ (Bowen, 1966)).

Although African dust outbreaks over Europe occur throughout the year, the frequency is lower in November and December. Intensive events are usually recorded in February and March, when Sahel dust is transported towards Europe over the Atlantic Ocean. The summer is another period with high probability of the occurrence of dust events, but in this case Sahara is the source land of the dust, which is transported over the Mediterranean basin.

2.7 Conclusions and recommendations

2.7.1 Conclusions

Particulate matter (PM), particles suspended in the atmosphere, is a complex entity and a product of several particle and gas sources and subsequent atmospheric processes. The processes are ongoing all the time, resulting in constantly evolving particles changing in size and chemistry. This gives rise to a number of questions, such as what are the major sources, how the concentrations should be measured and what are the epidemiologically relevant parameters, which can only be answered with a sufficient understanding of the characteristics of particulate matter.

The character of a particle depends on a number of parameters among which size and chemical composition (including chemical reactivity) play the most important role. It is important to note that, as there is no "gold standard" available, many decisions made in relation to particles are by convention.

The major difference with respect to other pollutants is that atmospheric particles are a mixture of pollutants distributed on particles ranging several orders in size, with different chemical composition, whereas most of gaseous 'classic pollutants' are monitored as single species. This mixture of pollutants, in many cases with very different origins, induces special features to atmospheric particles.

Examples of these features are:

- a) PM components are emitted by very different sources, which requires abatement strategies to be based on information on PM speciation.
- b) Different PM fractions may have large differences in atmospheric lifetime as a result of the different physical and chemical properties. Components of hydrophobic particles, in the size range of 0.1-1 μm , have the longest lifetime in the atmosphere ranging from days to weeks.
- c) Some natural and anthropogenic components are very stable under normal atmospheric conditions and may be transported over very long distances (>5000 km) from the emission sources (examples are African dust outbreaks, soot and persistent organic pollutants).
- d) Some of the components have relatively high elevated natural background levels, with strong geographical variations, that may decisively limit the relative impact of the control of the anthropogenic input.
- e) The impact on health may be selectively attributed to specific PM species (with particular chemical and physical characteristics) that may represent a relatively low proportion of the bulk PM mass and consequently the cause-effect relationship may be difficult to demonstrate if speciation of PM is not considered.
- f) The sampling and measuring conditions that may be applicable for a group of PM components may cause underestimation or overestimation of levels of other group of PM species, causing monitoring problems.
- g) For most gases, measurements can be traced back to primary standards based on SI units (volume, mass), but PM measurements for certain metrics are only traceable to conventional reference methods.

In Table 2.1 the particle size fractions of ambient particle mass affected by the different emissions sources are summarised. The indication is set to 100 when the PM mass resulting from the particulate emissions is fully within the range set by the definition of PM_x , i.e. the mass of particles with size less or equal to X. Less than 100 is a coarse indication of the relative fraction of the emissions in the PM fraction given. The classification is relative for every emission source but is not intended for comparison between the different sources.

Table 2.1 Order of magnitude indication of typical source contributions to PM mass fractions

Source	PM_{10}	$\text{PM}_{2.5}$	$\text{PM}_{1.0}$	$\text{PM}_{0.1}$
Traffic-exhaust	100	100	100	50
Road dust	100	10	1	0
Winter sanding	100	10	1	0
Combustion	100	100	100	50
Industry-primary PM	100	50	5	1
Industry-secondary PM	100	100	100	50
Fugitive emissions	100	10	1	0
Quarries	100	10	2	1
Agriculture ¹⁾	100	10	2	1
African dust	100	20	5	0
Marine aerosol	100	20	2	0
Biogenic sources ¹⁾	100	10	2	1

¹⁾ Including primary and secondary emissions

2.7.2 Recommendations

There is a need to fill in the considerable gaps that remain in scientific knowledge and evidence on PM characteristics, to help inform policy makers about future strategies for reducing the health impacts of PM. Examples are the few data available for urban areas on PM_{2.5} and PM_{1.0} concentrations, particle composition and number concentration of particles.

2.8 References

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3. Air quality assessment methods

3.1 Monitoring

3.1.1 Scope of Section 3.1

This chapter focuses on PM monitoring in Europe. It discusses the legislative requirements, the available methods and the methods that are routinely used by Member States. It describes the difficulties in the quantification of airborne particulate matter, the comparability of the various methods used in different Member States and the procedures in Member States and at the EU level to restore data consistency, in particular the correction factors or equations applied to automated non-reference measurements. It also discusses the consequences of a change from the PM₁₀ fraction to another metric.

3.1.2 General

The quantification of PM depends on the physical or chemical parameter of interest and the measuring method used. The wide range of different PM parameters has been discussed in Chapter 2. Mass concentration of specific size fractions such as PM₁₀ and PM_{2.5} is the most common way to quantify ambient air PM pollution levels; current EU legislation refers to this type of parameter.

Unlike for other air pollutants (e.g. most of the gaseous pollutants), there is no metrological reference available as a base for PM mass concentration measurements. Therefore, for PM₁₀-monitoring a reference method was developed by CEN (Comité Européen de Normalisation). A standard for PM_{2.5} measurements is currently being developed. Other methods used in networks to determine PM₁₀ levels must be compared to the reference method to show 'equivalent' results. Because intercomparison exercises of different PM₁₀ monitoring methods showed large deviations, it was recognised that results from non-reference methods have to be corrected and enhanced efforts to harmonise the PM measurements in the EU are needed.

Monitoring of "black smoke" as a surrogate for PM has been widely used for decades by some Member States. Especially with changes in emissions, the comparability of results has become questionable.

3.1.3 Reference methods

For PM₁₀, the European Reference Method is described in CEN standard EN 12341, adopted by CEN in November 1998 (CEN, 1998). It defines a PM₁₀ sampling inlet coupled with a filter substrate and a regulated flow device. The mass collected on the filter is determined gravimetrically by means of a micro-balance under well defined environmental conditions. This is the reference method under the First Daughter Directive; it gives, by definition, the "correct" PM₁₀ results. Further details are provided in Annex 2.

For the measurement of the PM_{2.5} fraction no European Reference Method has been established up to now. Such a standard is currently being developed by CEN (CEN TC 264/Working Group 15) under a mandate of the European Commission. As for PM₁₀, the method is based on the gravimetric determination of the PM_{2.5} fraction of particles in the air, sampled at ambient conditions. The work started in 2000 and included extensive field tests in different climatic regions of Europe until autumn 2003. Nine measuring sites were involved in this study located in Greece, Italy, Spain Austria, Germany, the Netherlands, the United Kingdom and Sweden. Generally, the PM_{2.5} test procedure will cover the same aspects as the PM₁₀ test procedure: establishment of an appropriate and practical PM_{2.5}

Reference Method (based on the gravimetric determination of PM sampled on a filter) as well as deriving a workable test procedure to prove equivalence of further candidate methods to the Reference Method. The final PM_{2.5} CEN standard method is not foreseen to be approved before the end of 2004. Further details are provided in Annex 3.

3.1.4 Monitoring methods

PM₁₀ measurement methods currently used in European countries for routine measurement of PM within networks are listed in Table 3.1. For most of the countries information was also available on PM_{2.5} methods used in networks. However, this table is not comprehensive and does not always present the latest state. Beta-absorption gauges, followed by TEOM instruments, are still the most widely used instruments. Some networks use gravimetric sequential samplers for compliance measurements, which give equivalent results to the PM₁₀ reference method or PM_{2.5} single channel samplers like the WINS (US EPA PM_{2.5} Reference Sampler) and other equivalent PM_{2.5} samplers. Some networks use co-located PM₁₀ continuous analysers to provide up to date public information only. Though the filters of sequential samplers must be weighed individually, the samplers can be continuously operated over a period of e.g. 14 days and thereby cover the complete period of one year without too high personnel cost for maintenance. The filters of manual gravimetric PM samplers can also be used for the subsequent analysis of PAH and/or heavy metals, in order to fulfil the requirements of the proposed Fourth Daughter Directive.

Table 3.1 PM₁₀ and PM_{2.5} measurement methods currently used in European countries

Country	PM ₁₀ (24h)		PM _{2.5} (24h)	
	Method	No. of meas. points	Method	No. of meas. points
Austria	Beta-absorption TEOM Gravimetry	31 12 12	Gravimetry	1
Belgium	Beta-absorption TEOM	17 21	TEOM	10
Bulgaria	Beta-absorption	8		
Czech Republic	Beta-absorption TEOM Gravimetry	55 2 10		
Denmark	Beta-absorption TEOM Gravimetry	10 2 10	TEOM	2
Estonia	Beta-absorption	3		
Finland	Beta-absorption TEOM Gravimetry	13 28 13	Beta-absorption	4
France	Beta-absorption TEOM	72 253	TEOM	43
Germany	Beta-absorption TEOM Gravimetry	254 16 169	Beta-absorption TEOM Gravimetry	2 2 19
Greece	Beta-absorption TEOM Gravimetry	14 3 1	Gravimetry	1
Hungary	Beta-absorption Gravimetry	22 2	Gravimetry	2
Iceland	Beta-absorption	1		
Italy	Beta-absorption Gravimetry	22 4		
Latvia	Beta-absorption	1		
Norway	Beta-absorption	10	TEOM	3

Country	PM ₁₀ (24h)		PM _{2.5} (24h)	
	Method	No. of meas. points	Method	No. of meas. points
	TEOM Gravimetry	10 2	Gravimetry	2
Poland	Beta-absorption Gravimetry	9 18		
Portugal	Beta-absorption	11		
Slovakia	TEOM	23	TEOM	6
Slovenia	Gravimetry	3		
Spain	Beta-absorption	194	TEOM	2
	TEOM	61	Gravimetry	11
	Gravimetry	75	Optical	10
	Optical	11		
Sweden	TEOM	18	TEOM	10
	Gravimetry	8	Gravimetry	8
Switzerland	Beta-absorption	10	Gravimetry	6
	TEOM	10		
	Gravimetry	35		
Netherlands	Beta-absorption	19	TEOM	5
United Kingdom	Beta-absorption	1	TEOM	4
	TEOM	69		
	Gravimetry	6		

PM₁₀ and PM_{2.5} measurements are conducted by means of the same types of automated instruments and manual samplers. For PM_{2.5} measurements the PM₁₀ inlet is replaced by a PM_{2.5} inlet. Generally, for PM₁₀ measurements impactor type inlets are used in connection with automated instruments and manual samplers as well. All these impactor type inlets should give equivalent results, when operated according to the requirements of EN 12341 (only equipped with filter holder and pump), though they are especially designed, e.g. for different flow rates. For PM_{2.5}, impactor type inlets and a special cyclone type inlet (Sharp Cut Cyclone – SCC) designed for a flow rate of 1 m³/h are employed for automated instruments and manual samplers. These different inlets should give also equivalent results under the above-mentioned conditions.

It is also possible to use automated optical instruments to measure suspended particulate matter. Remarkable progress was made over the last years in the development of optical instruments. Recently developed optical instruments use laser semiconductors as a source of light. In the measuring unit of such devices, the particles contained in the air sample cross the laser beam. The particles scatter the light onto a photo-diode. The signals of the photodiode can be evaluated in two ways, either as a sum signal to represent the total dust concentration or as single signals which are classified according to different particle sizes. In the first case, the inlet determines the measured size fraction, so a PM_{2.5} inlet must be used for measuring PM_{2.5} concentrations. The second version classifies the particles according to different size fractions such as PM₁₀, PM_{2.5} and others by means of its internal classifying system. The smallest particles determined by optical instruments equipped with a laser have a size of approximately 0.1 µm. Nevertheless it has to be mentioned that these methods do not measure the particle mass itself but derive it from other PM characteristics. It should be taken into account that the variability of particle composition can influence the results.

Also black smoke monitoring (OECD, 1964), long established in some Member States, is based on an optical method. Air is sucked through a filter, of which the density is measured by a reflectrometer. The density can be converted by a calibration curve into gravimetric TSP units. However, the conversion depends on the content of black particles within the suspended particulates and thus varies over time and between different types of monitoring sites. No validated international standard exists for this method, and the monitoring signal is not an exact measure of the content of elemental carbon in the aerosol.

A variety of methods is available for the measurement of the carbon content of the aerosol (Heintzenberg, 1991). Organic and elemental carbon can be determined coulometrically after removal of the organic carbon from the aerosol by extraction and thermic desorption and subsequent oxidation of the elemental carbon to CO₂ (VDI 1996, VDI 1999). These manual methods are rather accurate, but labour intensive. However, the distinction of organic and elemental carbon remains one of the critical issues and is determined by the analytical procedure of method itself. Continuous soot monitoring (black carbon) is based on an optical method, the aethalometer (Hansen (1984)), which compares the transmission of light through a filter loaded with particulates with the transmission through an unloaded part of the filter. Measurements of elemental and organic carbon are not only important from a health perspective, but are also indispensable as input for source apportionment (Chapter 6) and the validation of models (Section 3.2).

Ultrafine particles contribute strongly to the particle number, but not to the mass (see Chapter 2) and are therefore usually characterised by their number concentration rather than by mass. For measuring the number concentration of ultrafine particles the so-called Scanning Mobility Particle Sizer (SMPS) is used most frequently nowadays. This instrument allows a more precise and faster classification of ultrafine particles than the classical diffusion battery. In the SMPS, particles within a size range of approximately 5 to 800 nm can be classified. The principle of this device is to classify positively or negatively charged particles according to their electrical mobility and hence their size in a Differential Mobility Analyser (DMA). After exiting the DMA, the classified particles enter a condensation unit with saturated alcohol vapour in which the ultrafine particles become droplets large enough to be counted optically by a laser photo detector unit (for details see (Fuchs, 1963) and (Wiedensohler, 1988)). However, Scanning Mobility Particle Sizers are not yet used routinely as are automated PM₁₀ or PM_{2.5} instruments. Mostly they are operated in the framework of research projects and run by scientific staff.

3.1.5 Problems and steps taken on the way to resolve these

As mentioned earlier, the CEN standard EN12341 is the reference method for PM₁₀ under the First Daughter Directive. However, any other method can be used if a Member State can demonstrate that it gives equivalent results or displays a consistent relationship to the reference method. In the latter case, results have to be corrected by a correction factor to produce results equivalent to the reference method.

Intercomparison measurements of PM₁₀ TEOM and PM₁₀ beta-absorption instruments with the gravimetric reference method performed in 1996 in three cities (Madrid-Berlin-Birmingham study, coordinated by the European JRC, Ispra) as well as results from intercomparison measurements performed by the Member States and collected by the EC working group on PM (EC, 2001) demonstrated that no constant and uniform relationship of continuous monitors and the reference method could be established. There was a wide range of factors depending, inter alia, on the monitoring site, the composition of the aerosol, the type of instruments used, and the season. Typically, lower readings of the continuous monitors compared to the reference method were found during winter time and in North-western Europe (up to 30 % for seasonal means), whereas ratios from experiments performed in Southern Europe or on hot summer days could be close to one (EC, 2001). Similar results were also found in validation experiments for the CEN PM_{2.5} field test procedure.

It could be shown that the lower readings of continuous monitors compared with the reference method are predominantly caused by evaporation of semi-volatile aerosols such as ammonium nitrate on the filter or in the sampling tubes, which have to be heated to some extent in continuous monitors (Kuhlbusch, 2001). This heating of the sampling tube or the filter unit (typically up to 50 °C in older types of continuous instruments up to 50 °C) is necessary to remove moisture from the sample, to avoid condensation effects and to stabilize the measurements (TEOM). Without proper heating there is a risk of measuring artefacts.

Also the gravimetric reference method gives readings which do not necessarily reflect the "real" ambient PM mass at the time of sampling nor the dry mass. Due to the conditioning of the filters to 50 % RH and 20 °C before weighing, hygroscopic aerosols such as $(\text{NH}_4)_2\text{SO}_4$ collected on the filters may take up or loose water due to the change of relative humidity (Harrison et al., 2004). Investigations by Winkler and Junge (1972) and more recent studies by Neusüß et al. (2000 and 2002) have shown, that the mass increase of dry filter samples was in the range of 10 - 30 %, when exposed to relative humidities of 50 % to 60 %. This water content inherently complicates the comparison with other methods. Modelling and mass closure studies that do not take account of water will often miss a substantial contribution to total PM mass as obtained by gravimetric measurements.

Loss of volatile material can also occur in the PM_{10} Reference Samplers, e.g. on hot sunny days in summer. If the filter holder is exposed to strong solar radiation, the filter of the Reference Samplers can become heated up to 50°C or more. Depending on the measuring period (e.g. winter or hot sunny days in summer) and the amount of volatile particles present in the air, the concentration ratio "automated instrument/Reference Sampler" can vary within a wide range between 0.5 and approximately 1 for daily means. Therefore, it is very useful to have information on the amount of volatile particles in the air during sampling. For the determination of nitrate (most of the nitrate is usually available as ammonium nitrate, a highly volatile compound which can dissociate into gaseous nitric acid and ammonia), a newly-developed continuous analyser is now available that is based on flashing of a $\text{PM}_{2.5}$ sample under nitrogen and subsequent determination of NO_x .

Various steps have recently been undertaken by the manufacturers to reduce losses of semi-volatile particles. Some sequential samplers have been equipped with cooling devices for the stored filters and the sampling tubes. The TEOM instrument and beta-gauge instruments were modified to operate at lower heating temperatures.

In the case of the TEOM, a diffusion dryer can be installed before the filter unit which removes the moisture of the sample (TEOM SES). This provision allows a reduction of the filter temperature from 50°C to approximately 30°C. However, the diffusion dryer removes also some particles from the air sample, particularly in the coarse mode.

In addition, the new versions of TEOM have an automatic built-in correction of $1.03 [\text{TEOM reading}] + 3 \mu\text{g}/\text{m}^3$, to make them consistent with the EPA certification. However, recent intercomparison measurements show that these improvements reduce the losses in comparison with the reference method, but there is still a significant bias (see Table 3.2).

New versions of beta-gauge instruments (e.g. FH62IR) were equipped with heating systems (TRS) for the sampling tube and the filter which can be controlled in relation to the ambient temperature and/or relative humidity and keep the tube 8 °C and the filter 3 °C above ambient levels, respectively. This is a compromise between condensation of moisture on the one hand and evaporation of semi-volatiles on the other. Also for beta-gauge monitors, the bias to the reference method could be reduced, but not avoided (see Table 3.2).

A prerequisite for using these automated instruments is that the measuring unit with the filter or filter tape is installed in a cabin with a controlled temperature of around 20°C.

The Differential TEOM introduced in 2001 uses two TEOM sensor units as well as two electrostatic precipitators and a diffusion dryer (SES) upstream. The two precipitators are alternatively switched on and off out of phase, so that alternatively one sensor is driven with particle free air, the other with particle laden air. The sensor in the particle free air stream is measuring the "artefacts" (e.g. evaporation of semi-volatile material), and the signal of the particle laden sensor can be corrected accordingly. However, this system cannot compensate drifts of the sensors, which may be different from each other. Consequently, the differential TEOM was further improved using only one sensor unit, a diffusion dryer (SES) and a cooled particle filter system upstream of the sensor.

The air stream alternatively bypasses the purge filter or is cleaned from particles passing through the cooled filter. The same sensor unit thus alternatively measures the particle mass and possible artefacts (TEOM FDMS). No comprehensive intercomparison measurements with the reference method in Europe for PM₁₀ at several sites have been reported to date. The results of a first study at one monitoring station near Paris from Sept. 2002 until August 2003 look encouraging (Blanchard, 2003). Further studies at different types of sites and climatic conditions are needed to fully evaluate the performance of FDMS in relation to the reference method.

To harmonise the PM₁₀ data measured with automated instruments in the various Member States the Commission, supported by a Member States Working Group developed a guidance paper on PM₁₀ monitoring and intercomparisons with the reference method (EC, 2001). It gives guidance on a set of principles for the intercomparison measurements between automated PM₁₀ instruments and the PM₁₀ reference method to demonstrate equivalence or to establish a consistent relationship between automated instruments and reference method. Only as an *interim solution for those Member States who had not conducted such intercomparison measurements until now*, so-called default factors for the correction of the results from automated instruments were proposed in the report. A default factor of 1.3 for the correction of the results from TEOM and beta-absorption-instruments, for both daily averages and annual means, was proposed.

If seasonal correction factors (or equations) are necessary it is recommended that interpolation through moving averages of the factors are used to avoid discontinuities in the time series when changing from season to season. Alternatively, a Member State might use the more stringent factor throughout the year; this would be easier for network operation and management as well as erring on the safe side for reporting.

Since the problem was recognised, intercomparison exercises were carried out by a considerable number of Member States and other European countries (e.g. United Kingdom, Netherlands, Germany, Spain, Belgium, Austria, France, Switzerland) leading to similar results but also demonstrating a large range of possible correction factors. Below, examples of correction factors are given and, hence of the comparability of gravimetric and automated methods.

PM₁₀

At several Belgian sites (Flanders) comparison measurements were conducted between PM₁₀ Reference Samplers and different automated methods. On the basis of regression analyses the following correction factors were derived:

Beta-absorption gauge (FH62I-N)	: 1.37 (average for all stations)
Beta-absorption gauge (FH62I-R)	: 1.39 (average for all stations)
TEOM at 50°C	: 1.47 (average for all stations)

It should be noted that the calculation of the correction factors gave better results, when the data sets from all related sites were merged, than for a single site's data set. The variance coefficients R² for the data sets Reference Sampler/automated instruments, which contained each between 70 to 116 paired values, were around 0.85.

Figure 3.1 compares the annual mean values from the Reference Samplers with the corrected annual mean values from the automated instruments. The annual PM₁₀ limit value (40 µg/m³) is plotted in the graph as a straight line.

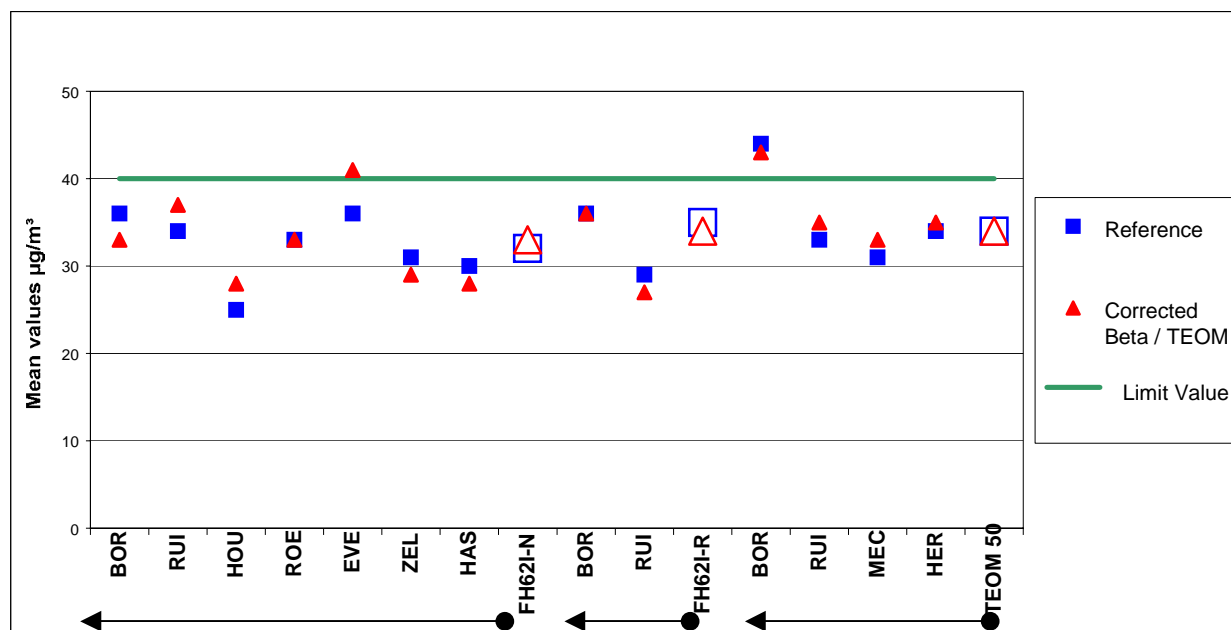


Figure 3.1 Comparison of the annual mean values from gravimetric samplers and corrected automated instruments for several Belgian stations (Flanders)

For the selected stations, the application of the correction factors to the data of the automated instruments gives quite reliable results on average. However, exceedances of the annual limit value cannot be established for each individual site by using a common correction factor for all related sites.

Figure 3.2 shows the number of exceedances of the 24h PM_{10} limit value of $50 \mu g/m^3$ during one year derived from the data measured with Reference Samplers and from corrected data measured with automated instruments.

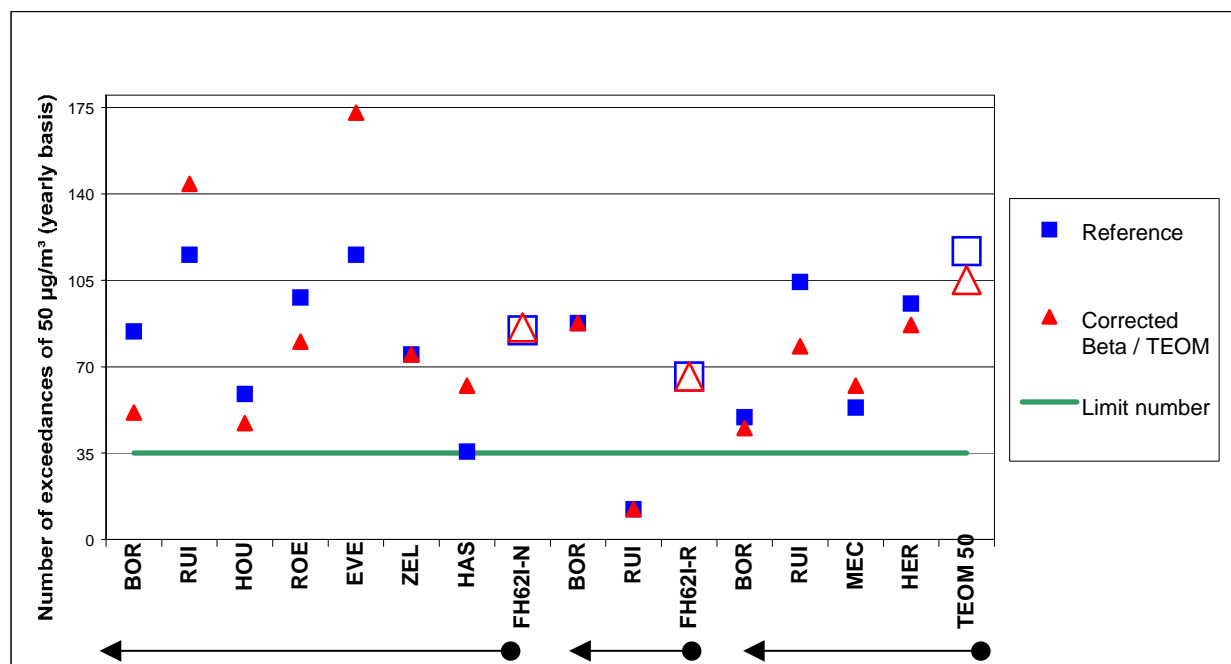


Figure 3.2 Number of exceedances of the 24h PM_{10} limit value derived from data of Reference Samplers and related data of automated instruments

The graph shows clearly that the application of correction factors to daily averages gives rather poor results. Obviously, the correlation between the data sets of the Reference Samplers and those of the automated instruments is not close enough.

Intercomparison measurements with more recent types of continuous instruments (TEOM with SES, run at 30 °C, and beta-gauge with TRS operated at approx. 3 °C above ambient temperature) and the gravimetric reference method (HVS Digitel DHA-80) were carried through from January to September 2003 at 19 stations in Germany (Northrhine-Westphalia). The results in Table 3.2 show a better correspondence with the gravimetric reference method, a good correlation (variance coefficients r^2 about 0.9) and quite consistent correction factors around 1.28 (TEOM) and 1.10 (FH62: beta-gauge), which did not vary systematically between different types of monitoring sites (traffic exposed (UT), urban background (UB) or industrial (UI)).

Table 3.2 Comparison of continuous PM_{10} monitors (TEOM and FH62 beta-gauge) with the gravimetric reference method at 19 sites in Germany, Jan.-Sept. 2003. N: number of daily means, r^2 : variance coefficients, y: daily mean measured with continuous method, x: daily mean measured with HVS Digitel DHA-80.

Station	Method	N	mean(x)	mean(y/x)	1/mean(y/x)	r^2
UI	TEOM	121	34.4	0.77	1.29	0.88
UB 1	TEOM	120	27.7	0.85	1.18	0.86
UB 2	TEOM	120	31.3	0.76	1.31	0.87
UB 3	TEOM	198	45.4	0.76	1.32	0.85
UB 4	TEOM	80	28.7	0.81	1.23	0.84
UB 5	TEOM	79	34.7	0.79	1.27	0.85
UT 1	TEOM	106	33.7	0.77	1.30	0.82
UT 2	TEOM	120	41.0	0.78	1.28	0.91
UT 3	TEOM	123	29.1	0.76	1.32	0.87
UT 4	TEOM	105	31.3	0.80	1.25	0.85
		Sum N = 1172	Mean value of correction factor b: 1.28			

Station	Method	N	mean(x)	mean(y/x)	1/mean(y/x)	r^2
UB 1	FH62	127	27.3	0.89	1.12	0.93
UB 2	FH62	112	30.3	0.86	1.16	0.90
UB 3	FH62	115	27.9	0.93	1.08	0.94
UI	FH62	221	43.9	0.97	1.03	0.89
UB 4	FH62	116	33.6	0.88	1.13	0.96
UB 5	FH62	121	31.1	0.88	1.13	0.87
UB 6	FH62	120	29.5	1.03	0.97	0.96
UT 1	FH62	117	31.7	0.89	1.12	0.89
UT 2	FH62	119	29.3	0.87	1.14	0.92
		Sum N = 1168	Mean value of correction factor b: 1.10			

These data clearly demonstrate that correction factors also depend on the types of instruments used, that some progress has been achieved by recent developments of the manufacturers, but that a significant bias relative to the reference method still exists.

A further step forward on the long and stony way to the comparability of PM monitors is the upcoming report "Demonstration of equivalence of ambient air monitoring methods" prepared by an EC Working group on Guidance for the Demonstration of Equivalence. The draft version of the report (January 2004) is out for external comments and is scheduled to be published in 2004. Inter alia, the report will describe a test programme (laboratory and field) for determining whether a PM candidate method (manual or automatic) is equivalent to the CEN reference method within the data quality objectives of the first daughter directive. It is planned to adopt this report also as CEN technical specification. Once available, the test procedure laid down in this report will replace the preliminary guidance from 2001 (EC, 2001).

The correction factors derived from the measurements conducted at the Belgian sites are more or less in the same range as the default factor of 1.3 for winter respectively cool seasons proposed in the Guidance Report. However, for single sites correction factors can deviate much from the value of 1.3. Some comparison measurements yielded correction factors below 1. The physical and/or chemical effects that cause the correction factors to be below 1, are not known at present.

According to the reports on air quality in 2001 by Member States to the Commission under the First Daughter Directive, correction factors have been applied at 72% of the monitoring sites (see Table 3.3). The correction factors reported in general are in the range of 1.0-1.3 for both beta-absorption gauges and TEOM¹². The correction factors are concentrated either around 1.0 or 1.3. It can be assumed that in a large number of cases the default correction factor (1.3) has been used. The indication of a correction factor of 1.0 could as well mean that no proper equivalence test has yet been carried out or finalised.

Table 3.3 Correction factors applied in 2001 to results from automated instruments in Member States according to the reports submitted under the First Daughter Directive. The percentage share of individual factors per method and Member State is shown in brackets.

Country	Method	Correction factor			
Austria	Beta-absorption	1.3			
	TEOM	1.3 (81%)	1.14 (14%)	1.22 (5%)	
Belgium	Beta-absorption	1.3			
	TEOM	1.3			
Germany	Beta-absorption	1.3 (45%)	1.0 (29%)	1.2 (14%)	1.15 (7%)
	TEOM	unknown (5%) 1.3 (39%)	1.0 (35%)	1.25 (23%)	1.2 (3%)
Denmark	Beta-absorption	1.0			
	TEOM	No measurement			
Spain	Beta-absorption	1.0 (42%)	1.13 (21%)	1.04 (14%)	1.2 (13%)
	TEOM	1.3 (5%) 1.3 (40%)	not reported (5%) 1.17 (26%)	1.13 (3%)	1.1 (31%)
Finland	Beta-absorption	1.0			
	TEOM	1.0			
Greece	Beta-absorption	1.0			
Ireland					
Portugal	Beta-absorption	1.3			
Sweden	TEOM	1.3			
Netherlands	Beta-absorption	1.33			
United Kingdom	Beta-absorption	1.0			
	TEOM	1.3			
France	Beta-absorption	1.0 ¹⁾ (85%)	1.0 (15%)		
	TEOM	1.0 ¹⁾ (81%)	1.0 (19%)		

¹⁾ In these cases France did not include this information as part of the annual report for the year 2001. However, it was reported to the Working Group that a correction factor of 1.0 had been applied so far. The French authorities are still investigating the matter and have not yet decided formally about correction factors.

Based on the experience in general, it can be concluded that there is a clear need to apply a correction to results from non-reference methods. However, the large variability of correction factors – depending inter alia on the location, the season and the instruments used – found in intercomparison measurements leads to the conclusion that *the application of a default factor is not a satisfactory solution.*

¹² In some cases correction factors below 1.0 are reported, which relate to TSP measurements.

PM_{2.5}

CEN WG 15 has recently performed field validation tests of PM_{2.5} samplers at nine European sites in the framework of establishing the upcoming CEN standard for the PM_{2.5} reference method. Inter alia, beta-gauge and TEOM (SES) instruments were compared with gravimetric samplers (45 - 90 pairs of daily means per monitoring site). The correction factors which can be extracted from the correlations varied considerably (range between 0.8 and 1.5), depending on the monitoring sites, the types of instruments and the seasons. As in the case of PM₁₀ instruments, measurements in warmer climates and/or during summer typically were at the lower end of the range, whereas rural monitoring sites with a high proportion of secondary aerosols ranged at the upper end. Application of PM_{2.5} correction factors to the means of the data sets of the automated instruments would again give reliable results but not for individual daily means, for which the relation between gravimetric samplers and automated instruments often showed a large scatter. By and large, it seems that the differences between automated PM_{2.5} instruments and gravimetric samplers are at least as large as for PM₁₀ instruments.

Conclusions on PM₁₀ and PM_{2.5}

The results for PM₁₀ and PM_{2.5} show that network users should follow the recommendation of the EC-Guidance Report to establish their own correction factors, rather than apply a default factor of e.g. 1.3. Once the test procedure for equivalency in the upcoming EC report will become available, networks should establish and demonstrate equivalency following the new guidance, which will replace the preliminary guidance issued in 2001 (EC, 2001). The correction factor problem becomes very visible when the correction is not applied consistently (e.g. in neighbouring Member States). The first reports submitted under the First Daughter Directive showed that a correction is not yet applied at all monitoring sites with equivalent methods in the EU.

Because of opposing factors, it is difficult to predict whether the problems of poor equivalency will increase or diminish when changing the PM metric from PM₁₀ to PM_{2.5}. On the one hand, slight deviations of the samplers from the ideal PM_{2.5} cut point will result in lower mass changes, as PM_{2.5} is close to a minimum of the particle mass distribution, whereas the PM₁₀ cut point is close to the maximum of the coarse mode. On the other hand, the overall PM mass decreases by about one third when going from PM₁₀ to PM_{2.5}, and the portion of secondary semi-volatile aerosols which is predominantly in the fine mode increases.

3.1.6 Further steps of harmonisation (QA/QC demands)

In general the QA/QC requirements described in Section 3.2.6 of the first Position Paper on PM of 1997 relating to the First Daughter Directive are still appropriate. These requirements cover all relevant items and can also be transferred to PM_{2.5} measurements. However, an additional requirement concerning the filter handling of manual samplers should be made. As sampled filters can lose volatile particulate matter during the transport from the site to the weighing facility under hot conditions, the samples should be maintained at a temperature close to the temperature of the weighing facility between retrieval from the sampler and storage in the conditioning room. If the sampled filters are not immediately transported to the weighing facility but stored at the site or stored in the laboratory for some days, they should be kept in a cool environment, preferably in a refrigerator.

Current experience shows that the required accuracy of 25% (Annex VIII of the First Daughter Directive) for continuous PM₁₀ measurements using non-reference methods can in many cases only be achieved if rigorous QA/QC-procedures are followed (UMEG et al., 2000). This includes frequent maintenance and checks of the performance of the monitor as well as comparison with the reference method.

As shown in the previous section, proper application of correction factors is indispensable when using non-reference methods. If this is not done, serious comparability problems and border inconsistencies in the range of 30% (in worst cases even up to 50%) for daily means between regions can be anticipated.

Harmonisation and quality control on the regional and European scale is especially important in view of the absence of any Standard Reference Material for PM. Therefore intercomparison exercises always require some field tests using the reference method that are often rather demanding in time and manpower. However, these kinds of exercises have to be carried out further in order to come to a better understanding, to solve problems and to achieve better harmonisation in the long run. AQUILA, the network of European reference laboratories, will start field intercomparison measurements with the PM₁₀ reference method in 2004, coordinated by JRC.

3.1.7 Monitoring strategy

The strategy for monitoring PM is on the one hand predefined by the requirements of the Air Quality Framework Directive and the First Daughter Directive, on the other hand it depends on the spatial variability and structure of the PM pollution. Additional pollutant-specific specifications are given in the First Daughter Directive such as requirements on the minimum number of stations per zone or agglomeration, the siting of monitoring stations, data quality objectives and the use of other tools than continuous monitoring. These provisions are largely based on the findings and recommendations in the first Position Paper on PM. As the basic ideas on monitoring strategy did not change since then, no general revision of this part was deemed necessary.

The general requirement in the Framework Directive that the air quality has to be assessed everywhere, should be seen in combination with Annex VI of the First Daughter Directive, which states that sampling points directed at the protection of human health should be sited to provide data which are representative for the exposure of the general population and where the highest concentrations occur to which the population is likely to be exposed for a period which is significant in relation to the averaging period of the limit value (hot spots). Guidance on this has been provided also in an EC-guidance report on assessment (EC, 2000).

The First Daughter Directive does not give indications for the spatial resolution of air quality assessment at hot spots around point sources. According to the EC-guidance report mentioned above a spatial resolution of 250x250 m² may be sufficient for a residential neighbourhood in the direct vicinity of an industrial area. In this report more guidance on other assessment issues can be found.

In the last few years, Member States have gained experience in applying these provisions, and in setting up appropriate networks for compliance checking. It is useful to review these experiences and the implications for the monitoring strategy, and to derive proposals to change or amend the legislation, if necessary.

In preparation of the implementation of the First Daughter Directive, Member States have reviewed their monitoring networks in order to review the network design with regard to the new requirements. Monitoring is obligatory in zones where the lower assessment threshold (LAT) is exceeded. Since the LAT for PM₁₀ (10 µg/m³ as annual average) is exceeded in most parts of Europe, monitoring is obligatory almost everywhere. In an EC-report on methods and results of the preliminary assessment (EC, 2002), there are indications that Member States have improved their network design. In many cases the number of monitoring stations changed, in some cases stations were relocated. As could be expected, there has been a major shift from monitoring Total Suspended Particles (TSP) to PM₁₀. According to the meta data submitted under the European Council Decision 97/101/EC on Exchange of Information (EoI) for 2001, PM₁₀ is measured at 63% of the stations compared to 37% for TSP. The relative share of station types was 43% for "urban background", 32 for "traffic", 9% for "regional background" and 9% for "industrial" (De Leeuw and Van Hooydonk, 2003). This distribution varies widely between Member States. This distribution varies widely between Member States, as can be taken from Table 3.4, where the number of stations per types for four Member States (MS) and one Accessing Country (AC) are compared with the average distribution in the EU.

Table 3.4 Number of rural, urban background and hot spot (traffic oriented and industrial) stations reported by selected Member States (MS), Acceding Countries (AC) as well as for the whole EU. The percentage of hot spot stations in the different monitoring networks was also reported (data from AIRBASE 2001).

Station type	MS1	MS2	AC	MS3	MS4	EU
Rural	16	3	19	0	8	97
Urban	13	45	29	6	84	417
Traffic	66	8	6	3	24	293
Industrial	23	4	1	1	19	77
Not defined	8	0	0	0	2	32
% hot spot of all stations	71%	20%	13%	40%	31%	40%

Table 3.4 demonstrates that there are still large differences in network design between the Member States. Whereas monitoring capacity in MS2 or AC is concentrated in the urban and rural background, where people are most likely to be exposed continually, the majority of stations in MS1 is concentrated at hot spots. Consequently, differences in mean PM₁₀ levels or total number of exceedances of limit values reported by the Member States cannot be compared directly and do not necessarily reflect the degree of the PM burden, without taking into account the different station mix. Since traffic can be identified as one of the major sources for PM emissions, which is ubiquitous, a better harmonization of the percentage of stations representing these hot spots and thereby the network design seems desirable.

Another problem which hampers the comparability of monitoring data are the somewhat vague siting criteria and description of monitoring sites in the Exchange of Information Decision, particularly for traffic exposed sites. Monitoring locations characterised as "urban traffic" (UT) can be within a street canyon, but also some 50 m away from busy streets. This may cause differences in the evaluation of the local PM₁₀ burden by traffic (40 % on average, see Chapter 4). Consequently, stricter siting criteria should be developed. As a first step, the exact location of traffic exposed sites should be reported.

As a result of the changed and enlarged PM monitoring networks in the Member States, the knowledge about the spatial structure and variability of the PM pollution in Europe has considerably improved, particularly for PM₁₀ (see also Chapter 4). The PM burden in most European cities and agglomerations can be characterised by the picture of an "island" of elevated PM levels (urban background) emerging out of a "sea" of regional background PM concentrations. The regional background differs within Europe, depending on different natural contributions or secondary aerosol formation and influenced also by long range transport, but on a city scale, it is quite uniform. On top of the "island" there are "peaks" of PM concentrations, corresponding to hot spot areas such as busy street canyons or certain types of industrial sources.

The general qualitative picture which was already an underlying assumption for the monitoring strategy of the first Position Paper has been confirmed. It is therefore reasonable to retain the four station categories: regional, urban background, road side and industrial. Only a few well placed monitoring stations per zone in most cases are sufficient to measure the regional and urban background, respectively. They should be representative, as stated in the First Daughter Directive, of at least an area of several square kilometres for urban background. Consequently, the minimum numbers of sampling points given in Annex VII of the First Daughter Directive for larger agglomerations (more than 1 million inhabitants) could be slightly reduced, the more so if PM_{2.5}, which tends to be somewhat more evenly distributed, will become the main parameter to be monitored. However, the major problem arises from monitoring hot spots. Their number can be legion in a large conurbation, bearing in mind the number of busy roads, and substantial concentration gradients have to be expected near such locations. A complete coverage of hot spots by measurements alone is practically impossible, and the combination of monitoring with model calculations is the only feasible option. The following steps provide a rather general outline that could be undertaken to design an appropriate network concerning in particular hot spots.

For streets:

- Diffusion modelling (simple street canyon models) of all relevant streets within a conurbation (screening). Relevant are at least all streets with adjacent residential premises and a moderate to high traffic flow.
- Select at least one or two of the highest polluted streets (taking into account also population exposure) for monitoring. Typically, these will be street canyons, and it may not be easy to find a siting place for the monitors. Specially designed "mini"-containers may be necessary. Place the monitors near to the building line within the streets following the siting criteria Annex VI of the First Daughter Directive.
- "Calibrate", if necessary, more sophisticated street canyon models, taking the real topography of the street and the resuspension into account, with the measurement results and apply the improved model to other relevant traffic exposed sites.
- Check the calculated exceedance of limit values at least by preliminary measurements, if the results of the model do not fulfil the data quality objectives laid down in the First Daughter Directive.

For industrial hot spots:

- Check the emission inventory and the experience gained by controlling industrial facilities for proper consideration of diffusive sources of particles.
- Especially if residential areas are exposed within a relevant neighbourhood (closer than 1 km from the source), place a monitor downwind of the source (with regard to the main wind direction) at the edge of the residential area being closest to the facility. Diffusion modelling may help to identify appropriate monitoring sites. However, diffusive sources are often not or only partly included in emission inventories, so that modelling results should be checked by measurements.

Setting up monitoring stations that fulfil the requirements of the legislation but also take practical and administrative considerations into account is often a matter of compromise. Despite some progress, it is therefore still not possible to propose simple recipes for the harmonisation of the network design. This might even require a detailed analysis of individual stations. In view of the importance of station siting for the comparability of data, reporting on the exact locations and surroundings of measurement stations by the Member States should be improved.

There is an increasing use of models as supplementary tool for the assessment of air quality. Several Member States are now using models even for compliance checking. This has consequences for the monitoring strategy, since it may influence the number and siting of monitoring stations. The exact relation between monitoring and modelling in the assessment of air quality is not clearly defined in the First Daughter Directive and is therefore still under debate. The uncertainties of models limit their application in compliance checking. On the other hand, models are necessary tools for assessing future concentrations. The extent to which models can be used is not clearly defined in the First Daughter Directive. The data quality objectives for modelling in the First Daughter Directive need further clarification.

Practical monitoring strategy will also be influenced by the choice of instrumentation. The use of the reference method implies a time lag of days between sampling and analysis of filter samples. This raises problems to comply with reporting requirements to the public (1999/30/EC, Art. 8), specifying that ambient concentrations of PM₁₀ shall be updated on a daily basis. The use of automated instruments on the other hand still raises problems in term of equivalence with the reference method (see previous section). A combination of both kind of instrumentation can be a possible solution to deal with the different requirements.

It should be mentioned that some Member States have national legislation beyond the air quality directives that requires a high time resolution (e.g. 1 hour) or that relates to short term actions and therefore makes it necessary to use automated instruments.

A change from PM_{10} to a different metric may lead to re-considerations with respect to the location of sampling sites as well as their total number per zone or agglomeration. Although the knowledge on $PM_{2.5}$ distributions is limited, there are indications for a more smooth spatial distribution compared to PM_{10} (see Chapter 4). A change from PM_{10} to $PM_{2.5}$ might make it possible to decrease the minimum number of sampling points for big conurbations.

3.1.8 Conclusions and recommendations on monitoring

3.1.8.1 Conclusions

Measurement of PM mass concentrations brings along considerable uncertainties mainly because of the risk of alterations of the sample during sampling itself, the handling of samples and during the measurement process. This highly depends on the environmental conditions and composition of the particles. Loss of semi-volatile particles is one of the major problems.

The PM_{10} Reference Method (EN 12341) was adopted 1998. The standard only applies to manual samplers, but it does not cover equivalence tests of automated instruments. Although the $PM_{2.5}$ Reference Method currently developed by CEN will not explicitly address automated monitoring methods, the statistical procedure given in that standard could still be applied to such methods.

The conflicting requirements of the First Daughter Directive with regard to monitoring (definition of the Reference Method based on gravimetric determination of the mass collected on a filter, implying a delay of days before the concentration is known) and information to the public (information to be updated at least daily) create a competing situation between the reference method and non-reference methods. Only using the reference method will lead to problems in complying with the public information requirements of the Directive, whereas only relying on non-reference methods could lead to problems with compliance checking of limit values if equivalence of the non-reference method can not be demonstrated.

The beta-absorption monitors and the TEOM are still the most commonly used methods to monitor PM in the Member States. However, sequential high or low volume samplers are nowadays also used to monitor the PM concentration, with a time resolution of one day, which is sufficient for compliance checking. A minority of networks address the problem by combining the telemetric approach for up to date public information with the permanent use of the gravimetric reference or equivalent method for compliance checking and/or onsite determination of correction factors throughout the year at some reference stations. However, there is a tendency in Member States to stay with automated instruments also because of lower maintenance costs (in terms of manpower) in most cases. In general Member States acknowledge that the results of continuous monitors have to be corrected by intercomparison with the reference method for compliance checking.

Progress has been made over the last years in the development of optical instruments, also to determine particle mass concentration. Reliable instrumentation for the measurement of particle number concentration and the size distribution of particles is available already since many years. However, due to the high costs and the sophisticated nature of this instrumentation, monitoring of particle number concentrations and size distributions is still mainly used for scientific applications.

A lot of effort has been made to compare results from commonly used non-reference methods with the reference method for PM_{10} . In most cases the results from beta-absorption instruments as well as the TEOM tend to underestimate the concentration substantially due to losses of semi-volatile particles. Therefore results from different methods can not be assumed to be comparable to the Reference Method without further proof, and usually a correction of the data will be necessary. These "correction factors" can vary substantially in space and even seasonally. Correction factors reported to the Commission based on the First Daughter Directive are generally in the range of 1.0-1.3. To harmonise PM_{10} and $PM_{2.5}$ measurements throughout the EU results must be corrected at all monitoring sites

where it is necessary. Despite progress, this objective is not yet achieved. The use of non-reference methods requires the rigorous application of appropriate QA/QC procedures to ensure comparability with the Reference Method and compliance with the data quality objectives of the directive.

A change in monitoring from PM₁₀ to PM_{2.5} would in the first place only require an exchange of the inlet (sampling head), regardless of the kind of measurement method. Because of opposing factors, it is unclear at present whether this change will increase the uncertainty of monitoring results. The number of monitoring sites necessary to cover the area of a Member State in a representative way might be lower compared to PM₁₀ because of the somewhat more uniform pattern of PM_{2.5}. The minimum number of sampling points for fixed measurement of PM in Annex VII of the First Daughter Directive can be slightly reduced for big conurbations, as urban background concentrations for PM_{2.5} can be expected to be rather uniform.

In view of the considerable concentration gradients around industrial sources with diffusive sources, additional guidance is needed, particularly on the size of the representative area to be assessed.

The ratio between hot spot and background stations will to a large extent determine the number of exceedances identified per Member State. As long as this "station mix" is not harmonized, comparisons of the number of exceedances per Member States may be misleading.

The combination of monitoring and modelling is of increasing importance for comprehensive air quality assessment. The uncertainty requirements for models are still not developed to the extent necessary. Additional guidance and clarification may be needed to improve this situation.

When using the reference method it is not possible to fulfil requirements of the First Daughter Directive to make data available to the public on a daily basis. Automated instruments on the other hand may need careful quality assurance to ensure that they meet the data quality objectives, especially the maximum uncertainty. Along with the development of technical solutions to improve the comparability of automated instruments with the reference method, the problems in meeting the requirements for public information when using the reference method should be addressed as well.

3.1.8.2 Recommendations

1. The Reference Method for PM₁₀ (EN 12341, adopted in 1998) should be reviewed as soon as possible taking into account the forthcoming Reference Method for PM_{2.5}. This is well in line with the regular review cycle of CEN (5 years).
2. Current automatic monitoring methods for PM are, in many instances, not equivalent to the reference method. It is considered important that PM measurements in the EU are harmonised. To help ensure this, the Commission has prepared a 'Guidance on Equivalency' (to be published in 2004). In accordance with the First Daughter Directive, this allows for corrections to data to be applied before checking that the remaining uncertainty meets the data quality objectives. Member States should apply this guidance once it is available.
3. Member States are encouraged to intensify QA/QC exercises within their State but also between different Member States.
4. The European Reference Laboratory for Air Pollution, of the Joint Research Centre (JRC/ERLAP) is encouraged to intensify its efforts to support the harmonisation of European PM measurements. AQUILA (European Air Quality Laboratories, the Network of National Reference Laboratories designated according to the Air Quality Framework Directive) should be active in this field.
5. In revising the First Daughter Directive it is important to address the current contradiction that the reference method cannot be used to meet the daily reporting requirements.

6. If new limit values are adopted for PM_{2.5}, it is recommended that careful consideration be given to establish appropriate PM monitoring and assessment strategies.
7. In view of the importance of station siting for the comparability of data it is recommended to review current practice in siting and to consider stricter siting criteria, particularly in the Exchange of Information Decision, which should be harmonized with the requirements of the First Daughter Directive.
8. To ensure better interpretation of results, it is essential to report a detailed description of monitoring sites used for compliance monitoring, including data on the exact locations, the surroundings and representativeness of the stations. Member States should clearly document and report any correction factors applied to the data.

3.2 Modelling of ambient particle concentrations

3.2.1 Scope of Section 3.2

This part of Chapter 3 gives a short introduction to models that are used for describing particle mass (PM) concentrations, uncertainties of such models and how they relate to legislation. This section also discusses the possibilities of modelling other metrics for particles, as number, surface, and specific chemical components as e.g. PAH, some of which have been put forwards as possible components linked to observed health effects.

3.2.2 General

Member States are required to assess the air quality throughout their territory, not only where there is risk of exceedance of thresholds such as limit values. The First Daughter Directive specifies a minimum number of monitoring stations for zones where monitoring is mandatory and where the assessment is solely done by measurements. If additional methods are used for the assessment, in particular computer models, this minimum no longer applies. Models are then used in combination with measurements to give a territory-covering description of the pollution levels. It is then left to the Member States to determine the number of stations needed for ensuring good quality of the assessment. Models will be used both to find possible areas risking non-compliance to be surveyed by measurements and to fill out areas between measurements sites.

Models are even indispensable for prognostic analysis. Prognostic calculations can be necessary to investigate whether limit values will be met in the future or to see how effectively certain emission reductions will bring future levels down. This type of analysis is important for national and local authorities when implementing the First Daughter Directive, but it is equally important for the development of European-wide air pollution abatement strategies and measures.

A third important field of model application is in aerosol research, where models can be used to mathematically express our knowledge of the processes that give rise to particulate matter in the atmosphere, thus enabling us to test, in validation studies, our understanding.

Modelling is a very versatile tool in achieving a fully covering description in time and space on how the air pollutants disperse from a source and form concentration fields over an area. However the capabilities and accuracy of the models are crucial for how extensive this modelling option can be applied. Of the factors affecting the accuracy of the results, the most important is often to provide a correct input to the dispersion models, in particular the emission data. Because different dispersion, transformation and deposition processes work on different time scales, their importance for modelling depends on the associated spatial scales. Only fast processes of less than hours are important on local

scale, while the slower ones have to be included when modelling on regional scale. For the larger scales, chemical or physical transformations significantly changing the characteristics of an air pollutant have to be described. Also deposition processes have to be accurately described at those scales, e.g. dry deposition depends on the surface characteristics and precipitation depends on cloud chemistry and physics. Concerning components with a long life time in the atmosphere, e.g. particles, it is necessary in estimating the local concentration at urban sites to take into account both the regional and the urban background.

In spite of the complexity and difficulties, modelling of gaseous components has shown to be quite successful. Particles, however, have a much more complex chemical and physical history than a usual gas molecule, e.g. they are formed and grow by gas to particle transformation in the atmosphere, which affects physical properties as number, surface and mass distribution. The chemistry of the single particles is changed with physical processes as coagulation and with condensation as secondary components are added. This change of size and chemistry strongly affects the deposition rate. Compared to the ordinary pollution gaseous components particles usually have a longer residence time in the atmosphere. At locations near PM sources, the observed regional contribution, i.e. long distance transported, to particle concentrations is large, over 50% for PM_{10} and $PM_{2.5}$ (see Chapters 4 and 6), implying the need to assess background contributions as well as the local. For prognostic analysis, the sources contributing to the regional as well as local levels can only be determined by a combination of measurements and modelling, as done within EMEP. It is important to note that, especially for shorter distances, the importance of processes may strongly depend on the PM fraction considered. The importance of formation of secondary aerosol on the local scale is not clear yet for the chemistry and number of ultrafine particles, but for PM_{10} these processes are not important. To reach a better understanding of the local situation both the regional and local situation has to be assessed, which only can be done by combining regional measurements and models and more local measurements and models.

3.2.3 Models for PM concentrations

3.2.3.1 Available models

The Model Documentation System (MDS) operated by the European Topic Centre on Air and Climate Change of the European Environmental Agency (<http://air-climate.eionet.eu.int/databases/mds.html>), lists more than twenty urban models dealing with PM_{10} , where the particles are then assumed to act as an inert substance. Some of these models are currently operative in the major European urban areas. A summary of aerosol models used in European dispersion models can also be found under Schlünzen (2001).

Several of the summarised models treat aerosol as inert but only a few include reactive aerosol (METRAS). A useful division of models is according to the spatial scale: large scale (global to regional scale) models, urban scale models and local scale (streets, individual sources). Models for the local scale, in particular models for streets, motorways and urban areas are mostly used only for PM_{10} and $PM_{2.5}$. They do not incorporate chemical reactions and aerosol formation. In practice, the main uncertainties here are probably the emissions and – for prognostic analysis, which cannot be based solely on measurements – the contribution from the regional and urban background. The importance of reaction and formation processes for the calculation of PM_{10} and $PM_{2.5}$ at the urban scale is not precisely known, however; e.g. during episodes of photochemical pollution formation may contribute significantly to the PM mass in large conurbations; formation processes are certainly important for ultrafine particles. An improved understanding of the underlying scientific processes occurring in polluted city environments is needed. For the large scales, aerosol physical and chemical processes must be parameterised or simplified so that it may be implemented in air quality models and used to improve their ability to act as forecasting and policy making tools and to provide relevant information for health impact studies. Many of the models developed for passive gases can also be used for particles, at least for the total mass of fine particles so small that their dynamic behaviour (turbulent

transport and dispersion) may be simulated as if they were gas-like passive tracers. However, this is not true for particles larger than one micrometer, especially those larger than 5 micrometers, for which their weight and their inertia in the turbulent motions are not negligible and gravity settling and cross-trajectory counter-diffusion should be modelled. On the other hand the fine particles molecular diffusivity is a function of their size/mass and this must be taken into consideration when simulating, e.g. surface transfer processes like deposition and coagulation.

The application of the most advanced models is very costly. For regular assessment purposes (policy development, assessment of compliance with limit values in past and future years) it is virtually impossible to apply such models for calculating the concentrations for a full year, so simpler models have to be used. Within CAFE, the integrated assessment model RAINS and the underlying EMEP model are the central models, which are consistently developed and tested. E.g. a dynamic aerosol module for describing the particle number concentration over Europe has been implemented and is presently tested (see Annex 4). For the urban scale, there is a lack of information at the European level; currently the City-delta project is carried out to improve the situation. A European-wide picture of road side and hot spots levels is virtually lacking. Very few national and hardly any local authorities have access to regional scale models, and they have to rely on the European modelling activities for their own regional scale prognoses. The practice of urban and local scale modelling in Member States differs widely. In a few Member States there are regular modelling activities, but in most Member States this happens at best only on an ad hoc basis.

3.2.3.2 *Model uncertainties and validation*

As mentioned above particles, are affected by a series of processes. Most of them are known, but not well quantified, the more of them involved the higher uncertainty is connected to the model. Of major importance is what particle characteristics are modelled and on what scale the modelling is done. One example is comparing mass and number. Coagulation can be an important process in the urban atmosphere, but it actually only redistributes particle mass, so it will not affect the particle mass, while particle number is strongly affected. However on regional scale the larger particles formed by coagulation are more available to deposition by precipitation and thus have a shorter residence time. The uncertainty increases the more the particle characteristic is depending on secondary processes. Mass is much less sensitive as, on a local scale, it depends mostly on emissions strength of the primary particles and deposition processes, that in turn depend on an accurate description or parameterisation of the wind, the turbulence and the particle size distribution of the mass. Secondary processes as condensation are less important as it is probably relatively small mass added compared to the original primary mass. However particle number is much more sensitive, as the smallest particles dominating the total number are strongly affected by secondary processes as nucleation, coagulation and turbulent deposition even on a local scale.

The regional background is in most parts of Europe important for PM mass fractions, while concerning particle numbers the regional background is mostly negligible in the urban environment. This is because secondary processes remove the small particles from the atmosphere and leave the larger accumulation mode particles to dominate the total number of particles. Concerning mass the same secondary processes add mass to the larger accumulation mode particles that are comparably slowly removed from the atmosphere, mostly by precipitation. One important aspect in this that adds considerably to the uncertainty for certain components, especially organics, is the unknown chemistry, including uncertainty of individual rate parameters, strongly affects the formation of secondary material, mainly giving uncertainty to particle size and mass. In total this means that properties dominated by large accumulation mode particles as mass and almost similarly surface in an urban site can be strongly affected by the regional background, i.e. long distance transported particles. Thus, boundary values are essential for describing the particle concentrations within an urban area if only operating the model on local scale.

The major concern in estimating uncertainty for a model prediction is the emission database. The emission database has to contain all different sources and the emission factors have to be accurate, as uncertainties in the emission database will directly be transferred into the results. Although this applies in principle for all scales of models, there are important differences in practice. In urban and local scale modelling, the background levels can for many applications be regarded as given, and then the uncertainties in the coarser fraction $PM_{2.5-10}$ is often dominant. As the urban models actually operate with in the source the spatial resolution becomes more important.

Unlike measurements, there are no agreed QA/QC procedures for model calculations. In view of all uncertainties, there is a generally recognised need to test PM models. A common way in testing models is by comparison of different models. The more direct comparison with measurements is usually connected with difficulties both in finding suitable data, concerning spatial and temporal resolution but also in quantifying the measurement errors and establishing how well the measurements in *e.g.* a point is representing the area covered by *e.g.* one grid box in the model. As comparisons with real measurements is crucial in determining the actual accuracy and precision in the model it is very important that model development and testing is planned and performed in close cooperation with different measurement networks.

A considerable effort on evaluating and improving urban scale models has been made within the EUROTRAC subproject SATURN. An other ongoing activity is the CityDelta project. In this project scenario calculations are done for a few selected cities, using different urban scale models. The models calculate the differences between the various scenarios. The purpose of the exercise is to circumvent some of the uncertainties by comparing and aggregating the differences predicted by the various models. Validation needed to establish confidence, even though a model set up showed give good agreement at a certain time and geographical conditions.

3.2.4 Models describing other metrics than PM_{10} and $PM_{2.5}$

To be able to describe metrics as number of particles it is necessary to implement description of several different physical and chemical processes as nucleation, coagulation, condensation and different cloud processes. Deposition, wet and dry, has to be described in more detail than usual. This kind of modelling is usually described as dynamic aerosol modelling.

In January 2002 an EMEP-workshop “Dynamic aerosol modelling: from box models to 3D transport models” was held to discuss the possibilities and difficulties with such models and to form recommendations for future use. The general conclusions and recommendations from the workshop are given below. Further to give an orientation on the status of the present modelling work, different specialist give short presentations on dynamical aerosol modelling describing the fundamentals, implementation and use on urban, regional and global scale presented in Annex 4.

The main conclusion of the EMEP Workshop was that, despite the recognized uncertainties in aerosol modelling, it is possible to get useful data from current models. The parameters considered to be of interest and possible to predict were: particulate mass of SO_4 , NO_3 , NH_4 , PM_{10} , $PM_{1.0}$, number of particles in Aitken and accumulation modes, optical properties as scattering and absorption. The workshop recommended the use of $PM_{1.0}$ instead of $PM_{2.5}$ in order reduce uncertainty as contributions from natural and anthropogenic events of dust are poorly described. However considerable and intensified attention is required in order to formulate and test better descriptions giving more reliable results. In this work the most important is validation against measured data, which should be pursued regularly.

A higher temporal and spatial resolution of measurements as well as the particle chemical composition is needed in order to validate model results. Practical and economical reasons are recognized and supposed to limit the measurement efforts. But some measurements can easily be performed with high time resolution for number and mass and are thus strongly recommended.

The most common aerosol model approaches are at the moment: sectional, modal and moment based. No clear difference in cpu were agreed on concerning the general approach, while differences exist due to different processes included and the level of detail with which they are described. The modal and moment based models need non-physical corrections. Several efficient modal models are available now.

In identifying the uncertainties in current aerosol models three main sources were found in current aerosol models. The emission inventories and emission model parameterisations should include sea spray, soil dust and other natural particle emissions, as well as provide characterisation of aerosol composition and size. The description of condensation processes can introduce an important source of error in the models. The partitioning between gas and particle for sulphate, nitrate and ammonium can be modelled by using equilibrium for daily averages but will have difficulties below that temporal resolution.

Thermodynamics are still not well known and are important for condensation, cloud processes and nucleation. This affects especially the treatment of organics, clouds and nucleation. The composition and characteristics of organic aerosols are largely unknown. This lack of knowledge severely limits the possibilities for applying and evaluating Secondary Organic Aerosols (SOA) models for policy applications. These areas need considerable and intensified attention to find better descriptions giving more reliable results. Still it is considered possible to have the current models give useful data. But more research on the current SOA formation descriptions is absolute necessary including continuous test and evaluation by comparing with relevant measurements.

3.2.5 Conclusions and recommendations on modelling

3.2.5.1 Conclusions on modelling

For the local scale, there exists an ensemble of dispersion models, that is mainly used for mass fractions (PM_{10} , sometimes also $PM_{2.5}$) that can at that scale be approximated as inert. For the urban scale, a set of models with varying complexity is currently developed and/or tested, e.g. within the CityDelta project. For the regional and global scale, fewer models are available; these include simplified and/or parameterised secondary processes.

Modelling for compliance checking under the First Daughter Directive is currently in its childhood, one of the reasons being that the framework for this is not well defined. Prognostic modelling for policy development is being done in a few Member State; for the European level, models are currently being developed, in particular under CAFE and the CLTRAP.

The major uncertainty for all models is the emissions database. The major uncertainty concerning mass on local scale today concerns the dust emissions that are not well described in current databases. As the scale increases, the secondary processes increase in importance. On the regional scale, the uncertainty in mass is mostly due to uncertainty in the emission databases, especially concerning dust emissions, but also concerning organic compounds, as the secondary formation is not well known. Models have still difficulties in describing some inorganic compounds as nitrate. Concerning metrics other than mass, the emission data base is at best in a development stage.

Models have to be compared with experimental values. Modellers strongly emphasise that there is a lack of experimental data and evaluation studies. As today's models for PM still have severe shortcomings, they need to be continuously checked and improved by comparisons with measurements.

3.2.5.2 Recommendations on modelling

1. Whilst considerable progress has been made in PM model development, there is still a need to improve, compare and validate models at all scales for a better understanding of PM and for provision of policy support.
2. As the uncertainty in model results is to a large extent determined by the uncertainty in anthropogenic and natural emissions of particulate matter, it is important to improve emission databases in Europe. For modelling there is a particular need for better emission data at the local authority level.

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4. Current concentrations and exposure

4.1 Scope

This chapter summarises our knowledge about the current PM concentrations and exposure. In particular, it discusses what current concentrations of PM₁₀ are compared to the limit values, and what is known about levels of other PM metrics (PM_{2.5}, PM_{1.0} and ultra fine particles). It investigates whether major differences between different regions in Europe exist. It characterises hot spots of the PM burden, including how confined or extensive in space they are. The uncertainty associated with the currently reported levels is reviewed. Finally, this chapter discusses the extent to which outdoor monitoring sites reflect personal exposure.

4.2 Current PM₁₀ concentrations in relation to the limit values

The following overview of PM₁₀ concentrations in Europe is primarily based upon PM₁₀ data reported by EU Member States to the European Commission (under the Exchange of Information Decision, 97/101/EC) and by other countries to the European Topic Centre Air Quality and Climate Change (ETC-ACC) (data from 25 countries in 2001). In addition, data have also been used which have been made available by country representatives of the PM Working Group.

The European PM₁₀ monitoring network is developing fast. From 2000 to 2001, the spatial coverage of the network has considerably improved, the number of stations (with at least 70% data coverage) rising from 355 to 718.

The majority of the reported data stem from continuous measurements (beta-absorption gauge and TEOM, see Chapter 3), only few data sets were measured by the reference method EN 12341 (CEN, 1998). The information at hand and time did not allow for an assessment of the data quality, in particular with respect to the application of correction factors (European Commission, 2002). The data were taken as reported to AIRBASE, without further quality considerations. This may particularly impair the comparability of data from the years 2000 and before, as EC guidance relating continuous measurements to the manual reference method was not published before 2001. An additional uncertainty affecting in particular the spatial distribution of PM levels over Europe originates from the different network design and "station mix" in the Member States. Whereas for example in Spain or Italy most stations measure at hot spots, other countries such as the United Kingdom or the Netherlands have placed the majority of their national stations at rural or urban background sites (compare Table 4.1). The different spatial representativities of the networks can give rise to considerable uncertainties when comparing PM levels from one country to another.

CAVEAT – Due to differences in calibration of the continuous monitors in relation to the reference method, and due to a differences in the "station mix" in the networks of the Member States, full comparability of PM₁₀ levels over Europe is not ensured.

Compared with the PM₁₀ limit values of the First Daughter Directive to be met in 2005 (Stage 1), PM₁₀ concentrations in parts of Europe are rather high. In 2001 (data for the year 2000 in brackets), the EU limit values were exceeded at 34% (34%) of the 718 (355) monitoring stations with at least 70% data coverage, especially at hot spot locations (see Section 4.2, Table 4.1 and Maps 4.1 - 4.3).

In 2001 (2000), PM₁₀ monitoring data in AIRBASE included around 750 (219) cities. The 24-hour limit value (not more than 35 exceedances of 50 µg/m³) was exceeded at least at one monitoring site in

179 (90) cities. A number of cities with a total of about 20 million people had exceedances on more than 45 days at least at one monitoring location.

The distribution of cities over Europe which have reported PM₁₀ data for 2001 to AIRBASE, and their PM₁₀ levels is presented in Maps 4.1 - 4.3. For Map 4.1 and 4.2, which show PM₁₀ concentrations at urban background and hot spot locations (exposed to traffic or industry, see Table 4.1), the 36th highest 24-hour mean was selected, as this characteristic exceeds the limit value to a larger extent than the annual means (see below). For rural sites, PM₁₀ annual means are shown in Map 4.3.

Table 4.1 *PM₁₀: Number of stations with concentrations higher than the limit values (LV) and with concentrations between upper assessment threshold (UAT) and LV (2001)*

Per country: total number of stations with >90% data capture (all stations R/UB/T/I/ndef) ^{1,2)}	Annual average			36 th highest 24 h value		8 th highest 24 h value	
	Number of stations with concentrations C (> 70% data) ³⁾		Maximum concentration (µg/m ³)	Number of stations with concentration C (>70% data)	Maximum 36 th highest 24h concentration (type of station ¹⁾) (µg/m ³)	Number of stations with concentration C (C > 50 µg/m ³)	Maximum 8 th highest 24h concentration (type of station ¹⁾)
	C > 40	24 < C < 40					
Austria 25 (8/12/23/7/0)	2	29	53.6	12	93.8 T	37	116 T
Belgium 20 (1/13/4/5/0)	0	16	36.8	6	59 I	20	105 I
Bulgaria 2 (0/10/0/0/0)	1	1	69.8	1	113.2 UB	2	137 UB
Czech Republic 56 (19/29/6/1/0)	11	29	64.3	30	107 RI	44	183 RI
Denmark 0 (1/0/3/0/0)	0	3	34.2	0	46.3 T	4	67 T
Estonia 1 (0/1/1/1/0)	0	1	29.9	0	47.0 T	1	69 T
Finland 20 (0/5/15/1/0)	0	1	25.0	0	48.1 T	7	75 T
France 105 (8/84/24/19/2)	1	33	43.6	2	60 T	43	75 T
Germany 247 (21/147/77/8/22)	4	135	45.5	40	73.6 NDEF	185	98 T
Greece 10 (0/4/8/2/0)	10	3	68.8	13	105.9 I	13	159 I
Hungary 1 (0/0/1/0/0)	1	0	44.0	1	74.9 T	1	103 T
Iceland 0 (0/0/1/0/0)	0	0		0			
Ireland 1 (0/2/4/0/0)	0	4	28.2	0	47 T	6	83 T
Italy 25 (2/13/28/1/0)	17	20	59.3	28	94.9 T	36	151 T
Latvia 0 (0/0/1/0/0)	0	0	(55.5) (<70%)	(1) (<70%)	(78.1) T	(1)	(120) T
Netherlands 19 (8/6/5/0/0)	0	19	37.0	9	60.9 R	19	88 R
Norway 1 (0/0/1/0/0)	0	0	23.1	0	39.7 T	1	
Poland 19 (4/23/2/3/0)	10	16	71.8	19	113 UB	27	178 UB
Portugal 5 (0/6/3/1/0)	3	6	54.3	8	83 T	9	115 T
Slovakia 5 (0/4/2/0/0)	0	6	39.6	3	58.4 UB	6	81 T
Slovenia 0 (0/2/1/0/0)	0	3	37.4	2	61.4 UB	3	88 UB
Spain 91 (16/13/66/23/8)	32	57	103.3	61	180 I	94	245 I
Sweden 4 (0/2/2/0/0)	1	1	51.1	2	115.4 T	2	208 T
Switzerland 19 (6/6/7/1/0)	0	6	25.5	3	53.7 UB	15	81 UB
United Kingdom 43 (3/45/8/4/0)	0	7	33.4	1	52.2 UB	8	88 UB
All countries 718 (97/417/293/77/32)	93	396	103.3	242	180 I	584	245 I

¹⁾ Station types: R: rural; UB: urban background; T: traffic; I: industrial; NDEF: non-defined

²⁾ Number of stations with more than 90% temporal data coverage. In parenthesis: all stations per type, irrespective of data coverage.

³⁾ Here, all stations with data coverage > 70% are counted

From the maps and Table 4.1 it can be concluded that:

- limit values were exceeded in 2001 (2000) at 242 (120) out of 718 (355) monitoring sites in 19 (14) out of 25 (17) countries which have reported data,

- the highest annual average in 2001 (2000) was 103 (70) $\mu\text{g}/\text{m}^3$ and the highest 36th 24-hour mean value was 180 (110) $\mu\text{g}/\text{m}^3$, both measured in Spain. This station represents a specific industrial hot spot, the second highest Spanish station shows considerably lower PM₁₀ levels in 2001 (annual mean: 78 $\mu\text{g}/\text{m}^3$; highest 36th 24-hour mean value 96 $\mu\text{g}/\text{m}^3$).
- whereas PM₁₀ levels did not exceed the limit values at urban background locations in Northern Europe (Scandinavia), in Western Europe (Ireland, France) and in the Alps (Austria and Switzerland), there were frequent exceedances even in the urban background in Spain, Italy, the Czech Republic and in Poland, and some exceedances in Portugal, Greece, the Netherlands, Belgium, Germany and Slovenia (Map 4.1). (It should be noted that no correction factor had been applied to the data from French stations).
- exceedances are more frequent at traffic exposed and industrial sites than in the urban background (Map 4.2). Even in countries which meet the limit values in the urban background, there were individual exceedances particularly at traffic hot spots (France, Sweden, Austria, Switzerland). The highest number of 24-hour exceedances occurred in Stockholm, probably due to the effects from studded tyres and resuspended street dust (the directive allows the limit values to be exceeded where this can be shown to be due to winter sanding),
- whereas PM₁₀ annual means at rural sites generally are below 30 $\mu\text{g}/\text{m}^3$ and at about half of the rural stations below 20 $\mu\text{g}/\text{m}^3$, they approach or even exceed the limit value in the Czech Republic and in Poland (Silesia). These rural sites near the "Black Triangle" are probably influenced by regional industrial emissions. High rural background levels above 30 $\mu\text{g}/\text{m}^3$ (annual mean) were also measured in parts of the Netherlands and Belgium and at one station in northwestern Spain. It should be noted that the indicative limit value for Stage 2 (annual mean of 20 $\mu\text{g}/\text{m}^3$) was exceeded at about half of the rural stations in 2001 and 2000.

The high rural background PM₁₀ levels in Eastern and Central Europe as well as in and around the Netherlands are also apparent from Figure 4.1. This figure presents the 8 rural stations in Europe (indicated by country name) exceeding the limit value for the 24-hour means (equivalent to the 36th 24-hour average above 50 $\mu\text{g}/\text{m}^3$). It has been mentioned already that the high concentrations near the Polish-Czech border are probably influenced by regional industrial emissions. Remarkably, also one rural station in Switzerland exceeded the short term limit value in 2001.

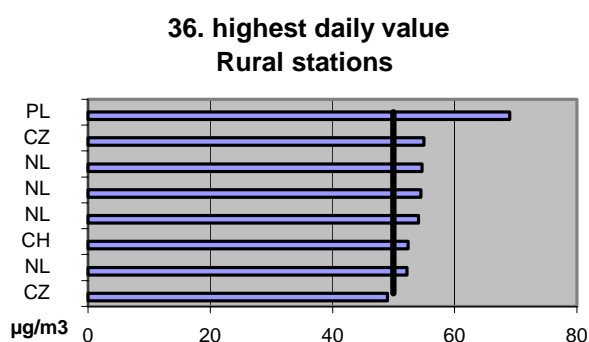
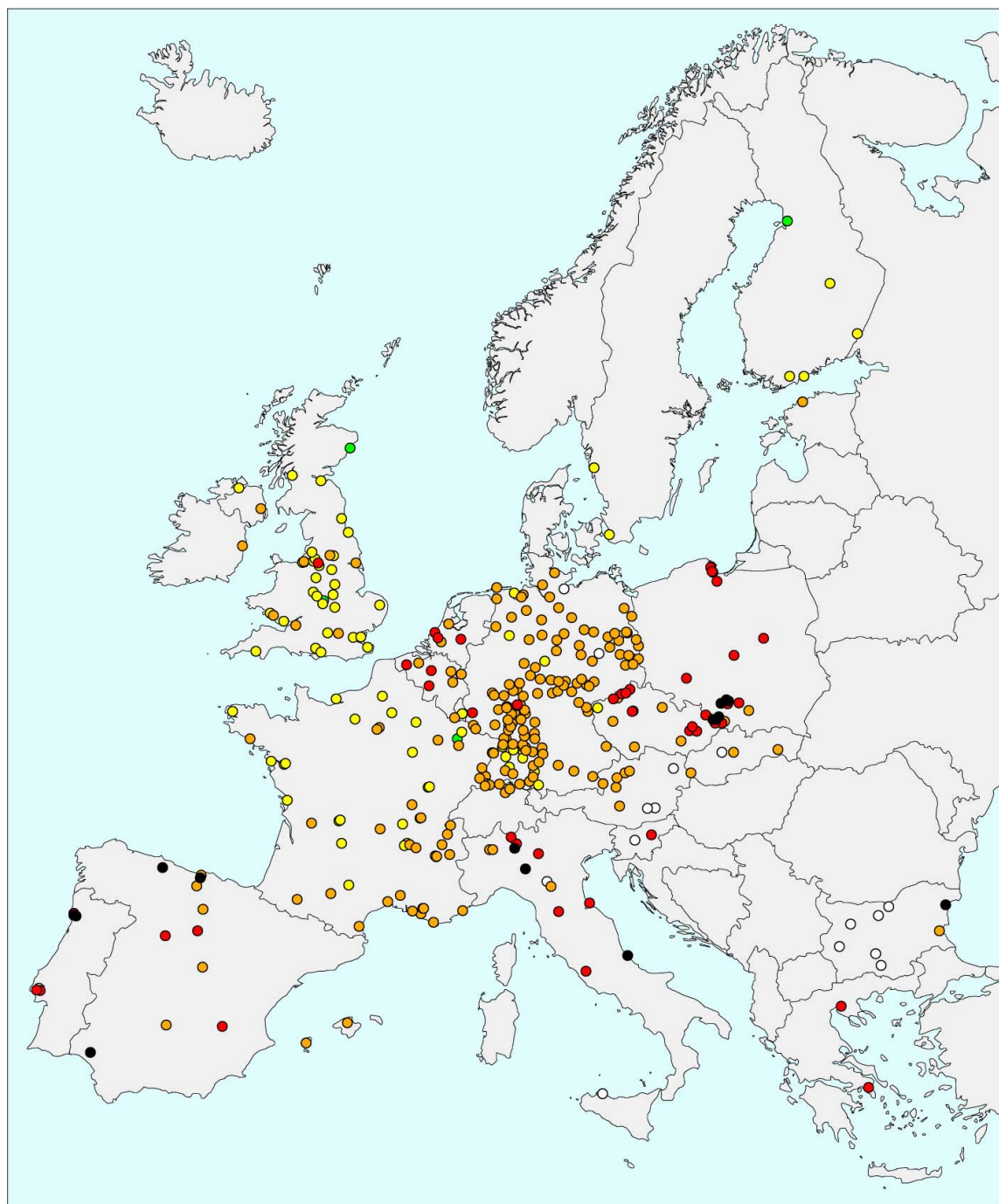


Figure 4.1 Rural background stations (indicated by country name) where the EU AQ limit value for 24-hour averages (36th highest 24-hour value of 50 $\mu\text{g}/\text{m}^3$) was exceeded in 2001. Source: AirBase.

Particulate Matter

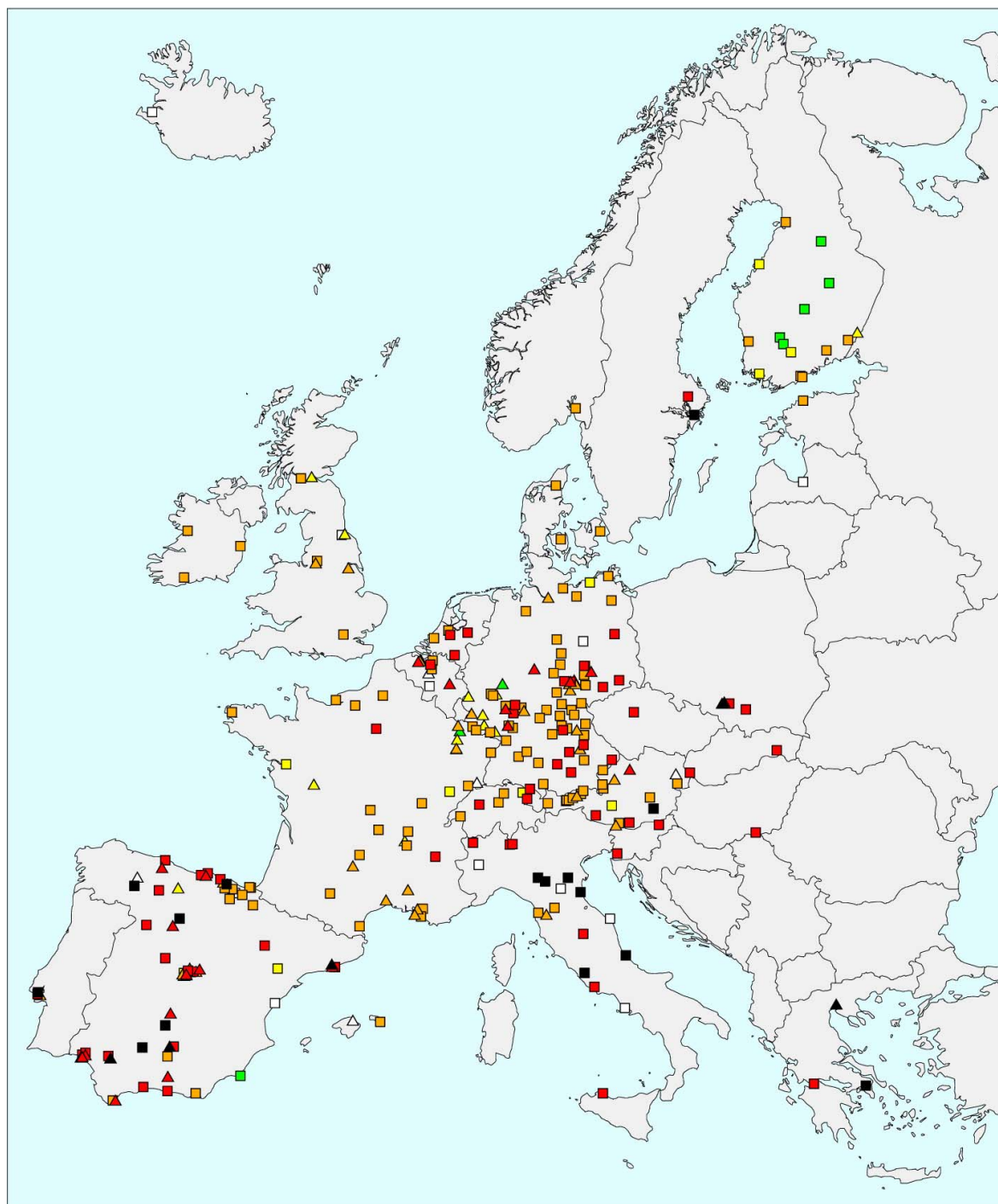


MAX36
Urban Background Stations

- Data Coverage < 70 %
- ≤ LAT
- > LAT and ≤ UAT
- > UAT and ≤ LV
- > LV and ≤ 50 % above LV
- > 50 % above LV

Map 4.1 Measured PM₁₀ levels(36th highest 24-hour mean) in Europe at urban background sites

Particulate Matter

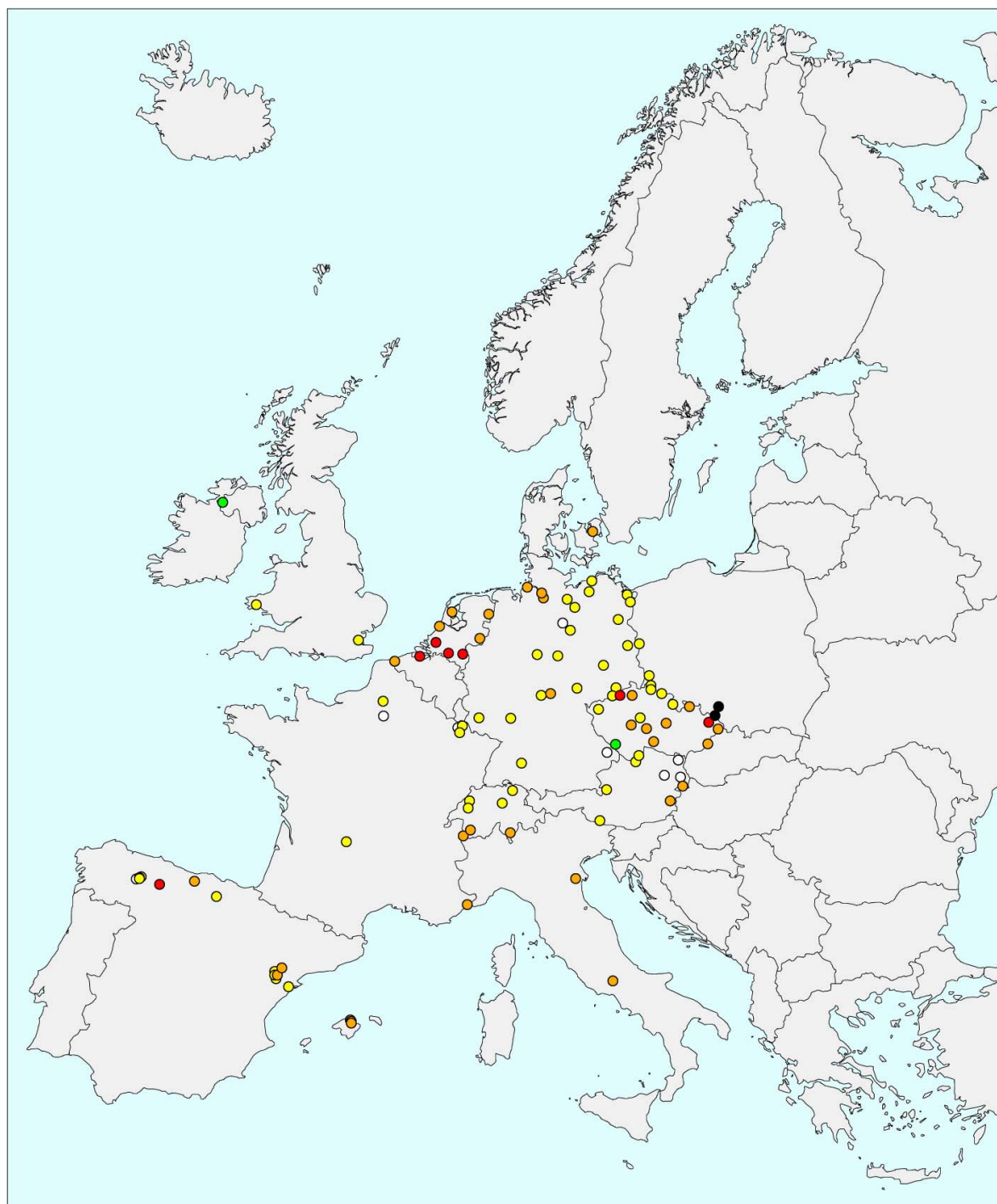


MAX36
Hotspot Stations

- Data Coverage < 70 %
- ≤ LAT
- > LAT and ≤ UAT
- > UAT and ≤ LV
- > LV and ≤ 50 % above LV
- > 50 % above LV

Map 4.2 Measured PM_{10} levels(36th highest 24-hour mean) in Europe at hot spot sites

Particulate Matter



Yearly Average
Rural Stations

- Data Coverage < 70 %
- ≤ 10 µg/m³
- > 10 µg/m³ and ≤ 20 µg/m³
- > 20 µg/m³ and ≤ 30 µg/m³
- > 30 µg/m³ and ≤ 40 µg/m³
- > 40 µg/m³

Map 4.3 Measured PM₁₀ levels (annual average) in Europe at rural sites

The number of European monitoring sites exceeding the EU limit values for 2005 and the indicative limit values for 2010 are presented in Figure 4.2a - 4.2c for the station types rural background, urban background and traffic, respectively (data from 2001). Each square (for Stage 1 limit values) and each rhombus (for Stage 2 indicative values) represents the position of one monitoring station on the surface which is spanned by the annual mean (right axis) and the percentiles corresponding to 35 24-hour exceedances (90.4 percentile) and 8 24-hour exceedances (97.8 percentile) respectively (left axis).

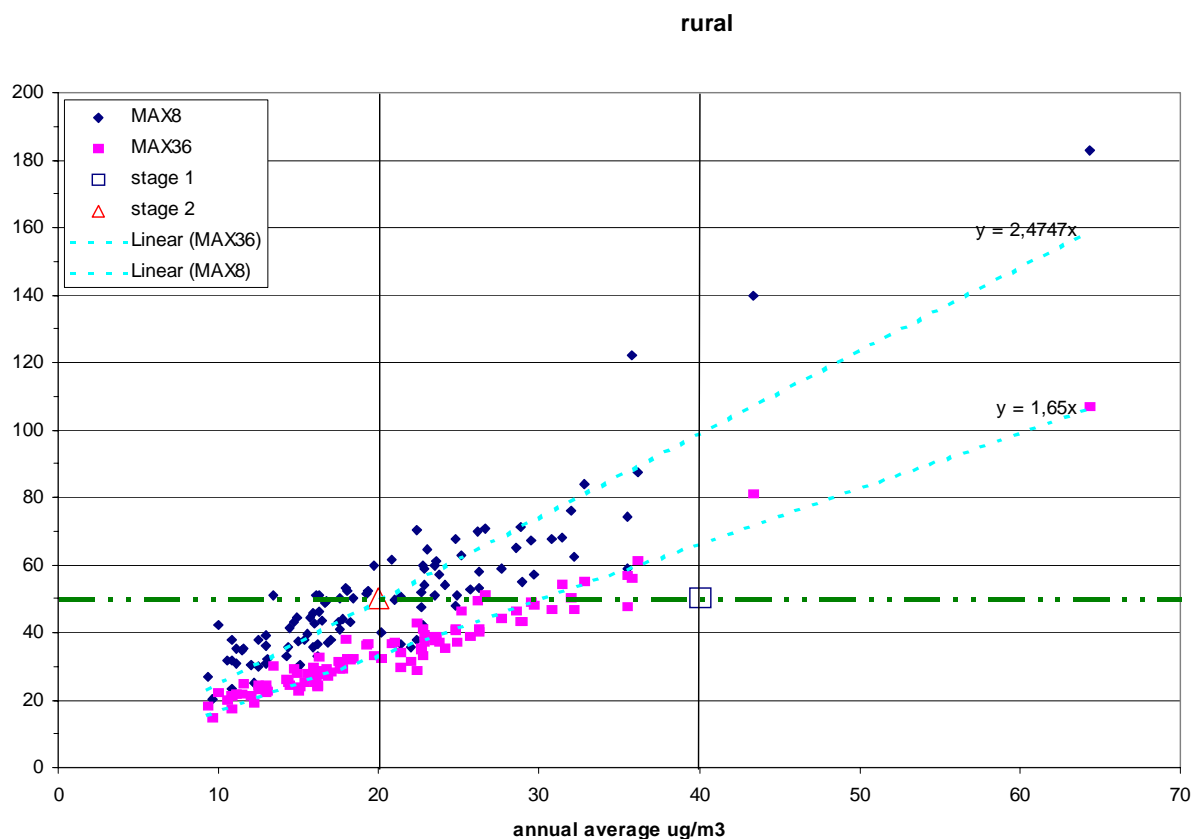


Figure 4.2a Exceedances of annual and short term limit values (Stage 1, squares, and indicative values of Stage 2, rhombus) at European rural monitoring stations for the year 2001. Plotted is the position of each monitoring station on a surface which is spanned by the annual mean (right axis) and the 90.4 and 97.8 percentile, respectively (left axis).

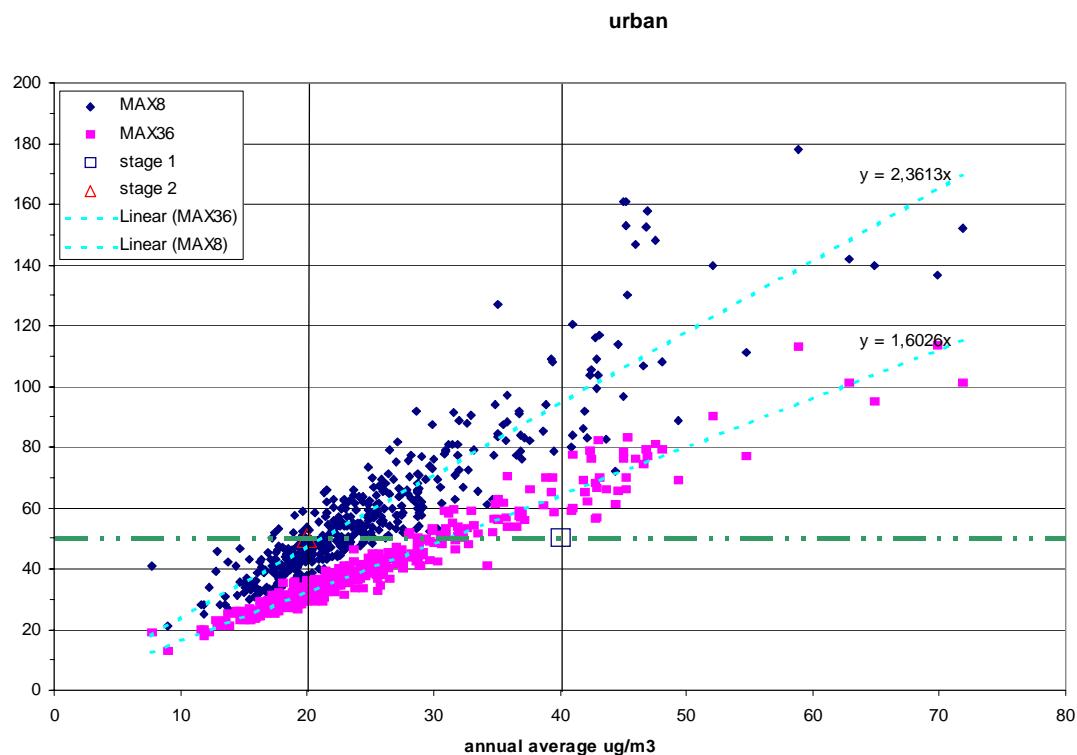


Figure 4.2b Exceedances of annual and short term limit values (Stage 1, squares, and indicative values of Stage 2, rhombus) at European urban background monitoring stations for the year 2001. Plotted is the position of each monitoring station on a surface which is spanned by the annual mean (right axis) and the 90.4 and 97.8 percentile, respectively (left axis).

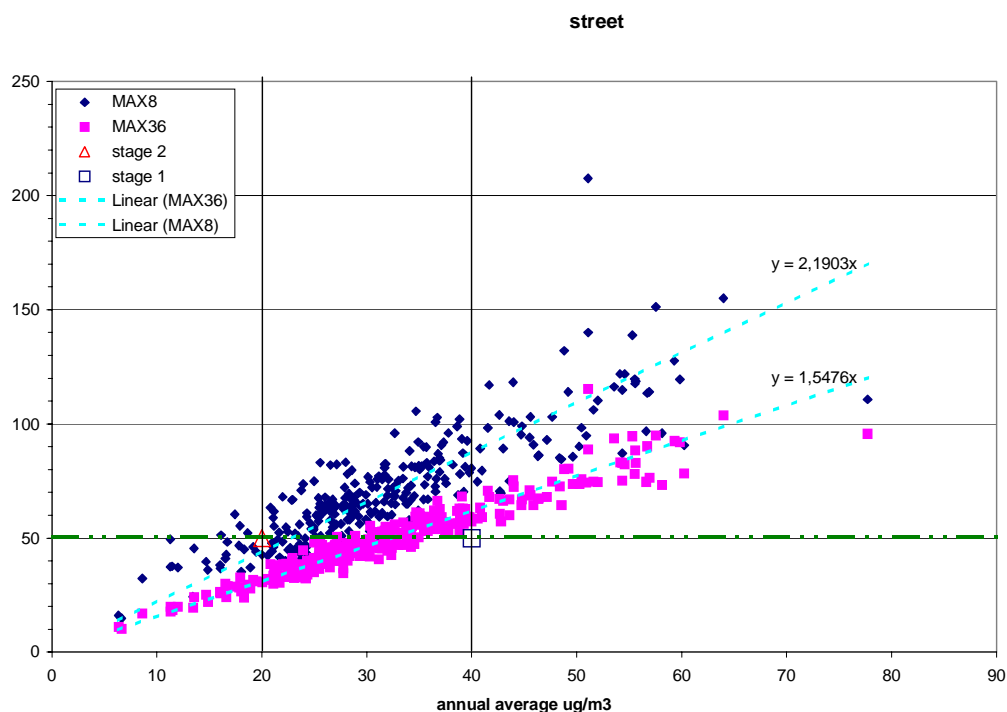


Figure 4.2c Exceedances of annual and short term limit values (Stage 1, squares, and indicative values of Stage 2, rhombus) at European street stations monitoring stations for the year 2001. Plotted is the

position of each monitoring station on a surface which is spanned by the annual mean (right axis) and the 90.4 and 97.8 percentile, respectively (left axis).

It can be concluded that the annual limit value of Stage 1 ($40 \mu\text{g}/\text{m}^3$) is only rarely exceeded at rural background stations. At urban background and traffic stations, this occurs more often (squares and rhombuses to the right hand side of the $40 \mu\text{g}/\text{m}^3$ line). The indicative limit value for the annual mean ($20 \mu\text{g}/\text{m}^3$) is exceeded at about half of the rural stations, and at the majority of urban background and traffic locations.

Exceedances of the limit values for the 24-hour means (squares and rhombuses above the $50 \mu\text{g}/\text{m}^3$ line) are even more frequent than those for the annual means. The regression lines for the limit values of Stage 1 have slopes around 1.6, indicating that the observed ratios of the two limit values (short term and annual) are considerably higher than the ratios calculated from the limit values itself (1.25). This demonstrates that the two limit values are not equivalent to each other, the short term value being stricter. It can be taken from the figures that an annual mean of $40 \mu\text{g}/\text{m}^3$ would be equivalent to a 90.4 percentile (corresponding to 35 days) of $60 - 65 \mu\text{g}/\text{m}^3$.

The indicative limit values of Stage 2 (annual and short term) are exceeded at the majority of stations. The observed ratios from the regression lines (between 2.2 and 2.5) differ little from the ratio of 2.5 which is calculated from the indicative limit values itself, indicating that they are broadly equivalent in their stringency.

The fact that the two limit values of Stage 1 (annual and short term) are not equivalent to each other holds true for all European countries, but has been more closely examined in the Netherlands (Buringh and Opperhuizen, 2002) and the United Kingdom (Laxen, 2003) by correlating PM_{10} yearly averages with number of days exceeding $50 \mu\text{g}/\text{m}^3$. In the Netherlands and in the United Kingdom, the short term limit value (35 days exceeding $50 \mu\text{g}/\text{m}^3$) roughly corresponds to an annual average of $30 \mu\text{g}/\text{m}^3$ (Buringh and Opperhuizen, 2002) (Laxen, 2003), although the correlations differed in detail (linear in the Netherlands, non-linear in the United Kingdom). The present limit value for the annual average ($40 \mu\text{g}/\text{m}^3$) would be roughly equivalent to 80 exceedances of a 24-hour mean of $50 \mu\text{g}/\text{m}^3$.

4.3 Hot spots of the PM_{10} burden

4.3.1 Traffic exposed sites

A comparison of Maps 4.1 and 4.2 suggested already that exceedances of the PM_{10} limit values were more frequent at traffic-exposed and industrial sites than at urban background or rural locations. This observation is corroborated by Figure 4.3, comparing PM_{10} annual means at rural, urban background and traffic sites within individual countries. Generally and as expected, the PM_{10} burden rises fairly substantially from rural via urban background to traffic exposed stations, particularly distinctly in countries such as the Czech Republic, Germany, Italy and the United Kingdom. Data from the Netherlands and Belgium indicate a much smaller difference, caused by an already quite high rural level. This rise in levels from rural to urban to hot spot is less pronounced than it is for purely primary pollutants such as SO_2 , and consistent with the relatively large importance of regional PM_{10} levels arising from secondary (mostly inorganic) particle formation.

Whereas the increase of the PM_{10} burden from rural to hot spot sites is obvious on a per country basis, it does not hold true generally on a European basis. Rural background levels for example in the Netherlands are higher than levels at kerbside stations in countries such as Finland, Norway or the United Kingdom. This underlines the importance of the regional aerosol background levels and has already been observed by an analysis of PM_{10} and $\text{PM}_{2.5}$ levels from 34 European research sites (Putaud et al., 2002).

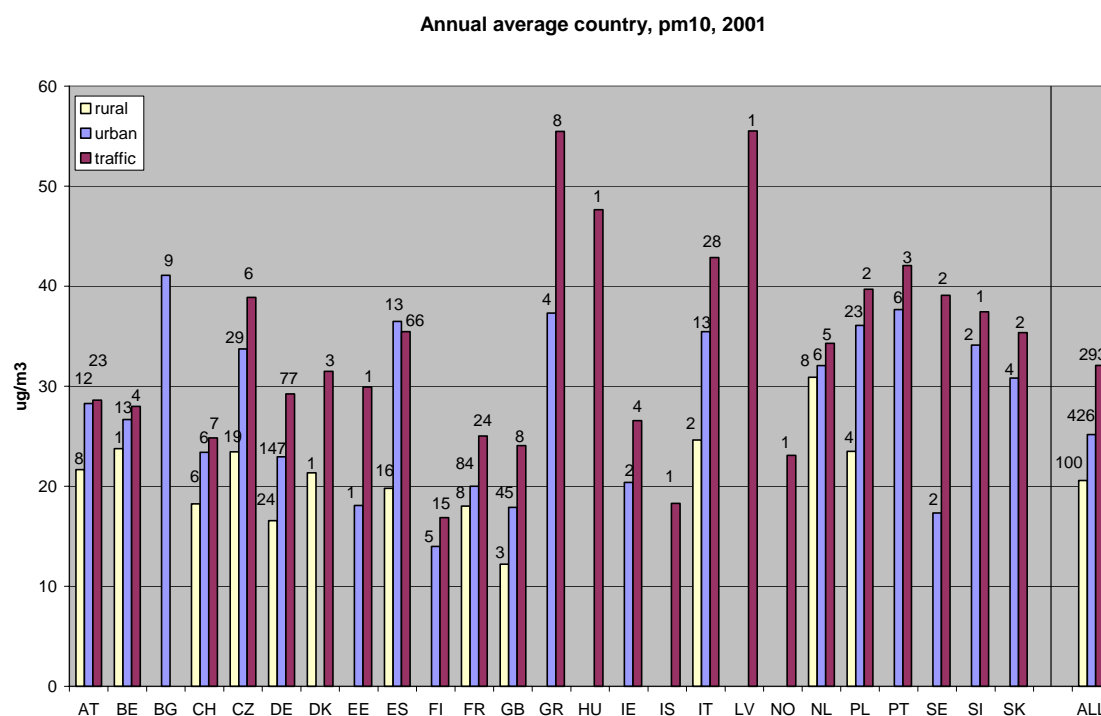


Figure 4.3 Country average PM_{10} concentrations (annual average) for rural, urban background and traffic stations, 2001 AirBase data (number of sites on top of bars)

Results from a more detailed study of Spanish PM_{10} data (1996 - 2000) are presented in Figure 4.4 (Querol et al.; 2003), both for uncorrected data from automatic monitors (Figure 4.4, left) as well as for corrected data (Figure 4.4, right). These data again show a substantial increase in PM_{10} levels from rural to urban background to traffic to intense traffic/industrial sites.

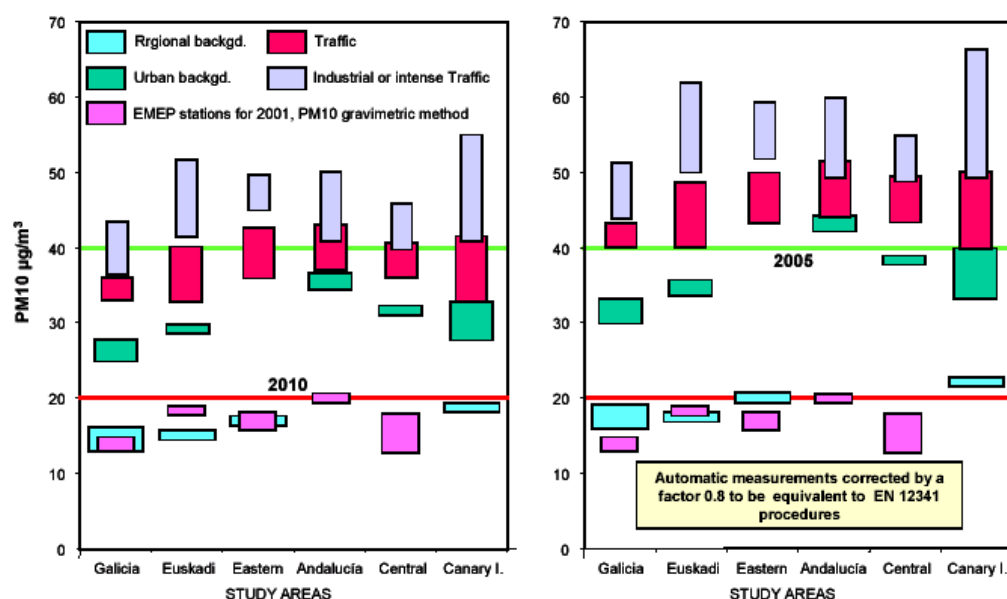


Figure 4.4 Range of annual PM_{10} mean levels at Spanish stations obtained for the 1996-2000 period by means of automatic monitors

In order to identify more clearly the additional PM_{10} burden at traffic exposed sites from AIRBASE and to avoid the ambiguity of comparing hot spot data from one city with background data from another, PM_{10} data (year 2001) were screened for pairs of measurements, where PM_{10} levels were monitored in the same city both at traffic exposed and urban background sites. From these corresponding 89 data pairs, PM_{10} ratios for annual means were calculated. For conurbations where more than one urban background site was available, the average of the sites was taken to represent the urban background. It should be born in mind that information about the distances of urban background locations in the city on the one hand and hot spot stations on the other was not used. They may be in the same area, and they may be fairly widely apart.

A frequency distribution of the ratios is presented in Figure 4.5. Although the ratios of the annual means span a considerable range from 1.9 to 0.7, the vast majority of the ratios are above 1, indicating a higher PM_{10} burden at traffic exposed sites compared to the urban background. The arithmetic mean of the ratios is 1.34 (1.3 in a similar evaluation from 2000 data pairs, $N = 37$) with a standard deviation of ± 0.25 .

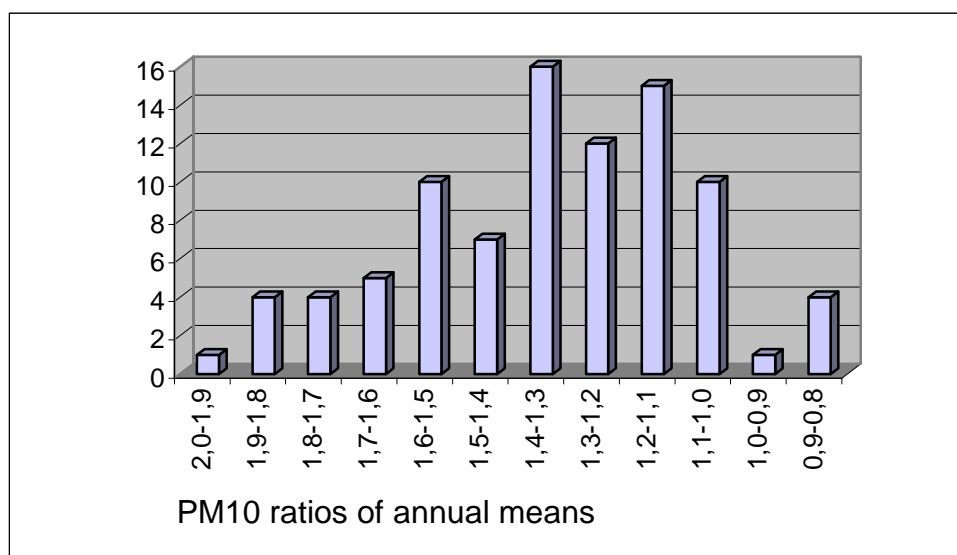


Figure 4.5 Frequency distribution of PM_{10} ratios. PM_{10} (annual means, $\mu g/m^3$) for traffic/urban background sites. Data from Air Base, 2001. Only pairs of data from the same city were taken into account. $N = 89$.

For the short term limit value, an evaluation of extra days of PM_{10} daily means $> 50 \mu g/m^3$ at traffic exposed sites compared with the urban background from data pairs monitored in the same city is presented in Figure 4.6.

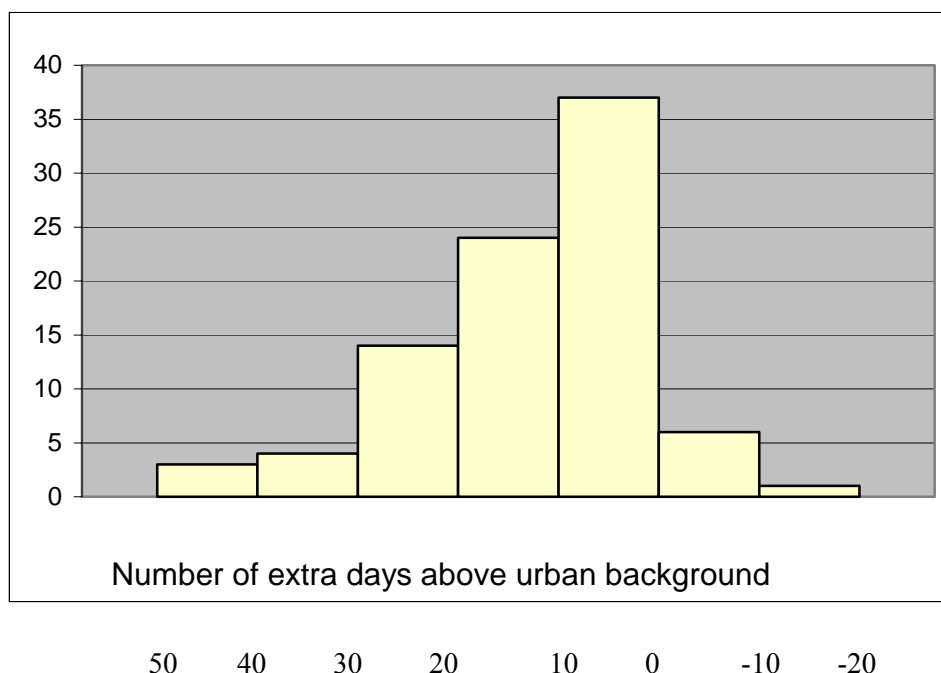


Figure 4.6 Number of extra days PM₁₀ exceeding 50 µg/m³ at traffic exposed sites versus urban background sites. Data from Air Base, 2001. Only pairs of data from the same city were taken into account. N = 89.

In the majority of cases, the number of exceedance days at traffic exposed sites is considerably higher compared with the urban background. On average, there were 12 extra exceedance days (range from -13 to 43 days).

The wide range of the ratios up to 1.9 for the annual means and for the number of extra days in exceedance of 50 µg/m³ is not surprising, bearing in mind

- the somewhat imprecise classification of monitoring stations in AIRBASE provided by the Member States,
- the considerable variation in the amount of traffic passing the hot spot stations and its increase over the urban background. The traffic data at each hot spot station have not been collected.

Sites classified as "traffic" can be situated in a street canyon as well as some 20 or even 50 m apart from major roads or junctions. PM₁₀ concentration gradients fall off rather rapidly with the distance from the kerbside. Data from the United Kingdom (Laxen et al., 2002) indicate that they reach urban background levels after more than 20 m. This compares well with Dutch model calculations of the local contribution of a highway in the Rotterdam area to PM₁₀ levels (Buringh and Opperhuizen, 2002). The local contribution was highest close to the axis of the highway (a surplus annual mean of 7 µg/m³), levelling quickly off to a surplus burden of 1 µg/m³ or less after 100 m distance from the axis of the highway. However, only primary emissions without resuspension were modelled, so that the absolute local contributions close to the highway can be significantly higher. Somewhat larger distances of 50 - 150 m for PM_{2.5} were found in Brisbane (Hitchins et al., 2000). Consequently, the considerable range of ratios probably reflects the different distances of the monitoring sites classified as "traffic" from the kerbside and the different types of streets and local conditions (street canyons versus open settings), as well as the range in amount of traffic at each site. A more detailed analysis would only be possible if more details of the monitoring sites were known.

Table 4.2 shows some examples of well characterized pairs of PM₁₀ measurements in more detail. These data confirm the distinctively higher PM₁₀ levels in street canyons (on average 58% for annual means) in this limited data set close to the kerbside.

Table 4.2 Examples of simultaneously measured pairs of PM_{10} data from well characterized traffic sites and urban background sites in the same conurbation

Station, type	Year	Measurement method	PM_{10} ann. mean ($\mu g/m^3$)	Ratios for ann. means	PM_{10} no. of days > 50 $\mu g/m^3$	Ref.
Düsseldorf, street canyon, 1 m from kerbside, 45.000 veh./day	2001	Grav.	43	2	98	¹⁾
Düsseldorf (urban background)	2001	Grav.	21		13	¹⁾
Berlin, street canyon, near kerbside, 64.000 veh./day	2001	Grav.	42	1.4	51 (β -abs.)	²⁾
Berlin (urban background)	2001	Grav.	29		20 (β -abs.)	²⁾
London, street canyon, 1 m from kerbside, 80.000 veh./day	2000	TEOM (corrected by 1.3)	48	1.7	43	³⁾
London (urban background)	2000	TEOM (corrected by 1.3)	28		0	³⁾
Zürich street canyon, 3 m from kerbside, 31.0000 vehicles/day	2000	Grav.	42	1.8	38	
Zürich urban background	2000	Grav.	24 ($PM_{2.5} = 20.5$)		12	
Bern street canyon, 2m from kerbside, 32.000 vehicles/day	2000	Grav.	39.5 ($PM_{2.5} = 24$)	1.6	51	
Bern urban background	2000	Grav.	25		19	
Basel street canyon, 4m from kerbside 19.000 vehicles/day	2000	Grav.	30	1.2		
Basel urban background	2000	Grav.	25		12	
Madrid street canyon, 3 m from kerbside	2001	Beta-abs.	44	1.4	116	questionnaire
Madrid urban background	2001	Beta-abs.	31		42	questionnaire

¹⁾ Landesumweltamt Essen, 2002

²⁾ Lutz, 2002

³⁾ Laxen and al., 2002

The surplus PM_{10} burden at traffic hot spots is not only due to direct exhaust emissions, but also caused by the resuspension of road dust (mineral dust from the street surface, abrasion of tyres) (Rauterberg-Wulff, 2000; Bächlin et al., 2003). The resuspended part enters predominantly into the coarse fraction $PM_{10} - PM_{2.5}$. However, also considerable higher numbers of ultrafine particles (UFP) have been observed at traffic exposed sites compared to the urban background (see Section 4.3).

It may be argued that these elevated PM_{10} levels in ambient air do not reflect relevant human exposures, because the time spent at these locations is short in comparison to the averaging time of the limit value. However, people living in houses directly adjacent to street canyons with high traffic density will be exposed to such or only slightly lower levels. In addition, hot spots from traffic in a conurbation are not only "dots", but numerous narrow "ribbons" extending for kilometres.

Consequently, a small (the "ribbons" being only 20 - 100 m broad) but significant part of the urban population will be exposed to elevated PM₁₀ levels from traffic well above the urban background.

4.3.2 Industrial hot spots

Many, if not most industrial PM₁₀ sources do not give rise to hot spot situations in ambient air, even at high emission fluxes, as long as their emissions are collected, cleaned according to Best Available Technology and emitted via high stacks (typical example: power plants). On the other hand certain types of industrial installations with significant diffusive dust emissions can have an impact on adjacent residential areas. Typical examples are steel mills, non ferrous metal smelters, open mining, cookeries, cement production, large uncovered stock piles or loading and unloading of dusty goods.

Table 4.3 shows some examples of PM₁₀ concentration levels encountered in the vicinity of industrial hot spots compared to nearly urban background levels.

Table 4.3 *PM₁₀ measurements at industrial hot spots and in the urban background of the same city*

Station, type	Year	Method	PM ₁₀ , ann. mean (µg/m ³)	PM ₁₀ , No. of days > µg/m ³	Ref.
Duisburg, adjacent to cookery (200 m), steel mill (1 km)	2001	Grav.	38	67	¹⁾
Duisburg, adjacent to steel mill (500 m)	2001	Grav.	40	87	¹⁾
Duisburg-Buchholz, urban background	2001	Beta-abs. (1.3)	26	15	¹⁾
Meuse valley, downwind (> 300 m) of various industrial sources incl. fertilizer and gypsum plant, lime kiln	2001	Beta-abs. (1.3)	47	53	²⁾
Meuse valley, Liege (urban background)	2001	Beta-abs. (1.3)	34	8	²⁾

¹⁾ Landesumweltamt Essen, 2002

²⁾ Belgian Questionnaire, 2002

The examples of Table 4.3 show that PM₁₀ levels near certain types of industrial installations can be as high as in street canyons, and can exceed the urban background by nearly a factor of 2. The areas of elevated PM₁₀ concentrations can reach one to several square kilometres. Within this zone, a spatial gradient of PM₁₀ levels occurs, with levels being highest close to the source for installations with low emitting (diffusive) sources generally levelling off to the urban background beyond approximately 1 km. As in the case of the traffic hot spots, only a small but significant part of the European population will be exposed locally. However, the population affected may be significant in number if one bears in mind that several hundreds if not thousands of such installations exist.

4.4 Current levels of other PM metrics (PM_{2.5}, PM_{1.0}, ultrafine particles)

4.4.1 Questionnaire on PM

As only very few PM_{2.5} data and virtually no data on other PM metrics than PM₁₀ are available in AIRBASE, the PM Working Group sent a questionnaire to the Member States and Accession Countries in mid 2002, asking for any available data. In parallel, an investigation was being carried out by the Joint Research Centre, based on a more detailed questionnaire sent to scientists in Europe (Putaud, 2002).

19 countries submitted data in response to the questionnaire of the PM Working Group. Of these, 14 countries supplied data other than PM_{10} , see Table 4.4.

Table 4.4 Responses to the questionnaire on PM metrics other than PM_{10} (number of station years per country and metrics)

	Number of station years					Time trends
	UFP number	UFP size distribution	$PM_{1.0}$	$PM_{2.5}$	PM_{10}	
Austria	0	0	0	1	2	-
Belgium	0	0	0	3	22	-
Czech Republic	0	0	0	0	123	-
Denmark	2	2	0	0	5	
Estonia	0	0	0	0	3	-
Finland	2	2	2	2	2	Reference of publication
France	0	0	0	17	7	-
Germany	9	8	2	19	418	1 site
Hungary	0	0	0	0	8	-
Italy	1	0	0	1	4	3 sites
Luxembourg	0	0	0	1	1	-
Norway	0	0	0	4	4	-
Portugal	0	0	1	3	88	-
Slovakia	0	0	0	0	21	-
Slovenia	0	0	0	0	5	-
Spain	0	0	10	24	24	-
Sweden	1	1	0	12	12	1 site
Switzerland	4	0	0	12	1	-
United Kingdom	2	2	0	20	20	-
Total	21	15	15	119	770	5 sites

It can be concluded that the data base particularly for UFP and $PM_{1.0}$ was limited to a few research stations in respectively 7 and 4 countries, which rendered any detailed analysis of spatial distributions not possible. Also the data set on $PM_{2.5}$ was limited (data from 13 countries). In addition, there were substantial differences between the data sets, which made direct comparison very difficult:

- In some cases the statistics referred to another period than the calendar years 2000 and 2001, as asked for in the questionnaire. The period was then either another year (not always a calendar year), or shorter than a year.
- The levels had been measured with different measuring methods; this is known to cause comparability problems (Chapter 3). Taking $PM_{2.5}$ as example, 8 countries used gravimetry, 6 countries TEOM instruments with correction factors (1.3 or 1.2) and one country TEOM instruments without correction factors. One country did not report on the monitoring method. As the majority of data is based on either gravimetry or TEOM instruments with correction factors based on gravimetry, the $PM_{2.5}$ data are in this respect more or less comparable, though.

In view of the scarcity of the data, it was not attempted to select only data with fully comparable characteristics, because this would yield data sets too small to give results with statistical significance. Thus, all data sets were included in the analysis, in spite of the shortcomings mentioned above. Consequently, uncertainties as to the representativeness of the data should be borne in mind, which may be more important even than the uncertainties in the measuring results as such. As a consequence, the analysis should be regarded as preliminary.

Black smoke levels which can be regarded as a proxy for particles emitted by combustion related sources were not included in the questionnaire.

4.4.2 PM_{2.5} Levels

Annual means

Figure 4.7 shows frequency distributions of PM_{2.5} annual means for the year 2001 at European stations classified as "urban background" (dark columns, at background distance) and as "urban traffic sites" (grey columns in the foreground). The majority of urban background sites (28 out of 38) met a threshold of 20 µg/m³ in 2001, whereas at traffic exposed sites, this threshold was exceeded in 15 out of 23 cases.

These data corroborate the assumption that urban background levels (around 12 - 22 µg/m³ in this data set) are generally lower than levels at traffic exposed sites (around 15 - 30 µg/m³). More significant conclusions, however, should be based on comparable station pairs in the same area. In the PM_{2.5} data base for 2001, there were 5 such pairs, consisting of an urban background and a traffic site in the same city: Grenoble (France), London (United Kingdom), Hannover and Essen (Germany) and Stockholm (Sweden). Here the ratios of PM_{2.5} annual means from traffic and urban background sites were 1.4 on average (between 1.2 to 1.7), indicating that the additional burden of PM_{2.5} at traffic exposed sites (around 40%) is comparable to the additional PM₁₀ burden discussed in Section 4.3.1. Apparently, the smaller share of resuspended dust in the PM_{2.5} fraction compared with the PM₁₀ fraction is compensated by the exhaust emissions, where fine particles prevail.

In measurements at three other sites (Munich, Stockholm and the Netherlands (Hoek et al., 2002), a somewhat smaller additional burden of 18% on average at traffic exposed sites was found compared with the urban background, but again the exact locations of the monitoring sites and important meta data such as traffic density or the geometry of the streets are not known.

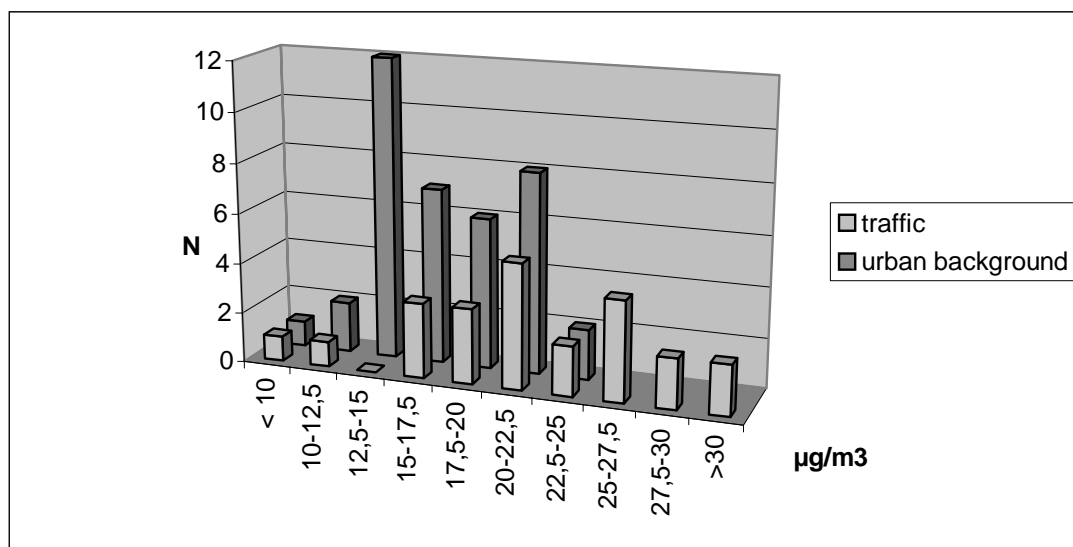


Figure 4.7 Frequency distributions of $PM_{2.5}$ annual means (2001) at European urban background stations (dark columns behind) and at European traffic sites (grey columns in front)

In Figure 4.8, average annual $PM_{2.5}$ concentrations (2001) at the three most important station types rural background (RB), urban background (UB) and urban traffic (UT) are plotted per country. Rural background concentrations seem to be on average quite uniform in Europe (between 11 and 13 $\mu\text{g}/\text{m}^3$) and considerably lower than urban background levels (around 15 - 20 $\mu\text{g}/\text{m}^3$), which in turn are lower than $PM_{2.5}$ annual averages at traffic exposed sites (typical range from 20 to 30 $\mu\text{g}/\text{m}^3$). However, in interpreting these figures the limited data set has to be borne in mind. Generally, the averages of the station types are based only on several cases, for some countries even on single stations. Consequently, spatial differences of $PM_{2.5}$ levels in Europe are not apparent from this limited data set, perhaps with the exception of a tendency for slightly lower $PM_{2.5}$ annual means in Scandinavian cities. Between individual sites, however, the $PM_{2.5}$ burden can differ considerably, ranging from 8.3 $\mu\text{g}/\text{m}^3$ (urban background, Umeå, Sweden) to 32.7 (traffic, Vienna) in this data set. An even greater variation (more than a factor of 10, compared with a factor of 4 here) has been observed at European research stations (Putaud et al., 2002).

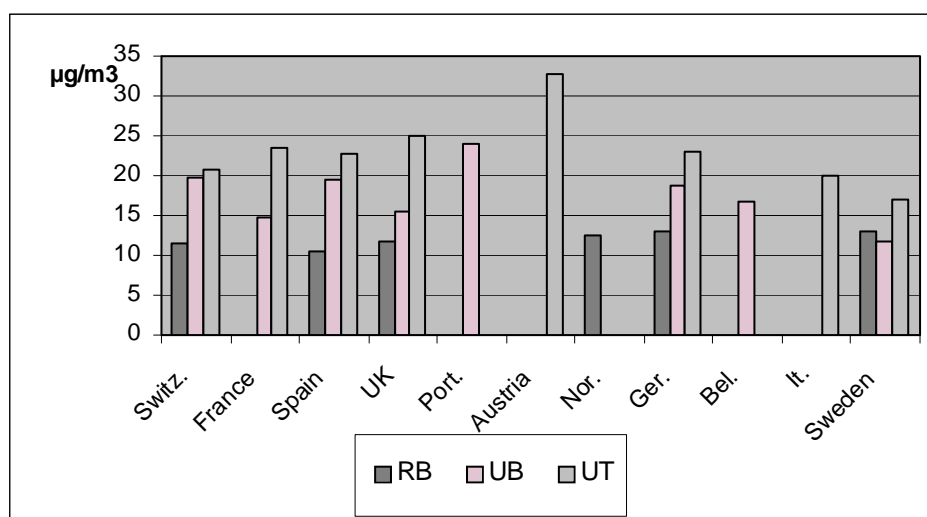


Figure 4.8 Annual $PM_{2.5}$ concentrations (2001, country-averages) for rural background (RB), urban background (UB) and urban traffic (UT) stations. Data from questionnaire on PM.

As only two industrial stations were contained in the data set, this station type could not be plotted in Figure 4.8. $PM_{2.5}$ concentrations at these two industrial sites were quite low.

90 percentile

Unfortunately, the 90 percentile was not asked for in the questionnaire, and from the statistics reported (annual arithmetic mean, 98 percentile and maximum value) it was not possible to estimate it accurately from an assumed frequency distribution (such as lognormal). However, there were 26 stations for which the frequency distributions were either known (5 stations in Germany) or the 90 percentile had been reported (Portugal: 1 station; Sweden: 14 stations; Spain: 6 stations). Correlating annual means with 90 percentiles from this data set in Figure 4.9 gives a straight line in good approximation ($r^2 = 0.91$). From Figure 4.9 it can be concluded that an annual mean of 20 $\mu\text{g}/\text{m}^3$ corresponds on average to a 90 percentile of 35 $\mu\text{g}/\text{m}^3$ (an annual mean of 15 $\mu\text{g}/\text{m}^3$ would be on average equivalent to a 90 percentile of 25 $\mu\text{g}/\text{m}^3$).

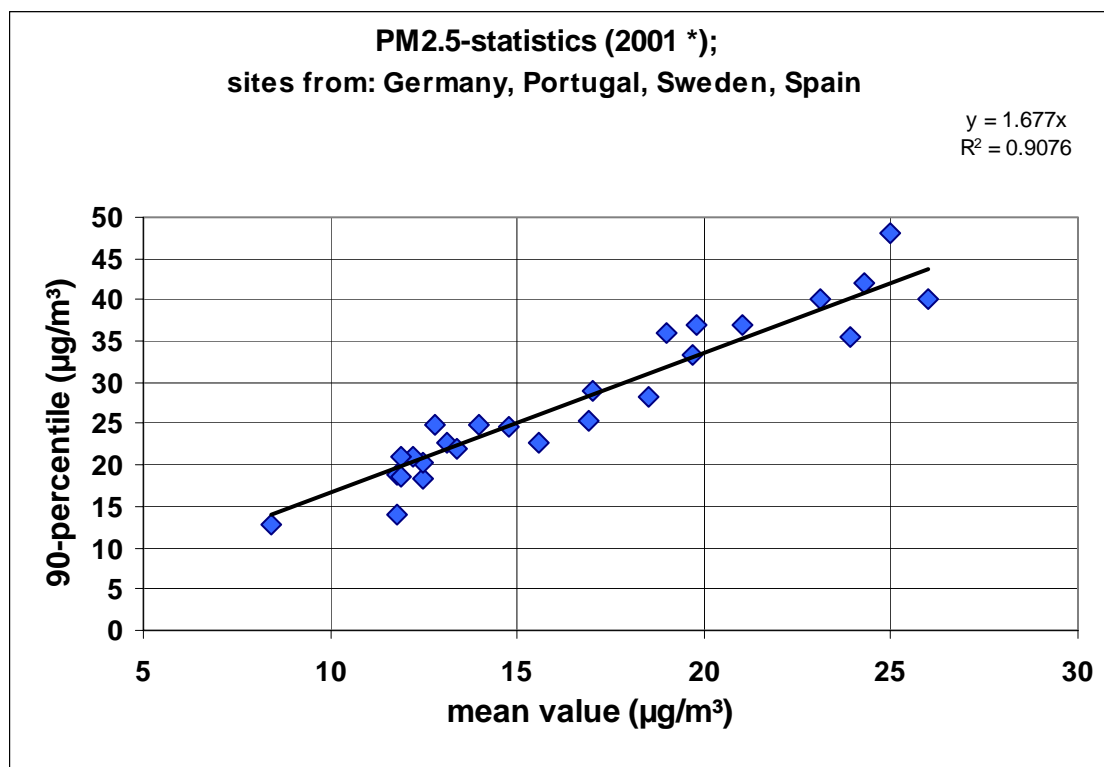


Figure 4.9 *PM_{2.5} statistics at 26 sites in Sweden, Germany, Portugal and Spain (German, Spanish and Portuguese data are from 2001, the Swedish data are from Sep. 99 - Aug. 01)*

98 percentile and maximum of 24-hour means

As should be expected, the range of PM_{2.5} 98 percentiles at European stations (17 - 121 µg/m³) is considerably broader than the range for annual means; it can also be seen that 24-hour means above 50 µg/m³ are no rare events at several stations (98 percentile above 50 µg/m³ at 19 out of 72 sites). Referring to daily maxima, a threshold of 50 µg/m³ was exceeded at the majority of stations (41 out of 72). On the other hand, an evaluation of the 98 percentile in relation to the station type shows a picture similar to that for the annual mean, suggesting that the ratio 98 percentile versus annual mean does not strongly depend on the station type, and consequently the dependence of PM_{2.5} levels on station type seems similar for the mean and high percentiles. This can also be concluded from the correlation of annual means and 90 percentiles in Figure 4.9.

PM_{2.5}/PM₁₀ Ratio

The ratios PM_{2.5}/PM₁₀ for measurements at the same station and in the same time period were reported as such by the countries in the questionnaire. Considering only data from 2001, the PM_{2.5}/PM₁₀ ratio centres around an arithmetic mean of 0.65, with a remarkably low standard deviation (range from 0.42 to 0.82, standard deviation $s = \pm 0.09$, $N = 72$). A slightly larger ratio of 0.73 ± 0.15 with a similar range from 0.57 to 0.85 has been found by Putaud et al. (Putaud et al., 2002). The PM_{2.5}/PM₁₀ ratios are quite uniform at the majority of European stations, with only slight tendencies towards somewhat higher ratios at rural background sites compared with urban traffic sites. The questionnaire data did not allow to draw conclusions on systematic differences between station types. Putaud et al. observed somewhat lower ratios for kerbside sites, suggesting large contributions of resuspended road dust to the coarse fraction.

Stations in Mediterranean countries (Spain and Portugal) did not generally show clearly different ratios compared to the other stations in Europe with the exception of some stations on the Canary Islands and Southern Spain with ratios close to 0.5 (significant contributions of natural sources in the coarse mode). The same can be said about the few data from the Nordic countries.

4.4.3 PM_{1.0} levels

The PM_{1.0} data set in the questionnaire of the PM Working Group was too small (15 station years) to detect statistically significant differences between station types (see Figure 4.10), although urban traffic sites tended to show somewhat higher PM_{1.0} levels than urban background sites. Regional differences across Europe were not significant either, particularly because twelve of the sixteen station-years came from the Iberian Peninsula.

The annual means ranged between 6 (Helsinki) and 21 µg/m³ (Llodio in Spain), but it should be noted that there are no rural background data. The ratio PM_{1.0}/PM_{2.5} varied around an average of 0.73, ranging between 0.48 and 0.87, with a standard deviation of ± 0.11. The ratio PM_{1.0}/PM₁₀ centred around 0.49 with a somewhat larger standard deviation of ± 0.16 (range from 0.21 to 0.68). The ratio 98-percentile/annual means varied around 3, again without a clear dependence on the site location. The maximum of daily means exceeded 50 µg/m³ at the majority of stations (highest value: 147 µg/m³).

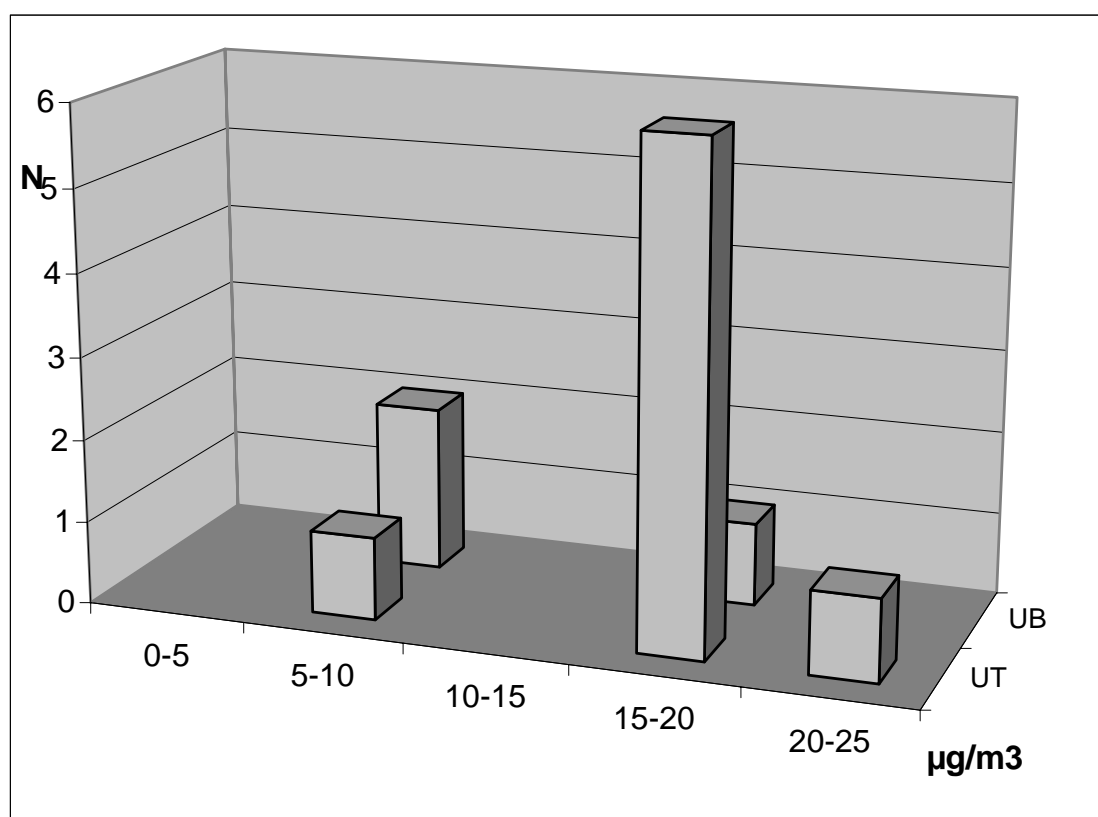


Figure 4.10 Frequency distribution of European PM_{1.0} annual means at urban traffic sites (UT, in front) and urban background sites (UB, behind). Most data stem from 2001 with the exception of Lisbon (2002) and Helsinki (1998/99)

4.4.4 Ultrafine particles

Number of ultrafine particles

Most of the UFP data received in the questionnaire of the PM Working Group were from urban traffic stations (7, with 17 station years in total); there were three urban background stations and three rural background station (from the United Kingdom, Sweden and Switzerland). The levels behaved as expected: lowest at rural background (annual mean 4000 - 10 000 particles per cm³), highest at traffic

stations (annual mean 10 000 - 80 000 particles per cm^3) (see Figure 4.11). The range was over an order of magnitude, so considerably larger than for PM mass concentrations, which is consistent with the picture that there are near sources relatively many very small particles with a limited lifetime. As a consequence, the ratio of particle number and mass concentration tended to decrease with distance to sources: at rural sites this ratio was considerably lower than at traffic stations both for PM_{10} and $\text{PM}_{2.5}$ (Figure 4.11). These findings are consistent with those of Putaud et al. (2002), who also found in their data that the particles < 100 nm consist at polluted sites mainly of carbonaceous material (black carbon and organic matter).

The frequency distribution of hourly means, characterised by the ratio 98 percentile/mean (see Figure 4.11), did not differ much from that of mass concentration metrics: the ratio was typically 2.8 (range 2.2-3.8). The steepness of the distribution did not visibly depend on the average level or on the station type.

Size distribution of ultrafine particles

As the size classes of the various data sets were different, a simple overview of the size class distribution could not be made. By calculating the total number of particles smaller than certain sizes, a cumulative graph was made (see Figure 4.12). As in some cases also the number of particles < 10 nm was not given, the data were not very well comparable. The fraction of very small particles found was higher close to sources: below 50 nm, 49% of the particles were found at the remote background station, while at the two urban background stations this size fraction accounted for about 63% and at street stations 63 - 79%.

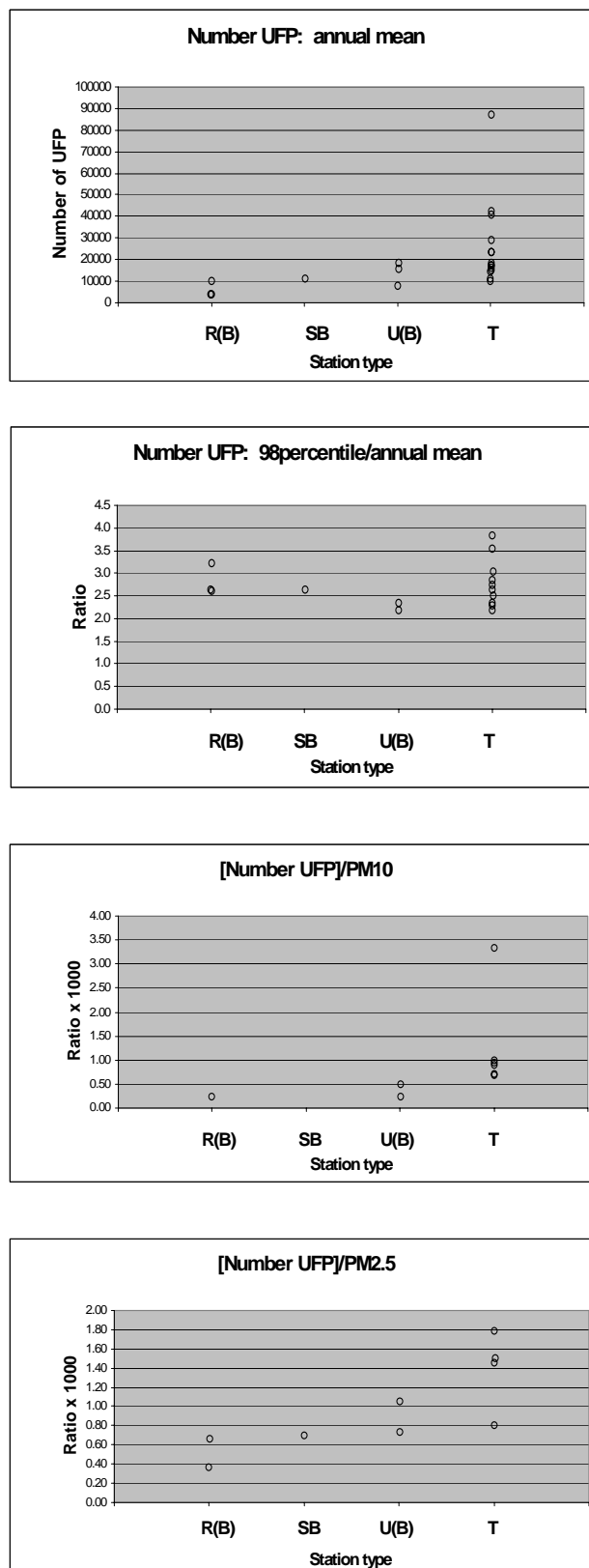


Figure 4.11 Dependence of UFP (number of particles per cm^3) on station type: annual mean; ratio of 98 percentile to annual mean; $\text{UFP}/\text{PM}_{10}$; $\text{UFP}/\text{PM}_{2.5}$. Station types: R(B): rural background; SB: suburban background; U(B): urban background; T: traffic.

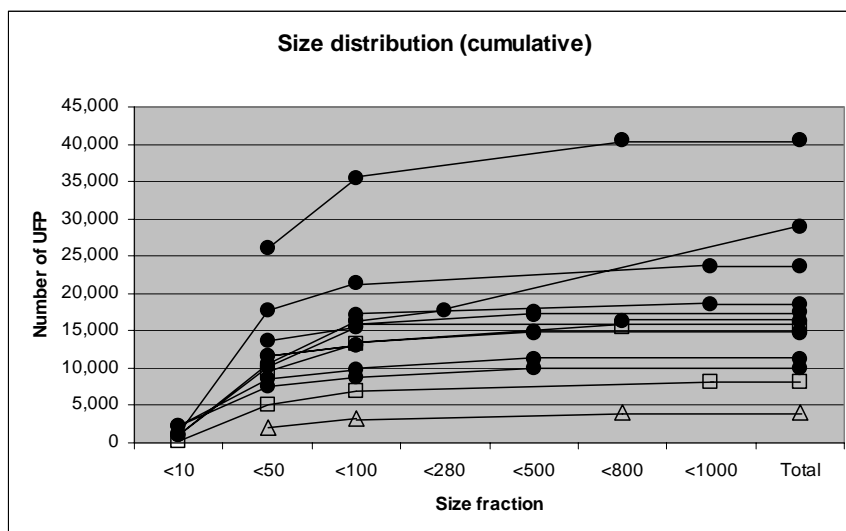


Figure 4.12 Cumulative UFP size distribution. Closed circles: traffic stations; open squares: urban background stations; open triangles: rural background stations.

4.5 Exposure

As is the case for other air pollutants, the total exposure to suspended particulate matter (of whatever size) of an individual is the result of contributions from the two microenvironments, outdoor air and indoor air. The indoor air compartment can be subdivided further into homes, restaurants, passenger cabins of cars, busses and aircrafts, workplaces etc. Consequently, in studies to detect and quantify the health effects of particles attention must be paid that exposure is characterised adequately.

Generally speaking, there are two different ways to obtain such characterization. One is by *measuring* total air exposure using personal sampling: the persons under study are provided with a personal sampler each which they have to carry on them or position as close to them as possible over the entire 24 h of the day. Since this is cumbersome for a study participant, the following alternative can be used: total exposure is *modelled* taking into account the time spent in the various microenvironments (indoors and outdoors) and the concentrations observed in these microenvironments.

Personal sampling provides a concentration level that represents integration over the concentration levels in all compartments visited by the study subject during the 24-h (or longer) measurement period and, thus, cannot detect the individual contribution of any compartment. In contrast, the modelling process using the combination of the pollutant concentrations in the different microenvironments and the time spent therein permits to assess the contribution to total exposure of each of these microenvironments. This kind of source apportionment can be of great help to decide what measures should be given preference in controlling pollutant concentrations.

Williams et al. (2000a) performed personal, indoor and outdoor measurements on 21 elderly persons living in a retirement facility in Baltimore. The study subjects spent about 95% of their time indoors. Mean $PM_{2.5}$ levels were 13.0, 10.0, 9.4, and 22.0 $\mu g/m^3$ for personal sampling, inside apartment sampling, central site in the building, and outdoor site (11 km from the building), respectively. The difference between personal and indoor concentrations is explained by what is called a “personal cloud” surrounding the individual. This is supported by findings of Luoma and Batterman (2001) who concluded that occupants' activities increased the concentration of particles of 5-25 μm by up to 10 $\mu g/m^3$ in the vicinity of the activity.

The influence of particle size on the relationship between personal, indoor and outdoor concentrations should not be underestimated. This influence may be one of the reasons why the personal/outdoor ratio as reported for PM_{2.5} by Williams (2000a) is <1 while the respective ratio for PM₁₀ was found to be >1 by Janssen et al. (1998). In their study on 37 adults, these authors found a mean outdoor PM₁₀ concentration of 42 µg/m³ whereas the mean personal concentration was 62 µg/m³. The median Pearson's R in this latter case was 0.50, and 0.71 if days with exposure to tobacco smoke were excluded. In an earlier study with PM_{3.5} samplers Spengler et al. (1985) had found much lower correlations between personal and ambient sampling. Correlation coefficients depended on the smoking status of the study participant, but did not exceed 0.28 in any case.

The results of Williams et al. (2000a and 2000b), together with those of others (e.g., Lioy et al., 1990), support the evidence that for an individual, indoor and personal PM concentrations can differ substantially from those measured at fixed sites outdoors although on a population average, there may be good correlations, e.g., $r = 0.89$ in the Baltimore study (Williams et al., 2000b). In a population-based PM_{2.5} exposure study on carried out in Toronto Pellizzari et al. (1999), 3-day average personal/residential outdoor and personal/ambient fixed site correlations were low ($r = 0.19-0.27$). Data for Helsinki obtained in the EXPOLIS study showed that PM_{2.5} concentrations measured at ambient fixed site monitors were poor predictors of personal exposures to PM_{2.5} (Kousa et al., 2002). WHO, in its 2003 report "Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide" (<http://www.euro.who.int/document/e79097.pdf>), concludes that "on a population level, personal PM of ambient origin 'tracks' ambient PM over time, thus measurements of PM in ambient air can serve as a reasonable 'proxy' for personal exposure in time-series studies".

In a recent publication on exposure to PM_{2.5} which describes the results of a model approach (Burke et al., 2001), it has been stated that the microenvironment "indoor-residential" had the greatest influence on total exposure to PM_{2.5}, compared to the other microenvironments considered, namely outdoor and non-residential indoor (office, school, store, restaurant, bar, in-vehicle). The outdoor compartment as such was found to make a *direct* contribution of around 5% on average. Another 35% was due to an *indirect* contribution via infiltration of outdoor air into indoor spaces. Thus, about 60% of the total exposure to PM_{2.5} could not be influenced by control measures taken to reduce outdoor air PM_{2.5} levels. For non-smoking homes only, the calculated ratio was about 50% : 50%.

In assessing exposure to indoor suspended particulate matter and the possible adverse health effects associated with such exposure, it should be kept in mind that the physical characteristics of suspended particulate matter encountered indoors differ from those present outdoors. In a recent review of the scientific evidence on associations between exposure to particles in buildings and health effects which was performed by an international group of experts (Schneider et al., 2003), only two cross-sectional studies and one longitudinal population qualified for in-depth review. However, for all three studies non-differential misclassification of exposure was likely to have played a role. In the longitudinal study, which addressed cardiovascular effects of PM_{2.5} on 26 elderly persons (Liao et al., 1999), in addition confounding by other environmental factors cannot be excluded, and no definite conclusion could therefore be drawn regarding the role of indoor PM_{2.5} on cardiovascular effects.

4.6 Conclusions and recommendations

4.6.1 Conclusions

PM levels

Compared with the limit values, PM₁₀ levels in parts of Europe are rather high. In 2001, the limit values for 2005 were exceeded at 242 out of 718 monitoring stations (34%) in 19 out of 25 countries which reported data. The short term limit value for 24-hour means was more frequently exceeded than

the limit value for the annual mean. These two limit values are not equivalent, the present short-term limit value roughly corresponding to an annual mean of 30 (instead of 40) $\mu\text{g}/\text{m}^3$, and the present annual limit value of 40 $\mu\text{g}/\text{m}^3$ roughly corresponding to 80 exceedances (instead of 35) of the 24-hour mean of 50 $\mu\text{g}/\text{m}^3$.

The indicative limit value (Stage 2) of 20 $\mu\text{g}/\text{m}^3$ for the annual average was exceeded at half of the rural background stations.

The available data for the other PM metrics ($\text{PM}_{2.5}$, $\text{PM}_{1.0}$ and number of ultrafine particles (UFP)) are still too scarce to draw firm conclusions. It is to be expected that the spatial distribution of $\text{PM}_{2.5}$ and $\text{PM}_{1.0}$ tends to be more homogeneous than for PM_{10} , the mean $\text{PM}_{2.5}/\text{PM}_{10}$ ratio being approx. 0.65 (with a range from 0.4 to 0.8). Whereas quite homogenous rural and urban background levels for $\text{PM}_{2.5}$ in Europe correspond to this expectation, the ratios between $\text{PM}_{2.5}$ levels at different types of stations (rural background, urban background and urban traffic) seem to be similar to the corresponding PM_{10} concentration ratios. An annual mean of 20 $\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ is met by the majority of urban background locations, but is exceeded at most urban traffic sites. Numbers of ultrafine particles span a range of one order of magnitude (from thousands/ cm^3 at background sites to ten thousands/ cm^3 near sources).

Regional differences

Urban PM_{10} levels indicate a spatial variation in Europe. The data reported to AIRBASE show highest concentrations in Southern, Central and Eastern Europe (some of the Acceding countries), moderately high levels in the densely populated parts of Central and Western Europe, already lower levels in northwestern Europe (United Kingdom, Ireland) and the Alpine regions and the lowest levels in Scandinavia, the annual averages from comparable types of areas spanning over a factor of 3. However, it must be taken into account that some of this spatial variation may be due to the different siting of monitoring stations in Member States (e.g. more hot spot stations in Spain compared to some other countries). Urban $\text{PM}_{2.5}$ levels show less variation in Europe, with a tendency to somewhat lower concentrations in Scandinavian cities.

Not only the urban concentrations, but also the rural background concentrations differ for PM_{10} , being particularly elevated in some of the eastern Acceding countries, but also in the Netherlands, Belgium and parts of northwest Germany. In Southern Member States, natural sources raise the rural background to an extent that it becomes comparable to background levels in Central Europe.

The rural background for $\text{PM}_{2.5}$ seems to be more uniform in Europe.

The spatial variations indicated above need to be confirmed based upon an analysis of the spatial representativeness of the stations in each of the areas and scales in Europe.

Hot spots

Typical hot spot areas with elevated levels of PM are traffic exposed sites and areas near certain industrial sources with fugitive dust emissions.

For the different PM metrics, the amount of the surplus burden at traffic exposed sites decreases in the following order: number of UFP > $\text{PM}_{10} \approx \text{PM}_{2.5}$. The surplus PM_{10} burden (annual means) centres around 35% of the urban background with a considerable range (up to about 100%), depending on traffic density, location (street canyon versus more open settings), distance of the monitoring station from the kerbside and urban background levels. A smaller data set with well characterized sites showed a PM_{10} surplus burden of 60%. $\text{PM}_{2.5}$ concentrations at traffic sites were on average about 40% higher than in the urban background, based on a limited number of data (range from 70% to 20%). The "ribbons" of elevated PM_{10} and $\text{PM}_{2.5}$ levels along busy streets are fairly narrow, reaching the urban background after a distance of 20 - 100 m (or more, at the most densely trafficked streets

and roads) from the kerbside. A significant, if not the greater part of the PM_{10} surplus burden near streets is due to resuspension, which predominantly adds to the coarse mode $PM_{2.5-10}$. High numbers of UFP (up to 80 000 particles/cm³ annual mean) have been measured in street canyons.

Industrial sources without a significant share of fugitive emissions and applying Best Available Technology for their stack emissions usually do not give rise to elevated PM_{10} and $PM_{2.5}$ levels in their surroundings. However, areas around industrial sources with high fugitive dust emissions can have elevated PM_{10} levels comparable to those near busy streets. PM_{10} concentrations are highest near the source and level off quickly, reaching urban background concentrations after approximately 1 km. The affected area can range from one to several km².

Uncertainty

The uncertainty associated with measurements using the reference method at a single station is fairly small, when an efficient quality control and assurance (QA/QC) system is in place at the responsible laboratory. The measurement uncertainty should then be smaller than $\pm 15\%$. When an automatic method is used, the possible off-set relative to the reference method, requiring a correction factor to be used, adds to this uncertainty. This offset results in annual means that are generally 0 - 30% lower than the concentrations obtained using the reference method, depending on the types of instruments, and the applied correction factors is often uncertain. For daily means, the uncertainty can be higher than 30%.

When using measurements at fixed stations to make an assessment of typical, or average, or maximum PM levels in a city or an area of the city, the issues of representative station locations and of spatial coverage of the area play an important role. The added uncertainty of an average PM concentration, derived from the concentrations measured at the stations in the area, depends naturally upon the number of stations and their locations and how well they represent the area. A general assessment of this added uncertainty cannot be given, but it may well range from as low as 10% (good spatial coverage and well placed network) to as high as 100% (few stations to represent a large inhomogeneous area). Guidance should be given to assess the spatial representativity of monitoring stations.

In view of these limitations, many of which cannot be quantified, the overview above must be interpreted with caution.

Exposure

As European citizens spend most of their time indoors, including compartments such as houses, workplaces and the interior of commuting vehicles, personal exposure is only partly and indirectly dependent on outdoor PM levels.

The penetration of fine particles into houses was found on average to be about 70% but spanning a wide range depending on the status of the buildings (e.g. air conditioned or not), the climate and the season (e.g. open windows in summer). In addition, indoor sources such as tobacco smoke can raise the personal exposure considerably. In first results of the EXPOLIS study it was shown that the personal exposure to traffic $PM_{2.5}$ was nearly twice as high compared with ambient air due to direct exposure during commuting. Nevertheless on a population average the concentrations measured in ambient air at fixed stations can be regarded as a reasonable approximation to characterise the health risks of those PM metrics that show a relatively smooth spatial distribution (like $PM_{2.5}$).

4.6.2 Recommendations

1. In view of the emphasis on PM_{2.5} in the new WHO recommendations, PM_{2.5} monitoring should be intensified as soon as possible to give representative data from Europe, covering all relevant area types.
2. There is limited information on other PM metrics or parameters related to PM, such as chemical composition including elemental carbon, PM_{1.0}, and the number concentration of ultrafine particles. It is recommended to set up some 20 monitoring stations throughout the EU where the different parameters are being monitored on a longer term. These stations should be set up and operated in a harmonized way to ensure comparability of data, and cover the relevant types of stations, e.g., rural, urban background and traffic sites. The data obtained from these monitoring sites should mainly serve to perform better source apportionment, validation models and trend analysis. They may also help to calculate correlations between the different parameters. It is recommended to aim this network also at facilitating better targeted future strategies for reducing the health impacts of PM and identifying the components and/or properties of PM which are particularly relevant for health effects.
3. Such data should be combined with health studies and source apportionnement studies to facilitate better targeting of future strategies for reducing the health impacts of PM and to help assess the attainability of possible future targets based on different metrics.
4. Indoors, where citizens spend much of their time, PM levels are only partially dependent on outdoor levels. This is also true for passenger cabins of vehicles. More research is needed in order to identify the contributions of outdoor, indoor and other (personal activities and community) sources to personal exposure to PM.

4.7 References

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5. Emissions

5.1 Scope

This chapter describes the emissions of particulate matter and particulate precursor pollutants across Europe. The anthropogenic and natural emission sources of these pollutants are discussed, together with an assessment of the progress made to date in establishing European particulate emission inventories. Issues that historically have affected the reliability of particulate emissions data are described, with particular reference made to the quality of the particulate data submitted in 2003 by Parties reporting to the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP).

The chapter also presents sector and country-specific emissions data for particulate, and particulate precursor species for 2000. An assessment is made of the extent to which anthropogenic emissions of particulates have decreased across Europe over the last decade.

5.2 Introduction

5.2.1 Primary and secondary PM

Particulate matter refers to those particles of varying size and chemical composition that are emitted to, or are formed via chemical reaction in, the atmosphere. *Primary* particulate matter comprises particles that are emitted directly to the atmosphere from natural and anthropogenic sources. Emissions may be separated according to their origin (anthropogenic combustion, anthropogenic mechanically generated; or natural processes) or by their specific emission source. Particulates are emitted from a wide range of emission sources, including fuel combustion, surface erosion and wind blown dusts, and mechanical break-up from for example, quarrying and construction activities. In contrast, *secondary* particulate matter refers to the particles of low volatility formed via atmospheric chemical processes from the gaseous emissions of precursor pollutants. Specifically, the formation of secondary particulate matter in the atmosphere occurs through the reactions of the precursor species sulphur dioxide (SO₂), nitrogen oxides (NO_x) and ammonia (NH₃) to form particulate sulphates and nitrates respectively, as well as organic aerosols formed from the photochemically-induced oxidation of some non-methane volatile organic compounds (NMVOCs).

Table 5.1 summarises the emissions of primary particulate matter of the 15 EU Member States (EU15), the 10 Acceding countries (AC10) and 4 EFTA Countries (EFTA4). Two columns of primary PM₁₀ data are shown. The first column shows the data that was available at the initial time of writing of this chapter and which is used as the basis of the primary PM₁₀ sectoral and trend analysis that follows in Sections 5.5 and 5.7. Unless indicated otherwise, the emissions data shown is from national data that was officially submitted in 2002 by countries to UNECE/CLRTAP. Where countries did not report values to CLRTAP for this year, missing data was filled with primary PM₁₀ data obtained from the Auto Oil II programme (European Commission 2000) for EU-15 countries, and for AC countries from CEPMEIP (2001).

The second column shows updated national estimates for PM₁₀ data from the 2003 UNECE/CLRTAP submissions that only became available in the final editing stages of this report. As described above, gaps in the reported PM₁₀ data were again filled with estimates from the Auto Oil II programme (European Commission 2000).

Table 5.1 *European Emissions of Primary PM₁₀ and PM₁₀ precursors, 2000 (in ktonnes). Updated emission estimates for primary PM₁₀ that became available in the final editing stages of this report are also shown.*

Country Grouping	Country	PM ₁₀		NO _x	SO ₂	NM VOC	NH ₃
		Data used in the following chapter sections	Updated data: 2003 CLRTAP submission				
AC	Bulgaria	93 ¹⁾	93 ¹⁾	184	982	120	56
AC	Czech Republic	125 ¹⁾	33	398	265	247	74
AC	Estonia	33 ¹⁾	33	41	95	34	9
AC	Hungary	62 ¹⁾	30	187	485	172	71
AC	Latvia	13 ¹⁾	13 ¹⁾	37	18	96	12
AC	Lithuania	20 ¹⁾	20 ¹⁾	48	43	61	25
AC	Poland	314 ¹⁾	296	922	1511	599	325
AC	Romania	186 ¹⁾	186 ¹⁾	319	912	638	221
AC	Slovakia	41 ¹⁾	41 ¹⁾	106	120	89	30
AC	Slovenia	13 ¹⁾	13 ¹⁾	58	97	40	19
AC	Total	900	758	2300	4528	2096	842
EFTA4	Iceland	0	0	40	44	10	0
EFTA4	Liechtenstein	0.1	0	0.4	0.1	0.5	0.2
EFTA4	Norway	0	66	223	27	372	305
EFTA4	Switzerland	26	26	96	19	159	68
EFTA4	Total	26	92	359	90	542	373
EU15	Austria	47	47	184	41	239	68
EU15	Belgium	59 ²⁾	64	289	181	248	100
EU15	Denmark	34	22	207	27	132	101
EU15	Finland	48	49	236	74	160	33
EU15	France	567	545	1432	659	1660	791
EU15	Germany	887 ²⁾	175 ³⁾	1637	832 ⁴⁾	1653	624
EU15	Greece	41 ²⁾	41 ²⁾	382	541	398	74
EU15	Ireland	19	19	125	131	90	122
EU15	Italy	204 ²⁾	204 ²⁾	1485	923	1672	448
EU15	Luxembourg	5 ²⁾	5 ²⁾	17	3	15	7
EU15	Netherlands	53	51	421	91	281	153
EU15	Portugal	21 ²⁾	21 ²⁾	369	375	484	103
EU15	Spain	115 ²⁾	115 ²⁾	1419	1535	1585	518
EU15	Sweden	30 ²⁾	66	247	58	418	56
EU15	United Kingdom	172	178	1513	1166	1499	297
EU15	Total	2303	1602	9963	6637	10533	3495

Source of data: Submissions to UNECE CLRTAP 2002, unless otherwise indicated:

¹⁾ Data from CEPMEIP (2001)

²⁾ Data from Auto-Oil II programme (European Commission 2000)

³⁾ Estimate of 175 kt PM₁₀ derived from 2003 UNECE/CLRTAP submission value for total TSP in 2000 of 251 kt. There is a large discrepancy between these two emission estimates for PM₁₀, however a review of the quality of the estimates made in the Auto-Oil II programme is beyond the scope of this review chapter.

⁴⁾ An updated value of 638 kt was reported by Germany to UNECE CLRTAP in 2003, after the original compilation of this data.

PM emissions inventories have been mainly produced for PM₁₀. The increasing interest in the correlation between PM_{2.5} (particles less than 2.5 µm) and health indicators has resulted in countries having developed and improved their PM_{2.5} national emission estimates. Total suspended particulates (TSP) is the other measure of particulate matter often reported by countries. In terms of quantitative analyses in this chapter, assessments focus on PM₁₀ emissions, primarily due to reasons of data availability.

5.2.2 Anthropogenic PM sources

There are a large number of sources of primary particulate matter, of both anthropogenic and natural origin. This section describes the main anthropogenic emission sources; natural emission sources are discussed further in Section 5.8.2.

Of the anthropogenic sources of PM, stationary combustion sources, production processes, the road transport sector, and agriculture are the most significant sources for primary PM as well as for emissions of precursor gases. Each of these respective sectors is described in more detail below (see Section 5.7).

Stationary Combustion

A major source of PM₁₀ is from the stationary combustion of fuels. Significant sources included under stationary combustion include power stations, other large industrial sectors such as iron and steel, and the residential sector. The size distribution of particles from combustion is dependent on the type of fuel burnt coupled with the nature of the combustion process itself. Emissions of particulate matter from combustion of solid fuels are generally of a coarser size range than those formed from the combustion of petroleum fuels, and which in turn are coarser than particles formed from gas combustion. Gas combustion produces less particulate mass emissions than combustion of solid and liquid fuels, and the particles produced are also entirely in the fine particle size range (<1 µm).

Production Processes

Significant sectors include the production of metals, cement, lime, coke, and chemicals, bulk handling of dusty materials, construction, and mining and quarrying. Emissions from these sources are difficult to quantify due to the contribution of fugitive emissions (i.e. diffuse emissions which are released directly into the atmosphere from a variety of points and areas connected to a process rather than being collected in a controlled manner and then vented to atmosphere through a vent or stack). Usually a substantial fraction of the particles from these sources is greater than 10 µm but the large quantities of particles emitted ensure that the fraction of particles less than 10 µm is nevertheless significant.

Road Transport

All on-road vehicles emit PM₁₀ from fuel combustion processes. However, diesel vehicles emit a much greater mass of particulate per vehicle kilometre than petrol-engine vehicles. Virtually all of the particles are less than 2.5 µm in size, and most of the mass is on even much smaller particles, in the size range below 0.5 µm. (USEPA 1995, TNO 1997). Emissions also occur from the wear of brake linings and tyres, and are considered to fall into a coarser size range than exhaust emissions. Particles are also emitted as a result of entrainment (or suspension and resuspension) of particles from road surface wear and previously settled dust on the road surface, although there are few estimates available on a European scale as to the extent to which resuspension contributes to total PM emissions. Resuspension is discussed in more detail in Section 5.8.1.

Figure 5.1 illustrates the contribution of different road transport sub sectors to PM fine (here indicating PM₁₀ and PM_{2.5}) and PM coarse (here indicating TSP) in Germany. It is clear that finer particles are emitted through from the combustion processes, while coarser particles are emitted as a result of mechanical processes i.e. from brake and tyre wear, and through resuspension.

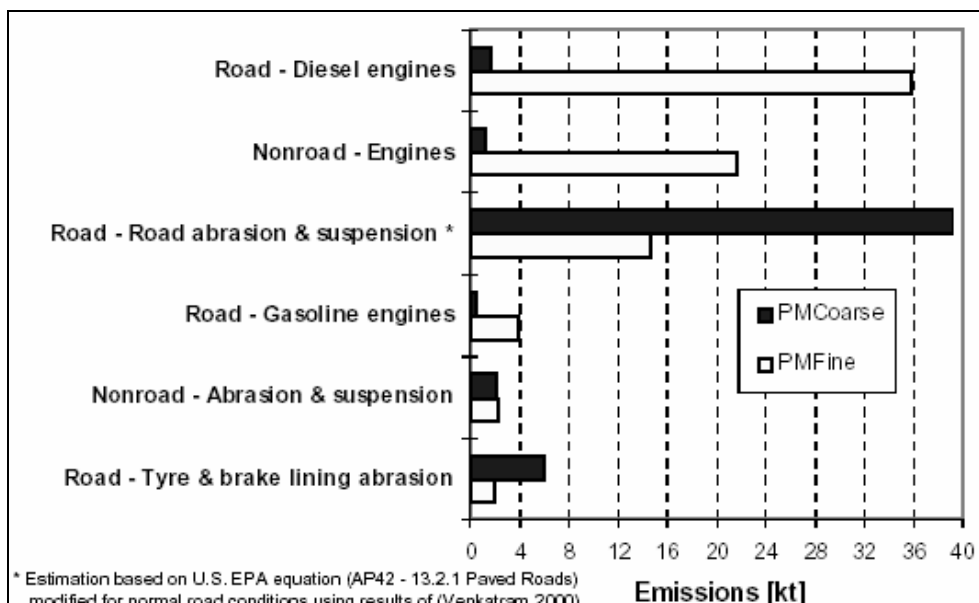


Figure 5.1 Contribution of road transport sub-sectors to TSP and PM₁₀ (Germany, 1998), Source: Pregger, T (2002).

Agriculture.

Emissions from agriculture occur from a range of sources including land preparation, fertiliser application and harvesting. Poultry and pig farming are also considered to be significant emitters of particulate matter, particularly the finer size fraction. Emissions will of course be dependent on the type of agriculture processes used in different countries.

5.3 Progress in establishing European primary PM inventories

PM emissions data have not been reported as consistently at the European level as other air quality pollutants e.g. NO_x and SO₂. Currently, there is no requirement within EU legislation to report data on PM emissions. However, the national emission ceilings directive (NEC D; 2001/81/EC) does request Member States to report emissions of the pollutant precursors SO₂, NO_x, NH₃ and NMVOC. Three examples of European inventories for PM are described in this section, which provides an overview of the progress made in establishing PM inventories.

5.3.1 UNECE Convention on Long Range Transboundary Air Pollution

Prior to 2000, parties to the UNECE Convention on Long Range Transboundary Air Pollution (CLRTAP) were not formally required to report PM data on an annual basis. In 2003, however, parties were requested to report PM emissions data, as TSP, PM₁₀ and PM_{2.5}. Nineteen of the forty-eight Parties to the Convention submitted PM emission data¹³ to the UNECE in 2003. All nineteen parties submitted some PM data for the year 2000 and nine parties reported some emissions data for 1995. Four parties submitted a full time series (1990-2001) of data. Out of the nineteen Parties submitting PM emissions data, six reported using the draft NFR format while sixteen used the current NFR

¹³ In this context, emissions data are defined as actual numerical values (greater than zero) and do not include reporting flags (e.g. NE (not estimated)).

format (with three Parties using a combination of the two formats). Thirteen parties reporting PM emissions data submitted some data on PM_{2.5}.

5.3.2 CEPMEIP

The Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP) is part of the activities aimed at supporting national experts in reporting particulate matter emission inventories to the EMEP programme under the UNECE CLRTAP. Within this work programme, an overview of particulate emission estimation methods was compiled (CEPMEIP, 2001). These were then used and applied to compile a European emission inventory for particulates for the base year 1995 (TNO, 2003). CEPMEIP information has been widely used in recent country reporting of PM under CLRTAP.

CEPMEIP was originally initiated as a joint activity by both the UNECE Convention (CLRTAP) and the European Environment Agency. This programme aimed to develop default methods and emission factors for the use of national experts when submitting particulate matter emission inventories within the CLRTAP/EMEP reporting framework. In addition to emission factor development, a second goal of CEPMEIP was the production of emissions on EMEP 50 x 50 km grid, suitable for modelling purposes. Compilation of the inventory drew on literature review and data provided by approximately 20 countries. Country data included measurement of concentrations and emission inventory estimates.

The CEPMEIP inventory was largely based on an earlier inventory developed by TNO in 1996, which was the first consistent gridded emission inventory for PM₁₀, PM_{2.5} and PM_{0.1} for the reference years 1990 and 1993 (TNO 1997). This dataset has since been widely used. During the years following 1996, several additional new studies on PM emissions were performed, enabling improvement of knowledge on primary PM emissions. These projects served as a key point of expertise on the field of primary PM emissions and provided linkages between experts in emission inventories, experimental research and PM modelling. The CEPMEIP project made use of this well-established network.

5.3.3 RAINS

The Regional Air Pollution Information and Simulation (RAINS) model for integrated assessment of pollution developed at the International Institute for Applied Systems (IIASA) includes modules for gaseous PM precursors and primary particulate emissions (IIASA 2001). The RAINS model is designed to provide a practical policy context so that cost-effective emission control strategies can be determined in a multi-pollutant/multi-effect framework. The model itself uses emission factors (from literature sources and from consultation with country experts), information on emission control options and activity data to derive emission estimates for Europe, for key air quality pollutants, including PM. As the data is calculated within a model, rather than being reported on a country-by-country basis, a complete inventory is available from 1990 (base-year) across all sectors.

5.3.4 Differences between inventories

EMEP (2002) has published a detailed comparison of the differences in available emission information for four different countries (Austria, France, the Netherlands and the United Kingdom) for the year 1995 from national submissions, the CEPMEIP project and estimates provided from scenario runs from the IIASA RAINS model. The evaluation showed large differences for several source categories and was intended as an illustration of the type of discussion that would eventually lead to the creation of a harmonized European-wide PM emission inventory. Currently, main sources like

resuspension of road dust, semi-anthropogenic and natural sources like forestry, natural soil dust emissions and soil dust emissions from agriculture and biological pollen emissions are not considered in all inventories.

Table 5.2 illustrates the differences in national totals across the existing European PM inventories for the selected countries (EMEP 2002). Some differences between the inventory estimates were significant. The RAINS scenario calculation for TSP produced a similar estimate to CEPMEIP, with a good match for Austria and the Netherlands but a large difference compared to the national (UNECE) estimate, for France. However, even for countries with similar national totals like Austria, large differences can be seen for the different sectors (for some sectors larger than a factor of three). In contrast, for PM₁₀, RAINS estimates are significantly lower than CEPMEIP and national estimates, while the differences for other countries between all inventories are less than ten percent. The same pattern, as for PM₁₀, is observed for PM_{2.5}.

Table 5.2 Comparison of 1995 National Totals across PM inventories. Source: EMEP (2002).

Year: 1995	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS	UNECE	CEPMEIP	RAINS
Units: Gg	PM2.5			PM10			TSP		
Austria	27.6	33.5	31.0	46.8	46.2	43.5	75.8	82.7	77.5
France	336.0	350.6	205.3	587.7	449.7	289.0	1525.0	693.3	527.3
Netherlands	n.a.	41.4	37.6	60.9	64.4	62.0	75.0	127.3	117.7
United Kingdom	132.3	164.4	154.7	237.9	259.6	261.1	n.a.	473.4	555.6

Some significant progress has been made over the last eight years in providing European-wide PM inventories. Increased numbers of countries are now also reporting particulate emission estimates to UNECE CLRTAP, although there are still significant gaps in reporting of country data, i.e. non-reporting of PM from likely emitting sectors, and also large uncertainties in the emission factors used to derive certain sector estimates.

5.4 Data Quality Issues

PM inventories are relatively new at a European level. For example, countries were only formally required to report PM emissions data under CLRTAP from 2000. Therefore, estimation techniques, and particularly the use of emission factors, have not been researched or developed to the same extent as has been the case for other pollutants. This means that certain data quality issues arise, particularly for estimations of PM from certain sectors and for determining estimates of particulate size fractionation. A further factor is that countries do not have the same levels of experience to be able to deliver a complete inventory on an annual basis for the different PM size fractions. Not all national PM inventories are subsequently complete when reported to EMEP under CLRTAP.

Thus two main issues arise – the quality of the estimates due to uncertainties, particularly with emission factors, and the completeness of data submissions. An example of uncertainties in PM inventories is shown below from work undertaken in the United Kingdom, that has attempted to quantify the uncertainties in the national emission estimates (Passant 2003). Table 5.3 outlines the estimated magnitudes of uncertainty in particulate matter estimates (expressed as 95% confidence intervals), and for the PM precursor gases NO_x, SO₂, NH₃ and NMVOCs. The estimated uncertainties for PM₁₀ are clearly significantly larger than for the precursor pollutants NO_x, SO₂ and NMVOCs. This is due to the relatively large degree of uncertainty in the emission factors used within the United Kingdom national inventory.

Table 5.3 Results of UK Uncertainty Analysis (emissions in ktonnes). Source: Passant (2003).

Pollutant	Inventory Year	Mean	95% confidence interval as % of mean
PM ₁₀	2000	172	+51%/-20%
NO _x	2000	1525	+/-7%
SO ₂	2000	1156	+4%/-3%
NH ₃	2000	320	+19%/-18%
NMVOCs	2000	1678	+11%/-9%

However it should be noted that uncertainties in trends are likely to be significantly smaller than uncertainties in absolute numbers (Eggleston et. al. 1998). Although the above tables reflect specific uncertainties for the United Kingdom, they are probably broadly indicative of the typical uncertainties in other national inventories. This is because the estimation of particulate matter around Europe will draw on similar emission factors that have similar level of associated uncertainty.

PM inventory data quality, in terms of completeness in reporting, has been assessed using CLRTAP data submitted in 2003. Much of the analysis is taken from a recent review by the European Topic Centre on Air and Climate Change (ETC-ACC 2003) of the 2003 submitted CLRTAP data. The aim of the review was to pilot a range of tests devised to assess the general quality of air emissions data reported.

Figure 5.2 shows an overview of the data density of the 2003 'new NFR' reported inventories showing sectors and pollutants having high, medium and low levels of reporting. It provides a broad indication of the comparative levels of reporting and indicates on the basis of reported data, which are the significant sources for each pollutant. Coupled with knowledge of what sectors might be expected to report emissions, this type of analysis can help identify where under-reporting may be occurring.

The reporting level of primary PM fractions is generally lower for all sectors than for precursor pollutants, as illustrated by the number of blue and yellow colours. For example, in the case of the agricultural sector, the level of PM reporting might be expected to be similar to the number of values reported for NH₃. Similarly, for the road transport sector, a lower level of PM₁₀ reporting than for NMVOC, NO_x and SO₂ is observed.

5.5 Progress in reductions of European PM₁₀ emissions

5.5.1 Methodology description

Emissions data shown in this, and following sections is from the national and sectoral emissions data that have been officially reported in 2002 by countries to UNECE/CLRTAP. Where not reported, primary PM₁₀ national totals were obtained from the Auto-Oil II programme (European Commission 2000), for EU15 countries and from CEPMEIP (2001) for AC countries. Sectoral splits were subsequently derived using sector weightings from CEPMEIP (2001). Data was gap-filled using established ETC-ACC methodologies where required, to obtain consistent and comparable time-series information derived from reported Party data to the UNECE Convention on Long-range Transboundary Air Pollution.

Further information concerning the data and methodologies used to compile the emission statistics is available in the Air Pollutant emissions fact sheet series from the EEA web site¹⁴.

¹⁴ www.eea.eu.int

5.5.2 Assessment

Figure 5.3 illustrates the reduction in emissions of PM_{10} between 1990 and 2000 for Europe 29 (an aggregated country grouping that includes the 15 Member States, the 10 Acceding countries and the 4 EFTA Countries). The reduction shown in the graph is primarily due to reductions in the EU15 grouping, where emissions have decreased by 18%. The Accession total shown are almost constant over the time period since, due to an almost total lack of reported data by countries in 2002, the time series for most AC countries are based on the 1995 CEPMEIP estimates. Still, the AC emissions are included here to give an impression of the total emissions and the magnitude from the various country groupings.

Sector Name and Code		NH3	NMVO	NOx	SOx	PM2.5	PM10	TSP	High Medium Low
Energy Production & Distribution	1 A 1 a	59	140	143	135	72	74	77	
	1 A 1 b	27	108	128	129	72	73	63	
	1 A 1 c	44	125	130	111	71	72	63	
Manufacturing Industries and Construction	1 A 2	44	108	110	113	45	47	46	
	1 A 2 a	41	95	97	96	58	58	49	
	1 A 2 b	30	82	84	93	59	58	48	
	1 A 2 c	30	80	82	82	44	44	46	
	1 A 2 d	28	80	83	81	44	44	46	
	1 A 2 e	28	79	82	82	44	44	46	
	1 A 2 f	54	96	98	97	56	57	47	
International Transport	1 A 3 a i (i)	12	99	99	98	19	31	20	
	1 A 3 a i (ii)	12	74	74	74	19	19	19	
	1 A 3 a ii (i)	12	129	129	119	64	64	55	
	1 A 3 a ii (ii)	17	92	93	78	50	50	50	
Road Transport	1 A 3 b	69	95	97	99	42	44	43	
	1 A 3 b i	105	96	108	108	59	59	51	
	1 A 3 b ii	104	95	107	107	59	59	51	
	1 A 3 b iii	103	106	106	106	59	59	51	
	1 A 3 b iv	89	94	105	93	54	54	42	
	1 A 3 b v	91				2	2	2	
	1 A 3 b vi					59	61	49	
Other Transport	1 A 3 b vii	31	33	33					
	1 A 3 c	53	137	137	138	72	73	64	
	1 A 3 d i	24	85	85	85	14	27	14	
	1 A 3 d ii	41	137	137	139	72	73	64	
	1 A 3 e	31	68	68	58	42	43	42	
	1 A 3 e i	12	49	51	26	29	29	29	
Commercial Residential and Agricultural Combustion	1 A 3 e ii	13	40	41	41	40	42	29	
	1 A 4 a	40	138	140	142	72	74	64	
	1 A 4 b	41	96	98	100	42	44	43	
	1 A 4 b i	49	106	106	106	59	60	50	
	1 A 4 b ii	24	88	88	76	56	56	44	
	1 A 4 c	67	80	82	82	35	36	35	
	1 A 4 c i	38	107	108	108	59	61	51	
Military	1 A 4 c ii	48	105	106	106	59	61	49	
	1 A 4 c iii	12	77	78	78	42	42	30	
	1 A 5 a	14	18	20	20	18	18	20	
Fugitive Emissions from Solid Fuels	1 A 5 b	24	49	49	48	39	41	29	
	1 B 1	37	49	49	50	19	20	20	
	1 B 1 a	2	2	2	1	16	16	17	
	1 B 1 b	52	69	51	52	40	40	28	
Oil and natural gas	1 B 1 c					12	12	12	
	1 B 2	17	81	69	83	37	38	38	
	1 B 2 a	14	62	52	52	12	12	12	
	1 B 2 a i	11	81	24	25	11	11	11	
	1 B 2 a iv	12	94	49	58	24	24	25	
	1 B 2 a v		108			1		1	
	1 B 2 b	9	88		29	11	11	11	
Mineral Products	1 B 2 c		72	75	59	38	38	26	
	2 A	19	80	45	82	43	44	45	
	2 A 1		13	15	61	30	30	31	
	2 A 2				1	1	29	29	
	2 A 3					6	6	7	
	2 A 4					12	12		
	2 A 5		39					1	
Chemical Industry	2 A 6		90	2	2	12	12	12	
	2 A 7	29	53	30	41	54	54	45	
	2 B	69	80	82	76	42	43	43	
	2 B 1	52	39	20	12				
	2 B 2	65	1	86		12	12	12	
	2 B 3		13						
Other Industrial Production	2 B 4		4	2	24	24	24	34	
	2 B 5	80	93	91	91	44	44	35	
	2 C	24	123	118	119	69	71	59	
	2 D	18	80	63	67	42	43	43	
	2 D 1	25	80	54	62	49	49	40	
	2 D 2	12	114	3	2	19	19	22	
Solvent Use	2 G	33	20	20	22	13	26	28	
	3 A	1	139	4	4	3	5	7	
	3 B		114	4	4	3	3	5	
	3 C	3	109	6	6	8	32	33	
Agriculture	3 D	22	126	1	2	31	32	33	
	4 B	82				10	10	10	
	4 B 1	61				24	24	12	
	4 B 1 a	74				20	20	20	
	4 B 1 b	74				20	20	20	
	4 B 13	79				4	4	4	
	4 B 3	91				18	18	18	
	4 B 4	65				17	17	17	
	4 B 6	92				17	17	17	
	4 B 7	36				12	12	12	
	4 B 8	102	1			39	39	27	
	4 B 9	102				39	39	27	
	4 C	12							
	4 D	68	39	25		1	5	5	
	4 D 1	103	65	25		30	30	18	
Natural Sources	4 F	37	30	53	24	17	17	13	
	4 G	19	18	9	2	36	36	36	
	5 B		4	24					
	5 E	12	60	36	12				
Waste Disposal and Treatment	6 A	39	85	14	14	14	14	14	
	6 B	26	32	7	8	6	6	7	
	6 C	52	99	97	99	53	55	43	
	6 D	46	37	23	10	25	25	25	
NATIONAL TOTAL		130	141	143	145	72	75	76	

Figure 5.2 NFR reporting data "density" from 1990 - 2001 for EU15 countries, showing the frequency with which emission values have been reported in a given sector. The values given in

the table show the count of emission values reported by countries for each sector between 1990 and 2001 (number of years x number of countries).

The EFTA emission total is small compared to the other country groupings, accounting for less than 1% of the Europe 29 total in 2000. A decrease of 18% is also observed for this country grouping.

If real time series data had been available for Acceding countries, the overall decrease would be expected to have been greater than the 18% indicated in Figure 5.3.

Figure 5.4 illustrates the changes in PM₁₀ emissions on a country-by-country basis, and shows that most EU countries have had significant primary PM₁₀ emission reductions over the period 1990-2000. Greece and Portugal had an increase of 5%, and Finland a very large increase of 61%. The reasons why the Finnish emissions have increased steeply over this period are not currently known. Reported emissions in the Finnish 2003 UNECE/CLRTAP submission are consistent with the values reported in 2002. Both the EFTA countries that have reported emissions data show significant reductions.

Sectors contributing most to these reductions are energy industries and industry sectors, and road transport. In energy industries, fuel switching to natural gas and the use of and improvements to technologies such as electrostatic precipitators has contributed to the decrease in emissions observed. In the road transport sector, tighter PM₁₀ emission regulations have been introduced for heavy duty vehicles and cars.

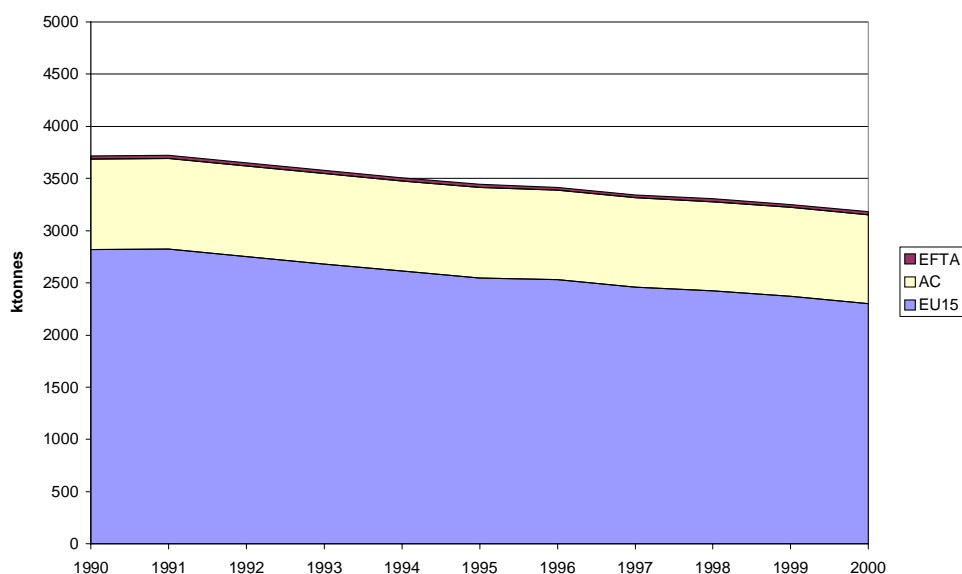


Figure 5.3 European PM₁₀ emissions 1990-2000

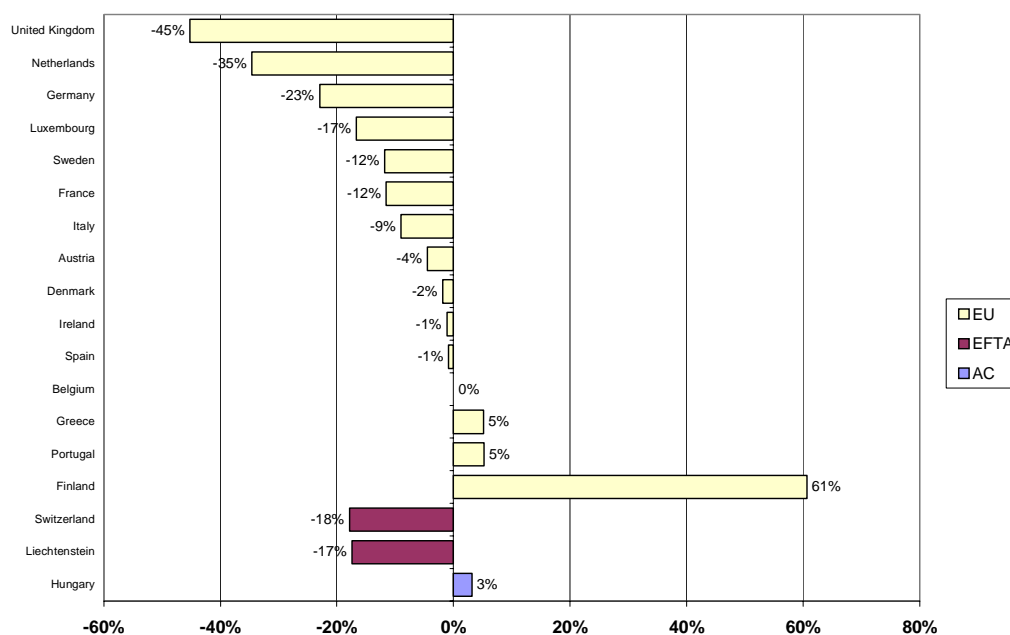


Figure 5.4 Percentage change in PM_{10} emissions 1990-2000.

5.6 Progress in reductions of European PM Precursor Gases

Figures 5.5 and 5.6 show the Europe 29 (EU15, EFTA4 and AC10) emission trend between 1990 and 2000 for Precursor Gases. SO_2 has the most significant reduction in emissions, predominantly due to a switch from high sulphur solid and liquid fuels to natural gas, in the energy industries, industry and domestic sectors, as well as construction of new power plant and the use of low sulphur coal and flue gas desulphurisation.

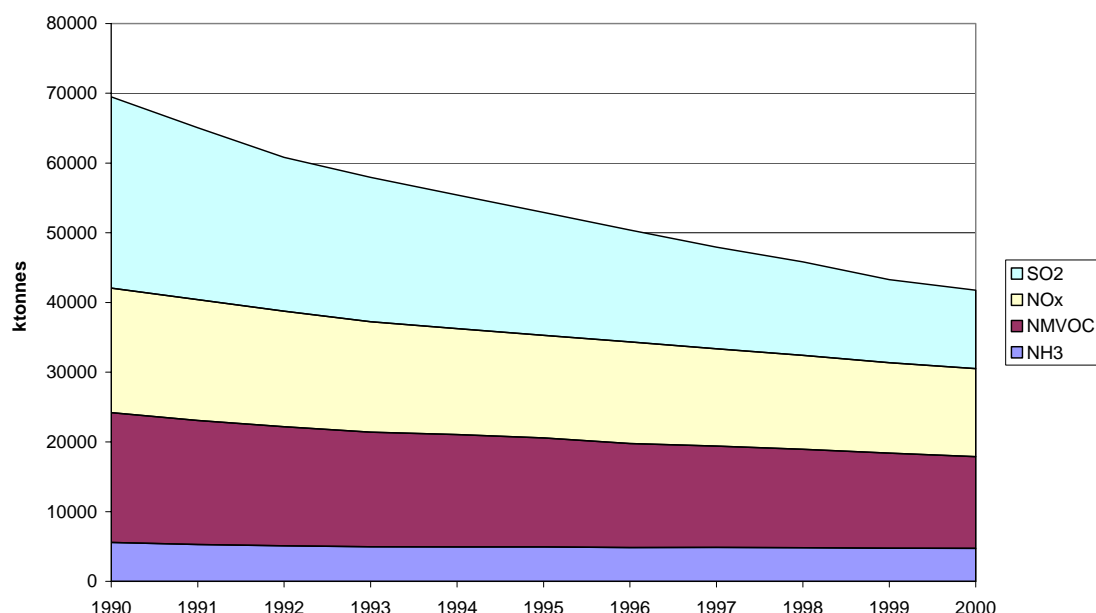


Figure 5.5 Emissions of Precursor Gases in Europe 1990-2000

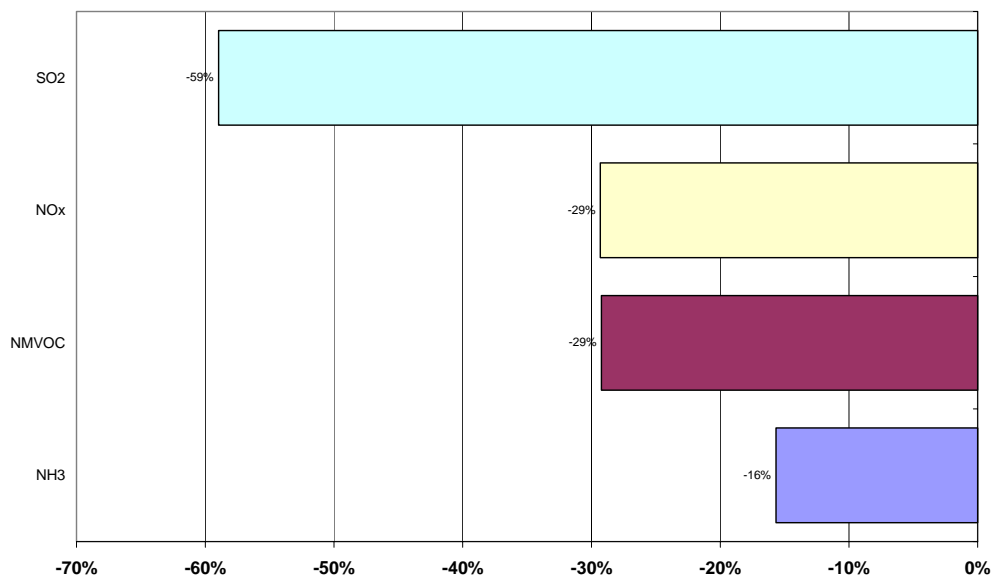


Figure 5.6 Reduction of Precursor Gas Emissions in Europe between 1990 and 2000

5.7 Sectoral emissions of primary PM and PM precursors in 2000

5.7.1 PM₁₀

As described in preceding sections, sectoral emissions data was obtained from officially reported data by countries to UNECE/CLRTAP in 2002. Where not reported, sectoral estimates were derived using CEPMEIP (CEPMEIP 2001) sector weightings applied to PM₁₀ national totals from the Auto-Oil II programme (European Commission 2000) for EU15 countries and from CEPMEIP (2001) for AC countries. Figures 5.7 and 5.8 illustrate the sector contributions to PM₁₀ and precursor gases for the EU15 and AC10 countries respectively.

In EU15, PM₁₀ emissions are dominated by sectors including road transport, industry and "other" (which is primarily fuel combustion in the residential sector). In the Acceding countries, the largest sectoral contribution of PM₁₀ emissions comes from the energy industries sector, with only a small contribution from road transport. This difference is probably due to smaller vehicle fleets in Acceding countries, the greater relative use of solid fuels in comparison to natural gas in power generation, and the less wide use of effective abatement technologies in the power generation sector. With state-of-the-art abatement equipment installed, this sector is only of minor importance relative to other emission sources.

Similar differences between EU15 and AC10 are also reflected in the NO_x emissions. In both areas transport and energy industries are dominating the emissions, but the transport sector is more dominant in EU15 while the energy sector is more dominant in AC10.

For SO₂, the energy industry is dominant in both areas, although more so in EU15 than in EAC10, where the "other" sector contributes more (probably small-scale coal burning for space heating).

For non-methane VOCs, the main contributions are from "other" (solvent use) and road transport, similar in both areas. For NH₃, agriculture (livestock waste) dominates completely in both areas.

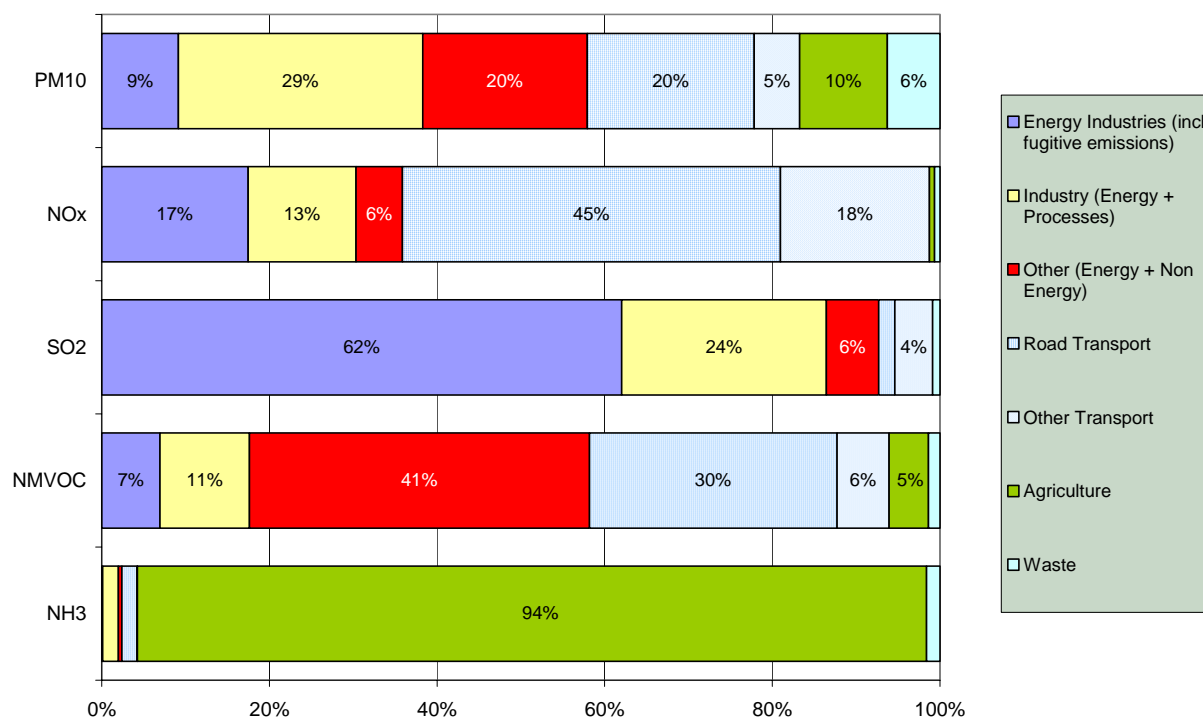


Figure 5.7 EU15 Sector Contribution to PM (as PM₁₀) and Precursor Gases

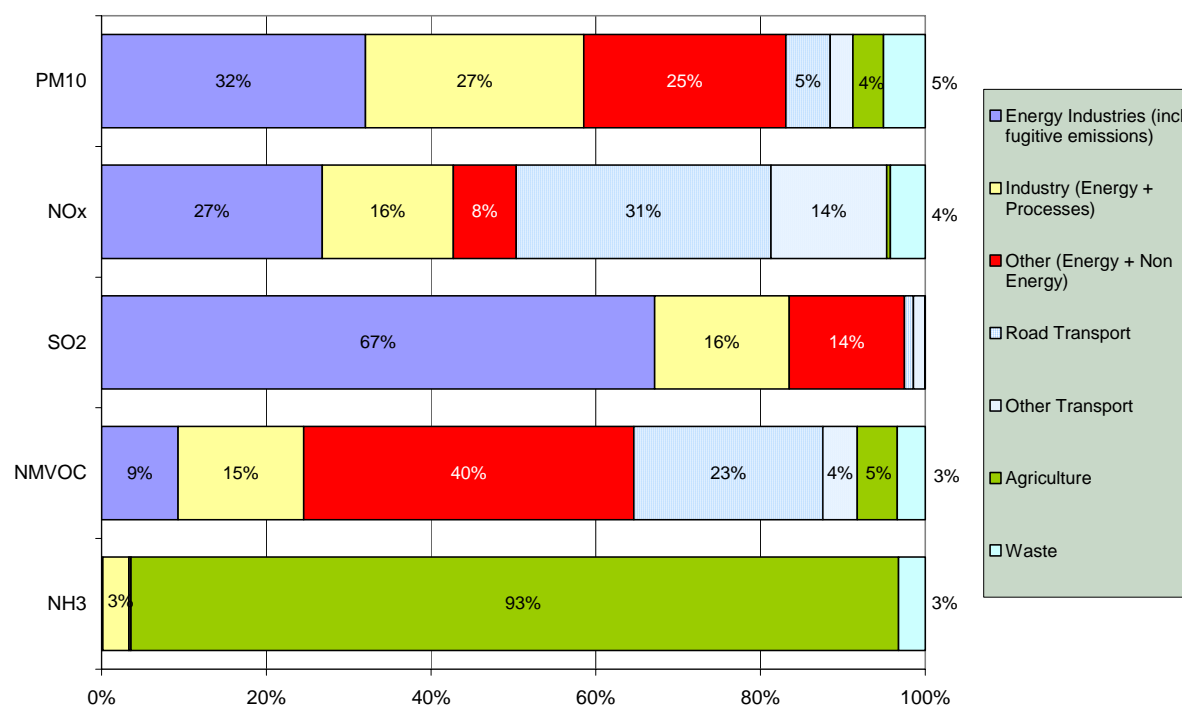


Figure 5.8 Acceding country Sector Contribution to PM (as PM₁₀) and Precursor Gases

5.7.2 PM_{2.5}

Parties under CLRTAP are also requested to submit national estimates of PM_{2.5} emissions. Under the 2002 submission, PM_{2.5} country inventories were much more incomplete than those for PM₁₀. Less

experience of estimating smaller size fractions may result in Parties focusing on delivering a complete PM_{10} inventory. For the 2002 CLRTAP submission, 7 countries submitted a dataset on $PM_{2.5}$. France had one of the more complete inventories. The sector breakdown for $PM_{2.5}$ and PM_{10} emissions in 2000 for France is shown in Figure 5.9.

The PM_{10} emission total is approximately twice the size of the $PM_{2.5}$ inventory, which is represented by the relative area of the two figures. For PM_{10} , the industry and agricultural fractions are much more significant, relative to other sectors, illustrating the significant sectoral differences between the size fractions.

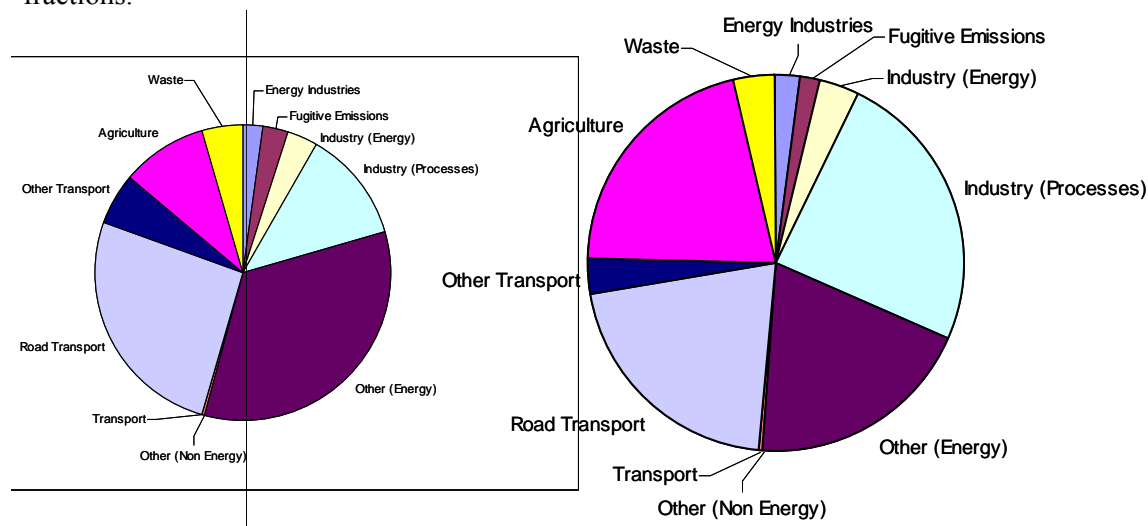


Figure 5.9 Emissions of $PM_{2.5}$ (left) and PM_{10} (right) by sector, France 2000.

5.8 Other sources of PM

5.8.1 Resuspension of particles from roads

Background

Particulate emissions from paved roads are due to direct emissions from vehicles in the form of exhaust, brake wear and tyre wear emission, as well as resuspension of loose material on the road surface. The road transport sector is a main source of "resuspended" particles. Other important sources of resuspension of particles are agriculture (this resuspension source is treated as an integrated part of the agriculture PM source, see elsewhere in this chapter) and wind blown dust, treated as part of the natural sources.

The term "resuspension" is commonly used for the particle source that the road surface represents, although "entrainment" of particles could be a better term. The particles originate from suspension of instantaneously generated particles from wear of the road surface, or from resuspension of particles previously deposited on the road surface, originating from the sources on the road itself or deposited from external sources. Both mechanisms are of importance. In the following text, 'resuspension' is used to describe both these mechanisms, unless specified otherwise.

Resuspension is much higher from unpaved roads than from paved roads. However, since the traffic activity on unpaved roads generally is only a minute part of the total traffic activity, and although PM problems may arise close to unpaved roads, this section concentrates on resuspension from paved roads.

Vehicles travelling along roads cause resuspension via one of three mechanisms. Particles are suspended instantaneously from the wear and shear of the road surface by the tyres. This wear associated with studded tyres (typically used in winter time in many Nordic countries) is at least one order of magnitude larger than from non-studded tyres. Another mechanism is the resuspension, during dry periods, of particles from the deposit of particles on the road, caused by the vehicle turbulence. The amount of particle deposit on the road surface is dependent upon many factors, the most important being the amount of traffic, the quality of the road surface, to what extent studded tyres are used, and the length of the wet period just prior to the dry period at hand, since the deposit on the surface increases especially when the surface is wet. Also, the road cleaning regime is of importance, especially for the resuspension of coarse particles. It is still a question as to what extent road cleaning reduces PM₁₀ resuspension, and indeed resuspension of PM_{2.5}.

So, in dry conditions, the mechanisms of vehicle-induced turbulence and instantaneous road wear are active. The induced turbulence is much larger for large heavy-duty trucks than for passenger cars, so the fraction of heavy goods vehicles in traffic is an important parameter. In wet conditions tyre spray is a potentially important resuspension route, as well as the instantaneous suspension of particles from road wear by the tyres which takes place also during wet conditions.

Contributions to the deposit of particles on the road surface come from many sources, such as the mentioned wear for the road surface, but also from exhaust particles, wear of brake and tyre material and other vehicle mechanical wear sources, friction-inducing material brought on the surface (sand, salt), particles from the nearby surfaces as well as particles from the goods and materials carried by trucks.

Recent sources of summarised information on the resuspension source from roads include the Workshop on Particles in the size of 2.5 to 10 micrometers in Urban Areas, held at the Freie Universität in Berlin, 4-6 November, 2002 (<http://www.trumf.fu-berlin.de/veranstaltungen/>), and a literature summary on "Emissions of road wear and resuspension particles in the road environment" by Gustavsson (2003).

Emission factors

As the emissions from resuspension depend on a number of different parameters, including traffic density, road surface material, state of the road, maintenance of the road, driving patterns, fleet composition and climatological factors, and there are large differences in all these parameters, it has not been possible to derive commonly accepted emission factors which are applicable at a large range of different situations.

Gustavsson (2003) has summarised the literature of studies of resuspension PM emission factors related to roads. The US EPA AP-42 method of 1997 and update in 2002 are included. This formula from EPA includes the average fleet weight and the so-called silt load factor (representing the surface loading) as the main determinants of the emission factor.

The studies and emission models available give emission factors varying over several orders of magnitude, from < 100 to several 1000 mg/veh-km for passenger cars, and up to several 10,000 mg/veh-km for heavy duty vehicles, dependent upon which PM fraction is considered, the traffic speed, type of road, use of studded tyres, etc.

Some studies have resulted in estimates of average emission factors for resuspension particles, valid for the road traffic activity of a country as a whole. Some methodologies, like CEPMEIP, considers only road abrasion, but not resuspension. For the United Kingdom, an average PM₁₀ emission factor of 40 mg/veh-km has been estimated for the road traffic vehicle composition as it exists (Nicholson,

2000). This emission factor is based on resuspension under dry conditions, not making any distinction between vehicle-induced turbulence and tyre shear.

For Sweden, a first estimate of an average resuspension emission factor for *winter conditions* has been made based upon PM measurements in a street (Hornsgatan) in Stockholm, of 209 mg/veh-km for PM₁₀ (Johansson, 2002; Foltescu et al., 2001). For Norway, a model has been developed for estimation of emissions of resuspension particles from road surfaces (Tønnesen, 2000). Applied to the whole of the Norwegian traffic activity, this model gives an average emission factor for resuspension of particles in the PM₁₀ range of approximately 130 mg/veh-km, valid for *winter conditions*. The increased resuspension emission factor in Sweden and Norway clearly shows the effect of use of studded tyres, which are used widely in both countries during winter months.

An estimate of the resuspension emission factor for PM_{2.5} has also been calculated in the Swedish study from Hornsgatan (Johansson, 2002). The estimate is 25 mg/veh-km, about one tenth of the PM₁₀ emission factor for resuspension.

Total emissions of resuspension particles from roads

PM₁₀

Examples of emissions inventories, where the resuspension source is specified, are shown here for Austria, Switzerland, Germany, the United Kingdom and Norway.

Winiwarter et al. (2002) have estimated that the Austrian PM₁₀ emissions from traffic caused by resuspension exceed those from tail-pipe emissions by a factor larger than ten, when using the US EPA AP-42 method. However, these results are classified as highly uncertain.

A preliminary emissions inventory for PM₁₀ compiled in Switzerland by Filliger et al (1999) indicated that the emissions from resuspension are more than three times higher than those from tail-pipe emissions.

For Germany (Pregger and Friedrich, 2002) the total resuspension PM₁₀ emissions were estimated to be about 25% larger than the exhaust particle emissions, using the modified US EPA AP-42 model (see Figure 5.1). The PM_{2.5} emissions from resuspension were calculated to be about 33% of the exhaust particle emissions. Duering and Lohmeyer (2003) derived emission factors for road traffic based on measurements in several locations. They concluded that the total PM₁₀ emissions were up to six times higher than those from exhaust alone, with a large range of variation between locations. They did not provide an estimate of total national resuspension emissions to be compared with exhaust PM emissions.

In the United Kingdom, estimates are made of the re-entrainment of dust (or resuspension) from road transport. Such estimates are not included in official reported estimates to avoid double counting. Double counting could be an issue given that particles that are re-entrained in the air have already been emitted and deposited. In 2000, the estimate for PM₁₀ in the United Kingdom for this source sector was 19.4 ktonnes. Other emissions from road transport were estimated to be 31 ktonnes (Goodwin *et al.*, 2002).

For Norway, where studded tyres are used extensively during the 5-6 month winter period, the vehicle exhaust PM₁₀ emissions were calculated to be 4.32 ktonnes in 1994, while the resuspension emissions of PM₁₀ were estimated to be about 2 ktonnes, about half of the exhaust particle emissions (Bang et al., 1999)

These examples indicate that there is a large spread in estimates of the strength of the resuspension source of PM₁₀, as the estimates are based on different models and assumptions. The examples give

the indication, however, that the PM resuspension from roads represents a PM source which is of the same order of magnitude as the exhaust particle source, in terms of total national PM₁₀ emissions.

PM_{2.5}

The dominant part of the mass of the particles from the resuspension source fall in the coarse fraction (PM₁₀-PM_{2.5}), although the Hornsgatan study in Stockholm (Johansson 2002) also gives an estimate of its contribution to PM_{2.5} mass. The study gave an emission factor for PM_{2.5} from resuspension of about 1/10th of the PM₁₀ emission factor. This estimate is valid for a situation with extensive use of studded tyres. In the AP-42 model, the base emission factor for resuspension from roads is about 4 times lower for PM_{2.5} than for PM₁₀ (Gustavsson, 2003). Thus, the resuspension source is also of importance for PM_{2.5}, especially in hot spot situations near roads during dry road conditions, and especially when studded tyres are used.

Summary

The results from the many studies and models on resuspension of PM from roads show a large spread in the results in terms of emission factors for various PM fractions, as well as in terms of calculated total national emissions. The uncertainty is very large. Estimates of the ratio of resuspension to tail pipe PM₁₀ emissions differ largely from a factor of 0.5 to 10. This ratio is considerably lower for PM_{2.5}, since all tail pipe emissions are in this size fraction, while the main part of resuspension falls under the coarse mode.

It is clear that the resuspension source is very important for PM concentrations in hot spot situations (close to streets with large traffic) during dry road conditions, especially when studded tyres are used extensively. More research is needed to clarify the strength of the resuspension source of PM, for various traffic, meteorological and road conditions.

5.8.2 Natural Sources

The main natural sources of PM₁₀ include wind-blown dusts and soils, sea-spray and biological matter such as pollens and fungal spores. Other sources of a more intermittent nature include forest fires and volcanoes. The particles generated by these sources mostly arise from mechanical attrition and are thus relatively large i.e. they are generally greater than 2.5 µm.

Breaking waves produce sea-spray which can dry and yield salt particles in the atmosphere, though these tend to be large and short-lived. A more significant source of sea salt particles are the minute bubbles that burst at the ocean surface. As this happens the film from the cap of the bubble shatters giving salt particles 1.5-8 micrometers across. These droplets from the ocean are mostly sodium chloride with traces of other salts, but there is potential for other substances to be incorporated as the bubble forms.

Biological particles in the atmosphere vary in size significantly. Most pollen grains are well in excess of 10 µm diameter, and even allowing for the fact that their aerodynamic diameter may be different from their geometric diameter (for example because they are hollow), few intact pollen grains are likely to be sampled within PM₁₀. There is clear evidence, however, that pollen grains can break up whilst in the atmosphere generating fragments in the PM₁₀ range extending right down to below 1 µm.

Many fungal spores also exceed 10 µm in diameter, but spores of some abundant species do extend in size into the 5-10 µm range, and hence contribute to coarse particle mass in the atmosphere. There is a typical seasonal pattern in fungal spore abundance with a maximum in the summer and a minimum in the winter.

Natural emissions of the secondary particulate precursor species also occur. For example, volcanoes and sea spray are typical natural sources of SO₂, while lightning is one of the main natural sources of NO_x. Important natural emission sources of ammonia include soils under natural vegetation, oceans and from excreta of wildlife species. There are large uncertainties associated with quantifying the levels of emissions from natural sources. However, for illustrative purposes, a global estimate of NO_x and NH₃ emissions has been compiled in FAO (2001). This indicates that natural sources accounted for 45% and 20% of global NO_x and NH₃ emissions in 1990, respectively.

5.9 Conclusions and recommendations

5.9.1 Conclusions

Progress has been made in establishing a single European format for reporting particulate inventory data, and increasing numbers of countries are reporting particulate emission estimates to UNECE CLRTAP. However, there remain problems with the gaps in reported country data (i.e. non-reporting of PM from likely emitting sectors). The number of reported primary PM_{2.5} and PM₁₀ emissions remains generally lower for all sectors in the 2003 UNECE/CLRTAP submissions than for the PM precursor pollutants. In 2003, the reporting response of primary PM_{2.5} emissions to UNECE CLRTAP was higher than for previous years, but was still lower than the reporting response for PM₁₀.

There are also large uncertainties in the emission factors used to derive certain sector estimates of particulate matter. In particular, PM_{2.5} emission factors are generally of higher uncertainty than those for PM₁₀, largely due to the lower level of experience countries have in reporting of smaller size fractions of particulate material. Similarly, there are large uncertainties associated with quantifying the levels of emissions of particulate and secondary particulate precursor species occurring from natural sources.

In EU15, PM₁₀ emissions are dominated by sectors including road transport (including exhaust emissions, break and tyre wear and resuspension, industry and "other" (which is primarily fuel combustion in the residential sector). In the Acceding Countries, the largest sectoral contribution of PM₁₀ emissions comes from the energy industries sector, with only a small contribution from road transport.

Significant progress has been made in reducing emissions of primary PM₁₀ and PM precursor pollutants in Europe between 1990 and 2000. Emissions of primary PM₁₀ have reduced by 18% across Europe as a whole, mainly as a result of reductions made in the road transport, energy industries and industry sectors. Emissions of precursors have also decreased substantially. The main factors behind the reductions observed are a switch to cleaner fuels across the energy and transport sectors, as well as improvements in pollution abatement techniques.

Best present estimates of the strength of the PM source that resuspension from roads represents, indicates that on a national, annual basis the emissions of PM₁₀ could be at least of the same magnitude as the vehicle exhaust particle emissions, but highly dependent upon the extent of use of studded tyres. Individual estimates from various studies range from 0.5 to 10 times the vehicle exhaust emissions. The PM_{2.5} emissions from resuspension could be 1/10th of its PM₁₀ emissions. Due to the nature of resuspension, with very large emissions during dry periods, this source could dominate the PM concentrations, especially PM₁₀, close to roads with large traffic loads.

5.9.2 Recommendations

1. Further measurements of particulate emissions are recommended to allow the development of more robust, country-specific emissions factors. Particular attention is also required to improve the emission factors for PM_{2.5}, to improve the quality of inventory reporting for this pollutant.
2. Countries are requested to improve emission reporting at European level, since these data are essential for the policy development process in Europe.
3. The UN/ECE Task Force on Emission Inventories and projection, in close collaboration with the EEA and national experts, is encouraged to work on improved inventories to get a harmonized European-wide PM emission inventory and projections.
4. Data on chemical composition of PM emissions is still scarce. Such data is needed for source apportionment and validation of PM emission inventories and models using monitoring data. Therefore, typical source profiles should be established for the main sources of primary PM.
5. Quantification of the resuspension source is still uncertain, and several models based upon different assumptions exist. Being an important source for near-road PM₁₀ occurrence and also relevant for PM_{2.5}, efforts should be put into better quantification of this source.
6. More information to quantify sources of both natural particulate emissions and natural secondary PM precursors is needed.

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6. Source apportionment

6.1 Scope

In addition to the local and regional anthropogenic particulate emissions, both the levels and composition of ambient air PM depend on the climatology (mainly temperature, humidity, photochemistry, resuspension of soil particles, rain scavenging potential, re-circulation of air masses, dispersive atmospheric conditions) and on the geography (mainly proximity to the coast, soil cover and proximity to arid zones) of a given region. Therefore, wide variations in PM levels and characteristics may be expected when considering different EU regions such as Southern Mediterranean and the Scandinavian countries with very different climatologic and geographical patterns. Consequently, due to the difficulty of reporting general PM compositional characteristics, in this chapter examples of source apportionment analysis performed in Germany, Spain, the United Kingdom, the Netherlands and Sweden are given to illustrate different PM situations in the EU. Detailed data are presented in Annex 5. The main objective of this chapter is not to cover all possible PM situations from different EU sites, but to compare and interpret PM characteristics and sources of five selected regions, having very different climatic patterns, within EU. Consequently, PM₁₀ levels and contributions in other countries and cities may well fall outside the general picture presented here. Putaud et al. (2002) recently compiled a comprehensive report on physical and chemical characteristics of particulate matter at kerbside, urban, rural and background sites in EU, and detailed studies on PM speciation and source apportionment analysis may be found in APEG (1999), Visser et al. (2002), Quass and Kuhlbusch (2002) and Querol et al. (2003), for the United Kingdom, the Netherlands, Germany and Spain, respectively.

The source apportionment analysis is a very useful tool to evaluate the different natural and anthropogenic contributions to bulk levels measured in ambient air. There are a number of techniques that may be used, and results may vary according the methodology used. These techniques may be classified according the following three main groups: a) methods based on numerical data treatment, b) modelling based on emission inventories, c) receptor modelling.

a) Methods based on the *evaluation of monitoring data*. This is a straightforward approach in which basic numerical data treatment is used to identify main sources of measured components. Examples are: (a1) correlation of wind direction with levels of measured components to identify the possible location of major sources; (a2) the correlation of gaseous pollutants (tracing a major emission source) with PM components to identify source associations; or (a3) subtraction of levels measured at regional background to those obtained at urban background and/or roadside levels to identify the contributions from the regional background, the city background and the load from the monitored street (see Lenschow et al., 2001 as an example). The main advantage is the simplicity of the methods and the related low impact of mathematical artefacts by the data treatment.

b) Source apportionment analysis may be based on *emission inventories and/or dispersion models* (such as Lagrangian trajectory models) to simulate aerosol emission, formation, transport and deposition (Eldering and Cass, 1996; Kleeman and Cass, 1998; Visser et al., 2001). Although in some cases these models may yield results consistent with the experimental data, it requires a detailed emission inventory, which is not always available. These methods are limited by the accuracy of emission inventories, especially when natural emissions are important. In some cases these limitations account for the low reproducibility of experimental data yielded by these modelling tools. A significant advantage of this method is that it may be used in scenario studies to evaluate the impact of emission abatement strategies on the anthropogenic contribution to ambient PM concentrations.

c) *Receptor modelling* techniques are based on the evaluation of PM chemical data acquired at receptor sites, and most of them do not require previously identified emission sources (Henry et al., 1984; Thurston and Spengler, 1985). These types of models have played a key role in the evaluation of

PM sources with respect to national air quality standards in certain countries. In the United States, the Chemical Mass Balance Model (Gertler et al., 1995; Chow et al., 1996, Wilson et al., 2002) has been widely used, whereas in Europe receptor modelling techniques have been mainly based on methodologies that do not require chemical profiles from source emissions (Harrison et al., 1997; Pio et al., 1998; Rodriguez et al., 2002). The main limitations of the latter methods are: (c1) not all emission sources are detected by the method, but only those with a significant influence on the variance; and (c2) the stability and production rate of secondary PM components such as sulphate, nitrate, ammonium, or organic carbon may strongly vary between seasons, which may result in identification of artefact source chemical profiles.

This chapter focuses on the PM₁₀ and PM_{2.5} source contributions in urban areas of EU and on the differences observed among the above EU regions. At industrial hot spots, the source contribution to PM₁₀ and PM_{2.5} may vary considerably as a function of the industrial process and the distance to the emission source. However, the conclusions obtained for traffic hot spots may apply to most of EU urban areas due to the similarity of the emission sources. Most of the differences observed among traffic hot spots may be attributable to traffic intensity but differences may also be due to climatic patterns such as frequency of rain (due to both scavenging and road dust wash out capacity) or dispersive conditions.

The data selected here were obtained from the following sources:

- PM data from Berlin in 1998, Germany from Lenschow et al. (2001). Data for 2000-2001 were obtained from Abraham (2001) and Abraham H.J. (Senate Department of Urban Development, Berlin, Germany, personal communication).
- PM data from rural and urban background and kerbside sites in Spain in the period 1999-2001 were obtained from Querol et al. (2001 and 2003), Artíñano et al. (2003) and Rodríguez et al. (2002) and from EMEP for 2001.
- PM data from Sweden for 1999-2001 were extracted from Areskoug et al. (2004) and Swietlicki et al. (2004).
- PM data from the Netherlands were obtained from different sources but compiled by E. Buringh of RIVM, National Institute of Public Health and the Environment in the Netherlands.
- PM data from the United Kingdom were derived from four paired urban background and roadside sites in London and Birmingham operated by the University of Birmingham (Harrison et al., 2003).

In most of these examples, source apportionment analysis has been carried out by considering the urban background as an "island" of elevated PM levels in a regional background and on top certain peak concentrations due to traffic hot spots at the road side sites (Lenschow et al., 2001). PM₁₀ and PM_{2.5} measurements performed in these three typical locations (regional background, urban background and roadside sites), including speciation, were used as basic information for source apportionment analysis. In this chapter the above mentioned source apportionment strategy a3 is used to compare the PM characteristics and sources of different urban scenarios in Europe. However, for some examples data obtained using modelling receptor data and emissions inventories are also presented.

Furthermore, data on chemical speciation of PM₁₀ and PM_{2.5} from regional background, urban background and road side sites from Switzerland and Austria (supplied by P. Strahl and J. Schneider) are also included for comparison.

The data of the examples selected are presented in detail in Annex 5. The chapter compares the data in the different geographical and climatic context represented by the examples and gives conclusions and recommendations. The results obtained on levels and source apportionment of PM₁₀ and PM_{2.5} at regional and urban background and roadside sites in the different EU examples considered are summarised in Tables 6.1 and 6.2 at the end of this chapter.

6.2 PM levels

In the selected EU examples (see Table 6.1 at the end of this chapter) at regional background sites, average PM₁₀ levels were found to range from 19 to 24 µg/m³, with the exception of Sweden, where measured PM₁₀ levels are lower (16 µg/m³). At urban background sites, the highest PM₁₀ levels were recorded in Spain and Berlin, where annual PM₁₀ mean varied from 28 to 42 µg/m³. In the other examples PM₁₀ levels reached around 25 µg/m³. The lowest PM₁₀ values were also recorded in Sweden (17-23 µg/m³). A similar variation was found for roadside stations. The highest values were obtained in Austria, Berlin and Spain (37-53 µg/m³). In the Netherlands and the United Kingdom, PM₁₀ measured at roadside sites varied from 30 to 35 µg/m³, and from 26 to 51 µg/m³ in Sweden.

Concerning PM_{2.5} (see Table 6.1 at the end of this chapter), at regional background sites average levels ranged from 8 to 20 µg/m³ for all examples. Maximum levels were measured in Austria (20 µg/m³) and minimum in Sweden (8 µg/m³). Similar levels were obtained in Spain and the Netherlands (14 µg/m³). At urban background sites, the highest PM_{2.5} levels were recorded in Spain and Berlin, where annual PM_{2.5} means varied from 20 to 30 µg/m³. In the other countries, PM_{2.5} levels ranged from 15 to 20 µg/m³, with the exception of Sweden (8-15 µg/m³). Finally, at roadside sites, PM_{2.5} levels were found to range from 25 to 40 µg/m³ in Spain, Austria and Berlin, 20 to 24 µg/m³ in the Netherlands and the United Kingdom and 13 to 18 µg/m³ in Sweden.

It should be mentioned that in the Netherlands there was very little difference between the PM₁₀ levels registered at urban background and roadside stations (29-37 µg/m³ and 31-37 µg/m³ in 2001, respectively), whereas in most of the examples elsewhere these levels were higher at roadside sites with respect to urban background by 30 to 50%. The PM₁₀ peculiarities described in detail in Annex 5 for the Netherlands may account for these low differences in PM₁₀ levels at urban and roadside sites. The small differences between PM₁₀ levels at regional and urban background and roadside sites might also be explained by the application of a uniform correction factor of 1.33 to all Dutch PM₁₀ data. From field experiments (Van Putten et al., 2002) there are clear indications that for the rural, more agricultural areas with high ammonia concentrations and low organic carbon this factor is too high; in urban area with considerable traffic the factor is too low. In the case that different factors, more representative for each of the different type of sites (for rural sites 1.15; for urban sites 1.45 and for traffic sites most likely even higher (Van Putten et al., 2002)), would be used a similar picture to the other European countries would emerge.

Considering the range of PM₁₀ and PM_{2.5} levels for the different types of stations, the regional, city background, and local traffic contributions to PM₁₀ at most of the EU roadside stations can be estimated to be 35-50, 20-35 and 30-50%, respectively (see also Chapter 4). However, in the Netherlands and Sweden other contributions are found: 50-75, 15-35, 1-20% and 30-65, 5-30, 35-55%, respectively. Similar conclusions can also be drawn for PM_{2.5} levels, given that the above contributions are estimated to be 34-50, 20-35 and 30-40% for most of the examples and 60-70, 5-25, 15-25% and 40-65, 5-30, 20-35% for the Netherlands and Sweden. The long range contribution is found to be stronger in Netherlands and Sweden (anthropogenic sources) and in Spain (African dust). In Sweden the most northerly stations are affected by very local sources.

The PM₁₀ regional background contribution measured at urban background sites may be estimated to be around 50-57% in most of the EU examples and 50-93% in Sweden and the Netherlands. For PM_{2.5} this regional background contribution to the urban background levels varied from 60 (Spain) to 95% (Sweden).

The ratios PM_{2.5}/PM₁₀ varies throughout the different EU regions depending on the type of site. Thus, at regional background sites PM_{2.5}/PM₁₀ were found to range from 0.7 to 0.8. At urban background sites the PM_{2.5}/PM₁₀ ratio ranged from 0.4-0.5 (in Canary Islands and Southern Spain) to 0.8 in the Netherlands, Berlin and Northern and Central areas from Spain. In the United Kingdom, Sweden and

Eastern Iberian Peninsula between 0.6 and 0.7 were measured. Finally, the $PM_{2.5}/PM_{10}$ ratios at roadside sites were usually 0.6-0.7. The lowest ratios (0.4) were obtained at some roadside sites in Sweden and at the Canary Islands, indicating the strength of road dust emissions (road and tyre abrasion, sanding, salting) and the African dust inputs, respectively.

6.3 PM speciation and source apportionment

6.3.1 Mean annual levels

Based on the results on chemical speciation of PM_{10} and $PM_{2.5}$ from the above examples, the following conclusions on source contributions may be drawn:

- The marine contribution to PM_{10} is estimated to be 2 to 4 $\mu g/m^3$ for most of the EU example cases selected. For $PM_{2.5}$, this contribution is down to 0.2 to 0.8 $\mu g/m^3$, but up to 2 $\mu g/m^3$ in southern Sweden.
- The mineral contribution to PM_{10} was very similar for all the EU examples selected with the exception of Spain, where the mineral contribution was usually 2-3 times higher. Thus, at regional sites the mineral contribution to PM_{10} was around 2 $\mu g/m^3$ for all countries with the exception of Spain, where contributions of up to 6 $\mu g/m^3$ were measured. At urban sites the mineral contribution ranges from 3-5 $\mu g/m^3$ in Central Europe and accounted for 7-9 $\mu g/m^3$ in Sweden and 10 $\mu g/m^3$ in Spain. At roadside sites, the mineral contribution accounted for 4-7 $\mu g/m^3$ in Central Europe, 16 $\mu g/m^3$ in Spain and 17-36 $\mu g/m^3$ in Sweden. The relatively high values recorded in Berlin (14 $\mu g/m^3$) are attributed to the fact that the mineral contribution, water and other unaccounted compounds were grouped. A clear differentiation was also evident for the mineral contribution to $PM_{2.5}$ at urban background stations, with contributions from 0.4 to 0.7 $\mu g/m^3$ for most EU examples, 2 $\mu g/m^3$ for the United Kingdom, 1-6 $\mu g/m^3$ for Sweden, and from 2 to 5 $\mu g/m^3$ in Spain, with the lowest levels recorded at regional background sites and the highest at roadside stations.
- The contribution of secondary inorganic aerosols, mainly from industrial, agricultural and traffic emissions, ranged from 6 to 13 and 5 to 11 $\mu g/m^3$ for PM_{10} and $PM_{2.5}$ in almost all the EU examples selected. The concentrations were some what lower, 3-5 $\mu g/m^3$, in Sweden.
- The contribution of organic and elemental carbon (OC+EC) to PM_{10} , mostly arising from traffic and specific industrial emissions, at regional background sites ranged from 1 $\mu g/m^3$ in Sweden, 3 $\mu g/m^3$ in Spain to 7 $\mu g/m^3$ in Austria. At urban background sites this contribution increased for all countries, with the exception of the Netherlands, where a range from 2 to 9 $\mu g/m^3$ was found. At roadside sites, the OC+EC contribution usually ranged from 13 to 21 $\mu g/m^3$, with the exception of Sweden and the Netherlands where it is estimated to be around 5-7 $\mu g/m^3$. The same trend is obtained for the OC+EC contributions in $PM_{2.5}$, with concentrations only around 10-20% below PM_{10} .

6.3.2 Exceedances of the PM_{10} 24-hour limit value

Studies on chemical speciation of PM_{10} during exceedances of the PM_{10} 24-hour limit value (50 $\mu g/m^3$) were only supplied to the PM Working Group by Spain. These studies showed the prevalence of three PM_{10} fractions at the different types of sites studied: mineral dust, secondary inorganic phases and carbonaceous components. At the Canary islands, the mineral fraction and the marine aerosol were clearly dominant during days with limit value exceedance, with mean levels of 36 $\mu g/m^3$ and 20 $\mu g/m^3$ (42 and 24% of the bulk PM_{10} levels), respectively. The proximity of the African continent and the Atlantic island features accounted for the very high natural contribution measured during days exceeding the limit values in this site. At the urban background sites with high industrial influence, the

fraction of secondary inorganic species accounted for 14-25 $\mu\text{g}/\text{m}^3$ of the bulk PM_{10} load for days exceeding the limit value. For days exceeding the limit value at roadside sites with industrial influence, in addition to the secondary inorganic species, mineral dust (24 $\mu\text{g}/\text{m}^3$, 38% of PM_{10} mass) and carbonaceous matter (12 $\mu\text{g}/\text{m}^3$, 19%) also reached high PM_{10} proportions. Finally, at kerbside sites with low industrial influence, it was the carbonaceous (22 $\mu\text{g}/\text{m}^3$) and mineral (22 $\mu\text{g}/\text{m}^3$) fractions that constituted 62% of PM_{10} in days exceeding the limit value, given that traffic was the main contributor to PM for those days.

The number of annual exceedances recorded simultaneously with African dust outbreaks over the Spain ranged from 63 at traffic influenced sites from Las Palmas to 12 at the urban and background sites from Central and Northern Spain.

By use of validated models and measured emission factors, the air pollution situation in Stockholm has been shown to strongly depend on long range transport and local suspension of road dust. It has a strong seasonal dependence with the maximum during February – May and is manifested by a high number of PM_{10} exceedances, more than 100 per year along the major roads inside and outside the city.

Modelling studies carried out in Oslo by the NILU indicated that wood burning (in small stoves) and road dust resuspension are the main contributors to PM_{10} exceedances in this northern city (S. Larsen, personal communication).

6.3.3 Source apportionment

Based on the above results the following quantitative estimations on source apportionment may be obtained:

- The annual mean marine contribution reached 2-4 $\mu\text{g}/\text{m}^3$ for most of the examples considered but, occasionally, it accounted for up to 7 $\mu\text{g}/\text{m}^3$ in coastal areas in the Netherlands and for up to 11 $\mu\text{g}/\text{m}^3$ on the Canary Islands. Furthermore, mean regional mineral contribution to PM_{10} (mainly of a natural origin) ranged from 5.5 $\mu\text{g}/\text{m}^3$ in the Eastern and Southern Iberian Peninsula to around 2 $\mu\text{g}/\text{m}^3$ for the remaining areas, with the exception of the Canary Islands (8 $\mu\text{g}/\text{m}^3$). Therefore it can be deduced that, as a mean, the natural input to PM_{10} in Europe varies from $\approx 8 \mu\text{g}/\text{m}^3$ in Spain to $\approx 4 \mu\text{g}/\text{m}^3$ in most of Northern and Central Europe. In $\text{PM}_{2.5}$ the marine and natural mineral regional contributions are significantly lower compared with PM_{10} , but it is still present in that fraction: around 3 $\mu\text{g}/\text{m}^3$ in Spain and $<1.5 \mu\text{g}/\text{m}^3$ in Central Europe. Sporadically, African dust outbreaks over Southern EU induce very high PM_{10} and $\text{PM}_{2.5}$ levels. Up to 70 $\mu\text{g}/\text{m}^3$ 24-hour $\text{PM}_{2.5}$ levels with more than 80% being mineral dust have been measured for some of those events over Spain.
- At urban background sites, measurements of the mineral city background contribution attributed to demolition, construction and road dust resulted in 5 $\mu\text{g}/\text{m}^3$ in Spain and Sweden, and 2-4 $\mu\text{g}/\text{m}^3$ in the remaining areas in Europe. In $\text{PM}_{2.5}$ this contribution was slightly higher in Spain and Sweden (1 $\mu\text{g}/\text{m}^3$) than in the other areas (0.4-0.6 $\mu\text{g}/\text{m}^3$).
- The local mineral contribution at road sites, which corresponds to the road dust emissions from the street where the monitoring station is located, accounted for 9-24 $\mu\text{g}/\text{m}^3$ in Sweden, 6 $\mu\text{g}/\text{m}^3$ in Spain and for 1-5 $\mu\text{g}/\text{m}^3$ for the rest of the examples considered. In $\text{PM}_{2.5}$ this contribution ranged from a mean of 2 $\mu\text{g}/\text{m}^3$ in Spain and Sweden to $<0.5 \mu\text{g}/\text{m}^3$ in the remaining examples. These differences in levels of crustal components may be attributed largely to the higher soil-resuspension effect during dry conditions in the Southern EU, while higher rainfall in Northern EU may result in higher frequency of wet road surfaces, resulting in lower dust emissions, and contribute to cleaning the road dust from streets. However, measurements in Sweden showed quite high road dust emission, giving 9-24 and 2 $\mu\text{g}/\text{m}^3$ contributions to PM_{10} and $\text{PM}_{2.5}$ annual mean

values, respectively, which was a result of the use of studded tires and sanding of roads during the winter and spring period.

- EC+OC regional contribution to PM_{10} ranged for most of the examples considered from $1 \mu\text{g}/\text{m}^3$ in Sweden through $2.5 \mu\text{g}/\text{m}^3$ in Spain to $5 \mu\text{g}/\text{m}^3$ (Berlin). The urban background EC+OC contribution, mainly related to traffic emissions varied from 1 (Sweden), 5 (Spain) to 8 (Berlin) $\mu\text{g}/\text{m}^3$. The local contribution of EC+OC at kerbside sites, related to traffic exhaust emissions, tyre and pavement abrasion, reached levels from 3 (Sweden), 6 (Spain) to 9 (United Kingdom) $\mu\text{g}/\text{m}^3$. Given that OC+EC are mainly accumulated in the $PM_{2.5}$ fraction, their contributions to $PM_{2.5}$ followed a similar pattern to that described for PM_{10} .
- The contribution of secondary inorganic phases (arising from traffic, industrial emissions including power generation and agriculture) reached levels from 3 to 9 $\mu\text{g}/\text{m}^3$ and 3 to 8 $\mu\text{g}/\text{m}^3$ at regional sites for PM_{10} and $PM_{2.5}$, respectively. In areas with high industrial influence there was an input from 2 to 5.5 $\mu\text{g}/\text{m}^3$ and 1 to 5 $\mu\text{g}/\text{m}^3$ of secondary inorganic aerosols, on top of the above contributions.

Studies of source apportionment carried out in Sweden demonstrated that a fraction of 8-16 $\mu\text{g}/\text{m}^3$ PM_{10} and 7-13 $\mu\text{g}/\text{m}^3$ $PM_{2.5}$ was attributable to long-range transport of foreign anthropogenic emissions. The range reflects lower values found in the north and higher ones in the south. Similar results were obtained in the Netherlands, where the transboundary contribution to PM_{10} was estimated to be 11 $\mu\text{g}/\text{m}^3$.

The studies on source apportionment analysis carried out demonstrated that at kerbside sites the major contribution sources are: traffic (including exhaust and abrasion material): 40-55% of PM_{10} and 45-60% of $PM_{2.5}$; and industry: 12-15% of PM_{10} and around 20% of $PM_{2.5}$. The value for traffic for $PM_{2.5}$ was somewhat lower in Sweden 22-37% due to relatively low emissions and the high fraction of long-range transport and resuspension.

Concerning the natural input, quantitative data from chemical speciation has only been supplied to the PM Working Group by Spain. In this Member State, on average, the natural input reached 17, 24 and 38% of PM_{10} , and 11, 16 and 26% of $PM_{2.5}$ measured respectively at kerbside sites, urban background sites and regional background sites of Spain. In some places such as the Canary Islands these contributions may even reach higher proportions (40-65%). Peak values up to 300 and 60 $\mu\text{g}/\text{m}^3$ PM_{10} and $PM_{2.5}$ daily levels attributable to African dust outbreaks have been reported in several Southern EU air quality monitoring stations. Also very high contributions of marine aerosols to PM_{10} may be reached in coastal areas of central and Northern EU (up to 7 $\mu\text{g}/\text{m}^3$ annual contributions have been reported for the Netherlands).

6.4 Conclusions and recommendations

6.4.1 Conclusions

Most of the conclusions summarised below apply for urban areas of EU. At industrial hot spots the source contribution to PM_{10} and $PM_{2.5}$ may vary considerably depending on the industrial process and the distance to the emission source. The conclusions obtained for traffic hot spots may apply to most of EU urban areas due to the similarity of the emission sources. Most of the differences observed among traffic hot spots may be attributable to traffic intensity but also to climatic patterns such as frequency of rain (due to both atmospheric scavenging and the washing efficiency of road dust during rain episodes) or dispersive conditions.

Keeping the above considerations in mind, the main conclusions of this chapter can be summarised as follows:

1. *Natural PM*

Marine aerosols and natural dust contributions from arid or semi-arid areas were found to be the major sources of natural PM. Other natural sources such as natural inorganic secondary species or natural biological PM had a lower contribution to the mean annual PM_{10} and $PM_{2.5}$ bulk mass concentration measured at different sites from EU. The marine contribution to PM_{10} reached 2-4 $\mu\text{g}/\text{m}^3$ on an annual average for most studies available in EU, but occasionally, it accounted for up to 7 $\mu\text{g}/\text{m}^3$ in coastal areas in the Netherlands and for up to 11 $\mu\text{g}/\text{m}^3$ on the Canary Islands. Mean natural mineral contribution to PM_{10} ranged usually from 2 to 6 $\mu\text{g}/\text{m}^3$. Thus, as a mean, the natural input to PM_{10} in Europe varied from 3 to 8 $\mu\text{g}/\text{m}^3$ in most EU regions, with the highest values recorded in Southern EU. In $PM_{2.5}$ the marine and mineral regional contributions were significantly lower compared with PM_{10} , but it is still present in a concentration range from 1-2 $\mu\text{g}/\text{m}^3$ in Central EU to 3-4 $\mu\text{g}/\text{m}^3$ in Southern and Northern EU. Peak values up to 300 and 70 $\mu\text{g}/\text{m}^3$ for daily levels of respectively PM_{10} and $PM_{2.5}$, attributable to African dust outbreaks, have been reported in several Southern EU air quality monitoring stations.

2. *Anthropogenic mineral dust*

Concerning major anthropogenic sources, urban mineral dust ranged from 2 to 4 $\mu\text{g}/\text{m}^3$ in PM_{10} and from 0.5 to 1 $\mu\text{g}/\text{m}^3$ in $PM_{2.5}$ at urban background stations. At road side sites, road dust accounted for an additional fraction of 3 to 6 $\mu\text{g}/\text{m}^3$ in PM_{10} and from <0.5 to 2 $\mu\text{g}/\text{m}^3$ of $PM_{2.5}$ in most regions of EU. The highest mean annual mineral dust contributions were reported for Southern EU. These differences in levels of crustal components may be attributed largely to the higher soil-resuspension effect during dry conditions in the Southern EU, while higher rainfall in Northern EU may contribute to reduce resuspension by washing road dust from the road pavement. In Nordic countries, road wear and sanding of roads substantially contributed to PM_{10} and $PM_{2.5}$ annual mean values, giving about 5 and 1 $\mu\text{g}/\text{m}^3$ at urban background sites and 10-25 and 3 $\mu\text{g}/\text{m}^3$ at road sites, respectively. The main problem in Sweden (and probably also in Norway and Finland) is not the yearly mean value, but rather the strong seasonal variation, with numerous exceedances (over 100 in the large cities) during winter and spring period.

3. *Carbonaceous PM*

Carbonaceous PM (both organic matter and elemental carbon), in urban areas mainly emitted by traffic, usually ranged from 7 to 13 $\mu\text{g}/\text{m}^3$, both in PM_{10} and in $PM_{2.5}$. At roadside sites, an additional 6 to 9 $\mu\text{g}/\text{m}^3$ input was usually observed both in PM_{10} and $PM_{2.5}$. Sweden is not included in this range; there, considerable lower inputs of 1 and 3 $\mu\text{g}/\text{m}^3$ at urban background and roadside sites were found.

4. *Secondary inorganic aerosol*

The contribution of secondary inorganic phases (arising from traffic, industrial emissions including power generation and agriculture) reached levels from 4 to 9 $\mu\text{g}/\text{m}^3$ and 4 to 8 $\mu\text{g}/\text{m}^3$ at most regional sites for PM_{10} and $PM_{2.5}$ respectively. In areas with high industrial influence there was often an input from 2 to 5.5 $\mu\text{g}/\text{m}^3$ and 1 to 5 $\mu\text{g}/\text{m}^3$ of secondary inorganic aerosols, on top of the above contributions for PM_{10} and $PM_{2.5}$.

5. *Long-range transport*

The importance of long range transport of anthropogenic pollutants is manifested in concentrations found in the Swedish regional background stations, ranging from 8 to 16 $\mu\text{g}/\text{m}^3$ PM_{10} and 7 to 13 $\mu\text{g}/\text{m}^3$ $PM_{2.5}$ (annual averages). The higher values were for the southern parts and the lower for the northern parts of Sweden. This implies a significant fraction of long range transported PM also for other European sites. Furthermore, the African dust outbreaks affecting Southern EU accounted for the increased concentrations of mineral dust in PM_{10} , but also in $PM_{2.5}$. These dust contributions had a major impact on the exceedances of the 24-hour limit value but also in the increased PM_{10} and $PM_{2.5}$ mineral background levels with a clear influence on the annual PM values.

6. *Traffic hot spots*

At roadside stations in most examples, local traffic accounted for the major contribution (including exhaust and abrasion products), with 40 to 55% of the annual PM_{10} levels and 45-60% of $PM_{2.5}$.

6.4.2 Recommendations

1. In rural areas, secondary inorganic species account for the largest contribution of PM_{10} and $PM_{2.5}$, both about 35-55%. Second in mass is elemental and organic carbon, accounting for about 15-35% of PM_{10} and 17-40% of $PM_{2.5}$. To reduce regional background concentrations, PM emissions abatement strategies should therefore focus on reducing primary carbonaceous compound emissions and gaseous precursors of secondary components of PM.
2. Road traffic is a major emissions source contributing to PM_{10} and $PM_{2.5}$ concentrations close to roads. Abatement strategies to reduce PM concentrations (especially concerning organic carbon, elemental carbon and mineral dust) close to roads should therefore focus on road traffic.
3. Natural PM sources can contribute significantly to $PM_{2.5}$, though less than to PM_{10} . It is recommended to include derogations for exceedances of a possible future $PM_{2.5}$ limit value due to natural sources.
4. It is recommended to develop PM source apportionment further for differentiation of sources such as diesel, gasoline and wood combustion to help target better abatement strategies.

6.5 References

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Table 6.1. Mean annual levels ($\mu\text{g}/\text{m}^3$) of PM_{10} , $\text{PM}_{2.5}$, organic and elemental carbon (EC+OC), mineral elements, marine contribution and secondary inorganic aerosols (SIA) recorded in regional background, urban background and roadside stations in different countries in the EU.

	PM_{10}					$\text{PM}_{2.5}$				
	Total	EC+OC	Mineral	Marine	SIA	Total	EC+OC	Mineral	Marine	SIA
AUSTRIA										
Regional background	24	7	1	2	9	20	7	0.4	0.2	8
Urban background	--	--	--	--	--	--	--	--	--	--
Roadside sites	53	21	4	2	11	38	17	0.7	0.6	9
BERLIN										
Regional background	19	5	3 ¹⁾	2	8	--	--	--	--	--
Urban background	28-38	8	5 ¹⁾	2	13	22-30	7	3*	1	11
Roadside sites	37-51	13	14 ¹⁾	2	13	25-39	10	3*	1	11
SPAIN										
Regional background	21	3	6	2	7	14	3	2	0.7	5
Urban background	31-42	7	10	2	7	19-25	6	3	0.7	8
Roadside sites	45-50	13	16	2	9	28-35	10	5	0.7	9
SWEDEN										
Regional background	8-16	1-2	2-4	1-4	3-5	7-13	1-2	1-3	1-3	3-5
Urban background	17-23	2-3	7-9	1-4	3-5	8-15	2-3	2-4	1-3	3-5
Roadside sites	26-51	5-6	17-36	1-4	3-5	13-18	5-6	4-6	1-3	3-5
THE NETHERLANDS										
Regional background	22	5	2	4	6	14	3	0.6	0.6	6
Urban background	25	5	3	4	10	20	5	0.6	0.6	9
Roadside sites	30	7	6	4	10	20	7	0.7	0.6	9
UNITED KINGDOM										
Regional background	--	--	--	--	--	--	--	--	--	--
Urban background	25	9	5	2	8	16	8	2	0.3	6
Roadside sites	35	18	7	2	8	24	16	2	0.3	6
SWITZERLAND										
Regional background	14-24	4.5	2	0.2-0.5	7	18	4	1	0.2-0.5	8
Urban background	24	6	2	0.2-0.5	9	19	5	1	0.2-0.5	8
Roadside sites	42	13	6	0.2-0.5	10	22	8	1	0.2-0.5	7

¹⁾ Unaccounted mass: mineral matter + water + other unaccounted components.

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Table 6.2 Mean annual levels of PM_{10} , $PM_{2.5}$, organic and elemental carbon (EC+OC), mineral elements, marine contribution and secondary inorganic aerosols (SIA) recorded in regional background, urban background and roadside stations in Central, Northern and Southern EU based on the data reported from the examples from Austria, Berlin, Spain, Sweden, Netherlands, United Kingdom, Switzerland.

		Central EU (based on examples from Austria, Berlin, Switzerland, Netherlands, United Kingdom)			Northern EU (based on examples from 13 sites in Sweden)			Southern EU (based on examples from 10 sites in Spain)		
		Regional background	Urban background	Roadside sites	Regional background	Urban background	Roadside sites	Regional background	Urban background	Roadside sites
		In $\mu\text{g}/\text{m}^3$								
PM ₁₀	Total	12-24	24-38	30-53	8-16	17-23	26-51	14-21	31-42	45-50
	EC+OC	4-7	6-9	13-21	1-2	2-3	5-6	2-5	4-9	10-18
	Mineral	1-2	3-5	4-8	2-4	7-9	17-36	4-8	8-12	10-18
	Marine	2-4*	2-4*	2-4 ¹⁾	1-4	1-4	1-4	2-4 ²⁾	2-4 ²⁾	2-4 ²⁾
	Secondary	7-9	7-13	8-13	3-5	3-5	3-5	5-9	6-11	6-11
PM _{2.5}	Total	12-20	16-30	22-39	7-13	8-15	13-19	12-16	19-25	28-35
	EC+OC	3-7	5-8	8-16	1-2	2-3	5-6	2-4	4-8	8-12
	Mineral	0.5-2	0.4-2	1-2	1-3	2-4	4-6	1-3	2-5	4-6
	Marine	0.2-1	0.2-1	0.2-1	1-3	1-3	1-3	0.2-1	0.2-1	0.2-1
	Secondary	6-8	6-11	7-11	3-5	3-5	3-5	4-8	7-10	6-10
		In %								
PM ₁₀	EC+OC	30-35	20-30	40-45				15-25	12-25	25-37
	Mineral	5-10	10-15	12-15				12-40	25-30	25-37
	Marine	5-20	5-12	5-8				5-20	5-10	3-8
	Secondary	35-55	30-35	25-28				35-45	20-27	13-25
PM _{2.5}	EC+OC	30-40	25-35	35-45				17-30	20-35	30-40
	Mineral	2-8	2-8	5				8-20	10-20	10-15
	Marine	2-5	1-3	1-2				2-5	1-3	1-2
	Secondary	35-55	35-40	27-35				17-30	20-35	30-40

¹⁾ 7 $\mu\text{g}/\text{m}^3$ in coastal areas of the Netherlands

²⁾ 11 $\mu\text{g}/\text{m}^3$ in the Canary Islands

7. Trends and projections

7.1 Scope

This chapter synthesises information on historic and projected trends of PM₁₀ concentrations in Europe. It attempts to quantify the trends and projected baseline emissions and ambient levels in Member States in 2005 and 2010 for PM₁₀. It investigates whether projections for alternative scenarios with additional abatement exist. Furthermore, the chapter discusses the extent to which expected trends in concentrations differ between the different spatial scales (rural/large scale, urban background, busy streets). It also identifies uncertainties in projections, including the major factors that determine these uncertainties. The data presented in this chapter are mainly on PM₁₀. Due to the scarcity of monitoring data on PM_{2.5}, an analysis of trends or sufficient validation of PM_{2.5} modelling is not yet possible. The uncertainty in PM_{2.5} projections is considered too high to justify an evaluation of projected future PM_{2.5} concentrations.

7.2 Introduction

To understand the likely future pollution climate for PM it is important to understand historic trends in PM emissions and concentrations. Historic trends are the starting point for projections. Projections of future ambient concentrations of particulate matter (PM) help policy makers assess the attainability of current limit values or alternative targets. Projections also help assess the likely impact of abatement measures on future PM concentrations.

It is desirable to model future concentrations at both the large and smaller geographic scale. Larger scale modelling provides projections of average background levels of PM over a wide area. These are useful to help estimate the future exposure of the population as a whole to PM. Europe-wide concentration projections have been generated by EMEP and REM3-models (Stern, 1994; Hass et al. 1997). Most Europe wide models calculate future 'background' concentrations (average concentration in grids of 30x30 or 50x50 km²). These models average the impact of PM sources over a wide area.

Additionally, it is important to be able to project PM concentrations at a smaller scale (cities or smaller) to better understand the (1) the average future exposure of people living in urban areas where there tend to be higher emissions of PM and (2) the impact of industrial point sources or traffic on concentrations at a local level.

Both large and small-scale projections are important to help assess the likely attainability of the current limit values and the potential impacts of abatement measures on PM concentrations.

Two Member States (the United Kingdom and the Netherlands) and Switzerland have completed national projections of ambient concentrations of PM. Similar studies for Germany and Denmark are underway. All projections are uncertain. Chapter 3 concludes that validation of the results of current large scale models against PM measurement data is lacking and so is a priority.

Projections are available for some cities in Europe but modelling methods and assumptions differ widely making comparisons difficult. The European City Delta project is currently carrying out PM concentration projections for a number of European cities using comparable methods and assumptions. Results from City Delta are not available at the time of writing but these will also provide a valuable addition to assessment of attainability of the limit values.

This chapter presents a number of projections on a European scale. In this context 'European' means that projected emissions in the whole of Europe are taken into account and not just in one country,

though the model output can be restricted to one country. It also presents the key results from the few available national projections.

7.3 Historic trends in ambient PM concentrations

Analysis of PM data until 2001

Reliable and representative concentration data from a substantial number of stations are mainly available for PM₁₀ in AirBase¹⁵ since 1996/1997. The number of annual time series has increased from 106 in 1996, to 193 in 1997, steadily to 532 in 2000 and with a large step to 909 stations in 2001. Thus, a time series of 5 years is available (6 years for some stations in some countries).

Figure 7.1 shows the annual average and 36th highest 24-hour value per year (corresponding to the limit value definition of the EU Directive) from 1997 to 2001, as mean value for each of the following station classes: rural, urban background and street (mainly traffic influenced). Only stations with at least 4 years of data have been included.

Figure 7.1 shows that on average, for the stations involved in this analysis, there was a downward tendency between 1997 and 1999, and a slight increase between 1999 and 2001. The reduction over the whole period was of the order of 20%. Further data will be needed to confirm whether this reflects a change in emissions or the natural variation from year to year due to meteorology.

Table 7.1 shows the percentage of monitoring stations with downward and upward trends (significant or non-significant, according to the Mann-Kendall test, at 90% confidence level). For the period 1997-2001, there was a non-significant reducing trend at close to 50% of the stations. There was a significant reducing trend at about 30% of stations; and there was a non-significant increasing trend at about 20% of the stations. The latter are mostly urban background and street stations.

Table 7.1 Historic trends in PM₁₀ concentrations, 1997-2001. Source: ETC/ACC.

Statistic	Number of stations	Reducing trend % of stations		Increasing trend % of stations	
		Significant	Non-significant	Significant	Non-significant
Annual average					
Total stations included	183	27%	53%	2%	18%
Urban stations in trend-line	73				
Street stations in trend line	40				
Rural stations in trend line	31				
36th highest 24-hour					
Total stations included	193	30%	45%	2%	23%
Urban stations in trend-line	79				
Street stations in trend line	43				
Rural stations in trend line	30				

¹⁵ <http://etc-acc.eionet.eu.int/databases/airbase.html>

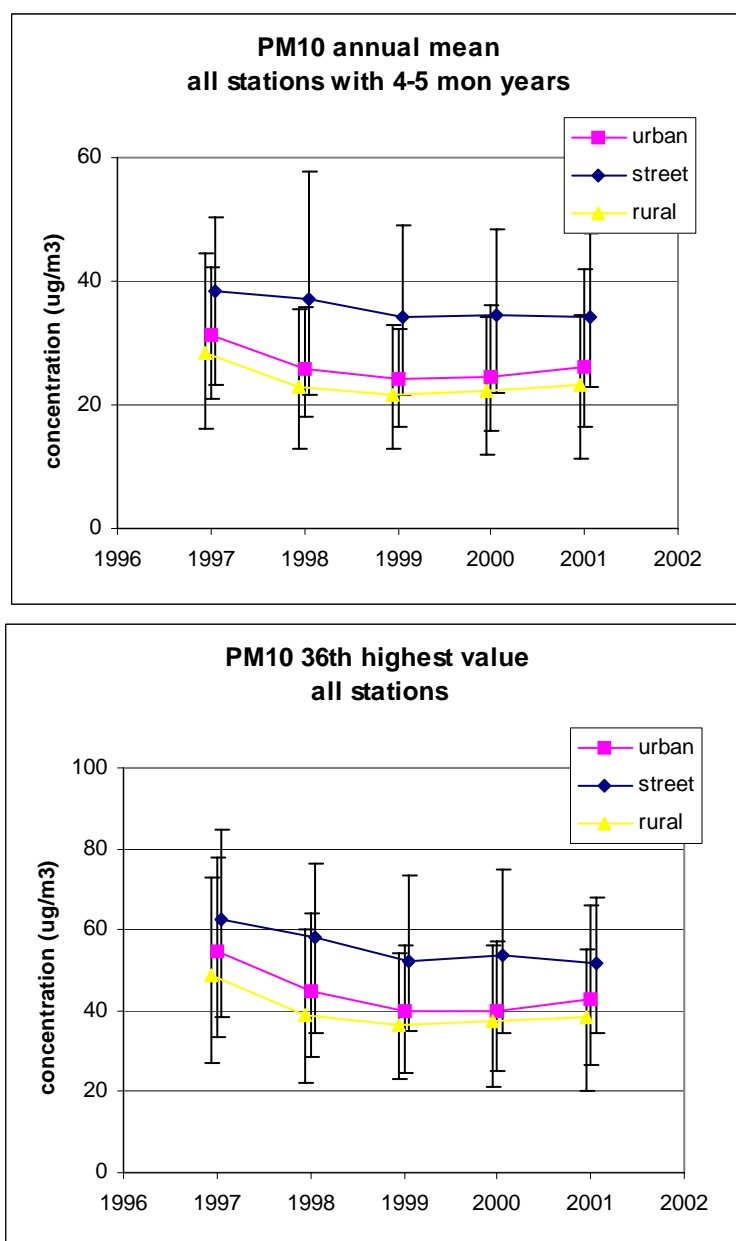


Figure 7.1 PM_{10} , inter-annual variations, 1997-2001. Annual average, and 36th highest 24-hour value. Average over all stations with reported data, per station type. The vertical bars indicate the 10th and 90th percentiles. Source: ETC/ACC.

The number of stations with data available for such a trend analysis is limited to about 190 stations of various types. Figure 7.2 shows the location and type of these stations on a map. 11 countries are represented in these trend lines, with many stations in Belgium, the Netherlands, the Czech Republic, Spain, Switzerland and the United Kingdom. This sample of countries and stations reflect the extent of reporting by the countries of historic data to AIRBASE. Most stations record a reducing trend; but many also record an increasing trend, so the overall picture is not uniform over Europe. Also, the influence of interannual meteorological variations has not been accounted and corrected for in this analysis. Thus, even if the PM_{10} concentrations have decreased over the period 1997-2001, as an average over the available stations, a clear conclusion as to the Europe-wide extent and significance of this downward trend cannot yet be drawn from the AIRBASE data.

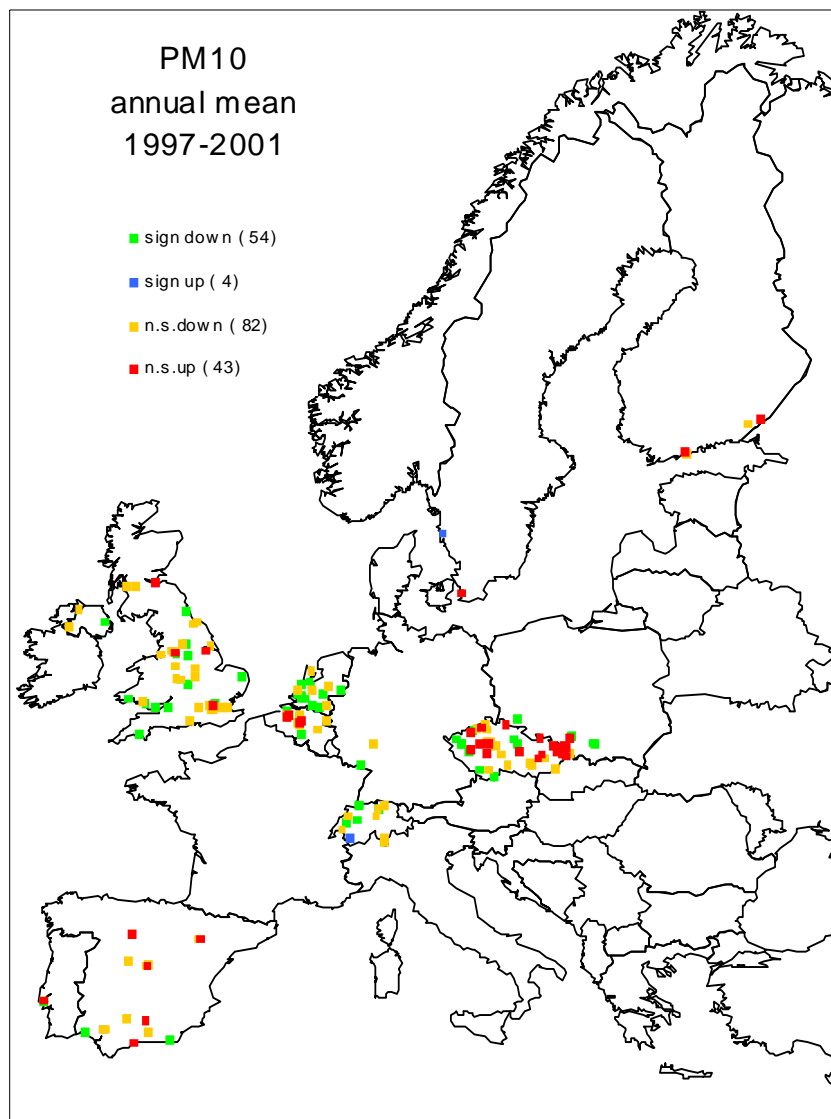


Figure 7.2 *PM₁₀ stations with at least 4 years of data during 1997-2001. The map shows location and significance of trends at each station.*

Apart from showing the significance of the rural PM₁₀ concentrations compared to the concentrations in urban areas, Figure 7.1 shows that the average changes from year to year are almost the same for urban and rural stations, and the street stations also follow the rural trend fairly closely. This indicates that the urban background (as well as the traffic hot spot) PM₁₀ concentrations are influenced strongly by the PM₁₀ mass entering urban areas. Consequently abatement measures aimed at reducing rural background PM concentrations also benefit urban and even street PM₁₀ levels. Even so, particularly the street level PM₁₀ concentrations are considerable higher than the rural (and urban) background levels, indicating that local PM emission contributions and their control are important as well. This is discussed in more detail in Chapter 8 on abatement.

The relative importance of secondary inorganic particles originating from precursor gases (SO₂, NO_x, NH₃) can be evaluated from Figure 7.3. The measurements of particulate sulphate at EMEP stations show a decreasing trend since 1996 to 2000. (Preliminary analysis of 2001 data shows an increase from 2000). For 27 stations in 10 countries (DK, FI, ES, FR, UK, CH, NL, PL, SK, NO) with full data sets for 1996-2000, the annual average sulphate concentration (measured as S) was reduced by 0.43 µg/m³, or about 38% since 1996. Figure 7.3 shows that the degree of reduction varies between areas in Europe. For nitrate the Europe-wide data set is much smaller than for sulphate. Section 5.6 on emissions of PM precursor gases (SO₂, NO_x and others) show that SO₂ and NO_x emissions in Europe

were reduced by about 18% and 12% respectively between 1996 and 2000. The measured sulphate-S concentration reduction 1997-2000 translates to roughly to about $1.7 \mu\text{g}/\text{m}^3$ as e.g. ammonium or calcium sulphate. The reduction of the nitrate part of PM can be expected to be of about the same magnitude as for sulphate. The PM reduction corresponding to the sulphate and nitrate reduction is thus of the same magnitude as the measured reduction and annual changes in rural (as well as urban) PM_{10} . Thus, the reduction in secondary inorganic aerosol is large enough to make up a significant part of the PM_{10} reductions. The interannual changes in PM_{10} is also, though, influenced by other parameters, such as meteorological conditions.

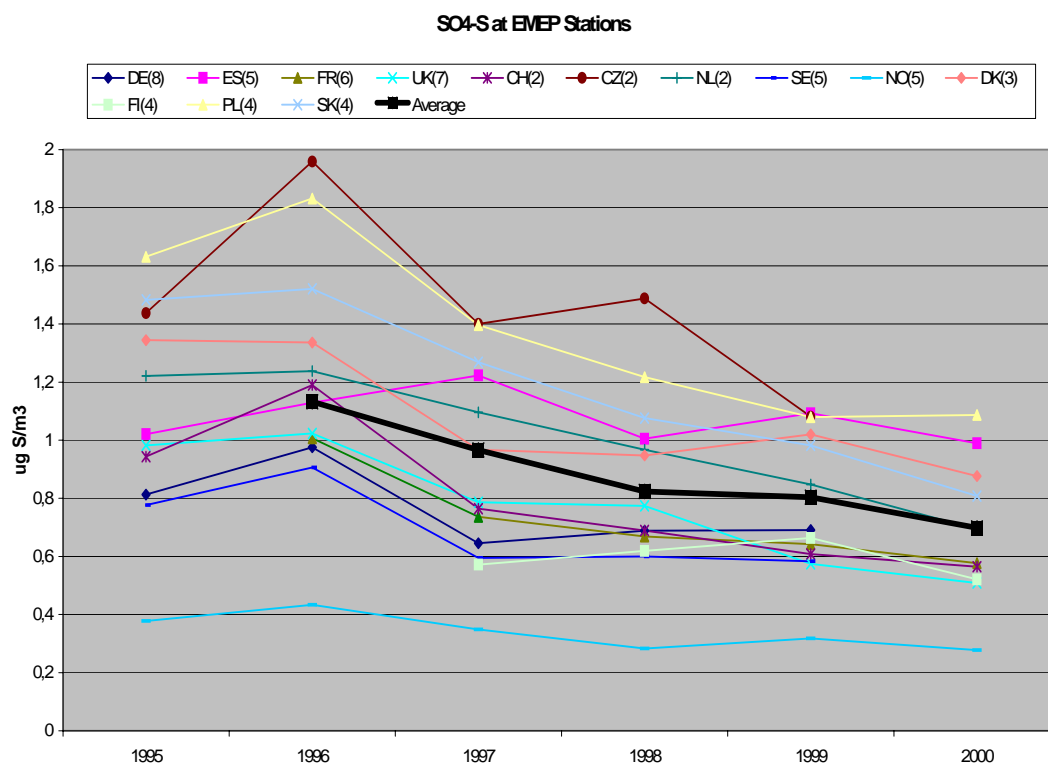


Figure 7.3 Sulphate-S measurements at EMEP stations. The "average" line represents 27 stations in the 5 countries with full data coverage from 1996 to 2000. Source EMEP/CCC.

To broaden the basis for evaluating the significance of the changes in PM_{10} , Figure 7.4 shows average PM_{10} changes compared with changes for other compounds, namely SO_2 , NO_2 and ground level ozone, also based upon data in AIRBASE.

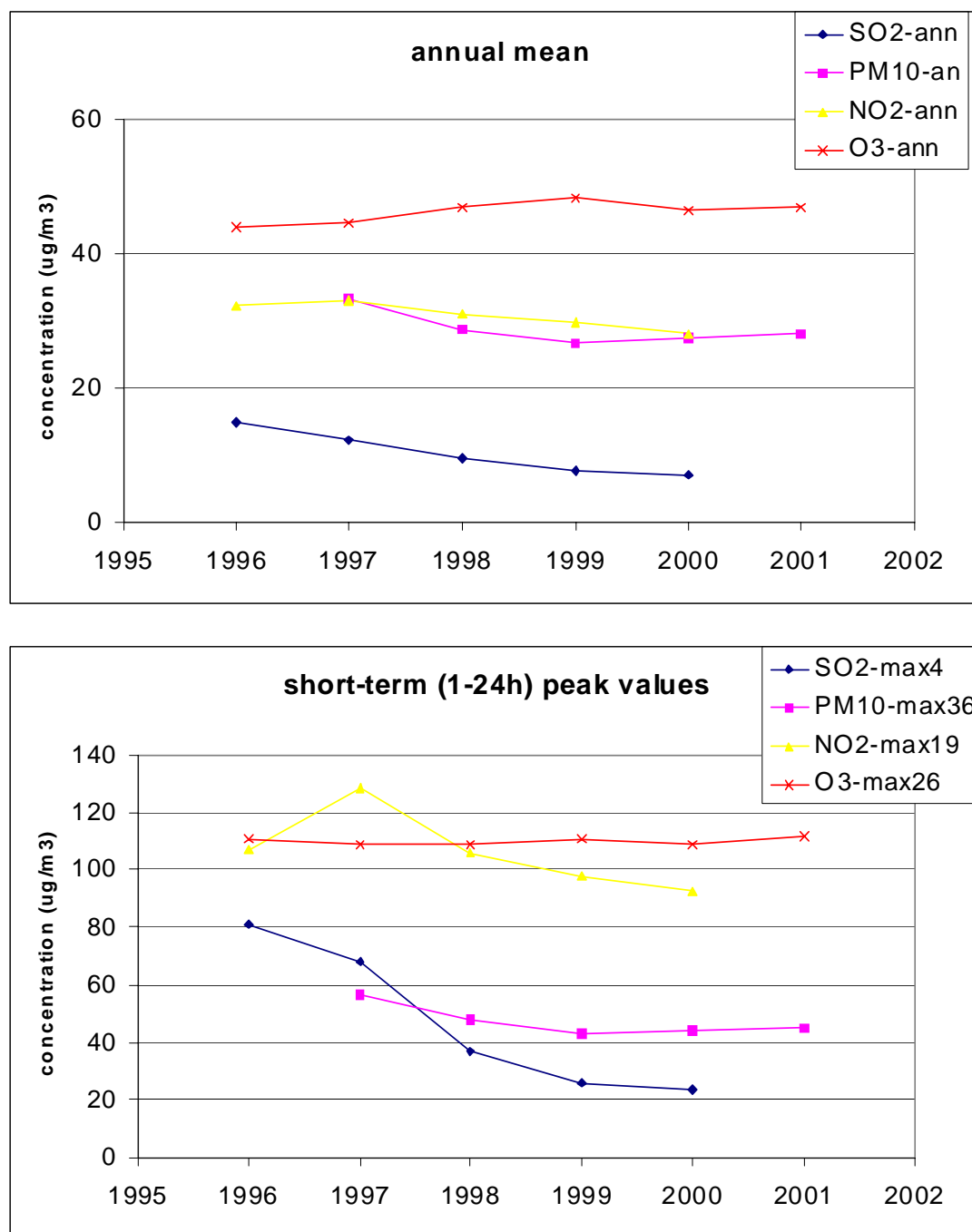


Figure 7.4 Summary of changes in measured concentrations of SO_2 , NO_2 , PM_{10} and ozone in Europe, all stations. Annual mean and high short-term values. $\text{SO}_2\text{-max4}$: 4th highest 24-hour average SO_2 concentration; $\text{PM}_{10}\text{-max36}$: 36th highest 24-hour average SO_2 concentration; $\text{NO}_2\text{-max19}$: 19th highest hourly average NO_2 concentration; $\text{O}_3\text{-max26}$: 26th highest 24-hour 8-hour max O_3 concentration.

Figure 7.4 includes data for 1997-2001 (except for NO_2 , where 2001 data still have not been included); all station types have been aggregated. The reducing trend for PM_{10} is similar to that for NO_2 , up to 2000. SO_2 shows an even stronger downward trend, while ozone shows a stable (short term peaks) or increasing (annual average) concentration level.

Figure 7.5 (1997-2000 data, all station types aggregated) shows that many European countries have a similar reducing PM_{10} concentration trend since 1997. Spain, however, has an almost unchanged level since 1997. (In some other countries, the number of stations with data reported to AIRBASE is very

small, so the reported development in PM₁₀ level may not be representative for the general situation in the country.) In all countries, the levels were largely unchanged from 1999 to 2000, and even a small increase is indicated.

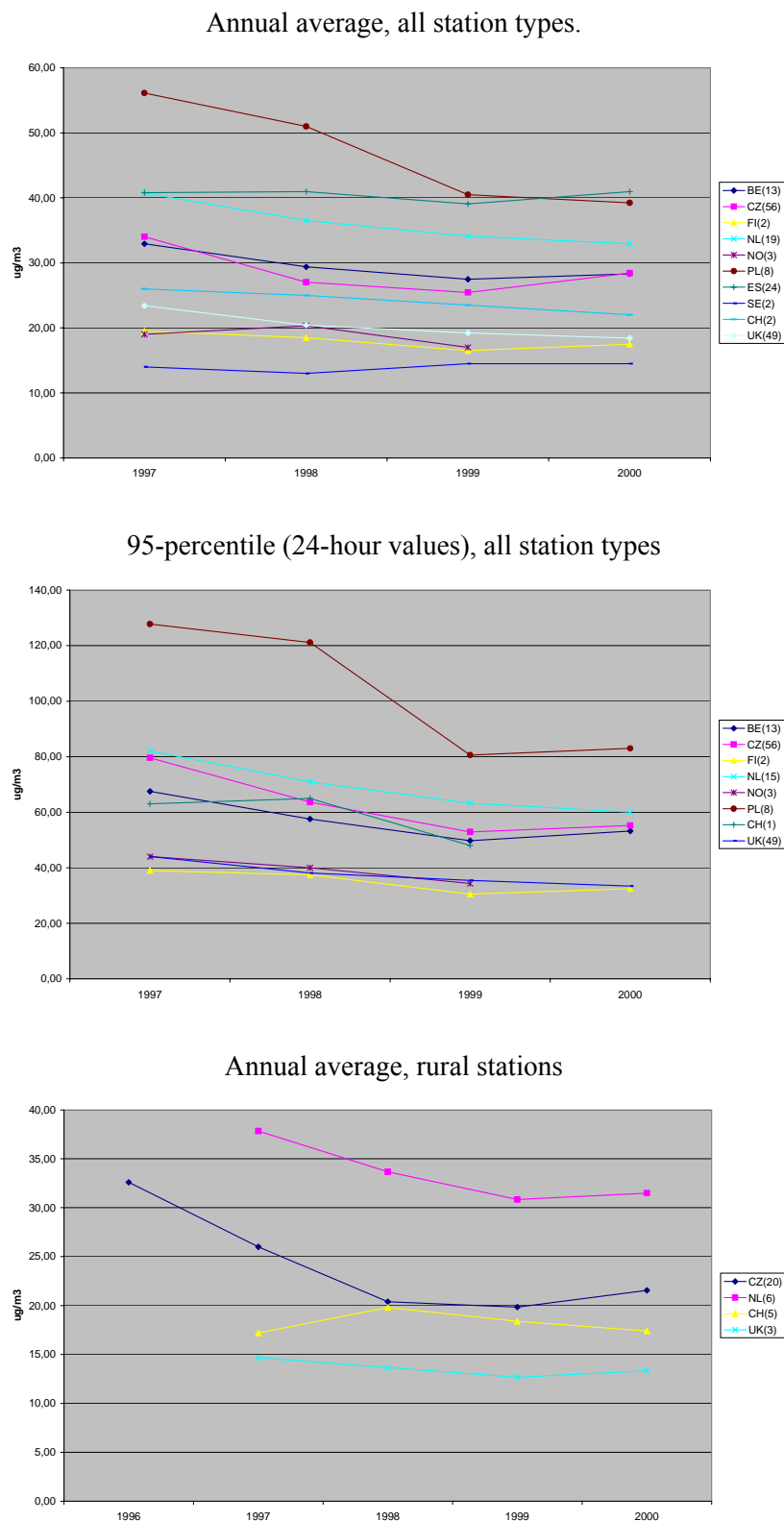


Figure 7.5 PM₁₀ trends in some European countries, 1997-2000 (number of stations in parenthesis). Source: ETC/ACC

For rural areas there are only four countries with time series in AirBase since 1996/1997 (CH, CZ, NL, UK). No tendency can be seen for Switzerland and the United Kingdom, while rural PM₁₀ has decreased in the Czech Republic and the Netherlands. Similar to the urban situation, rural PM₁₀ has increased somewhat from 1999 to 2000.

The 95th percentile of 24-hour values (corresponding approximately to the 18 highest day annually) shows a stronger decrease since 1997 of about 28%, although there has been little change from 1999 to 2000. This suggests that episodic high PM₁₀ concentrations are being reduced more effectively than the annual averages.

Annex 6 presents further data provided by countries.

Recent data on 2002 and 2003

There is limited evidence from some Member States on recent PM₁₀ trends up to 2003. Two examples are presented in Figure 7.6, which show monthly moving PM₁₀ annual averages from 5 stations in Germany and 4 stations in Spain. The German data were measured with the manual reference method and thus are not influenced by any changes in monitoring techniques or correction factors.

Figure 7.6 corroborates the finding that recent PM₁₀ trends are not uniform in Europe. The Spanish stations exhibit a more or less ongoing decrease of PM₁₀ levels since 1996, perhaps with a stabilisation at urban stations in 2002 and 2003. PM₁₀ levels at German stations show a net increase in 2002 and particularly in 2003, with a minimum in 2001. Taking the whole period from 1998 onwards, no clear trend is apparent.

The net increase of PM₁₀ levels in 2002 and 2003 seen at the German stations has also been observed in Belgium (Flandres), Switzerland (Straehl, 2004), Denmark and parts of the Czech Republic, especially in Silesia, where recent PM₁₀ levels are now comparable again with those measured in 1996 (<http://www.chmi.cz/uoco/isko/groce/gr02e/aobsah.html>). It thus seems that the recent increase of PM₁₀ levels at German stations presented in Figure 7.6 may be representative for the situation in parts of Western and Central Europe. However, one should be very cautious in drawing conclusions from changes over short time periods, because interannual variability of PM₁₀ levels due to favourable or unfavourable dispersion conditions may mask the structural trend.

Consequently, it is an important but open question whether the recent increase of PM₁₀ levels in some countries in Western and Central Europe is only due to meteorological conditions or also to some extent to changed emissions. Emissions inventories for 2002 and 2003 are not available at the time of writing. Adverse dispersion conditions with long lasting periods of stagnant air masses particularly in 2003 definitely play an important role in those countries. This is corroborated by rising levels of other pollutants such as NO₂ in 2003. At least in the Czech Republic and Poland, however, there are also some indications for again rising emissions from industry due to increased production of coke and iron and from domestic heating.

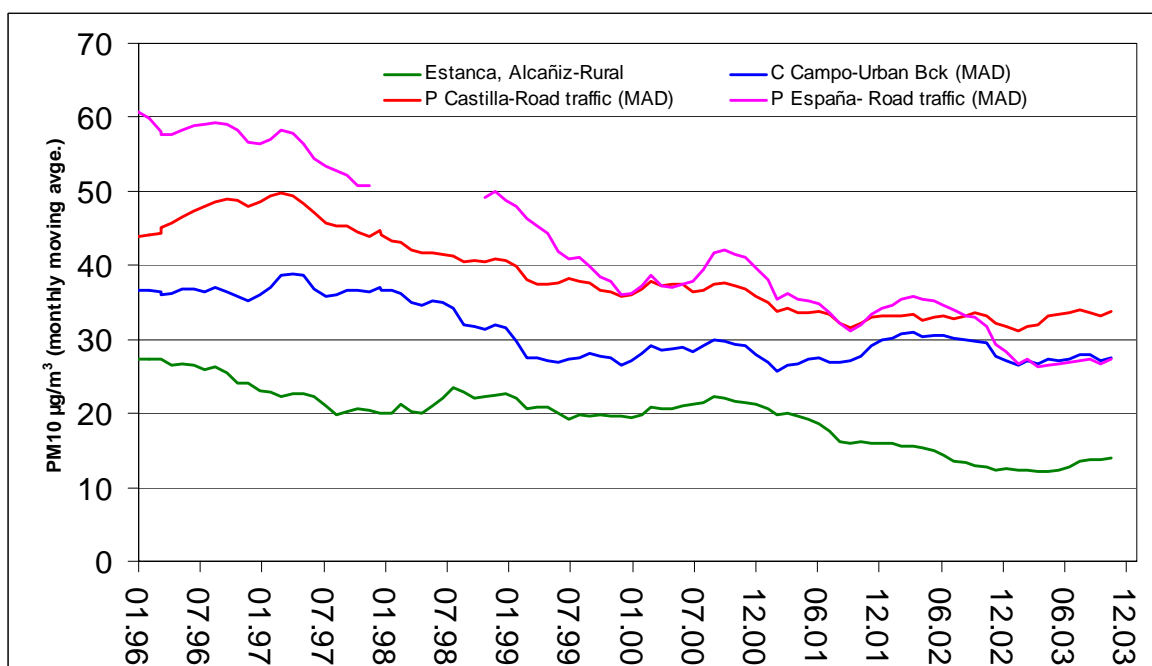
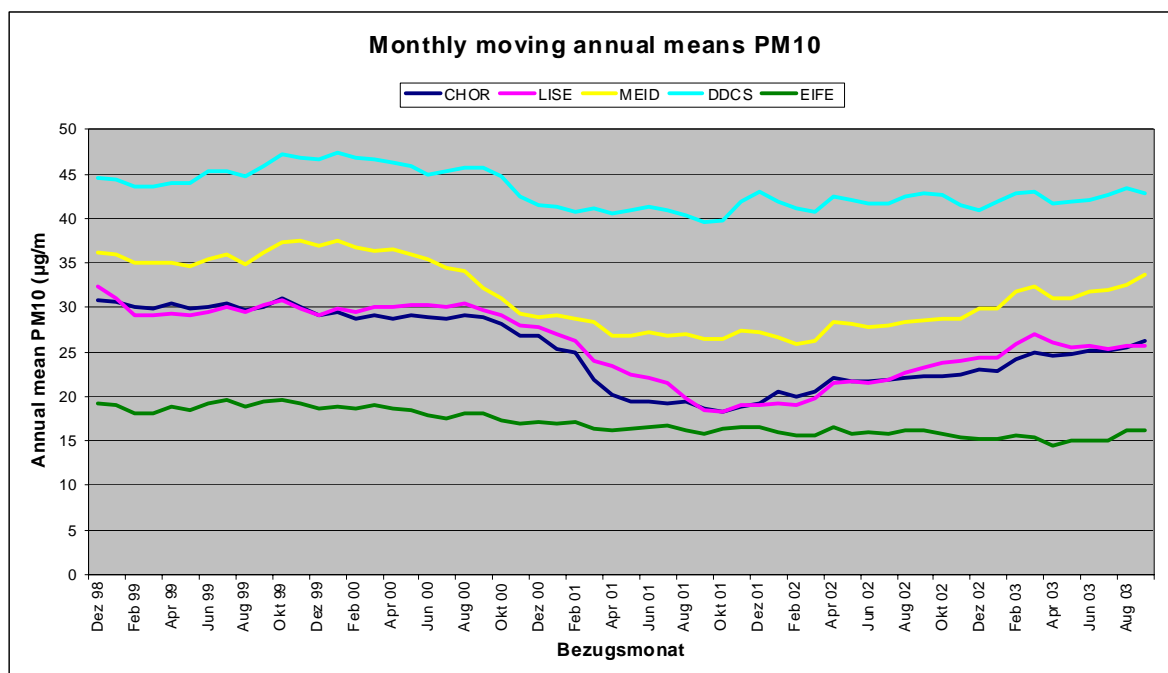


Figure 7.6 Recent PM_{10} trends (monthly moving annual averages) at 5 German stations above (CHOR, LISE, MEID: urban background, DDCS: traffic, EIFE: rural) and 4 Spanish stations below (rural, urban background and traffic).

7.4 Future projections of ambient concentrations of particulate matter

7.4.1 Europe wide model projections of PM_{10}

This section presents concentration estimates from the EMEP and REM3-models. Figures 7.7 and 7.8 present the yearly average modelled PM_{10} concentration in 1999 for the two models: EMEP (Tsyro et al., 2001) and REM3 (Stern, 2003). These can be compared to measured concentrations to help

validate the model outputs and give an idea of the possible uncertainty in projections. Modelled concentrations include both primary PM and Secondary Inorganic Aerosol (SIA).

The concentration estimates for 1999 are based on internationally reported emissions for Europe (UN-ECE for NO_x and SO₂, and the CEPMEIP-inventory for primary PM emissions – see Table 7.2).

The models estimate well the spatial scale and coverage of PM concentrations but model concentration results for many geographic areas are much lower than measured levels.

A quick scan for the models reveals that the correlation coefficients between measured and observed time series of daily averaged concentrations for a number of stations in Germany and the Netherlands range between 0.4 and 0.8, with the correlation coefficients at the higher end in rural areas and at the lower end at locations close to cities. Hence, despite the gap between the absolute modelled and observed levels, the models are able to follow the temporal behaviour of PM₁₀.

Table 7.2 Annual emissions (in ktonnes) for the 40 countries considered for the base year and two scenarios.

	Base Year (1999)	CLE (2010)	CLE + MFR for PM (2010)
PM _{coarse}	2531	1698	638
PM _{2.5}	2833	2161	833
SO ₂	24574	14750	14750
NO _x	20527	15552	15552
NM-VOC	18667	13410	13410

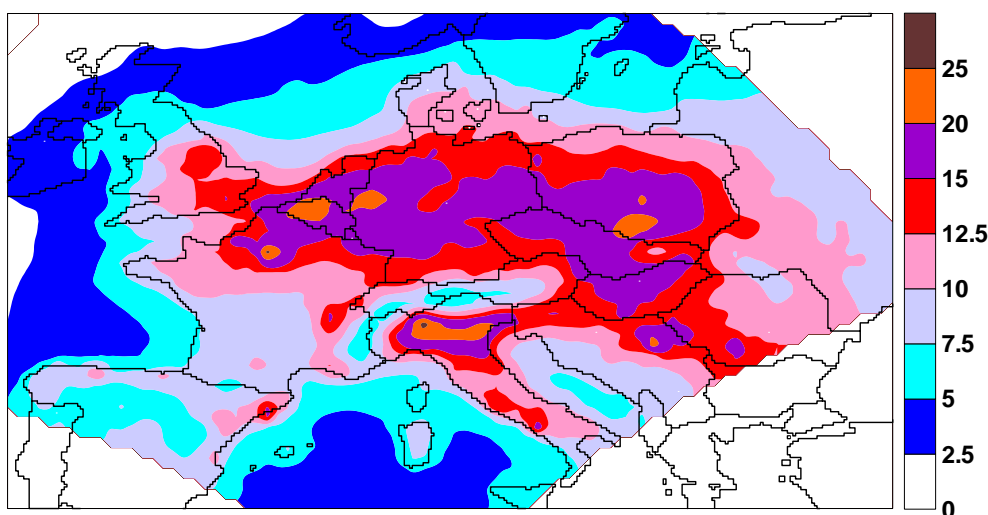


Figure 7.7 Annual average PM₁₀ concentrations in 1999 (µg/m³) as computed by the EMEP model.

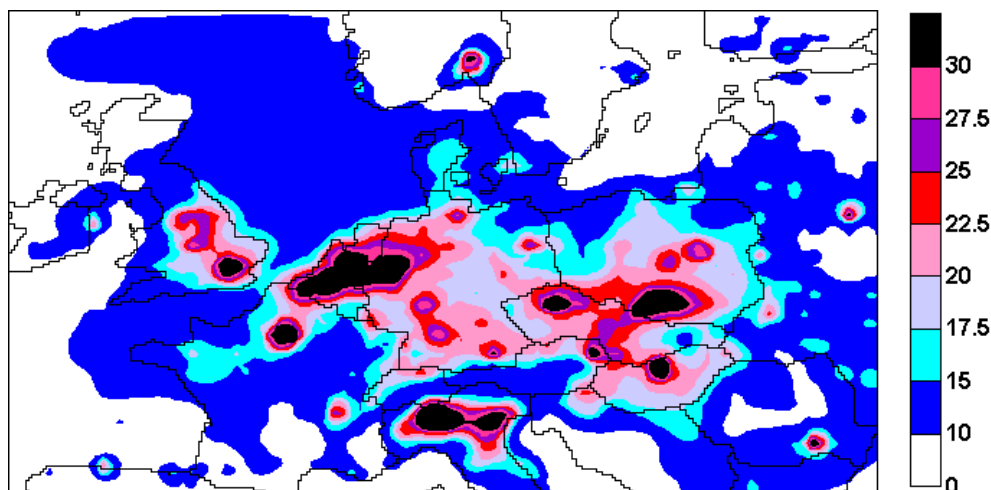


Figure 7.8 Annual average PM₁₀ concentrations in 1999 ($\mu\text{g}/\text{m}^3$) as computed by the REM3 model

Projections PM₁₀ with the EMEP model

Projections for 2010 have been estimated using two emissions scenarios prepared by IIASA (ref) for the gaseous components as well as for the particulates. Table 7.2 shows the total emissions of the 40 countries for the 1999 base year and the two future scenarios. The CLE (Current Legislation) scenario includes emissions of both gaseous compounds and particulates assuming current policies and measures. The CLE + MFR (Maximum Feasible Reduction) for PM scenario assumes that all technologically feasible abatement measures are applied to primary sources of PM (both fine and coarse). The emissions of the gaseous components remain the same as in the CLE scenario.

Figures 7.9 and 7.10 show the annual average *difference* (decrease) in PM₁₀ concentrations calculated using an earlier version of the EMEP-model (the only version for which projections are available), based on the emissions scenarios in Table 7.2. The EMEP model has been developed further recently so the EMEP computations presented here are therefore only indicative. Since currently projections for 2010 are only available from computations with the older model-version, the data presented here are related to that earlier model. For both scenarios the emissions reductions result in annual average background concentration of PM₁₀ below the Stage 2 indicative limit value of 20 $\mu\text{g}/\text{m}^3$ (resulting concentrations not shown here).

There are significant uncertainties in these projections. As discussed above, the EMEP model (in common with other large scale models) underestimates total PM₁₀ compared to measured values. This is partly because some PM sources – for example wind blown dust, sea salt and secondary biogenic aerosols are not (yet) included in the model. There are also indications that the model underestimates the anthropogenic primary component of the PM mix. This is illustrated in a recent Status Report of EMEP, in which modelled concentrations explain only half or less of the measured concentration, both at locations of rural and urban character (EMEP, 2003). However, these differences are smaller for PM_{2.5}, indicating that the model performance is significantly better for the fine than for the coarse fraction. It is to be expected that the model will underestimate future PM concentrations as well. At the same time, the model is likely to underestimate the effects of PM abatement measures from primary sources. Consequently caution should be used when assessing the likelihood of attaining the Stage 2 limit value.

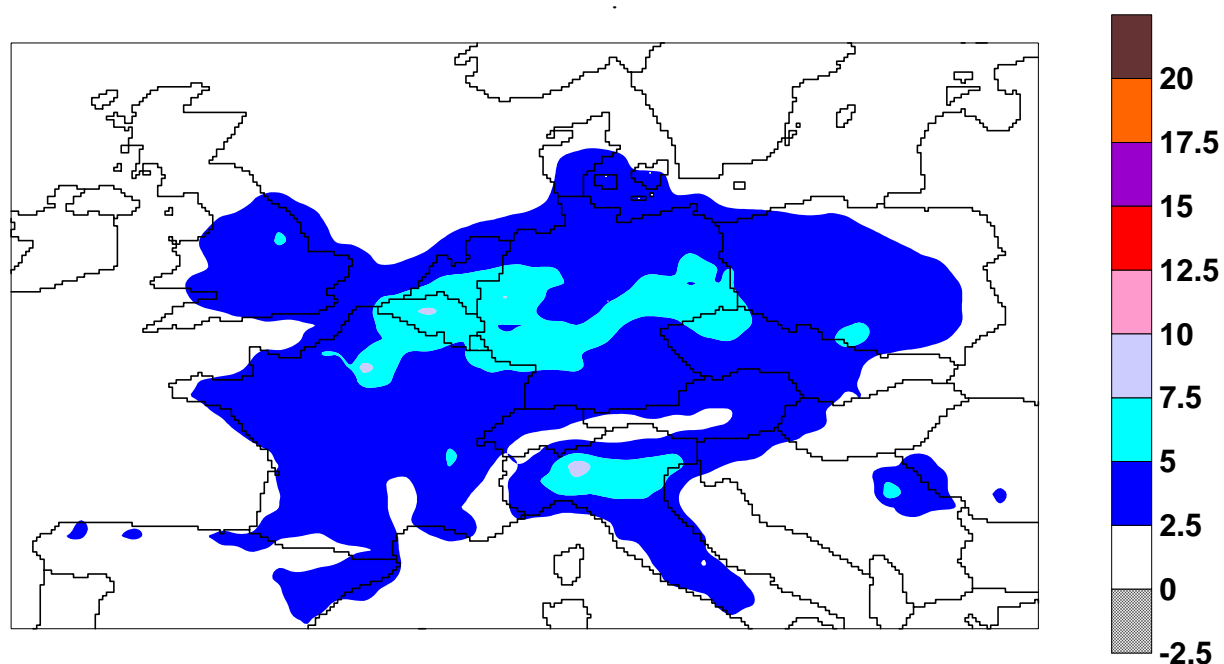


Figure 7.9 *Difference in the annual average PM₁₀ concentrations (µg/m³) between the base year (1999) and 2010-CLE as computed by the EMEP model.*

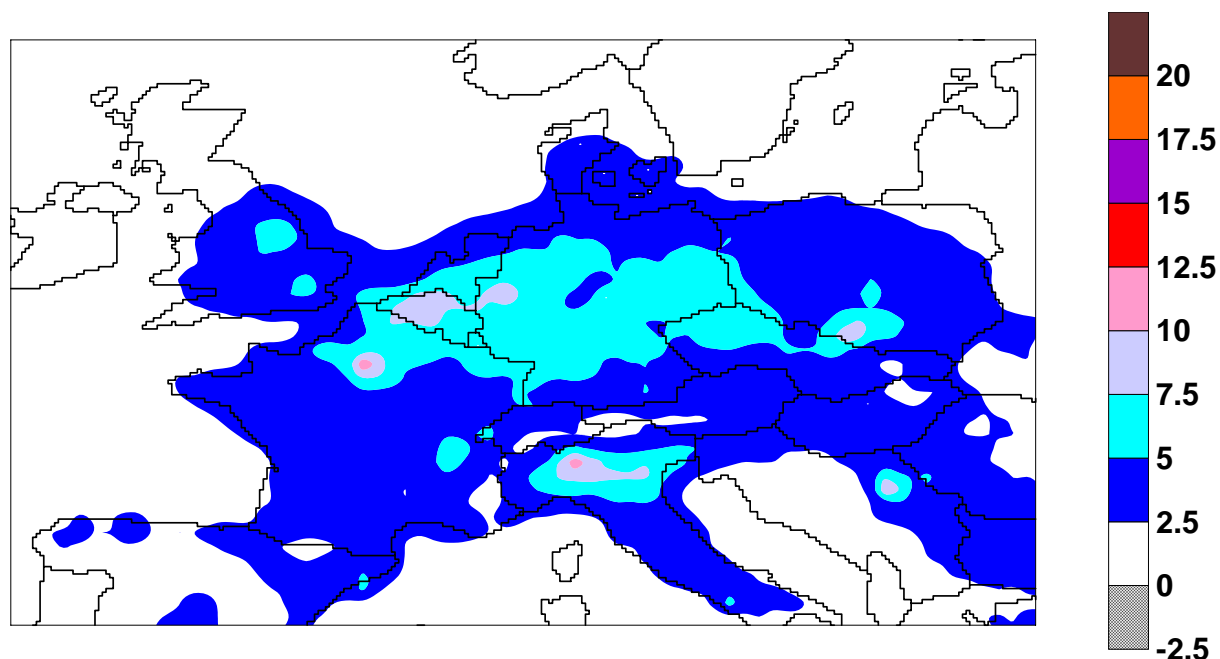


Figure 7.10 *Difference in the annual average PM₁₀ concentrations (µg/m³) between the base year (1999) and 2010-CLE + MFR for PM emissions as computed by the EMEP model.*

The EMEP model provides estimates for background concentrations averaged over a wide area. Clearly, it is not possible to estimate from these large scale levels if the Stage 2 indicative limit value will be met at all locations in Europe.

Table 7.3 aims to assess the impacts of the CLE and MFR scenarios at specific monitoring sites. The table shows the current (1999) measured concentrations at a range of monitoring sites in Europe. The changes in average concentration estimated by the EMEP model in 2010 have then been subtracted from these measured values to arrive at an estimate of the PM₁₀ concentration at that site in 2010.

The locations have been selected because they have a reasonably long time series of daily average measurements. These time series may not be representative for the whole of Europe.

Table 7.3 Annual average PM_{10} concentrations in 1999 (observed) and 2010 (projected) from EMEP calculations and number of days exceeding $50 \mu\text{g}/\text{m}^3$ in those years at a number of rural and suburban sites in Germany, the United Kingdom and the Netherlands. (MFR stands for CLE + MFR for PM in the header of the table)

Country	Station location	Yearly average concentration			Number of days > 50 $\mu\text{g}/\text{m}^3$		
		1999 (obs)	2010 CLE	2010 MFR	1999 (obs)	2010 CLE	2010 MFR
DE	Finsterwalde	40	36	35	65	45	42
	Neubrandenburg	14	11	11	1	1	1
	Guestrow-Guelzow	27	23	22	19	12	11
	Schwerin	29	25	24	36	23	21
	Stralsund	21	18	17	6	3	1
	Loecknitz/Mewegen	15	11	10	6	3	2
	Goehlen	15	11	10	2	1	0
	Jork	26	22	21	18	12	11
	Westpfalz-Waldmohr	20	17	15	1	0	0
	Zartau/Waldstation	18	14	13	8	3	3
	Deuselbach	10	11	10	0	1	1
	Neuglobsow	11	12	11	0	1	1
	Zingst	9	10	10	0	1	1
	Melpitz	17	18	17	2	7	7
UK (rural sites)	Lough Navar	9	8	8	0	0	0
	Narberth	13	11	11	0	0	0
NL	Biest Houtakker	37	32	31	51	37	35
	Breukelen ¹⁾	40	35	35	60	38	37
	De Zilk	30	27	26	22	12	11
	Eibergen	30	25	24	30	15	15
	Philippine	34	28	25	42	25	19
	Vredepeel	35	30	29	55	29	22
	Westmaas	35	30	29	45	25	23
	Wieringerwerf	33	29	29	38	23	23
	Wijnandsrade	30	24	22	17	7	7
	Witteveen	27	23	22	27	11	11

¹⁾ In the vicinity of a highway

The site-specific projections suggest that many sites will not meet the Stage 2 indicative limit values, even if maximum feasible reduction measures are implemented. The additional benefit of the MFR scenario appears small. It is important to emphasise that these projections are uncertain. It is possible that they underestimate the potential concentration reductions for the two emissions scenarios. The model may be underestimating the contribution from primary anthropogenic emissions sources. If this is the case, it will also underestimate the reduction in concentrations resulting from abatement measures applied to these sources.

REM3 model projections (including comparison with EMEP model results)

Similar projections have been performed with the REM3 model (Stern, 1994; Hass et al., 1997) for the base year (1999) and the CLE scenario in 2010 (Figure 7.11).

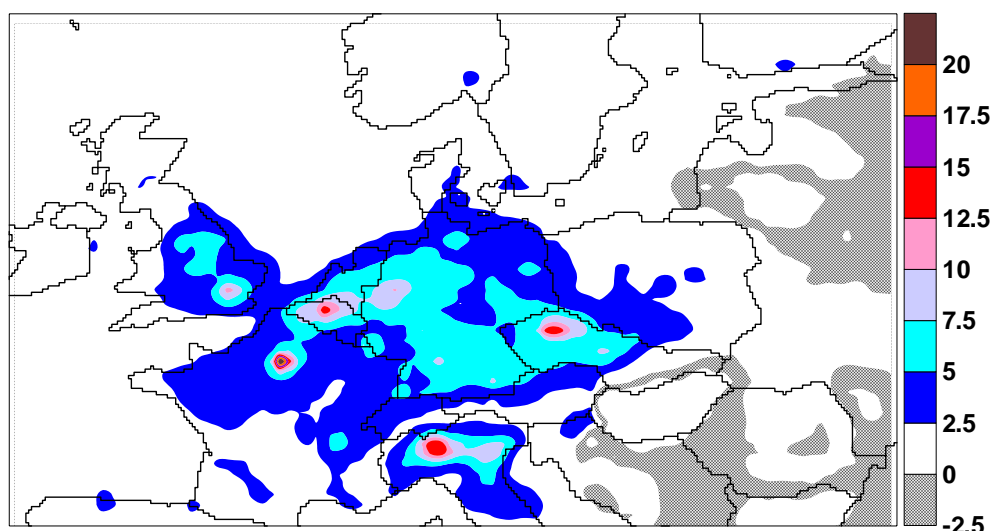


Figure 7.11 *Difference in the annual average PM_{10} concentrations ($\mu\text{g}/\text{m}^3$) between the base year (1999) and 2010-CLE as computed by the REM3 model.*

Comparing the computed differences for the CLE-scenario from REM3 and EMEP shows similar reductions in PM_{10} concentrations. This is notable, because REM3 assumes a greater contribution from primary PM. On the other hand the EMEP model appears to estimate slightly higher reductions in secondary aerosols compared to REM3 (see Figure 7.12). The estimated resulting average total reduction in PM_{10} concentrations in 2010 as computed by REM3 and EMEP is shown in Figure 7.13. Comparison of Figures 7.12 and 7.13 suggests that the major part of the reduction is related to secondary inorganic aerosols (SIA).

For computing the number of exceedances of the indicative limit value for the daily average the same approach was followed as with the EMEP model. The difference between the base year results and the CLE scenario results were subtracted from the measured time series and the number of exceedances subsequently counted. Table 7.4 shows the results.

Table 7.4 Annual average PM_{10} concentrations in 1999 (observed) and 2010 (projected, CLE) from REM3 calculations and number of days exceeding $50 \mu\text{g}/\text{m}^3$ in those years at a number of rural and suburban sites in Germany, the United Kingdom and the Netherlands.

Country	Station location	Yearly average concentration		Number of days $> 50 \mu\text{g}/\text{m}^3$	
		1999 (obs)	2010 CLE	1999 (obs)	2010 CLE
DE	Finsterwalde	40	37	65	48
	Neubrandenburg	14	11	1	1
	Guestrow-Guelzow	27	22	19	10
	Schwerin	29	25	36	24
	Stralsund	21	17	6	1
	Loecknitz/Mewegen	15	13	6	3
	Goehlen	15	11	2	1
	Jork	26	21	18	10
	Westpfalz-Waldmohr	20	15	1	0
	Zartau/Waldstation	18	14	8	4
UK (rural)	Lough Navar	9	8	0	0
	Narberth	13	12	0	0
NL	Biest Houtakker	37	30	51	33
	Breukelen ¹	40	34	60	38
	De Zilk	30	25	22	9
	Eibergen	30	23	30	15
	Philippine	34	28	42	23
	Vredepeel	35	28	55	27
	Westmaas	35	30	45	26
	Wieringerwerf	33	30	38	28
	Wijnandsrade	30	23	17	9
	Witteveen	27	22	27	13

¹⁾ In the vicinity of a highway

Comparing the results for CLE from REM3 in Table 7.4 with the corresponding results from EMEP (Table 7.3) shows that the projected annual averages and the number of exceedances agree well. Some stations that are present in Table 7.3 are absent for REM3 due to the smaller domain definition.

Consequently the conclusions from the two models are similar: they project large reductions in PM_{10} concentrations between 1999 and 2010 for both scenarios, but also that the Stage 2 limit value is unlikely to be met at a large number of locations in Europe even if maximum feasible emissions abatement measures are implemented.

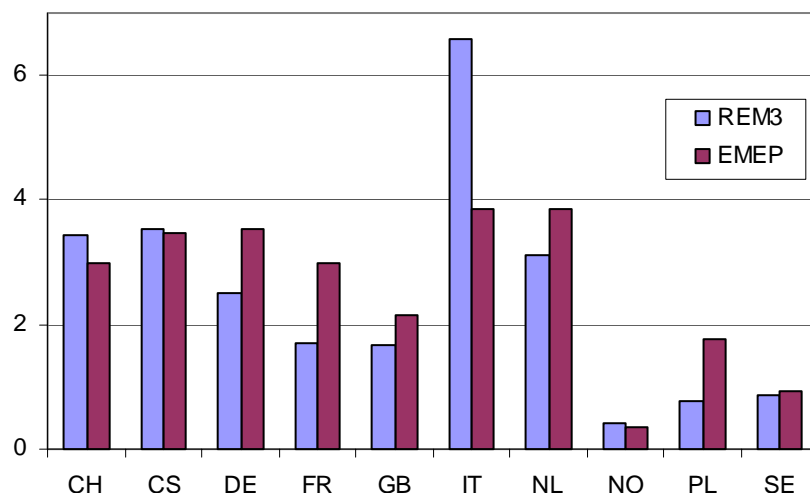


Figure 7.12 Average reduction in secondary inorganic aerosol (SIA) concentrations (in $\mu\text{g}/\text{m}^3$) in 2010-CLE as computed by REM3 and EMEP

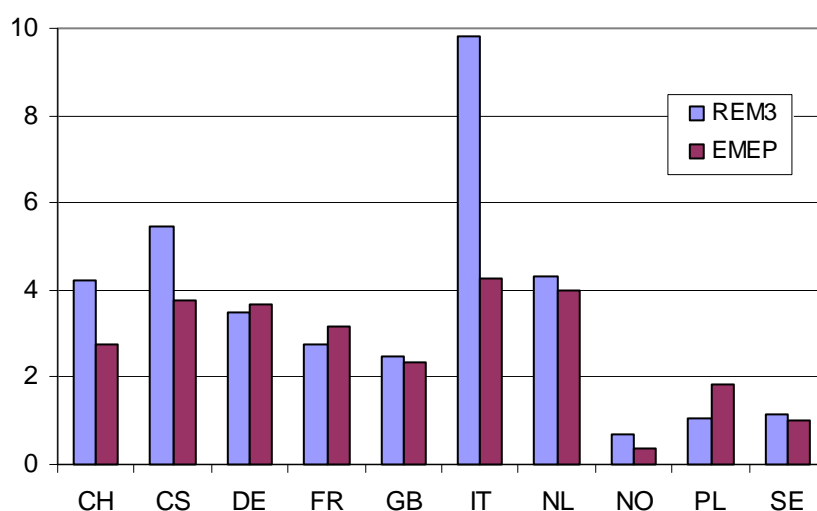


Figure 7.13 Average reduction in PM_{10} concentrations (in $\mu\text{g}/\text{m}^3$) in 2010-CLE as computed by REM3 and EMEP

7.4.2 Case studies for individual Member States and for cities

Country specific scenario studies have been performed in the United Kingdom, the Netherlands and Switzerland. Studies for Germany and Denmark are underway. In summary, the national studies substantiate the picture that emerges from the EMEP and REM3 studies.

7.4.2.1 Projections for the United Kingdom

Site-specific projections for background locations in the United Kingdom have been performed using a combination of various types of modelling and statistical analysis (Stedman et al., 2001a/b, 2002). The results obtained for the sites were then used to derive background concentration maps (i.e. at non-roadside locations) as well as roadside concentration maps. The concentration projections are based on

emissions projections for a baseline (current policies) and an illustrative scenario incorporating a package of possible additional measures to reduce emissions (Figure 7.14).

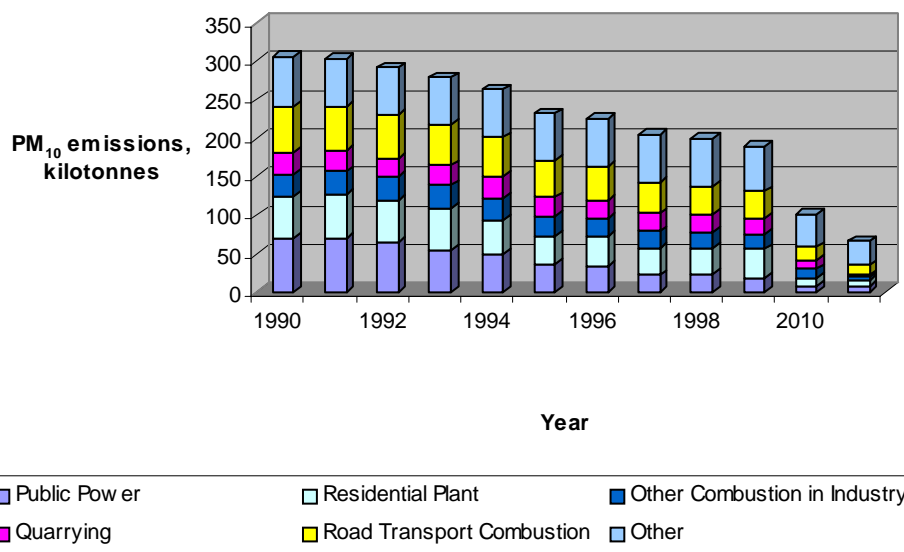


Figure 7.14 Trend in PM emissions in the United Kingdom 1990 - 2010 (for 2010 two scenarios are included: baseline and an illustrative package of additional measures)

Figure 7.15 shows the improvement in annual average background PM₁₀ concentration between 1999 and 2010 for the baseline scenario. For the baseline scenario the projections suggest that nearly all the United Kingdom – except perhaps Inner London – is likely to meet by 2010 at background sites the Stage 2 annual average limit value of 20 µg/m³.

Estimated annual mean PM₁₀ concentrations for 1999 and for 2010
baseline scenario (ugm-3, gravimetric), Ref NETCEN 31/08/2001 census_scen

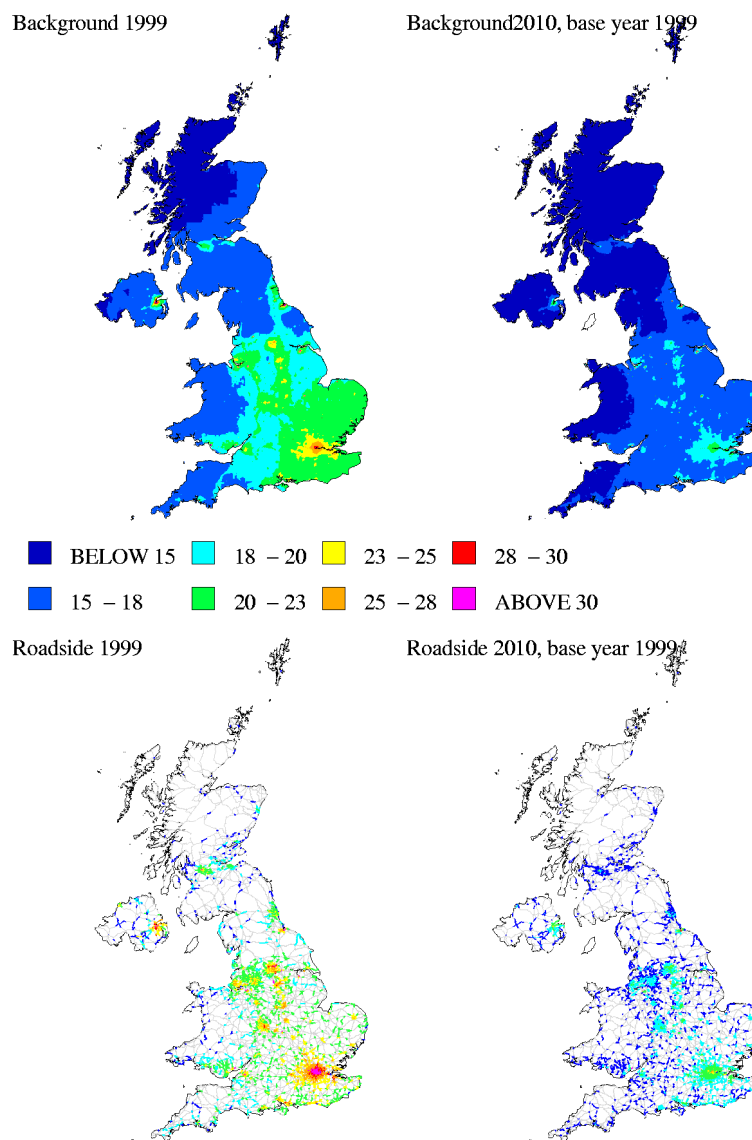


Figure 7.15 Estimated annual mean PM₁₀ concentrations for 1999 and for 2010 baseline scenario.

Similarly, roadside PM₁₀ concentrations are predicted to be much lower in 2010 than in 1999. Exceedances of 20 µg/m³ annual average are however still expected to be common at the roadside for the baseline scenario, especially in England. Exceedances of 25 µg/m³ are largely confined to Inner London. The illustrative package of possible additional measures is predicted to have a significant impact on the number of road links with estimated annual mean PM₁₀ concentrations in excess of 20 µg/m³, based on the gravimetric method, particularly in Outer London and the Rest of England (Table 7.5).

Table 7.5 The number of built-up major road links with estimated PM_{10} concentrations greater than or equal to various thresholds in 2010 for the base year 1999 for the baseline and illustrative scenarios

Year		1999				2010 Baseline				2010 Illustrative Scenario			
Threshold ($\mu\text{g}/\text{m}^3$, gravimetric)		28	25	23	20	28	25	23	20	28	25	23	20
Geographical area	Total												
Scotland	548	0	0	1	8	0	0	1	8	0	0	0	3
Wales	282	2	2	2	5	2	2	2	5	0	1	1	2
Northern Ireland	101	0	1	4	12	0	1	4	12	0	0	1	9
Inner London	760	0	44	333	760	0	44	333	760	0	0	44	709
Outer London	789	0	2	13	694	0	2	13	694	0	0	0	181
Rest of England	4700	5	12	35	708	5	12	35	708	0	0	0	55
Total	7180	7	61	388	2187	7	61	388	2187	0	1	47	959

Figure 7.16 below shows examples of measured trends and projections for two background urban sites in the United Kingdom (London and Belfast). Projections are presented for four different base years (1996 to 1999).

The figures demonstrate the reducing trend in measured and projected concentrations at these sites. Using different base years indicates the range in uncertainty for the projections associated with (1) different meteorological conditions – PM_{10} levels were generally higher in 1996 because of the influence of easterly winds that year – and (2) the measured levels from which projected values are derived; a higher starting point results in higher projections, all other assumptions remaining the same.

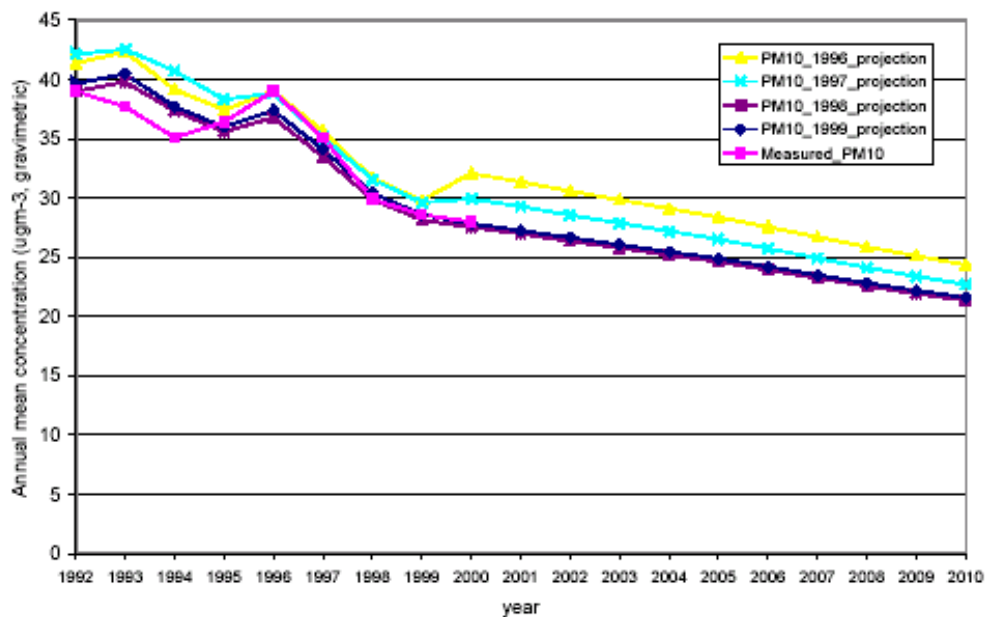


Figure 7.16a Measurements and projections at UK sites: London Bloomsbury

Figure 7.16a shows that despite reducing trends annual average PM_{10} concentrations are unlikely to meet the Stage 2 indicative limit value by 2010 at this background monitoring site in London.

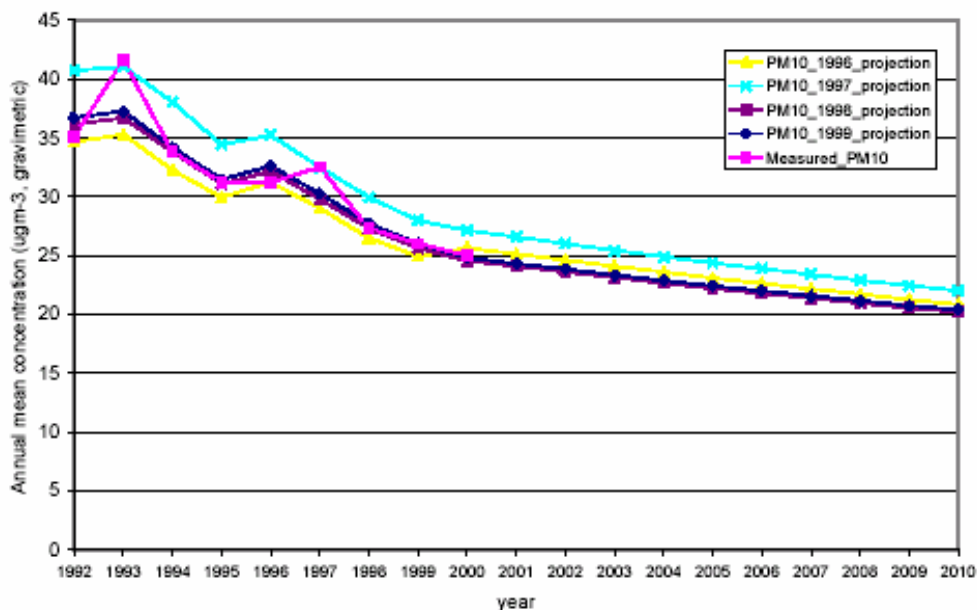


Figure 7.16b Measurements and projections at UK sites: Belfast Centre

Figure 7.16b shows that annual average PM_{10} concentrations at the monitoring site in Belfast city centre are likely to be close to the Stage 2 indicative limit value in 2010.

Table 7.6 shows population weighted mean concentrations for the base year (1999) and for 2010 for the baseline and illustrative package of measures scenarios. The largest change in population weighted mean concentration for the illustrative package of measures is in London; the rest of England is similar to the value for the United Kingdom as a whole, which is $-0.811 \mu\text{g}/\text{m}^3$ (gravimetric).

Table 7.6 Population weighted annual mean background PM_{10} concentrations ($\mu\text{g}/\text{m}^3$) for the base year 1999 and for 2010 for the baseline and illustrative scenarios

Year	1999	2010 Baseline	2010 Illustrative Scenario
Scotland	17.867	15.168	14.688
Wales	19.218	16.316	15.550
Northern Ireland	20.472	16.476	15.919
Inner London	27.396	21.012	19.910
Outer London	25.038	19.745	18.771
Rest of England	21.355	17.627	16.796
United Kingdom	21.451	17.616	16.805

Projections have also been carried out at a smaller scale for Greater London, using a different modelling methodology and emissions inventory than the United Kingdom scale modelling above.

Figure 7.17 shows projected annual mean PM_{10} concentrations in Greater London in 2004, assuming 1999 meteorology (a typical year dominated by westerly and south westerly air flows) and current policies. Figure 7.18 shows projected 90.4 percentile (35 days exceedance) for the same assumptions. The modelling suggests that London is likely to meet the Stage 1 limit values in 2004, assuming it is a typical meteorological year. Further modelling (not shown) indicates that if a year has a higher than usual number of days with easterly air flows, London is unlikely to meet the limit value.

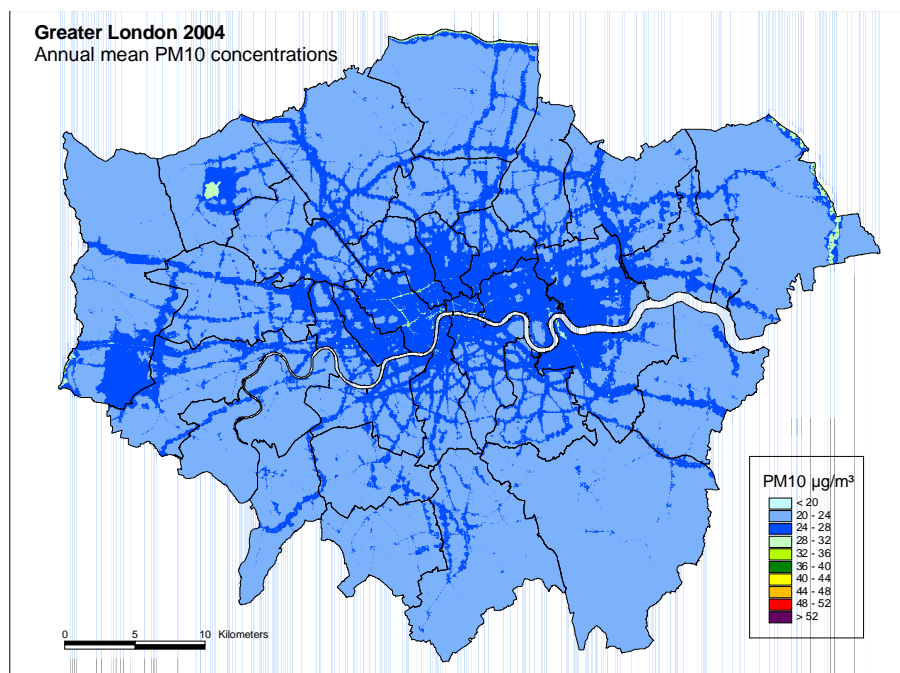


Figure 7.17 Annual average PM₁₀ concentrations for Greater London, 2004. Stage 1 limit value: 50 µg/m³.

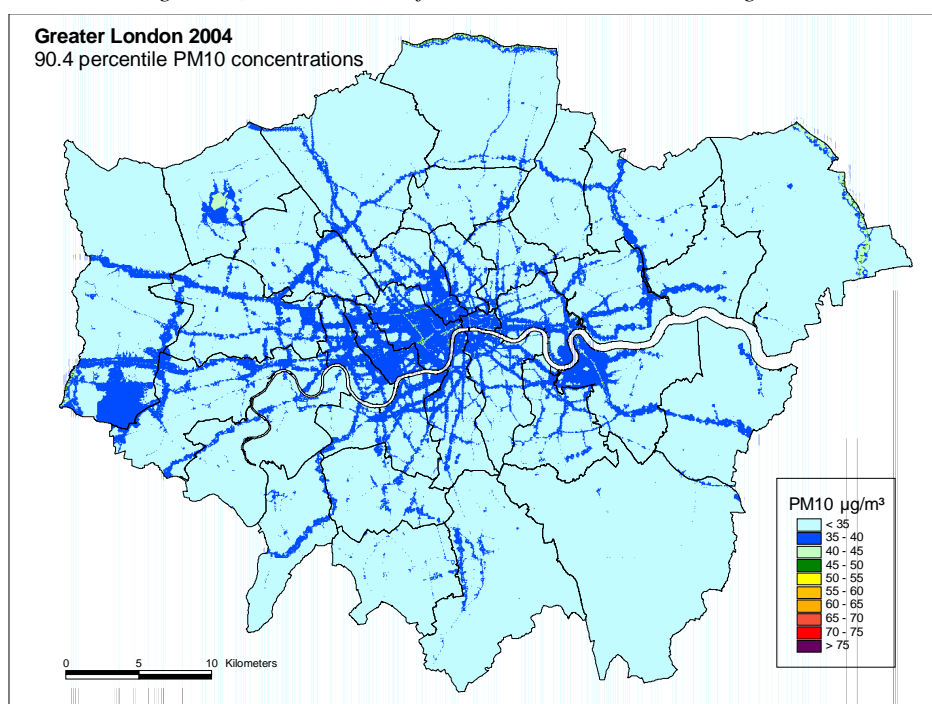


Figure 7.18 90.4 percentile (35 days exceedance) PM₁₀ concentrations for Greater London, 2004. Stage 1 limit value: 50 µg/m³.

Figure 7.19 shows projected annual mean PM₁₀ concentrations in Greater London in 2010, assuming 1999 meteorology and current policies. The modelling indicates that large areas of London are unlikely to meet the Stage 2 annual average indicative limit value in 2010 at background locations in addition to traffic-related hot spots.

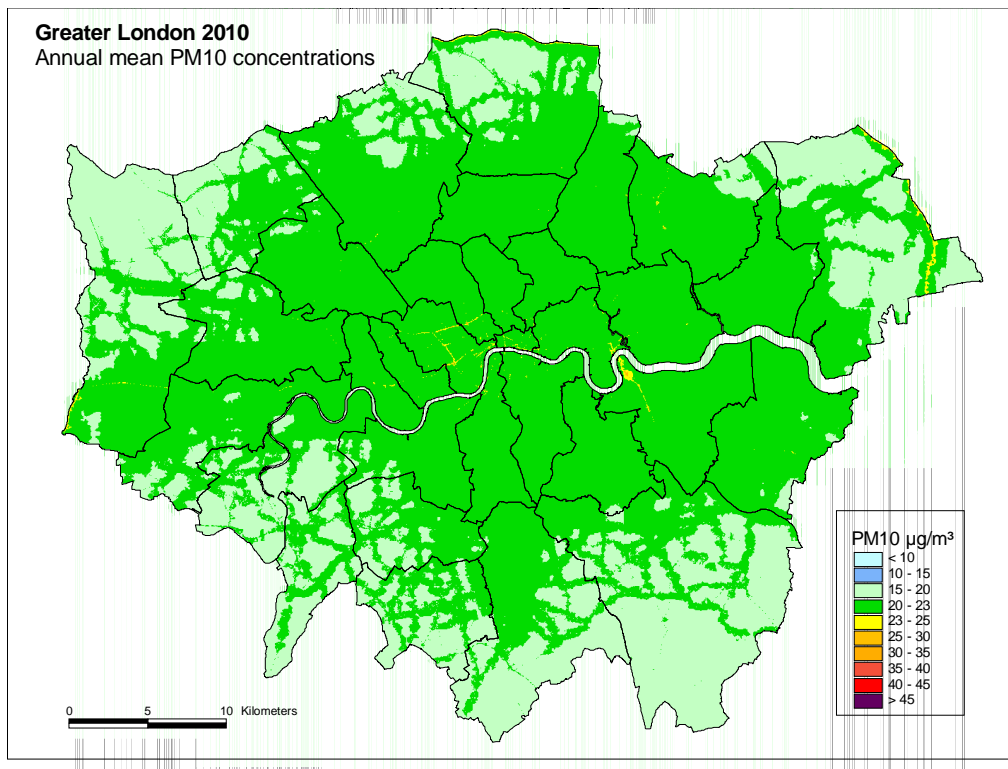


Figure 7.19 Annual average PM_{10} concentrations for Greater London, 2010. Stage 2 limit value: $20 \mu\text{g}/\text{m}^3$.

To conclude, modelling suggests that the United Kingdom is likely to meet the indicative Stage 2 annual average limit value by 2010 at background for most of the country with current measures, but is unlikely to meet the limit value for wide areas in Greater London – or at many locations close to busy roads, even with additional measures.

7.4.2.2 Projections for the Netherlands

Future PM_{10} concentrations have been estimated using two different types of models: LOTOS from TNO (Schaap, 2000) and OPS from RIVM (Van Jaarsveld, 1995). The estimated size of the average decrease in PM_{10} concentration to be expected between 1995 and 2010 at current control policies (CLE) is very similar for both models, in the order of $5\text{--}7 \mu\text{g}/\text{m}^3$. Similar as for the EMEP and the REM3 models, both LOTOS and OPS underestimate, to a smaller or larger extent, the measured PM_{10} levels. A substantial part of this is due to natural contributions that are not taken into account (sea salt, wind blown dust), but it cannot be excluded that the models are also excluding an anthropogenic primary contribution. In general, secondary inorganic aerosols (SIA) are well represented by the models.

The OPS-model attempted to solve the apparent underestimation by adding measured differences to the computed concentrations. These differences are estimated from the differences between calculated and observed concentrations in the period 1994-1999. This difference – known as the ‘non-modelled’ part – is then added to the calculated concentrations. The implicit assumption is that the ‘non modelled’ part remains constant in the future. At present the concentration of this non-modelled fraction is on average $18 \mu\text{g}/\text{m}^3$. This approach is comparable with the approach followed for LOTOS, where the differences between the base case and the 2010 projection were subtracted from the measurements.

With OPS, in 2010 all Dutch grids ($5 \times 5 \text{ km}^2$) are currently found to show yearly average PM_{10} concentrations below $40 \mu\text{g}/\text{m}^3$ (Figure 7.20). Thus, by extrapolation, compliance with the 2005 yearly

average limit value seems feasible in the Netherlands although local exceedances at 'hot spots' cannot be ruled out.

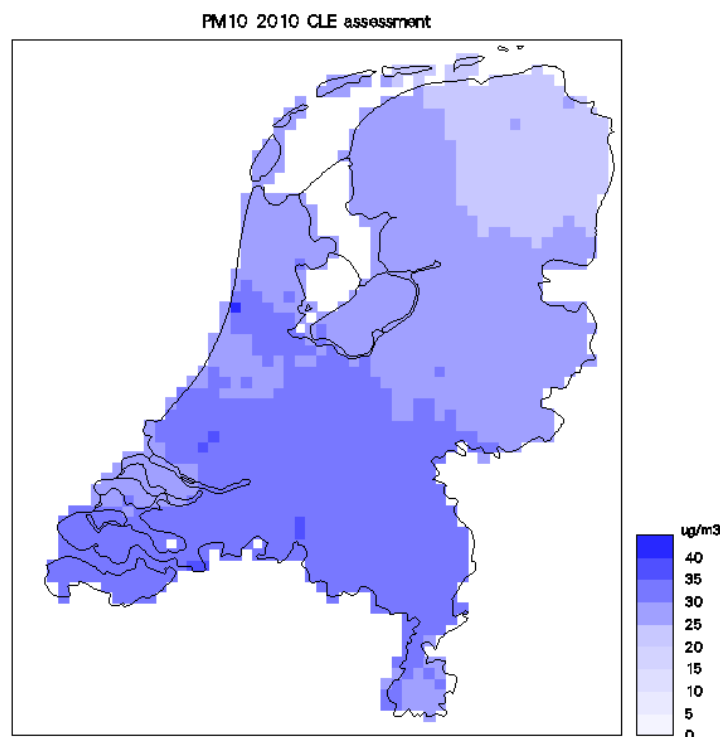


Figure 7.20 Projected PM₁₀ concentrations in the Netherlands in 2010 (CLE-scenario).

The available scenario calculations with the LOTOS model using the CLE scenario show similar results (Figure 7.21). With the LOTOS model it is also possible to make projections of the number of days with a daily average above 50 µg/m³ (Figure 7.22). It becomes clear that at the locations studied the proposed Stage 2 limit values in 2010 for both the annual average (20 µg/m³) and the number of exceedances in relation with the daily standard (7) are likely not going to be met. As observed also with the EMEP and REM3 models, even the 2005 24-hour limit value is still expected to be exceeded in 2010.

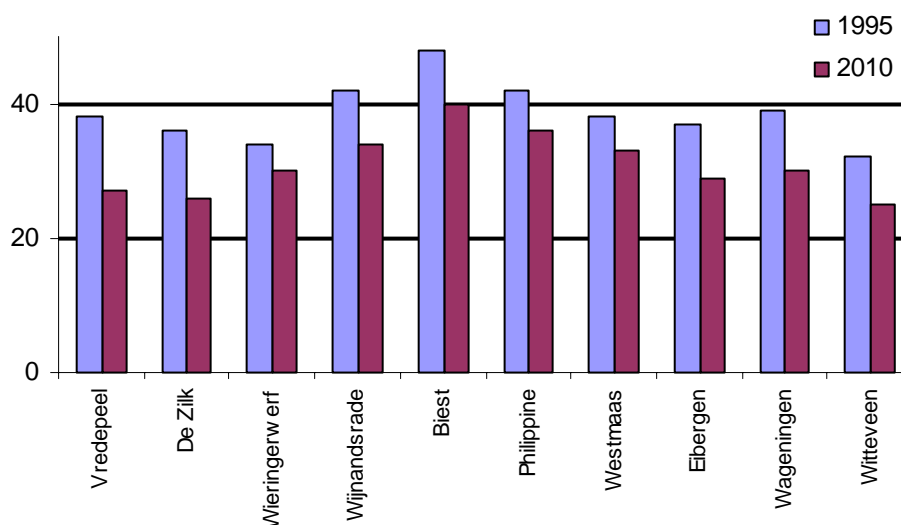


Figure 7.21 Annual average in 1995 (measured) and 2010 (projected with LOTOS) at various locations in the Netherlands

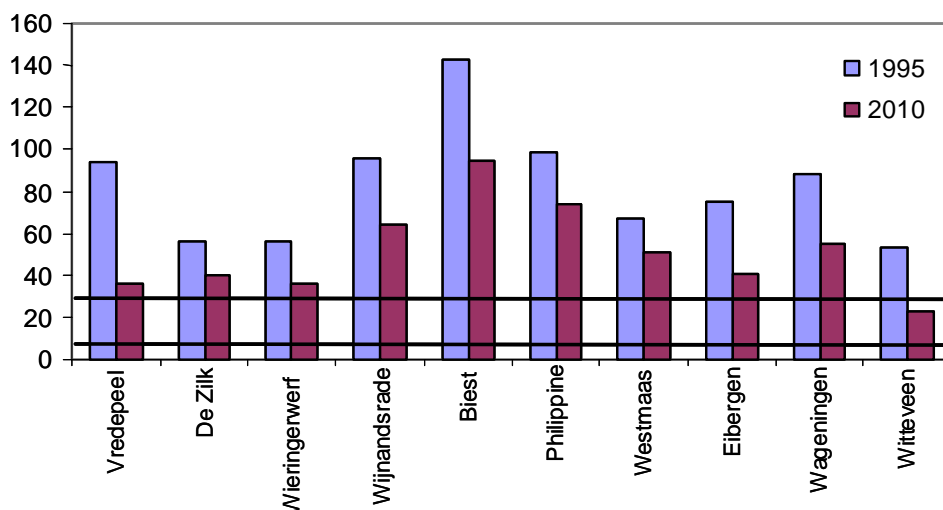


Figure 7.22 Annual number of days for which the limit value of $50 \mu\text{g}/\text{m}^3$ is exceeded at various locations in the Netherlands, as measured in 1995 and projected with the LOTOS model for 2010.

In addition, data from the OPS-study suggest that the impact of an additional emission reduction of 60% in 2010 of primary particle emissions (Maximum Feasible Reduction (MFR) scenario) compared to the CLE scenario PM_{10} concentration, will be limited to an average reduction of about $1 \mu\text{g}/\text{m}^3$ (Table 7.7). This is consistent with the small impact found in the EMEP MFR-scenario calculation, and the impact of the UK illustrative package of possible additional measures. For the Netherlands it would imply that even with maximum national emission control, the 2010 Stage 2 indicative annual average limit value of $20 \mu\text{g}/\text{m}^3$ is not attainable everywhere.

Table 7.7 Averaged results of PM_{10} calculations with OPS for the Netherlands using two 2010 scenario variants (CLE and MFR). The total including the non-modelled fraction of $18 \mu\text{g}/\text{m}^3$ is shown in brackets.

Year	Dutch primary PM_{10} emissions in ktonnes	Modelled average concentration of primary PM_{10} ($\mu\text{g}/\text{m}^3$)	Modelled average total (prim. + sec.) anthropogenic PM_{10} concentration ($\mu\text{g}/\text{m}^3$)
1980	116	11.4	29.7 (47.7)
1995	61	6.0	16.5 (34.5)
2010-CLE	42	4.8	11.1 (29.1)
2010-MFR	18	3.6	10.0 (28.0)

On average over the Netherlands, the impact of the emissions reduction between 1995 and 2010 is estimated to be $5.4 \mu\text{g}/\text{m}^3$. However, based on measurements an even higher concentration reduction can be deduced: between 1995 and 2002 the average Dutch concentration decreased by approximately $7.5 \mu\text{g}/\text{m}^3$. This suggests that the modelled projection underestimates the real decline in PM_{10} concentration. Projections with the LOTOS-model for Germany showed similar results when the calculated decrease was compared with the actual observed decline (data not shown here).

7.4.2.3 Switzerland

PM_{10} and $\text{PM}_{2.5}$ concentration maps have been computed for Switzerland for the year 2000, and also for the year 2010 in two scenarios ("business as usual" and "maximum possible reductions"). For all major source groups (road traffic, off-road traffic, industry, households, etc.), separate countrywide emission inventories with a spatial resolution of 200m were used. Emission modelling was performed for primary particles and for the gaseous precursors of secondary particles; the imported background concentration was parameterised.

For 2000, models estimated that annual average PM₁₀ concentrations were above 20 µg/m³ over 4.2% (1757 km²) of the total surface of Switzerland; 42.8% of the population (2.81 million) resides in this area.

For 2010 in the "business as usual" scenario, these numbers reduce to 1.2% (510 km²) of the surface and 21.8% (1.25 million people) of the population. For the "maximum feasible reduction" scenario, the corresponding numbers are 0.1% (39 km²) for the surface and 4.8% (0.11 million people) for the population. Table 7.8 shows the average PM₁₀ population exposure, split by contributions from source groups.

For PM_{2.5}, the part of the population in 2000 that lives in regions where 20 µg/m³ (annual average) is exceeded amounts 8.6%. This reduces to 4% of the population in 2010 with the current legislation scenario ("business as usual"), whereas this would be near 0% for the maximum feasible reduction scenario.

Table 7.8 Average PM₁₀ population exposure, split contributions from source groups.

Scenario	Source group	Average PM ₁₀ population exposure (µg/m ³)		
		Primary	Secondary	Total
2000	Anthropogenic, Swiss	6.62	3.25	9.86
	Anthropogenic, foreign	3.03	5.35	8.37
	Biogenic and geogenic	1.01	0.39	1.40
	Total	10.65	8.99	19.64
2010 CLE	Anthropogenic, Swiss	6.13	3.17	9.31
	Anthropogenic, foreign	2.47	4.61	7.08
	Biogenic and geogenic	1.01	0.39	1.40
	Total	9.61	8.18	17.79
2010 MFR	Anthropogenic, Swiss	3.92	2.91	6.83
	Anthropogenic, foreign	2.07	4.08	6.16
	Biogenic and geogenic	1.01	0.39	1.40
	Total	7.00	7.39	14.40

7.4.2.4 City experiences

At the Workshop on PM in Stockholm (20-21 October 2003) various examples of experiences in cities were presented (Berlin, Stockholm, Madrid, Milan, London and Duisburg). The major focus of the presentations was on traffic-related sources; long range transport (e.g. Berlin, London and Stockholm) and sometimes wood burning (Stockholm) were considered to be major sources as well. The general finding was that local traffic measures have limited impact on PM₁₀ concentrations. Exhaust emissions may be important from a health point of view, but constitute a comparatively small part of the total PM₁₀ concentration. It was expected that exhaust emissions would be better regulated at EU level. Local hot spots might to a large extent be caused by resuspension of road dust. However, it was also found that cleaning of roads virtually had no impact on PM₁₀ concentrations. These issues have been discussed more extensively elsewhere in this position paper, in particular in Chapters 4, 6 and 8.

7.4.3 Uncertainties

As discussed above, many air quality models appear to underestimate significantly PM₁₀ concentrations compared to measurements. It seems that annual average Secondary Inorganic Aerosol

(SIA) is quite well reproduced by the models; the contribution of primary emitted particles may be underestimated. It is not known fully why this is. It may be that emissions inventories underestimate PM sources; it may be that the models do not accurately convert emissions to concentrations; or it may be that measurements are overestimating PM concentrations.

Because of this possible underestimation – and by the nature of modelling any future scenarios – it should be borne in mind that projections are uncertain. It is possible that models (EMEP, REM3 and the Netherlands' models) underestimate the effects of abatement measures because they do not take full account of the primary component of the PM mix. It is this component that is affected most by national and local actions. Indeed there are examples that the models have not predicted well the rate of the recent reducing trend in PM concentrations. Projected PM concentrations that have been corrected to take account of current differences between modelled and measured results, may tend to be conservative (i.e. overestimate future concentrations). Thus, in the Dutch model the non-modelled part of PM₁₀ is assumed to remain constant in the future, whereas part of it may be anthropogenic and potentially to be influenced by measures. In this case the annual average of the non-modelled part is large, in size comparable to the indicative limit value for the annual average for 2010 (which is 20 µg/m³).

7.5 Conclusions and recommendations

7.5.1 Conclusions

Past and present trends

PM₁₀ data in AirBase show that on the average over the about 190 stations with data for all the years 1997-2001, there was a decreasing tendency from 1997 to 1999, and a slight increase between 1999 and 2001. For the period as a whole, the concentrations were reduced by about 15-20%, somewhat less for the annual average than for the 36th highest 24-hour concentration. This pertains to all three types of stations considered (rural, urban, street). Although the majority of the stations have a downward tendency (for most of them this tendency is non-significant, at 90% confidence level), about 20% of the stations have an increasing tendency (mostly street and urban background stations).

The results from analysis of the AirBase data are backed up by PM data from the EMEP database. Both AirBase and EMEP data indicates that PM₁₀ is not reduced all over Europe. E.g. Spain and Switzerland have a largely unchanged level over the analysed period. Other evidence is available from analysis of national trends. For example, PM₁₀ concentrations in the Netherlands have been reducing on average by 2 to 3% per year over recent years (1993-2002), after correction for variations due to meteorological conditions.

The reduction in rural secondary inorganic aerosols (sulphate and nitrate), as assessed from EMEP data, is of the same magnitude as the measured PM₁₀ reductions. Interannual variations in meteorological conditions also influence the levels of PM₁₀ and secondary aerosol, modifying the relationship between PM₁₀ and secondary aerosol.

Taking into account the influence of interannual variations in meteorological conditions which have not been accounted for, and also the relatively few stations (about 190) with long enough time series to be included in the analysis, one should be careful to attach significance to the indicated 15-20% reduction as a representative average figure for the European area in 1997-2001. The decreasing tendency occurs mainly in the early part of this period; while there is an increasing tendency in the later part of 1997-2000. There is also evidence from several countries (e.g. Germany, Belgium, Czech Republic, Switzerland) that PM₁₀ concentrations in 2002 and particularly in 2003 show an increase. Although this increase is mainly due to meteorological conditions, it cannot be excluded that increasing emissions in some parts of Europe may play a role as well.

Although it seems clear that concentrations have indeed been reduced in most of Europe before 2000 – and this can be explained at least partly by abatement of sources, particularly large combustion sources resulting in significantly reduced emissions e.g. of precursors of secondary PM –, the extent of PM₁₀ concentration reductions in Europe still cannot be accurately quantified.

Projections

Limited available projections of PM₁₀ estimate that concentrations will decrease significantly between now and 2010. Many of the projections are based on continental or national scale models, designed for background locations. In general, modelling remains confined to estimation of annual averages, since daily concentrations are more difficult to predict. Case studies are available only from a few European countries (United Kingdom, Netherlands and Switzerland; for Germany and Denmark studies are underway).

Based on the picture that emerges from the available scenario studies some indicative conclusions may be drawn:

- Projections indicate that it is unlikely that the Stage 2 indicative limit values for 2010 will be attained everywhere at rural or urban background sites in Europe, even if additional abatement measures (maximum feasible reduction) are implemented.
- Stage 2 limit values are unlikely to be attained in many European countries at a large number of hot spots in cities, close to busy roads and close to some industrial sources. Although the number of locations not attaining the Stage 2 limit values is likely to be high, in part of the countries the areas of these exceedances are likely to be small.
- The data suggest that the 2005 yearly average limit value may be attainable for most locations, but that the 2005 24-hour limit value in parts of Europe will not be attained by 2005, possibly even by 2010.
- Projections are significantly uncertain due to uncertainties in the modelling methodologies, assumptions and emissions projections. Most projections tend to underestimate the impact of abatement measures and so overestimate projected concentrations.
- The contribution of ‘natural’ sources like sea salt, wind blown dust and secondary biogenic aerosols to the total PM₁₀ concentration is highly uncertain.
- Because of their large scale, most models are also likely to underestimate – or do not estimate at all – the impacts of local measures on urban background and hot spot PM concentrations.
- As it is not possible to say whether the maximum feasible reduction scenarios would or would not allow the limit values to be met, it is difficult to propose limit values that are both challenging and feasible.

7.5.2 Recommendations

1. Member States should report PM₁₀ measurements for previous years to the full extent that data are available, to increase the basis for assessing historic trends and its comparison with historic emissions trends. Historic time series must be quality assured, particularly as concerns the correction factor used for measurements performed without using the reference method.
2. The historic time series should be analysed in terms of the influence of interannual meteorological conditions and other parameters, in order to separate the effect of emissions changes.

3. With some exceptions, there is a lack of PM concentration projections at the national and city scale. This hinders the ability to judge attainability of current and proposed PM targets and development of targeted abatement strategies. Member States are recommended to undertake their own national and city scale projections where they have not already done so.
4. Uncertainties in current projections should be addressed, particularly the underestimation of current PM₁₀ concentrations by some models as well as the possible underestimation of the impacts of abatement measures and consequent possible overestimation of future concentrations.

7.6 References

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8. Abatement

8.1 Scope

This chapter addresses the availability of sufficiently demonstrated and well-established control techniques for the different kinds of PM sources and metrics. It also considers cost-effectiveness and contains a brief discussion of possible priorities for abatement measures in terms of exposure and health effects.

8.2 Abatement techniques

8.2.1 Introduction

As covered in previous chapters, ambient PM consists of primary and secondary components. Primary PM is emitted directly from sources, whereas the secondary component is formed in the atmosphere from gaseous precursors including NO_x, SO₂, NH₃ and certain VOCs. Even though secondary PM contributes significantly to the overall ambient PM concentrations, this section contains only a short summary on abatement of these pollutants and concentrates on primary PM emissions, including control options for fugitive emissions. Only major sources of primary PM emissions are considered in this chapter. It should also be noted that a significant part of ambient PM may result from non-anthropogenic emission sources such as wind blown crustal material (e.g., Sahara dust), secondary organic aerosols formed from biogenic emitted VOCs (including mono terpenes), plant debris, etc. These sources are usually not under human control and are therefore not considered in this chapter.

8.2.2 Reduction of PM precursor emissions

Control options for NO_x, SO₂, NH₃ and VOCs are usually well established and may be found elsewhere, e.g., the existing position papers, for industrial installations in the BREF documents (<http://eippcb.jrc.es/pages/Boutline.htm>) established under the IPPC directive (96/61/EC), which also cover primary PM emissions (see Section 8.2.6 for further discussion).

8.2.3 Road traffic

Road transport contributes significantly to overall PM emissions (Chapter 5) and is at an urban scale often the dominating source for ambient PM as shown by source apportionment studies (Chapter 6).

Tail-pipe emissions are of special interest, since they almost exclusively contribute to fine PM (PM_{2.5} and below). Control options aim at an improved combustion, fuel-quality to minimise PM emissions (e.g., low S fuels), and filtering techniques. Filtering techniques include CRT (continuous regenerating traps), which combine particle traps with oxidising catalytic converters. Several European Directives have been put into force to regulate emissions from vehicles (light vehicles: Directive 98/69/EC; heavy duty vehicles: Directive 1999/96/EC) and fuel quality (Directive 98/70/EC¹⁶). Particulate traps have proved to be very efficient to reduce PM emissions also from cars (see Figure 8.1), but are not necessarily required to meet current emission standards (EURO III and EURO IV). Therefore, further tightening of emission limit values to levels equivalent to those achieved by particle traps seems a promising policy option.

¹⁶ Council and Parliament have agreed in late 2002 to a gradual introduction of sulphur free petrol and diesel fuels starting in 2005 and to be complete as from 1st of January 2009.

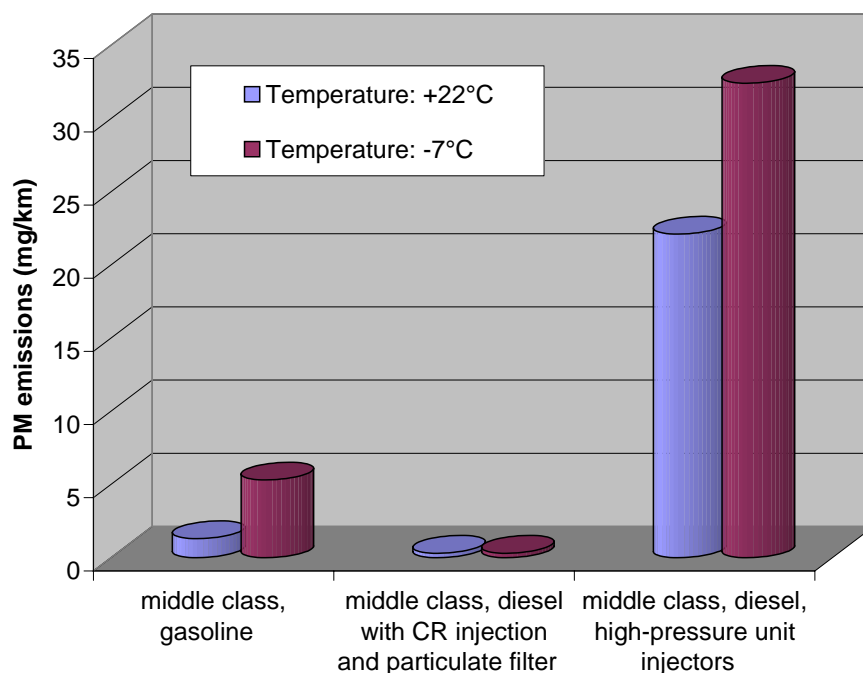


Figure 8.1 PM emission of a middle class gasoline and diesel passenger car with and without particle trap (Swedish National Road Administration, 2002). Note that PM emissions of diesel cars equipped with particulate traps are lower than those from (comparable) petrol cars.

A summary of current PM emission standard in the European Union is given in Table 8.1.

Table 8.1 Emission limit values for vehicles in the EU

	EU standards	PM emission standard
Diesel-passenger cars and Light Duty Vehicles (LDV) GVW < 1305 kg	Euro I - 1992 / 94	0.14 g/km
	Euro II - 1996	0.08 g/km
	Euro III – 2000	0.05 g/km
	Euro IV – 2005	0.025 g/km
Diesel LDV GVW 1305 to 1760 kg	Class II – 1994	0.16 g/km
	Class II- 2001	0.07 g/km
	Class II - 2006	0.04 g/km
Diesel LDV GVW > 1760 kg	Class III - 1994	0.25 g/km
	Class III - 2001	0.10 g/km
	Class III - 2006	0.06 g/km
Diesel Heavy Duty Vehicles (HDV) and buses	Euro I - 1992, <85 kW	0.61 g/kWh
	Euro I - 1992, >85 kW	0.36 g/kWh
	Euro II - 1996	0.25 g/kWh
	Euro II - 1998	0.15 g/kWh
	Euro III - 2000	0.10 g/kWh
	Euro IV and V - 2005 & 2008	0.02 g/kWh

The table below summarises the potential for reducing emissions.

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
HDV, tail pipe	Yes	no	Very high
LDV, tail pipe	Yes	no	Very high

Traffic emissions are also caused by **abrasion** of tyres, brakes and the road surface. However, little is known about the abatement options for these emissions. More information on the influence of different materials, information on different techniques is needed. This information should also cover reduction potentials, costs, side effects (e.g., on road safety) etc.

According to the CEPMEIP database established by TNO (2001):

- All emissions of tyre wear and road abrasion are in the coarse fraction. Only 5 % of the overall total suspended particulate (TSP) emissions are in the fraction from PM_{2.5-10}, while no material is emitted as PM_{2.5}. However, more recent studies have indicated that this is a simplification, since these sources can also give rise to particles <2.5 µm.
- In contrast, most emissions from brake wear are in the PM_{2.5} fraction.

Large uncertainties also exist concerning the (traffic induced) **resuspension**. One of the main factors influencing the amount of resuspension from **paved roads** is the silt loading. Control techniques for paved roads attempt either to prevent material from being deposited onto the surface (preventive controls) or to remove from the travel lanes any material that has been deposited (mitigative controls). Covering of loads in trucks, and the paving of access areas to unpaved lots or construction sites, are examples of preventive measures. Examples of mitigative controls include vacuum sweeping, water flushing, and broom sweeping and flushing. Actual control efficiencies for any of these techniques can be highly variable.

The effect of road washing on PM₁₀ concentrations: a case study in Milan

In Milan, the capital of the Lombardy Region, the Regional Environmental Agency for Lombardy undertook a field test in winter 2002 aimed at checking if any reduction of PM₁₀ concentrations may be obtained by washing regularly the roads (watering the road before brushing it mechanically). An area of 1 km² in the city centre was washed many times every night for ten days between February and March 2002. The variation of PM₁₀ both in concentration and composition at two different heights (2 and 25 meters) was investigated and compared with a reference site outside the test area.

The main conclusions drawn from the experiment may be summarized as follows:

- even when extending the washed area, no substantial reduction in PM₁₀ concentrations are to be expected;
- between the ground and 25m above, a vertical concentration gradient of about -10% exists but it is not influenced by the road washing;
- the anthropogenic sources are responsible for the 90% of the PM₁₀ concentrations, so that the road washing is not able to change the relative rate between anthropogenic and crustal sources. The meteorology can alter the components ratio not more than 3%.

Road sanding and PM emissions: a case study from Finland

The emissions from road dust are a particular problem in Scandinavian countries. High concentrations occur usually in early springtime, when snow has melted and roads are dry. This phenomenon has been extensively investigated in Finland (Kupiainen et al., 2003). Important findings are:

- The size fraction of road abrasion dust has been estimated as: TSP = PM₁₀ x 4; PM₁₀ = PM_{2.5} x 10.
- There is a good correlation between the amount of traction sand and PM₁₀ concentrations, at least under experimental conditions
- Pavement wear is strongly increased by sanding material ('sandpaper effect').

- It has been suggested that high-quality anti-skid aggregates in road material (high resistance to fragmentation) can reduce the amount of urban PM, compared to hard minerals (such as quartz) with low resistance to fragmentation.

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
Tyre wear, road abrasion	Small	Yes	Uncertain
Brake wear	Yes	No	Uncertain
Resuspension	(Yes)	Yes	Uncertain

A wide variety of options exist to control emissions from **unpaved roads**. The coarse fraction contains a considerable part of the PM₁₀ emissions from unpaved roads (according to EPA (2001) 75 %, see also Chapter 5). Control options fall into the following three groups (US EPA, 2001):

Vehicle restrictions that limit the speed, weight or number of vehicles on the road;

Surface improvement, by measures such as (a) paving or (b) adding gravel or slag to a dirt road; and

Surface treatment, such as watering or treatment with chemical dust suppressants.

A relationship between the moisture ratio "M", which is found by dividing the surface moisture content of the watered road by the surface moisture content of the uncontrolled road, and control efficiency is given in Figure 8.2.

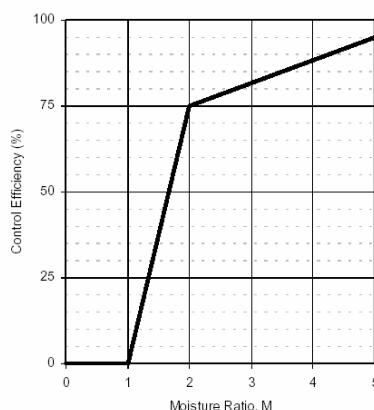


Figure 8.2 Dependence of control efficiency on surface moisture ratio (see text)

In addition, measures **to reduce road traffic volume** are effective for all mentioned emissions. There include economic (incentives), fiscal (taxes; road pricing), voluntary, regulatory (ban), informative (campaigns) and educational measures.

- Designation of zones with reduced emissions (low-emission zones)
A feasibility study for a low emission zone in London (AEA, 2003) revealed an appreciable potential for reduction PM limit value exceedances, depending on the stringency in excluding high emitting vehicles.
- Development of traffic reduction schemes to accompany new by-passes or other free-flow traffic measures
This could be a quite effective measure especially for small and medium-sized towns in order to exclude through-traffic from town or housing areas. A recent study in the effects of a bypass road revealed a fall of more than 30% of the PM pollution in the city centre of Nauen (a town near Berlin, Germany) (Lutz, 2003).
- Traffic management measures (traffic calming, road closure, re-routing, 30 km/h zones etc.)
A recent test of a temporary truck rerouting and of a speed limit (from 50 to 30 km/h) in a busy road in Berlin (HEAVEN, 2003) showed a mitigating effect of the truck ban on total PM pollution of around

7%. The impact of the speed limit on PM concentrations was marginal, but nevertheless positive. It should also be noted that blocking roads or rerouting traffic needs very careful examination of the situation in those roads bearing the rerouted traffic, so that the PM exposure of the population living there does not increase.

- Reallocating road space e.g. High Occupancy Vehicle Lane, Bus Priority Scheme, Cycle Lanes
- Park and Ride schemes
- Pedestrianisation of town centres
- Price Charging– at congested points in urban areas (like the congestion charge in London) or at tolls along motorways or major routes
- Integration of environmental concerns into transport policy at European, national and municipal level
- Promotion of policies that will advance a modal shift from private vehicle to public vehicle use
- Implementation of green travel plans.

Especially the last five examples of measures are associated with long-term planning, infrastructure investment and policy processes which require a time frame much longer than the attainment period of the PM Stage I limit values. So, their contribution to compliance with Stage I limit values is fairly limited.

It is possible to indirectly evaluate the theoretical potential of such measures by assessing the weekly variations in PM concentrations with significantly lower levels on weekends (due to reduced activity levels) at monitoring sites which are predominantly affected by traffic emissions. An illustrative example of daily variations of PM₁₀ on different weekdays is shown in Figure 8.3 for the Austrian site Lienz. While the night-time PM levels are practically unchanged, the peaks during morning and afternoon hours are sharply decreased. It should be noted, however, that weekends reflect an unusual situation in the sense, that traffic demand is a lot lower everywhere compared to working days. It is obvious that a durable large-scale reduction of traffic demand to weekend levels is hardly possible in practice.

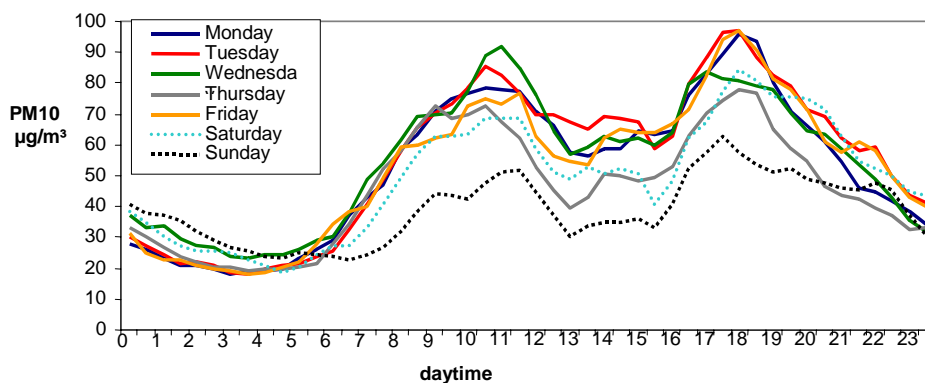


Figure 8.3 Diurnal variation of PM₁₀ on different weekdays at an Austrian traffic related site (Lienz, Tyrol; winter 2001/2002)).

There is extensive experience in Italy on the effect of short term actions for reducing PM₁₀ concentrations; these measures were primarily issued for traffic (ARPA Lombardia, 2003a,b; ARPA ER 2003). A detailed description of the areas and the measures is given in Annex 7.

Short term actions for reducing PM₁₀ in Italy

Both in Lombardy and in Emilia Romagna, the effectiveness of the measures has been recently assessed by the Regional Environmental Protection Agencies (ARPA Lombardia 2003 and ARPAER 2003), paying particular attention to the confounding effect of the meteorological changes which can obviously lead to misinterpretation.

In Lombardy the analysis showed that the measures reduce the concentrations of the primary pollutants, but have only limited influence on the PM₁₀ concentrations. The small size of the

effects may be explained considering that the agglomerations affected by the traffic limitations were surrounded by a territory with more than 2,200,000 of inhabitants within an area of about 4500 km², characterized by important sources of PM₁₀ and its precursors such as NO_x, SO_x and NH₃.

A more detailed comparison among the provisions lead to the conclusion that a pre-EURO vehicles ban is not very effective: the average reduction of the PM₁₀ concentrations was around 9% during the ban period, with a decrease in kilometres driven below 5%.

An alternate odd-even license plate ban appears a slightly more efficient, with a reduction of PM₁₀ concentration of about 12-14% and kilometres driven reduced by the same extent. During a complete ban of the vehicles circulation – with an evident decrease of primary pollutants concentration – the reduction of PM₁₀ concentration still depends on meteorological conditions and may reach 35-45%. The PM₁₀ composition changes during alternate odd-even plates bans are still under evaluation.

In Emilia Romagna, the analysis of the trend in PM concentrations at the monitoring stations of the regional network showed an average reduction of PM₁₀ concentrations on the Thursdays between October 2002 and March 2003 of about 10%. In contrast, the concentrations on Sundays are not far from those of a normal Sunday with typical weekend traffic levels. On a regional basis, the average reduction of the circulating vehicles during the regulated days has been nearly 15%, while the number of passengers on the public transportation system increased even more (30%).

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
Local traffic management to reduce traffic	Yes	Yes	Variable

8.2.4 Off-road transport

The control options possible for mobile road sources with diesel engines do in principle also apply for off-road diesel machinery. Filtering techniques are available (a list can be obtained from BUWAL (1999) and are often well suited for retro-fitting.

Railways may also contribute significantly to PM emissions. These include not only emissions from diesel driven engines, but also abrasion from breaks and tracks. Diesel emissions can be avoided by switching to other engine types or by technical abatement measures (particle filters). Less is known about measures to reduce emissions from break wear and tracks (BUWAL, 2002).

Abatement options for **ships** include the switch to low sulphur fuel. In addition, an optimisation of fuel combustion conditions can reduce emissions; in principle, also secondary abatement techniques (e.g., catalytic converters and filters) are applicable.

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
Off road machinery with diesel engines (filters)	Yes	No	Very high
Railways: Diesel engines (Substitution; filters)	Yes	No	Very high
Railways: Break and tracks	No	(Yes)	Uncertain

8.2.5 Domestic sources

The most important (small) **domestic sources** are those operated with solid (coal, wood and biomass) and liquid fuels. Possible measures can aim at

- Increasing the thermal standard for buildings
- Increasing energy efficiency
- Provide energy from installations with better controlled emissions (district heating)
- Use fuels with low specific emissions (e.g., gas)
- Use only burning devices, which are type approved. Type approval should be linked to compliance with certain emission standards.

An illustrative example of emission reduction potentials, in mass/unit mass dry wood burned, for various alternatives to conventional stoves and cordwood is shown in Table 8.2 (EPA, 1998).

Table 8.2 *Emission factors and reduction potential of wood stoves (EPA, 1998).*

Appliance	Particulate Emission Factor (g/kg)	Reduction in %
Conventional	18.5	-
Non-Catalytic	6	68
Catalytic	6.2	65
Pellet	2	89
Masonry Heater	3	84
Conventional with densified fuel	14	24

Measures that increase energy efficiency (e.g., increasing thermal standards of buildings) will also reduce emissions of greenhouse gases (GHG). However, there is a potential conflict with measures that increase the use of renewable fuels to decrease the net emissions of GHG, since biomass burning in very small installations may cause significant emissions of PM.

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
Domestic sources using solid fuels (Fuel shift; increase thermal standard.)	Yes	No	Substantial

8.2.6 Industrial sources including power generation

According to IPPC Directive (96/61/EC), new and existing large industrial installations (from 2007 on) have to apply Best Available Techniques (BAT). To assist Member States in assessing BAT under the IPPC Directive 96/61/EC, reference documents (BREFs) for certain industrial installations have been prepared by technical expert groups, which must be taken into account when authorities of Member States determine conditions for IPPC permits. These are available at <http://eippcb.jrc.es/pages/Boutline.htm>.

For emissions from industrial **combustion processes** control techniques are generally well established. These include electrostatic precipitators and tissue filters, scrubbers and for smaller installations also cyclones. These techniques differ substantially in their efficiency at different size fractions. An illustrative example indicating the large differences in efficiency is shown in Figure 8.4 (from <http://www.scheuch.at>).

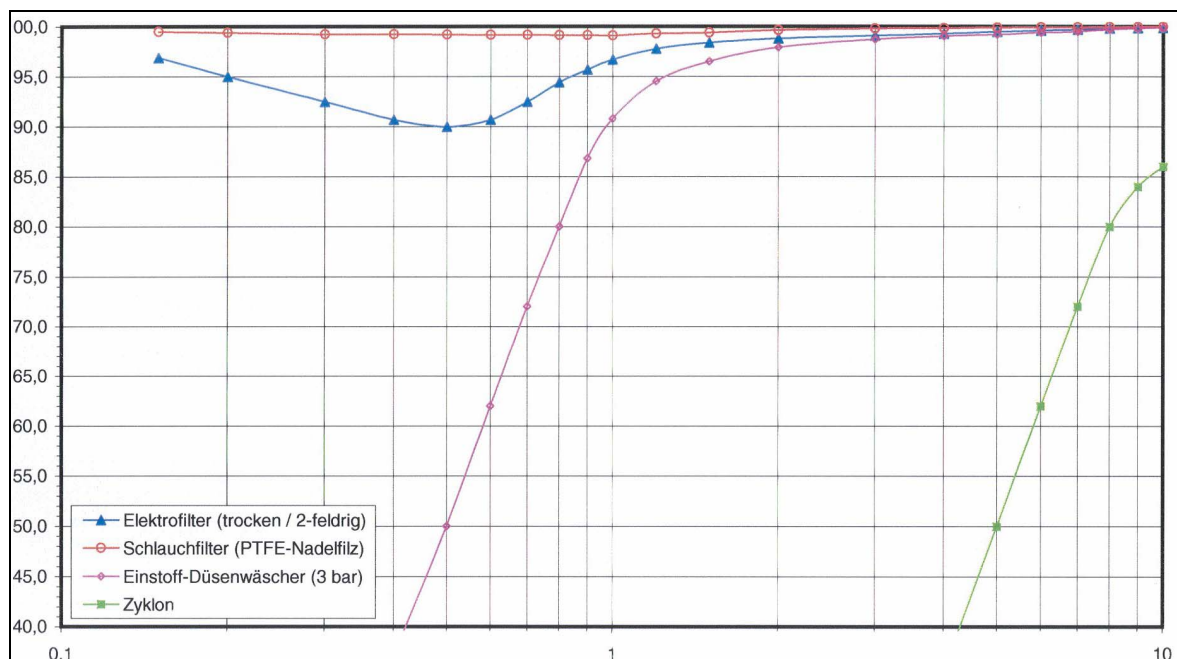


Figure 8.4 Removal efficiency (in %, on y-axis) of 2 stage electrostatic precipitator ('Elektrofilter'), tissue filter ('Schlauchfilter'), spray scrubber ('Einstoff-Düsenwäscher') and cyclone ('Zyklon') in relation to particle size (x-axis, in μm and logarithmic scale).

Hoods, enclosures and housings may capture fugitive emissions. In addition, systems can be installed to capture fugitive emissions. These captured streams can subsequently be ducted to an emission control system, e.g., the electrostatic precipitator, scrubber, bag house or cyclone. In addition, **good operational practice** to prevent or reduce fugitive losses can minimize these emissions.

Efficiencies and applicability varies largely between different processes/industrial branches. Additional information can be found at EPA (2001).

8.2.7 Commercial sources

8.2.7.1 Combustion sources

For combustion sources, in principle similar options exist as for domestic and industrial sources (technical measures such as the use of cyclones, electrostatic precipitators and filters; use of different fuels).

8.2.7.2 Construction, material handling

Measures to reduce the emissions of PM during construction activities include the use of low emission machinery. This may also be necessary for workplace security.

Fugitive emissions may be reduced during different processes including

- Processing, management and transport of materials (watering of materials; optimised logistic concepts; see also measures listed under unpaved roads; early paving of permanent roads);
- Storage of materials (coverage of materials, wet suppression, chemical stabilization);
- Deconstruction (watering).

However, monitoring the enforcement of these measures is difficult.

Source	Effect on		Reduction potential
	Fine PM (PM _{2.5})	Coarse PM PM _{2.5-10}	
Transport, storage and handling of materials; construction work;	No	Yes	Substantial

8.2.8 Agricultural Sources

Reduction of emissions of the PM precursor NH₃ is potentially the most important measure in the agricultural sector. Measures to reduce PM emissions from (diesel) engines are described in the sections on transport. In addition, measures for certain processes are available:

- Abatement options in **animal housing** include filtering and optimised ventilation;
- Measures in the agricultural sector include a **ban of stubble burning**;
- Soil resuspension can be reduced promoting **plant coverage** of unused fields;
- **Prevention of fires** in (managed) forests is necessary.

8.2.9 Reduction efficiencies used in the integrated assessment model RAINS

Table 8.3 summarises the emissions reduction potentials of the abatement options used in the RAINS model. It is important to note that these reduction potentials refer to the unabated case. It has to be emphasised that for many of the sources included abatement measures are already applied and that further reduction potentials are therefore often considerably smaller.

Table 8.3 Emissions reduction potentials (%) for different sources in the RAINS model (IIASA, 2003¹⁷)

Control Option	PM _{fine}	PM _{coarse}	PM _{>10}
Electrostatic precipitator: 1 field - power plants	93	95	97
Electrostatic precipitator: 2 fields - power plants	96	99	99.9
Electrostatic precipitator: more than 2 fields - power plants	99	99.9	99.95
Fabric filters - power plants	99	99.9	99.98
Cyclone - power plants	30	70	90
Wet scrubber - power plants	96	99	99.9
Electrostatic precipitator: 1 field - industrial combustion	93	95	97
Electrostatic precipitator: 2 fields - industrial combustion	96	99	99.9
Electrostatic precipitator: more than 2 fields - industrial combustion	99	99.9	99.95
Fabric filters - industrial combustion	99	99.9	99.98
Cyclone - industrial combustion	30	70	90
Wet scrubber - industrial combustion	96	99	99.9
Electrostatic precipitator: 1 field - industrial processes	93	95	97
Electrostatic precipitator: 2 fields - industrial processes	96	99	99.9
Electrostatic precipitator: more than 2 fields - industrial processes	99	99.9	99.95
Wet electrostatic precipitator: industrial processes	99	99.9	99.95
Fabric filters - industrial processes	99	99.9	99.98
Cyclone - industrial processes	30	70	90
Wet scrubber - industrial processes	96	99	99.9
Good housekeeping: industrial oil boilers	30	30	30
Good practice: industrial processes - Stage 1 (fugitive)	40	40	40
Good practice: industrial processes - Stage 2 (fugitive)	80	80	80
Fireplaces, catalytic insert	47	47	47

¹⁷ <http://www.iiasa.ac.at/rains/index.html>

Control Option	PM _{fine}	PM _{coarse}	PM>10
Fireplaces, non-catalytic insert	44	44	44
New domestic stoves (wood): non-catalytic	63	63	63
New domestic stoves (wood): catalytic	65	65	65
New domestic stoves (coal): Stage 1	30	30	30
New domestic stoves (coal): Stage 2	50	50	50
New domestic boilers: (coal)	40	40	40
New medium (automatic) size boilers: (wood chips, pellets)	89	89	89
New medium boilers: (wood chips, pellets) with end-of-pipe abatement	99	99.9	99.98
Cyclone for medium boilers in domestic sectors	30	70	90
Bag house for medium (automatic) boilers in domestic sector	99	99.9	99.98
Good housekeeping: domestic oil boilers	30	30	30
EURO I - 1992/94, diesel light-duty and passenger cars	61	61	61
EURO II - 1996, diesel light-duty and passenger cars	75	75	75
EURO III - 2000, diesel light-duty and passenger cars	86	86	86
EURO IV - 2005, diesel light-duty and passenger cars	93	93	93
EURO V - diesel light-duty and passenger cars, post-2005, Stage 1	99.95	99.95	99.95
EURO VI - diesel light-duty and passenger cars, post-2005, Stage 2	99.99	99.99	99.99
Construction and Agriculture Off-road - 1998, as EURO I for HDV	20	20	20
Construction and Agriculture Off-road - 2000/02, as EURO II for HDV	50	50	50
Construction and Agriculture Off-road; as EURO III for HDV	85	85	85
Construction and Agriculture Off-road; as EURO IV for HDV	97	97	97
Construction and Agriculture Off-road; as EURO V for HDV	97	97	97
Construction and Agriculture Off-road; as EURO VI for HDV	99.95	99.95	99.95
Rail and Inland Waterways Off-road - 1998, as EURO I for HDV	20	20	20
Rail and Inland Waterways Off-road - 2000/02, as EURO II for HDV	50	50	50
Rail and Inland Waterways Off-road; as EURO III for HDV	85	85	85
Rail and Inland Waterways Off-road; as EURO IV for HDV	97	97	97
Rail and Inland Waterways Off-road; as EURO V for HDV	97	97	97
Rail and Inland Waterways Off-road; as EURO VI for HDV	99.95	99.95	99.95
EURO I - 1992, heavy-duty diesel vehicles	45	45	45
EURO II - 1996, heavy-duty diesel vehicles	77	77	77
EURO III - 2000, heavy-duty diesel vehicles	85	85	85
EURO IV - 2005, heavy-duty diesel vehicles	97	97	97
EURO V - 2008, heavy-duty diesel vehicles	97	97	97
EURO VI, heavy-duty diesel vehicles, post-2008	99.95	99.95	99.95
EURO III, gasoline direct injection engines	50	50	50
EURO IV, gasoline direct injection engines	50	50	50
EURO V, gasoline direct injection engines	51	51	51
EURO VI, gasoline direct injection engines	52	52	52
EURO I, Light-duty, spark ignition engines: 4-stroke, not DI	50	50	50
EURO II, Light-duty, spark ignition engines: 4-stroke, not DI	50	50	50
EURO III, Light-duty, spark ignition engines: 4-stroke, not DI	50	50	50
EURO IV, Light-duty, spark ignition engines: 4-stroke, not DI	50	50	50
EURO V, Light-duty, spark ignition engines: 4-stroke, not DI	51	51	51
EURO VI, Light-duty, spark ignition engines: 4-stroke, not DI	52	52	52
Motorcycles and mopeds 2-stroke, Stage 1	30	30	30
Motorcycles and mopeds 2-stroke, Stage 2	70	70	70
Motorcycles and mopeds 2-stroke, Stage 3	71	71	71
Motorcycles 4-stroke, Stage 1	50	50	50
Motorcycles 4-stroke, Stage 2	51	51	51

Control Option	PM _{fine}	PM _{coarse}	PM _{>10}
Motorcycles 4-stroke, Stage 3	52	52	52
Heavy-duty, spark ignition engines, Stage 1	50	50	50
Heavy-duty, spark ignition engines, Stage 2	51	51	51
Heavy-duty, spark ignition engines, Stage 3	52	52	52
Combustion modification: ships (medium vessels)	20	20	20
Combustion modification: ships (large vessels - fuel oil)	40	40	40
Combustion modification: ships (large vessels - diesel)	20	20	20
Good practice: storage and handling	10	20	50
Feed modification (all livestock)	10	35	45
Hay-silage for cattle	10	40	70
Free range poultry	5	15	40
Low-till farming, alternative cereal harvesting	5	15	40
A generic option for 'other animals' - good practice	5	15	40
Good practice in oil and gas industry - flaring	5	15	40
Ban on open burning of agricultural or residential waste	100	100	100
Good practice in mining industry	25	47	54
Spraying water at construction places	10	20	50
Filters in households (kitchen)	10	20	50

8.3 Reduction potentials

A few local and national studies are available within which the reduction potential of measures to abate PM are assessed. Exceedances of the 24-hour Stage 1 limit value are frequent in Europe (see Chapter 4) and hence there is a widespread necessity to reduce PM₁₀ levels. The highest exceedances usually occur at hot spot locations in large agglomerations. City authorities are often confronted with challenging tasks in designing measures to comply with the PM₁₀ limit values of the First Daughter Directive, since high background concentrations of PM, out of control for local measures, occur in many areas in Europe. Figure 8.5 gives an illustration of the different contributions in Berlin.

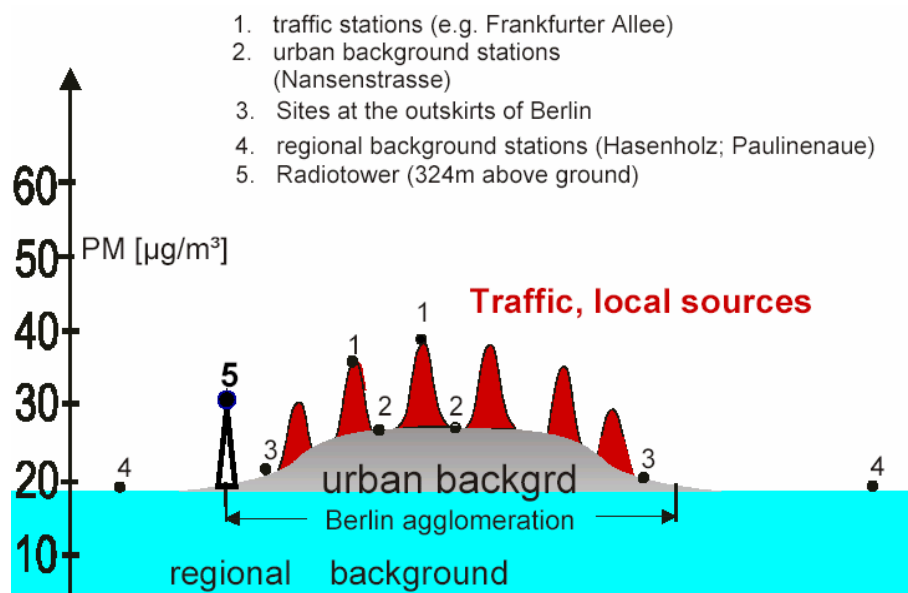


Figure 8.5. Schematic illustration of different contributions to PM₁₀ levels in Berlin (from Lenschow et al., 2001).

This section presents below illustrative examples of studies carried out in Berlin (Germany), the Netherlands and the United Kingdom.

8.3.1 City of Berlin

At traffic related sites in Berlin, more than 100 exceedances of the 24-hour limit value of $50 \mu\text{g}/\text{m}^3$ were recorded in 2003. More than 40 exceedances were measured at some of the urban background locations. A recent assessment of data from 2002 indicated that almost 60 % of the levels at traffic sites originates from regional background, while local traffic makes up about one quarter of the levels. Episodes of high PM_{10} levels are often linked with an even larger share of regional/large scale transport of PM, contributing up to 80% of the total PM_{10} burden (Lenschow et al., 2001; Lutz, 2003).

Some rough and preliminary estimates of the effect of different measures are given in Table 8.4 (Lutz, 2003).

Table 8.4 Estimates of the effect of abatement measures for Berlin (Lutz, 2003)

Measures	Estimated reduction of PM levels in 5 years
<i>EU-wide measures (current legislation):</i> Emission limits for mobile sources; fuel quality; NEC; IPPC, LCP	8 % (compared to 2000)
<i>Berlin related measures that are already under way:</i> Particle traps for all public transport buses, pilot program for CNG-vehicles, substitution of coal by natural gas, oil and district heating, BAT for industrial installations, investments in public transport & cycling infrastructure, bus lanes	4% (compared to 2000)
<i>Possible Berlin related measures under consideration in the future:</i>	
– accelerated turnover of the diesel fleet: enhanced introduction of CNG-vehicles in captive fleets, promotion of retrofit with PM traps, requiring EEV/EURO V standards for public buses, low emission zone	<8%
– Optimised traffic management for hot spots	<4%
– traffic and city planning aimed to shift modal split: enlarged zones with parking fees, optimising the public transport network, further investments in cycling infrastructure	Little effect on the short time
– BAT for stationary sources	Potential almost exhausted

According to city authorities, these reductions are not sufficient to comply with the Stage 1 24-hour limit value for PM_{10} , in particular in years with adverse meteorological conditions. It was concluded that local measures:

- are necessary, but have a limited effect on total PM_{10} concentrations at urban hot spots,
- would have to focus on motor traffic emissions as the main inner-city source,
- have to be complemented by European and national measures, such as stricter vehicle and fuels standards (e.g. EURO5 and EURO6),
- need to be accompanied by EU-wide efforts to further reduce the large-scale PM background concentrations.

This has been confirmed in a resolution signed by eight European metropolises (London, Paris, Warsaw, Prague, Rome, Stockholm, Stuttgart and Berlin), adopted at a conference in Berlin in November 2003 (SenStadt, 2003).

8.3.2 The Netherlands

In the Netherlands (Opperhuizen et al., 2003) emissions of PM₁₀ for different scenarios were derived (see Table 8.5). The CLE scenarios (CLE stands for **c**urrent **l**egislation) account for measures that were already decided upon within the European Union or national legislation. In contrast, the MFR (maximum feasible reduction) scenario includes all measures that are technically feasible (but not necessarily cost effective).

Table 8.5 *Trend and scenarios for Dutch PM₁₀ emissions*

Year	Dutch PM ₁₀ emissions in ktonnes
1980	116
1995	61
2005-CLE	48
2010-CLE	42
2010-MFR	18

Only moderate reductions are predicted without new measures in the period from 2005 until 2010. Additional measures on top of current legislation could reduce PM₁₀ emissions by more than 50%, however, at increasingly high costs.

8.3.3 United Kingdom

The Interdepartmental Group on Costs and Benefits (DEFRA, 2001) has investigated the costs and benefits of possible measures to further reduce PM₁₀ levels in the United Kingdom in addition to a CLE scenario that includes measures that are already agreed.

According to these estimations, major emissions reductions are expected in the period from 1999 to 2005 (mainly in the sectors power production, residential plants, quarrying and combustion from road transport, see Table 8.6) while no substantial reductions are predicted for the period beyond 2010. This group also identified possible additional measures (an illustrative scenario). The potential impact of these measures on industrial and domestic sources was an additional cut of emissions by 40% compared to the emissions in 2010 of these sources. Technical measures for road traffic combustion (S-free fuels; particle trap from 2006 for new light duty and heavy duty vehicles; retrofit programme for certain areas) would cut these emissions by an extra around 15% in 2010.

Table 8.6 *UK PM₁₀ emissions in a CLE scenario*

Year	PM ₁₀ emissions in ktonnes
1999	186
2005	111
2010	98
2015	93
2020	93

8.3.4 Europe

IIASA estimated the emissions of European countries for 1990 and 2010. For 2010 a current legislation (CLE) and a maximum feasible reduction (MFR) scenario was analysed (IIASA, 2002). However, it has to be highlighted that the figures, shown in Table 8.7, are preliminary, and more robust figures are currently (spring 2004) prepared by IIASA as part of the baseline development for CAFE.

Table 8.7 *PM_{2.5} emissions in 1990 and 2010 (CLE and MFR). Provisional data*

	National PM _{2.5} emissions in ktonnes		
Country	1990	2010 CLE	2010 MFR
Austria	35	22	14
Belgium	36	19	19
Denmark	17	11	8
Finland	41	25	18
France	184	95	94
Germany ¹⁾	464	105	116
Greece	51	51	26
Ireland	13	6	6
Italy	127	68	60
Luxembourg	4	2	1
Netherlands ¹⁾	29	16	18
Portugal	22	19	11
Spain	100	79	63
Sweden	45	22	18
UK	152	70	65
EU-15	1320	611	537
Czech Republic	205	81	26
Estonia	54	17	5
Hungary	65	21	8
Latvia	14	8	3
Lithuania	20	10	3
Poland	333	259	92
Slovakia	59	24	11
Slovenia	16	12	4
EU-23 ²⁾	2086	1043	689
Albania	13	8	2
Belarus	47	27	8
Bosnia-H.	36	23	5
Bulgaria	130	98	24
Croatia	18	12	4
Norway	13	9	7
Moldova	14	8	3
Romania	175	107	24
Russia	953	489	148
Switzerland	11	7	7
Macedonia	12	7	2
Ukraine	714	288	76
Yugoslavia	61	36	8
Total	4283	2162	1007

¹⁾ The lower number for CLE compared to MFR was derived from the IIASA report.

²⁾ Currently, no data are available for Cyprus and Malta.

According to IIASA's assessment, measures on top of the CLE scenario have only limited potential to further reduce primary PM_{2.5} emissions in existing EU Member States. However, the situation is different in Acceding countries and other non-member states. There are, however, significant

differences in the national estimates done by the United Kingdom and the Netherlands and those of IIASA in relation to further measures on top of CLE. At this stage, it is unclear if these differences are due to the different metrics (PM₁₀ versus PM_{2.5}) or different assumptions in the modelling approach.

IIASA has also prepared an analysis of emissions of PM precursors in 1990 and reduction potentials for 2010 ('maximum feasible reductions', MFR, i.e. reductions which can be achieved applying existing technical abatement measures; IIASA, 2002). In Table 8.8, the emissions were normalised to the emissions in 2010 under the current legislation scenario (CLE).

Table 8.8 Emissions of PM precursor, normalised to the predicted emissions in 2010 under current legislation

	SO ₂			NO _x			NH ₃		
	1990	2010 CLE	2010 MFR	1990	2010 CLE	2010 MFR	1990	2010 CLE	2010 MFR
Europe	2.80	1	0.32	1.69	1	0.59	1.22	1	0.70
EU-15	4.05	1	0.46	2.01	1	0.72	1.17	1	0.69
EU-23 ¹⁾	3.47	1	0.44	1.89	1	0.68	1.16	1	0.70

¹⁾ No data are available for Cyprus and Malta.

For NMVOC (also a precursor of secondary aerosols) IIASA has estimated that emissions might be reduced another 41% by applying maximum feasible reduction measures.

IIASA's assessment indicates that there is substantial further reduction potential for PM precursors in comparison to current legislation. IIASA is currently updating its emissions projections for PM and PM precursors as part of work under a contract from DG ENV to develop a baseline scenario for CAFE. Some of the trends in PM levels reported in Chapter 7 are at least partly attributable to past reductions in the emissions of precursors of secondary PM, thereby demonstrating the efficiency of such measures (even though those measures were predominantly not taken to reduce PM levels).

8.4 Cost-effectiveness of measures

The cost-effectiveness of measures depends on several different issues like the contribution of different sources to pollution levels, the reduction potential and the costs of additional measures. In addition, it is necessary to define

- the metric under consideration (like PM₁₀, PM_{2.5}; the latter requires more emphasis on combustion sources)
- the scale of the problem (reduction of the background level versus reduction of hot spot concentrations)
- the objective to be achieved (compliance with LV at hot spots; reduction of health effects).

Few studies have been conducted to investigate the cost-effectiveness of different measures. In general, studies are hampered by uncertainties in emission estimates, problems in modelling PM levels accurately and uncertainty on the efficiency of measures, in particular on those affecting non-combustion sources.

In the United Kingdom, the Interdepartmental Group on costs and benefits has (DEFRA, 2001) investigated a number of measures on the industrial, domestic and transport sector. Overall, the transport measures in the package appeared to be less cost effective than many of the potential industrial measures in terms of reducing background PM₁₀ concentrations. However, it was found that the transport measures were better targeted at reducing roadside concentrations and were more effective at this than the industry measures in London, where the highest roadside concentrations are found.

Much more information on local and regional measures, their costs and their effectiveness can be expected in the coming years. Member States had to implement the first air quality daughter directive (1999/30/EC). Under this directive, plans and programs have to be developed, implemented and reported to the Commission to reduce future PM_{10} levels if the sum of limit value and margin of tolerance is exceeded. If such exceedances occurred in 2001, plans and programmes have to be forwarded to the Commission at the end of 2003.

The highest exceedances occur in hot spot locations such as traffic related sites and industrial sites (see Chapter 4). In addition, the source apportionment points at strong contributions from long-range transport and traffic sources, and sometimes industrial sources. These sources will have to be addressed to affect PM levels significantly. In addition, it has to be kept in mind that the advice given by WHO suggests that tailpipe emissions from traffic and other combustion sources are critical sources in terms of health effects (see Chapter 10). This is a clear indication that combustion sources should be considered with some priority when assessing abatement strategies. In terms of exposure, those sources are of most interest which give rise to high concentrations in areas where people are likely to be exposed, such as road traffic within cities.

On the European level, IIASA has been contracted to develop the baseline scenario (which includes the establishment of emission and ambient air quality projections, including a current legislation scenario until 2020). This scenario will provide the basis for further analysis of possible emission reduction scenarios to achieve certain targets cost-effectively. Results of these analyses are expected in 2004.

8.5 Conclusions and recommendations

8.5.1 Conclusions

Chapter 5 provides an overview of the main emission sources of primary PM and secondary PM. In addition, there is a clear relationship between past measures and decreasing trends in PM levels in ambient air, demonstrating the efficiency of several measures (Chapter 7). For all the combustion sources mentioned in this chapter, numerous control options are available. These include technical measures, but also non-technical measures. The size fraction affected depends on the sources characteristics and the measure applied, but mainly the fine fraction is affected. The removal efficiency of the different measures is usually well established.

For non-combustion sources, also numerous measures have been described and tested, even though studies showing the quantified effects of such measures are scarce. However, the overall importance of these sources is decreased if the metric of interest is $PM_{2.5}$ rather than PM_{10} .

IIASA has estimated the reduction potential of additional measures on top of scenarios including currently agreed abatement measures for 2010. In general, a large potential exists for precursors of secondary aerosols, in particular for SO_2 , but also NO_x , NH_3 and NMVOC. The results for reduction potentials on top of current legislation scenarios for primary PM are somewhat conflicting. However, in general larger reduction potentials for primary PM on top of current legislation can be expected for Accession Candidate Countries and other non-EU countries compared to current Member States.

An assessment of the reduction potentials in a large agglomeration in Western Europe confirmed that measures to reduce local sources, in particular road traffic, are necessary, but have a limited effect on total PM_{10} concentrations even at urban hot spots. Local measures have to be complemented by European and national measures, such as stricter vehicle and fuels standards (e.g. EURO5 and EURO6) and need to be accompanied by EU-wide efforts to further reduce the large-scale PM background concentrations.

There is no simple answer on the cost effectiveness of measures, since this depends crucially not only on the availability and effectiveness of measures, but also on the PM metric under consideration, the scale (e.g. local hot spot versus urban or regional background) and the objectives (e.g., compliance with limit value versus reduction of health impact). Source apportionment studies have shown the important contributions of secondary inorganic aerosols from NO_x and SO₂ emissions and primary emissions from traffic to ambient PM levels. In addition, WHO has identified traffic and other combustion sources as critical to health impacts. Consequently, these sources should in any case be primarily considered in designing abatement strategies.

A comprehensive assessment of cost-effective abatement strategies to reduce PM impact on human health will be conducted by IIASA. These results are expected to be available in 2004. However, this integrated assessment will presumably cover only regional and urban background concentrations.

8.5.2 Recommendations

1. The First Daughter Directive (1999/30/EC) requires Member States to develop, implement and report air quality management plans to ensure that the limit values will be met in 2005. It can be expected that such plans will be developed for local hot spots, and also for urban regions. The experiences gained in developing such plans should be exchanged and assessed in the coming years.
2. PM pollution has a strong regional component, but also local contributions. This poses problems for air quality managers, since they have to handle different scales concurrently in developing abatement strategies. Therefore, robust European scenarios on emissions and ambient (background) levels of PM should be made available to support air quality management at a local level.
3. Some of the measures (such as more stringent emission limits for mobile sources) are most effectively taken at a European scale. The Commission is therefore requested to investigate further measures on a Community level to support Member States in reaching the PM limit values and targets.

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9. Attainability

9.1 Scope

This chapter synthesises information from elsewhere in the Position Paper (mainly Chapters 6, 7 and 8) to assess the attainability of current targets in the First Daughter Directive. It discusses whether Member States are able to meet the goals in the First Daughter Directive, and if not, why not and what abatement options are. It gives preliminary recommendations on what Member States and the Commission can do about attainability.

9.2 Are Member States able to meet goals in the First Daughter Directive?

Stage 1 Limit Values

Annual average concentrations of PM₁₀ have generally decreased since the late nineties. However, there is considerable variation between different monitoring sites, and not all locations display this downward tendency. We do not know the exact reasons for this general decrease. There are however strong indications that the reducing trend in rural and urban background has been driven mainly by reductions at the European level in precursor emissions of SO₂ and NO_x.

Despite the improving situation, there are currently exceedances of the Stage 1 annual and 24-hour limit values in most Member States. In 2001 the limit values were exceeded at 34% of the 718 monitoring sites¹⁸ reported under the Exchange of Information Decision (see also Chapter 4). According to a preliminary overview by the Commission of the first annual reports under the First Daughter Directive by 13 Member States on their air quality in 2001, 11 Member States reported exceedance of a Stage 1 limit value (the annual limit value in 8 and the 24-hour limit value in 10 Member States). The higher limit values plus margin of tolerance¹⁹ were exceeded in 7 Member States (the annual limit value in 6 and the 24-hour limit value in 7 Member States). There is not enough information currently to tell to what extent natural events and resuspension of particulates following the winter sanding of roads contribute to the observed exceedances²⁰. The overwhelming part of annual average PM levels at least in mid latitudes Europe away from the influence of marine aerosol seems to originate from anthropogenic emissions (see Chapter 6).

The short-term limit is exceeded more than the annual average LV and is a more stringent target (Chapter 4).

Only few Member States have modelled projections for PM₁₀. Such calculations are complex and the results are uncertain (Chapter 7) and have to be interpreted with care.

Keeping in mind this caveat, we can draw some conclusions from existing studies:

1. Background concentrations will likely continue to decrease to at least 2010. This decline will be driven by further reductions in the emissions of PM and its precursors SO₂, NO_x and NH₃, as required under EU legislation such as the National Emission Ceilings Directive, the Large Combustion Plant Directive and Directives on mobile sources.

¹⁸ With greater than 70% data capture

¹⁹ For 2001, the limit value plus margin of tolerance was 46 µg/m³ for the annual mean and 70 µg/m³ with 35 allowed exceedances for the daily mean PM₁₀ concentration.

²⁰ In such cases, limit values may be exceeded without the necessity to take actions

2. Results from selected Member States indicate that the Stage 1 annual average limit value can be met by 2005 at most locations with current measures or moderate additional measures.
3. The situation is less clear for the Acceding Countries. No national projections were available to the Working Group from these countries.
4. The most polluted monitoring sites – most of them traffic-related sites (see also Chapter 4) – in the EU and Accession Countries recorded concentrations around $60 \mu\text{g}/\text{m}^3$ and higher as annual mean in 2001. From statistical assessment (see Chapter 4) it is known that the 24-hour limit value (35 exceedances of a 24-hour PM_{10} level of $50 \mu\text{g}/\text{m}^3$) corresponds roughly to an annual mean of $30 \mu\text{g}/\text{m}^3$. Therefore, a reduction of PM_{10} levels in the order of 50% is needed in the most polluted sites within two or three years to comply with the more stringent 24-hour limit value. The Working Group considers it unlikely that pollution levels can be reduced sufficiently at all of these sites by the 2005 attainment date. To meet the 24-hour limit value, additional measures would have to be taken at different scales (local, regional and European) immediately and simultaneously, since emissions from these different scales contribute to the observed PM_{10} levels. The regional, urban background, and local traffic contributions to PM_{10} at the EU roadside stations can be estimated to account for 35-40, 10-35 and 30-50%, respectively. However, in the Netherlands and Sweden other contributions are found: 75, 10, 25% and 30-45, 5-20, 40-65%, respectively (see also Chapters 4 and 6). Measures would need to cover all major sources of PM and its precursors. In addition, several types of measure require considerable time to be implemented and to take effect.
5. The Working Group however believes that – in addition to compliance issues – PM_{10} levels at the most polluted sites are a serious threat to human health, and therefore measures should be taken as soon as possible to reduce these levels as far as practically possible.

Stage 2 Indicative Limit Values

There are currently widespread exceedances of the indicative limit values all over Europe (Chapter 6). This level is even exceeded significantly at some rural background sites. Recent trends and projections strongly suggest that the annual average and 24-hour Stage 2 indicative limit values will not be met in most Member States.

Furthermore preliminary modelling results from EMEP (Chapter 7) suggest that the Stage 2 indicative limit values will not be met widely even if Member States implement all technologically feasible abatement measures applied to primary sources of PM^{21} (both fine and coarse).

9.3 Why can't all limit values be met?

A large proportion of PM in Member States arises from outside their borders and so is beyond their control. For example, 8 to $16 \mu\text{g}/\text{m}^3$ annual average PM_{10} measured at Swedish regional background sites arises from long-range transport (Chapter 2). Similarly, PM concentrations in London are highly influenced by precursor emissions in Europe. For example, preliminary source apportionment modelling suggests that secondary PM (from outside London, including Europe) will contribute 26% of PM_{10} at an urban background, and 22% at a busy roadside site in 2010 (see Figure 9.1).

²¹ This scenario assumes that current measures continue to apply to PM precursor emissions.

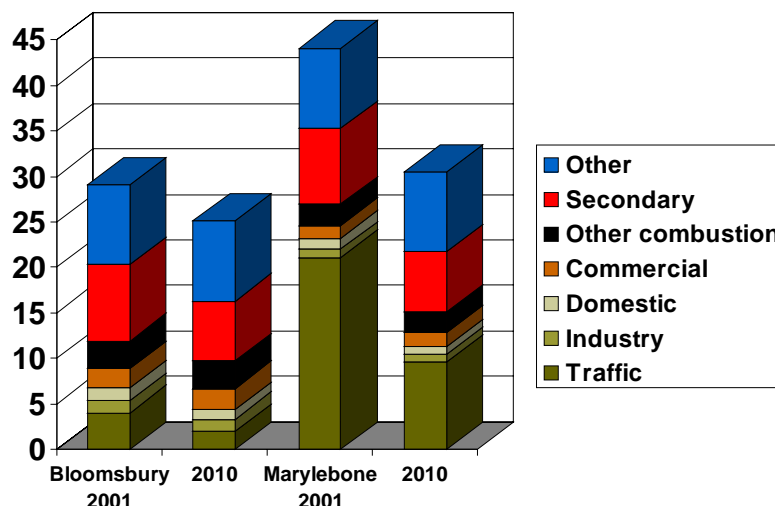


Figure 9.1 Modelled annual average PM₁₀ source contributions (µg/m³) at Bloomsbury (urban background) and Marylebone Road (roadside) locations in the UK in 2001 and 2010

There are even rural background sites (with no major emission sources within 20-30 km) in Central Europe that exceed the Stage 1 24-hour limit value. Consequently there is a limit to the ability of national and local measures to meet the limit values.

There are additional reasons why the Working Group believes that it is unlikely the Stage 1 24-hour limit value will be attained everywhere in 2005:

1. The current areas of exceedance are large and widespread although the highest exceedances usually occur at hot spots, for example near busy roads. Consequently any local measures to attain the LV must be applied at a large number of locations simultaneously. It is not simply a case of targeting a few hot spots. Simple relocation of emissions sources (e.g. by re-directing traffic) will often not be effective.
2. Many available measures take time to become effective. Even though several additional control options exist for some of the main emission sources, these measures take time to be implemented and become effective. As an example, the setting of new emission limit values for vehicles will only affect new vehicles (with the exception of retro-fitting programmes, which are anyway less practical for large fleets).
3. We know little about the contribution of resuspended road dust to PM₁₀ concentrations near roads, nor about the effectiveness of measures to reduce this source. Some studies indicate that commonly recommended measures (like frequent cleaning of roads) have a very limited effect on the amount of resuspension. If these results are confirmed, one of the major sources of urban PM can only be controlled effectively by non-technical measures.
4. Cost of additional measures. The few studies that have been conducted to estimate the cost-effectiveness of different measures indicate that additional measures to further reduce PM₁₀ emissions and concentrations are expensive, although they can yield large benefits to public health.

For example, the United Kingdom Government carried out an assessment of the costs and benefits of an illustrative package of national measures to achieve further reductions in PM₁₀ emissions and concentrations²². The illustrative package of measures²³ is estimated to lead to a 0.75 µg/m³ reduction

²² <http://www.defra.gov.uk/environment/airquality/igcb/index.htm>

²³ Considered to represent an upper bound to the measures that are likely to be implemented.

in UK population-weighted concentration levels in 2010. The potential abatement measures are spread across industry, the domestic sector and transport sectors, as well as the public sector. The total annual cost of this illustrative package of national measures in 2010 is estimated at between £785 million and £1,115 million. The application of the illustrative package of measures to the projections models reduces the annual mean PM₁₀ concentrations to below the Stage 2 indicative limit value at all but two monitoring sites in the United Kingdom.

There would be considerable health benefits of the reduction. A 0.75 µg/m³ reduction in particles from the proposed additional measures is predicted to lead to a gain of 278,000 to 508,000 life years for the UK population over the years from 2010 to 2110. After discounting, this is equivalent to 81,000 to 212,000 life years gained. In addition, this reduction is predicted to lead to 25200 fewer respiratory hospital admissions (additional or brought forward) in the United Kingdom. Consequent UK reductions in sulphur dioxide will lead to 4820 fewer deaths brought forward (2510 after discounting) and 3690 fewer respiratory hospital admissions (additional or brought forward).

A Netherlands study²⁴ has concluded that the cost of achieving a 60% reduction (the ultimate technical potential) – in addition to currently agreed measures – would be about 6000 million Euro per year. This would result in a concentration reduction of 1.1 µg/m³ averaged across the Netherlands compared to the base case. A reduction of 25% in emissions could be achieved at a cost of 200 million Euro per year and result on average in a reduction of 0.3 µg/m³. Much greater reductions would be achieved at hot spots.

Furthermore, IIASA has identified a range of additional technical options to mitigate PM emissions. Its maximum feasible reduction (MFR) emissions scenario assumes that all technologically feasible abatement measures (such as those described below) are applied to primary sources of PM. EMEP modelling, using the MFR scenario, indicates that even if Member States were to apply MFR, regardless of cost, the Stage 2 limit values would not be met widely in Europe (Chapter 7).

9.4 What are the abatement options?

9.4.1 Precursor emissions

Control options for emissions of PM precursors (NO_x, SO_x, NH₃ and VOCs) are well established. Precursor emissions reductions resulting from implementation of the National Emissions Ceilings Directive and other legislation will reduce secondary PM and so help Member States work towards attaining the limit values in the First Daughter Directive.

9.4.2 Primary emissions

There are numerous control options available for combustion and non-combustion sources of PM. Chapter 8 identifies abatement options with substantial emissions reduction potential from road transport, off-road transport, industrial and commercial sources.

The Netherlands has identified additional measures in addition to current legislation that would reduce primary PM₁₀ emissions by more than 50%. The United Kingdom has similarly identified additional measures that could reduce industrial and domestic emissions by 40% in 2010 and traffic measures that would reduce emissions by an additional 15% in 2010. Both countries identify significant additional costs to make these reductions.

²⁴ On health risks of ambient PM in the Netherlands (RIVM, 2002)

Effects on PM levels

Robust calculations are needed to assess the effect of different European emission reduction scenarios on regional PM levels for 2005, 2010 and later. IIASA will be modelling the potential impacts of reductions of primary and secondary PM on PM₁₀ concentrations as part of work under a contract to the Commission's DG-Environment to develop a baseline scenario for CAFE.

9.5 Conclusions and recommendations

9.5.1 Conclusions

The Stage 1 annual average limit value is likely to be attained in most Member States except at highly polluted locations or some urban background areas. In those areas, additional measures will be necessary to comply with the limit value in 2005.

Some Member States will encounter severe difficulties in attaining the Stage 1 24-hour limit value in 2005. One of the main problems is the high regional background concentration of PM₁₀, which is often caused by long-range air pollution.

Stage 2 indicative limit values are unlikely to be met widely throughout Europe. Attainability is not limited by the availability of technical options to reduce PM emissions. However, the concentration of PM in Member States is again strongly influenced by transboundary PM and attainability is outside the control of individual Member States. Europe-wide action on further reducing precursor emissions is needed if the Stage 2 indicative limit values are to be met. EU-wide measures are needed to further reduce emissions of precursors and/or further reduce emissions of primary PM from road traffic and other important sectors.

Additional techniques for primary and precursor emissions mitigation are available to help Member States meet current limit values. There is comparatively little information on the most effective balance of local, regional, national and measures and measures on a European scale.

In some areas, reductions of PM₁₀ levels of up to 50% would be required within 2-3 years in order to comply with the 24-hour limit value in 2005. This is unlikely to be achievable on this timescale because mitigation measures that have an immediate effect on emissions would be required simultaneously at a local, regional and international level.

In all cases, significant additional cost would be incurred to meet the Stage 2 indicative limit values at all relevant locations. However, there would be considerable health benefits to the European population if these indicative limit values were attained. These costs and benefits have been estimated only by a few Member States.

Chapter 11 gives some specific recommendations on the Stage 2 indicative limit values.

9.5.2 Recommendations: what can Member States and the Commission do about attainability?

1. Several Member States will have major problems complying with the Stage 1 24-hour limit value, despite implementing many measures to reduce PM concentrations. Member States should document their attainment difficulties when preparing *plans and programmes* according to the First Daughter Directive.
2. CAFE and the Commission should synthesise and assess information about attainability provided by Member States in their *plans and programmes* – and the information in this Position Paper – to inform a framework for Europe-wide additional actions and measures to help Member States meet the LVs in the First Daughter Directive.³ Member States should also consult with each other to find a solution for significant pollution originating in other Member States²⁵. However, since levels of (secondary) PM are high throughout large parts of (Central) Europe, this consultation should not be bilateral, but should include all Member States, Acceding Countries and the European Commission.
4. During this consultation (preferably within the CAFE framework), the effectiveness of measures at different scales should be investigated; in particular, measures on the Community level on precursor emissions and primary PM from the traffic and other sectors should be assessed.
5. The Commission should consider changes to the First Daughter Directive to ensure that targets are (1) challenging and achievable and (2) that Member States focus abatement strategies on achieving maximum health benefits from reductions in PM concentrations.

One possible option to consider is to extend the date for compliance for the more stringent Stage 1 24-hour limit value, whilst maintaining the current date for the Stage 1 annual average limit value.

In revising the current targets, the Working group recommends to take account of the recommendations formulated in Chapter 11.

²⁵ According to Article 8.6 of the Framework Directive 96/62/EC. The Commission could be represented at such consultations.

10. Conclusions of the World Health Organization

10.1 Scope

Under an agreement between the European Commission and the World Health Organization in support of the CAFE process, WHO provided updated information on health effects of air pollutants.

WHO has established a project “Systematic Review of Health Aspects of Air Quality in Europe” in the course of which the current state of knowledge concerning health effects has also been reviewed for airborne particulate matter. To focus the information as much as possible on their needs, the CAFE Steering Group had asked WHO 12 questions. These questions have all been answered in a recently published report “Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide” prepared by WHO. In addition to the answer as such, a detailed rationale has been given that justifies the answer. In the following, in addition to the introduction of the PM chapter of the WHO report, only the questions and answers related to PM are quoted from the report. The full report is available from the web (<http://www.euro.who.int/document/e79097.pdf>).

Based on this report, the European Commission submitted a set of follow-up questions to WHO in spring 2003, addressing some further points where additional advice from WHO was needed. A WHO Working group meeting agreed on answers to these questions in early 2004. Some of them are of high relevance also for the Working Group on PM and this position paper. These questions and answers are therefore also included in this chapter. The full report will be available on the WHO web page in spring 2004 (<http://www.euro.who.int/air>).

10.2 Findings from the WHO review

Introduction

Airborne particulate matter represents a complex mixture of organic and inorganic substances. Mass and composition in urban environments tend to be divided into two principal groups: coarse particles and fine particles. The barrier between these two fractions of particles usually lies between 1 µm and 2.5 µm. However, the limit between coarse and fine particles is sometimes fixed by convention at 2.5 µm in aerodynamic diameter (PM_{2.5}) for measurement purposes. The smaller particles contain the secondarily formed aerosols (gas-to-particle conversion), combustion particles and recondensed organic and metal vapours. The larger particles usually contain earth crust materials and fugitive dust from roads and industries. The fine fraction contains most of the acidity (hydrogen ion) and mutagenic activity of particulate matter, although in fog some coarse acid droplets are also present. Whereas most of the mass is usually in the fine mode (particles between 100 nm and 2.5 µm), the largest number of particles is found in the very small sizes, less than 100 nm. As anticipated from the relationship of particle volume with mass, these so-called ultrafine particles often contribute only a few % to the mass, at the same time contributing to over 90% of the numbers.

Particulate air pollution is a mixture of solid, liquid or solid and liquid particles suspended in the air. These suspended particles vary in size, composition and origin. It is convenient to classify particles by their aerodynamic properties because: (a) these properties govern the transport and removal of particles from the air; (b) they also govern their deposition within the respiratory system and (c) they are associated with the chemical composition and sources of particles. These properties are conveniently summarized by the aerodynamic diameter, that is the size of a unit-density sphere with the same aerodynamic characteristics. Particles are sampled and described on the basis of their aerodynamic diameter, usually called simply the particle size.

The size of suspended particles in the atmosphere varies over four orders of magnitude, from a few nanometres to tens of micrometres. The largest particles, called the coarse fraction (or mode), are

mechanically produced by the break-up of larger solid particles. These particles can include wind-blown dust from agricultural processes, uncovered soil, unpaved roads or mining operations. Traffic produces road dust and air turbulence that can stir up road dust. Near coasts, evaporation of sea spray can produce large particles. Pollen grains, mould spores, and plant and insect parts are all in this larger size range. The amount of energy required to break these particles into smaller sizes increases as the size decreases, which effectively establishes a lower limit for the production of these coarse particles of approximately 1 μm . Smaller particles, called the fine fraction or mode, are largely formed from gases. The smallest particles, less than 0.1 μm , are formed by nucleation, that is, condensation of low-vapour-pressure substances formed by high-temperature vaporization or by chemical reactions in the atmosphere to form new particles (nuclei). Four major classes of sources with equilibrium pressures low enough to form nuclei mode particles can yield particulate matter: heavy metals (vaporized during combustion), elemental carbon (from short C molecules generated by combustion), organic carbon and sulfates and nitrates. Particles in this nucleation range or mode grow by coagulation, that is, the combination of two or more particles to form a larger particle, or by condensation, that is, condensation of gas or vapour molecules on the surface of existing particles. Coagulation is most efficient for large numbers of particles, and condensation is most efficient for large surface areas. Therefore the efficiency of both coagulation and condensation decreases as particle size increases, which effectively produces an upper limit such that particles do not grow by these processes beyond approximately 1 μm . Thus particles tend to “accumulate” between 0.1 and 1 μm , the so-called accumulation range.

Sub micrometre-sized particles can be produced by the condensation of metals or organic compounds that are vaporized in high-temperature combustion processes. They can also be produced by condensation of gases that have been converted in atmospheric reactions to low-vapour-pressure substances. For example, sulphur dioxide is oxidized in the atmosphere to form sulphuric acid (H_2SO_4), which can be neutralized by NH_3 to form ammonium sulfate. Nitrogen dioxide (NO_2) is oxidized to nitric acid (HNO_3), which in turn can react with ammonia (NH_3) to form ammonium nitrate (NH_4NO_3). The particles produced by the intermediate reactions of gases in the atmosphere are called *secondary particles*. Secondary sulphate and nitrate particles are usually the dominant component of fine particles. Combustion of fossil fuels such as coal, oil and petrol can produce coarse particles from the release of non-combustible materials, i.e. fly ash, fine particles from the condensation of materials vaporized during combustion, and secondary particles through the atmospheric reactions of sulphur oxides and nitrogen oxides initially released as gases.

Recently a comprehensive report on PM phenomology in Europe was compiled. Sulfate and organic matter are the two main contributors to the annual average PM_{10} and $\text{PM}_{2.5}$ mass concentrations, except at kerbside sites where mineral dust (including trace elements) is also a main contributor to PM_{10} . On days when $\text{PM}_{10} > 50 \mu\text{g}/\text{m}^3$, nitrate becomes also a main contributors to PM_{10} and $\text{PM}_{2.5}$. Black carbon contributes 5–10% to $\text{PM}_{2.5}$ and somewhat less to PM_{10} at all sites, including the natural background sites. Its contribution increases to 15–20% at some of the kerbside sites.

Because of its complexity and the importance of particle size in determining exposure and human dose, numerous terms are used to describe particulate matter. Some are derived from and defined by sampling and/or analytic methods, e.g. “suspended particulate matter”, “total suspended particulates”, “black smoke”. Others refer more to the site of deposition in the respiratory tract, e.g. “inhalable particles”, which pass into the upper airways (nose and mouth), and “thoracic particles”, which deposit within the lower respiratory tract, and “respirable particles”, which penetrate to the gas-exchange region of the lungs. Other terms, such as “ PM_{10} ”, have both physiological and sampling connotations.

Question 1: Is there new scientific evidence to justify reconsideration of the current WHO Guidelines for the pollutant?

Answer:

The current WHO Air quality guidelines (AQG) provide exposure-response relationships describing the relation between ambient PM and various health endpoints. No specific guideline value was

proposed as it was felt that a threshold could not be identified below which no adverse effects on health occurred. In recent years, a large body of new scientific evidence has emerged that has strengthened the link between ambient PM exposure and health effects (especially cardiovascular effects), justifying reconsideration of the current WHO PM Air quality guidelines and the underlying exposure-response relationships. The present information shows that fine particles (commonly measured as PM_{2.5}) are strongly associated with mortality and other endpoints such as hospitalization for cardio-pulmonary disease, so that it is recommended that Air quality guidelines for PM_{2.5} be further developed. Revision of the PM₁₀ WHO AQGs and continuation of PM₁₀ measurement is indicated for public health protection. A smaller body of evidence suggests that coarse mass (particles between 2.5 and 10 µm) also has some effects on health, so a separate guideline for coarse mass may be warranted. The value of black smoke as an indicator for traffic-related air pollution should also be re-evaluated.

Question 2: Which effects can be expected of long-term exposure to levels of PM observed currently in Europe (include both clinical and pre-clinical effects, e.g. development of respiratory system)?

Answer:

Long-term exposure to current ambient PM concentrations may lead to a marked reduction in life expectancy. The reduction in life expectancy is primarily due to increased cardio-pulmonary and lung cancer mortality. Increases are likely in lower respiratory symptoms and reduced lung function in children, and chronic obstructive pulmonary disease and reduced lung function in adults.

Question 3: Is there a threshold below which no effects on health of PM are expected to occur in all people?

Answer:

Epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on health. It is likely that within any large human population, there is such a wide range in susceptibility that some subjects are at risk even at the lowest end of the concentration range.

Question 4: Are effects of the pollutant dependent upon the subjects' characteristics such as age, gender, underlying disease, smoking status, atopy, education etc? What are the critical characteristics?

Answer:

In short-term studies, elderly subjects, and subjects with pre-existing heart and lung disease were found to be more susceptible to effects of ambient PM on mortality and morbidity. In panel studies, asthmatics have also been shown to respond to ambient PM with more symptoms, larger lung function changes and with increased medication use than non-asthmatics. In long-term studies, it has been suggested that socially disadvantaged and poorly educated populations respond more strongly in terms of mortality. PM also is related to reduced lung growth in children. No consistent differences have been found between men and women, and between smokers and non-smokers in PM responses in the cohort studies.

Question 5: To what extent is mortality being accelerated by long and short-term exposure to the pollutant (harvesting)?

Answer:

Cohort studies have suggested that life expectancy is decreased by long-term exposure to PM. This is supported by new analyses of time-series studies that have shown death being advanced by periods of at least a few months, for causes of death such as cardiovascular and chronic pulmonary disease.

Question 6: Is the considered pollutant per se responsible for effects on health?

Answer:

Ambient PM per se is considered responsible for the health effects seen in the large multi-city epidemiological studies relating ambient PM to mortality and morbidity such as NMMAPS and APHEA. In the Six Cities and ACS cohort studies, PM but not gaseous pollutants with the exception of sulfur dioxide was associated with mortality. That ambient PM is responsible per se for effects on health is substantiated by controlled human exposure studies, and to some extent by experimental findings in animals.

Question 7: Which of the physical and chemical characteristics of particulate air pollution are responsible for health effects?

Answer:

There is strong evidence to conclude that fine particles ($< 2.5 \mu\text{m}$, $\text{PM}_{2.5}$) are more hazardous than larger ones (coarse particles) in terms of mortality and cardiovascular and respiratory endpoints in panel studies. This does not imply that the coarse fraction of PM_{10} is innocuous. In toxicological and controlled human exposure studies, several physical, biological and chemical characteristics of particles have been found to elicit cardiopulmonary responses. Amongst the characteristics found to be contributing to toxicity in epidemiological and controlled exposure studies are metal content, presence of PAHs, other organic components, endotoxin and both small ($< 2.5 \mu\text{m}$) and extremely small size ($< 100 \text{ nm}$).

Question 8: What is the evidence of synergy / interaction of the pollutant with other air pollutants?

Answer:

Few epidemiological studies have addressed interactions of PM with other pollutants. Toxicological and controlled human exposure studies have shown additive and in some cases, more than additive effects, especially for combinations of PM and ozone, and of PM (especially diesel particles) and allergens. Finally, studies of atmospheric chemistry demonstrate that PM interacts with gases to alter its composition and hence its toxicity.

Question 9: What is the relationship between ambient levels and personal exposure to the pollutant over short-term and long-term (including exposures indoors)? Can the differences influence the results of studies?

Answer:

Whereas personal exposure to PM and its components is influenced by indoor sources (such as smoking) in addition to outdoor sources, there is a clear relationship on population level between ambient PM and personal PM of ambient origin over time, especially for fine combustion particles. On a population level, personal PM of ambient origin “tracks” ambient PM over time, thus measurements of PM in ambient air can serve as a reasonable “proxy” for personal exposure in time-series studies. The relationship between long-term average ambient PM concentrations and long-term average personal PM exposure has been studied less. Contributions to personal PM exposure from smoking and occupation need to be taken into account. However, the available data suggest that imperfect relations between ambient and personal PM do not invalidate the results of the long-term studies.

Question 10: Which are the critical sources of the pollutant (or, for PM, its components) responsible for health effects?

Answer:

Short-term epidemiological studies suggest that a number of source types are associated with health effects, especially motor vehicle emissions, and also coal combustion. These sources produce primary as well as secondary particles, both of which have been associated with adverse health effects. One European cohort study focused on traffic-related air pollution specifically, and suggested the importance of this source of PM. Toxicological studies have shown that particles originating from internal combustion engines, coal burning, residual oil combustion and wood burning have strong inflammatory potential. In comparison, wind-blown dust of crustal origin seems a less critical source.

Question 11: Have positive impacts on public health of reductions of emissions and/or ambient concentrations of the pollutant been shown?

Answer:

Positive impacts of reductions in ambient PM concentrations on public health have been shown in the past, after the introduction of clean air legislation. Such positive impacts have also been reported more recently in a limited number of studies. Toxicological findings also suggest that qualitative changes in PM composition could be of importance for the reduction of PM-induced

Question 12: What averaging period (time pattern) is most relevant from the point of view of protecting human health?

Answer:

As effects have been observed from both short-term and long-term ambient PM exposures, short-term (24 hours) as well as long-term (annual average) guidelines are recommended.

Follow-up question 1: What is the health relevance and importance of short-time exposure to high peak levels or exposure in hot spots?

Answer:

Adverse health effects have been documented after short-term exposure to peaks, as well as long-term exposure to relatively low concentrations of PM, ozone and NO₂. A direct comparison of the *health relevance* of short term and long-term exposures has been reported for PM, but not for ozone and NO₂. For PM, long-term exposure has probably a larger impact on public health than short-term exposure to peak concentrations.

Some studies have documented that subjects living close to busy roads experience more short-term and long-term effects of air pollution than subjects living further away. In urban areas, up to 10% of the population may be living at such 'hot spots'. The public health burden of such exposures is therefore significant. Unequal distribution of health risks over the population also raises concerns of environmental justice and equity.

Follow-up question 2.1: Consideration of publication bias in the review²⁶

Answer:

Publication bias occurs when the publication process is influenced by the size or direction of results. The bias is usually towards statistical significant and larger effects. It can be detected and adjusted for using statistical techniques. Bias may also occur when literature is selectively ascertained and cited.

²⁶ The question was received as follows from the European Commission: *It appears possible that studies that have found no associations between particulate matter concentrations and mortality or morbidity have not been published. How has the expert group tackled the issue of a potential publication bias?*

This review used a systematic approach to identify all short-term exposure studies, but it did not formally investigate publication bias. The reviewers were aware that evidence of publication bias has been identified in meta-analyses of single city time series studies, but when estimates were corrected for this bias, significant positive associations remained. Furthermore, the multi-city time series studies, which have published results from all participating cities and are free from publication bias, have reported significant positive associations.

Because of the size and experience of the review group and referees, it is unlikely that any important published long-term study has been missed. Formal assessment of a possible publication bias has not been undertaken. Every effort was made to systematically ascertain long-term exposure studies.

Follow-up question 2.2: Consistency of epidemiological and toxicological evidence in defining thresholds²⁷

Answer:

Multiple factors determine whether a threshold is seen and the level at which it can occur. Exposure-response curves depend on the age and gender of the subjects, their health status, their level of exercise (ventilation) and, especially the health effect selected. For highly uniform population groups, with a specific exposure pattern, a full range of concentrations, and a specific health outcome, one could identify a specific threshold. However, when there are different exposure-response curves for different groups, thresholds are harder to discern in population studies, and may ultimately disappear. Therefore, the evidence coming from the epidemiological and toxicological studies is not contradictory.

Most epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on mortality and morbidity. It is likely that within any large human population, there is a wide range in susceptibility so that some subjects are at risk even at the low end of current concentrations.

Follow-up question 2.3: Contribution of different sources to PM-related health effects²⁸

Answer:

Only few epidemiological studies have addressed source contributions specifically. These studies have suggested that combustion sources are particularly important.

Toxicology, because of its simpler models and potential to tightly control exposures, provides an opportunity to determine the relative toxic potency of components of the PM mix, in contrast to epidemiology. Such toxicology studies have highlighted the primary, combustion-derived particles having a high toxic potency. These are often rich in transition metals and organics, in addition to their relatively high surface area. By contrast several other components of the PM mix are lower in toxic potency e.g. ammonium salts, chlorides, sulphates, nitrates and wind-blown crustal dust such as silicate clays.

Despite these differences among constituents under laboratory conditions, it is currently not possible to precisely quantify the contributions from different sources and different PM components to health effects from exposure to ambient PM.

²⁷ The question was received as follows from the European Commission: *In some areas there appears to be evidence pointing in different directions thus an indication of the certainty of the conclusions would be desirable. An example would be the issue of threshold for effect due to exposure to ozone where some epi-studies have not been able to identify a threshold whereas thresholds have been found in toxicological studies. The issue of thresholds could be re-assessed for different health endpoints.*

²⁸ The question was received as follows from the European Commission: *The WHO first report put a clear emphasis on the health effects of small PM originating from combustion sources. Can these relationships be quantified giving the source contribution to health effects? How may uncertainties in the source apportionment and the particle characterisation (size and composition) influence the quantitative assessment of pinpointing a source as being the contributor to health effects? Also, is there information and associated uncertainty on the health effects of specific secondary particle mass, such as the particle mass fraction due to agriculture activities leading to ammonia containing particles.*

Follow-up question 2.4: Impact of methods of analysis used in epidemiological studies²⁹

Answer:

This answer addresses matters relating to uncertainties in methods of analysis used. Epidemiological studies use statistical models of various types, including Poisson and logistic regression. The estimates of effect provided by air pollution studies are generally accompanied by confidence intervals. These convey the precision of the estimate or statistical uncertainty that arises because the analyses are subject to a degree of random error. To a varying degree, the results of these analyses are sensitive to the details of the model and the specification of confounding and interacting factors. Extensive sensitivity analyses have shown that associations between air pollution and health remain irrespective of the methods of analyses used.

Follow-up question 2.5: Possible regional differences in effects of air pollution?³⁰

Answer:

Potentially this could be a very influential issue since the characteristics of populations, environments and pollution (including particle concentration, size distribution and composition) vary throughout Europe. However, at this stage there is not sufficient evidence to advocate different guidelines for particles or other priority pollutants in different parts of Europe.

Several studies on short and long-term effects of particulate matter have consistently reported an association between pollution levels and mortality; however, there are differences in the size of the estimated effects of PM according to geographical region or according to the levels of other variables (potential effect modifiers). For example, it has been reported that the short-term effects of PM₁₀ are greater where long term average NO₂ concentration is higher, when the proportion of the elderly is larger and in warmer climates. Modification by socio-economic factors, such as the level of education, has also been reported. Plausible explanations for some of these observations have been proposed.

Effect modification, for example by the age distribution in a population and by climate should, if possible, be taken into account in sensitivity analysis of health impact assessments or risk assessments. Possible effect modifiers of other criteria pollutants have not been investigated to any extent so far.

Follow-up question 2.6: What is the evidence for adverse effects of coarse particles?³¹

Answer:

There is a large number of epidemiological studies showing that PM₁₀ (which includes both fine and coarse particles) has adverse health effects. Although smaller in number, the existing studies on the fine particle fraction (PM_{2.5}) show that there are also health effects from this fraction. Only recently have investigators begun to separately address health effects of coarse particles (PM_{10-2.5}). There is limited evidence that coarse particles are associated independently of PM_{2.5} with mortality in time series studies. A few studies have investigated the effect of long-term exposure to coarse particles on life expectancy without producing evidence of altered survival. There is evidence that coarse particles are independently associated with morbidity endpoints such as respiratory hospitalisations in time series studies. Considerations of particle dosimetry, chemistry and toxicology provide further evidence

²⁹ The question was received as follows from the European Commission: *In the review of the guidelines a systematic assessment of the uncertainties (such as confidence intervals) of the relative risks would give a better understanding of the degree of uncertainty. This item should also include the uncertainty in the application of different models (including GAM).*

³⁰ The question was received as follows from the European Commission: *The assessment of the risks builds on a concentration response relationship based on a number of studies from the US and Europe. However, different parts of Europe have different mixes of air pollution due to differences in sources, climate and so forth. To what extent may uncertainties of the applicability of these relations influence the risk assessment due to particles and other priority pollutants?*

³¹ WHO did not formally receive this question from the CAFE secretariat, but it was discussed at CAFE Workshop on Particulate Matter in autumn 2003 in Stockholm; these discussions clearly indicated its importance for the CAFE process. Therefore, the Scientific Advisory Committee (SAC) recommended to investigate this additional question.

of adverse health effects of coarse PM. Therefore, there is sufficient concern about the health effects of coarse particles to justify their control.

10.3 Recommendation

Even though the evidence on the relationship between exposure to PM and health effects has increased dramatically over the past few years, there are still large uncertainties and considerable gaps in knowledge. These gaps can only be reduced by targeted scientific research. Areas where such research is urgently needed include: exposure to different PM components; dosimetry; toxicity of different components; mechanisms of injury; susceptible groups, etc. The development of methods for measuring particle size and type is still necessary as well as the development of atmospheric models and particle emissions inventories able to support PM policies and of any new standard. The European Community and national institutions are requested to make appropriate funding available to facilitate the corresponding studies.

11. Recommendations concerning the PM metric and PM levels

11.1 Scope

The following two questions are of major importance:

1. What is the appropriate metric for PM?
2. What PM concentration levels should preferably be considered for the CAFE process?

Based on the material presented in the preceding chapters and the discussions within the Working Group, the following answers can be given to these questions.

11.2 PM metric

During the preparation of the first PM Position Paper of 1997, both PM₁₀ and PM_{2.5} were considered as a possible metric for limit values, but it became apparent that there was not sufficient information to justify recommending PM_{2.5}. Since then, WHO – within its activity „Systematic assessment of health aspects of air pollution in Europe“ – has stated that „fine particles (commonly measured as PM_{2.5}) are strongly associated with mortality and other endpoints such as hospitalization for cardiopulmonary disease“ (cf. Question and Answer No. 1 in Chapter 10) and has recommended the development of air quality guidelines for PM_{2.5}. At the same time, it was stated that „continuation of PM₁₀ measurement is indicated for public health protection“ and that the coarse fraction of PM₁₀, i.e. PM_{2.5-10}, cannot be considered innocuous given that there is some evidence that this coarse fraction also has some effects on health.

In the light of these health-related findings the PM Working Group recommends the use of PM_{2.5} rather than PM₁₀ as the principal metric for assessing exposure to particulate matter.

The Working Group also recognises the health relevance of the coarse fraction and recommends therefore to retain a standard on PM₁₀, but not to introduce a separate PM_{coarse} standard. However, the Working Group recommends reclassifying the PM₁₀ standard to become a target value and to relax the current monitoring requirements for PM₁₀. This will ensure that

- the additional burden for Member States in terms of monitoring PM_{2.5} and PM₁₀ will be manageable and
- at the same time the large data sets on PM₁₀ and the trend information will not be lost.

The Working Group considered also in depth the pros and cons of retaining PM₁₀ as the main metric. In terms of measures to achieve the limit value, the new metric will put more emphasis on anthropogenic combustion sources, and less on particles created by mechanical processes and due to natural sources. The Working Group is well aware of this consequence.

The Working Group has taken account of the argument that currently less information is available on PM_{2.5} than on PM₁₀ (for example availability of information on current air quality and future trends across Member States and Acceding Countries) and that this would hamper the ability to base legislation on this metric. However, there is currently more information available on PM_{2.5} than was available for PM₁₀ when the First Daughter Directive was established; in addition, experience shows that only legislation will significantly enhance the efforts to assess a specific air pollution indicator.

It is judged to be too early to consider regulation of PM_{1.0} and ultrafine particles. However, Member States should be asked to carry out more research in the coming years to establish more valid

information on these PM metrics with regard to both concentration levels and adverse health effects. The Commission should collect and review the results of this work within five years.

In conclusion, the majority of the WG recommends that once PM_{2.5} limit values have come into force and have replaced the Stage 1 PM₁₀ limit values, the PM₁₀ indicative limit values currently set for Stage 2 in the First Daughter Directive should be reclassified as target values with the aim to help control the coarse fraction, PM_{2.5-10}. The value for the PM₁₀ target value should be reconsidered in view of the results of the integrated assessment modelling. It would be sufficient to carry out PM₁₀ measurements at some 20% of the PM_{2.5} monitoring sites. Reasonable distribution of the PM₁₀ measurements should be ensured among the various types of sites (urban background, traffic, etc.), particularly where the coarse fraction may comprise a significant part of the mix.

11.3 PM concentration levels to consider for the CAFE process

The information assembled by the Working Group has led to the realisation that limit values may not provide the best means of optimising health protection from ambient PM. This arises from the confirmation from the health-related research reviewed by WHO that there is no evidence of a threshold for health effects. Limit values play an important part in the framework that has delivered improved air quality in Europe. They also have their limitations, for example, reliance on limit values constrains the system to what can be reasonably achieved at the worst hot spot location in Europe. This could be a roadside location in one city³². The substantial natural component of ambient PM, which is variable across Europe, will also influence where this hot spot is located. A limit value set in this way will result in many urban background locations, and in some cases the whole territory of a Member State, experiencing concentrations currently below the limit value. In these areas it will not be necessary to implement measures to reduce exposure to ambient PM, even though such measures would lead to substantial improvements to the health of the population.

Thus, while it is recognised that limit values have an important role to play in helping drive down exposure, the Working Group recommends that the Commission consider the use of alternative approaches, such as gap closure, emissions ceilings or targets, to supplement (but not replace) the use of limit values.

The Working Group has not given detailed consideration to the gap closure or target approach, but some initial ideas and principles are worth setting out as an example. The objective of this approach would be to reduce *total population exposure*, thereby maximising the public health benefits of actions to improve air quality. It would rely on the progressive closure of the gap between the *natural background* concentration and the *current population weighted exposure*. For example, the gap closure approach could be operated on the basis of monitoring and/or modelling of annual mean concentration values, or it could be used to develop emission reduction targets, so as to parallel the National Emissions Ceiling (NEC) Directive approach. It would be consistent with the approach to cost/benefit analyses being carried out as part of the integrated assessment procedure for ambient PM.

The population-weighted average PM concentration for a Member State would be strongly influenced by concentrations in urban areas, especially the larger urban areas. The background average concentration for an urban area could be determined either by modelling (validated against measurement) or by measurement. The requirement would be to consider all 'agglomerations' in the calculation of the population-weighted average. The contribution of exposure outside of urban areas could either be left out of the equation, or based on measured rural background concentrations at an

³² Although this may seem extreme, it is an inevitable consequence of a system of legally binding limit values – if they are set at a level below what can be achieved, then the Member State with this location will be in breach of the limit value, although it can do nothing reasonable to avoid this position. This is already leading to problems with the current PM limit values, where it appears that even with maximum feasible reductions the limit values are likely to be exceeded in some parts of Europe.

agreed number of sites or on modelling of the rural background. The 'natural' background could be determined by monitoring, coupled with source apportionment.

Targets could also be used to supplement limit values. One option could involve the application of target values, as used for ozone, to reduce exposure to PM at urban background locations. This would help drive down concentrations in the areas where the greatest population average exposure occurs. As there would not be legal penalties for not meeting the target value, it could be set at a stringent level. Member States would be required to prepare plans and/or programmes to attain the target value.

11.4 PM_{2.5} levels for input to the integrated assessment procedure

If the limit value approach is to be followed, the Group found it reasonable to start considerations for PM_{2.5} levels from the advice developed by WHO.

The results from studies of long-term exposures to PM_{2.5} suggest that an exposure-response relationship down to the lowest observed levels seems to be appropriate. Graphs presented in the recently published further follow-up of the ACS cohort³³ suggest that for cardiopulmonary mortality, and especially for lung cancer mortality, the risk was elevated even at (long-term) PM_{2.5} levels below 10 µg/m³. Also the size of the effects is of considerable concern. Therefore, the majority of the Working Group concludes that a reduction of exposure to fine PM up to the lower end of the concentration range investigated in the ACS study would be desirable from the health point of view. However, the Working Group also noted the necessity to take other considerations into account like feasibility when establishing a legally binding limit value. Taking further into account the currently observed levels, the anticipated decreasing trends of fine PM, in particular the secondary inorganic PM, and that the attainment date for a new PM_{2.5} limit value would not be before 2010, the Working Group concluded that such a limit value should preferably not exceed 20 µg/m³. The limit value would apply where people are likely to be exposed for a period that is significant in relation to the averaging period of the limit value. Given that in setting a final level, the attainability of any such level would have to be taken into account, the Working Group concludes that no single PM level should be recommended at this time. Rather, values within the range 12 to 20 µg/m³ should be used as an input to the integrated assessment procedure. Consideration will also need to be given to the appropriate attainment date.

According to WHO both a short-term (24-hour) and a long-term value (annual average) should be established for PM_{2.5}. From the available information about the frequency distribution of 24-hour values – which is scarce – the majority of the WG recommends that for the 24-hour average limit value a range (20 to 35 µg/m³, not to be exceeded more than 10% of the days of the year) corresponding to the range given for the annual limits seems reasonable as a starting point.

The value for the PM₁₀ target value should be reconsidered in view of the results of the integrated assessment modelling.

11.5 Rationale for the PM metric

There are a number of aspects discussed by the Working Group that have an impact on the decision concerning the PM metric to be selected. Besides health considerations, these aspects include uncertainties related to the measurement, the information available on current concentration levels of the PM metric under question, the possibilities for abatement and the cost for monitoring that result from the selection of a metric. Each of these aspects has been discussed in the Working Group in the light of the current knowledge of the different PM metrics.

³³ Pope, C. A. et al. (2002). Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. Journal of the American medical association, 287: 1132 - 1141.

Some health studies suggest a higher risk for individuals living closer to traffic sources. However, there have been concerns about the appropriateness of $PM_{2.5}$ (similar to PM_{10}) as indicator for traffic related PM pollution. Therefore, WHO recommended to re-evaluate the usefulness of black smoke or any other metric specific for combustion-related PM from traffic sources. Even though black smoke is a constituent of $PM_{2.5}$, it does not necessarily correlate well with $PM_{2.5}$. In particular, there are considerable differences in the spatial variability. However, the Working Group expects black smoke levels to substantially reduce in the forthcoming decade as a result of current and expected emission reductions, particularly from road vehicles.

Chapter 8 of this Position Paper identifies particulate traps as a particularly effective technology for reducing particulate emissions from diesel vehicles but notes that they are not necessarily required to meet current Euro III and Euro IV emissions standards. Controlling PM emissions from diesel vehicles will be in particular effective to control black smoke emissions. The Working Group therefore recommends that the European Commission and MS consider setting more stringent emissions limits that ensure particulate traps, or equivalent technology, are fitted to all new diesel road vehicles.

Consequently, whilst recognizing the important health implications of black smoke, the group considers that setting a separate air quality standard is not appropriate at this time, but recommends that the anticipated effects of the described measures should be closely followed by monitoring of an appropriate PM metric such as elemental carbon or Black Smoke. Should however emissions from diesel vehicles not be controlled adequately over the next decade, the Working Group recommends that an air quality objective for black smoke should be considered.

Furthermore the Working Group recommends that the integrated assessment modelling explore Euro V and Euro VI scenarios requiring emissions limits for PM for all new diesel road vehicles that are equivalent to emissions from the same vehicles fitted with particle traps, or using an alternative technology or measure that would achieve equivalent PM emissions reductions.

With regard to the uncertainty of PM measurements, it is clear that in the gravimetric determination, the uncertainty of a result increases as the mass deposited on the collection filter becomes smaller. Thus, the uncertainty of measurement increases from PM_{10} to $PM_{1.0}$. This is less the case for the new generation of automatic monitors such as the TEOM and beta-absorption monitors. However, for these monitors the correction factor used to establish equivalency to the reference method is likely to become larger (see Chapter 3). For ultrafine particles it was felt that this PM metric has not yet reached the status of a measurement method suitable for large-scale routine use in networks, though a number of monitors are run in Europe.

As concerns information on current PM concentrations, the requirements of the First Daughter Directive have caused PM_{10} to be the main parameter of interest. However, the availability of information on $PM_{2.5}$ concentrations has also grown over the last years. Much less is known about the concentrations the other PM metrics.

Changing the PM metric has also an impact on the kind of abatement measure to be applied. It has to be considered if and to what extent, measures at the local or regional level can decrease ambient concentrations of the chosen metric. Although the results of a recent International Workshop³⁴ have not yet been evaluated thoroughly, it should be noted that changing the PM metric from PM_{10} to $PM_{2.5}$ may change the potential of local or small-scale abatement measure, as local concentration increases of PM near traffic or industrial sources depend on the metric. The importance of this is difficult to evaluate as long as it is unclear whether health effects are locally increased or not.

³⁴ The International Workshop of the German Federal Environment Ministry on Air Pollution Abatement Planning in Europe (Berlin, 1 - 3 April 2003), organised by the German Federal Environmental Agency with the sponsorship of the European Commission.

The WG has considered the coarse fraction ($PM_{10-2.5}$) as a possible alternative for the PM_{10} metric. Basing a target on the coarse fraction has the conceptual advantage that it does not overlap with $PM_{2.5}$ and can consequently be regarded as a separate pollutant when considering emissions reduction measures. This would be consistent with USEPA proposals for PM. Such a target may have some advantages for mitigation strategies as this allows policymakers to focus on reducing $PM_{2.5}$ concentrations, the fraction with the most important health impacts.

However, there is less evidence on health effects of the coarse fraction than for that of PM_{10} and fine particles.

The Working Group also considers that there are two significant technical disadvantages to replacing a PM_{10} target with a $PM_{10-2.5}$ target.

Firstly, there are currently no standard instruments in use in Europe that directly measure the coarse fraction, so $PM_{10-2.5}$ would need to be determined by calculation from separate PM_{10} and $PM_{2.5}$ measurements. Consequently, co-located instruments will be necessary. Calculation of the coarse fraction based on two sets of measurements will increase the significant uncertainties already associated with PM measurement.

Secondly, a number of transition problems can be expected when changing over monitoring regimes from PM_{10} to $PM_{10-2.5}$. For example, trend data for the coarse fraction would be scarce so there would be no trends on which to base policy decisions and mitigation strategies. Furthermore, a whole new set of empirical data on the correction factor of equivalent instruments would have to be established.

Given the technical concerns, the Working Group has a preference for proposing PM_{10} as the second metric.

If it comes to estimating the consequences for monitoring networks that are associated with the use of one or the other PM metric, it is evident that the least financial needs arise from staying with PM_{10} because the Member States' networks are already equipped with the corresponding instruments. Given that under the requirements of Article 5 of the First Daughter Directive Member States have to provide information on $PM_{2.5}$ levels, a number of $PM_{2.5}$ monitors is also already available. A CEN standard is available for PM_{10} measurements, a draft exists for $PM_{2.5}$ measurements. It is emphasised in this context that a PM_{10} monitor can easily be converted into a monitor for smaller particles down to e.g. $PM_{1.0}$ by simply changing the air inlet. However, the effort for calibration and maintenance of the instruments will rise, as the mass will become smaller and the share of semivolatile secondary aerosol higher.

The Working Group also discussed a possible new short-term limit value. If there were a clear correlation between the annual average and the 24-hour average, then the latter would not be needed in addition. There is the need of ongoing information of the public, though for short-term public information purposes alone, a smaller number of allowed exceedance days, such as 7 corresponding to the 98-percentile, may be more suitable than the 35 days (90-percentile) of Stage 1. WHO recommends establishing not only a long-term (annual average) but also a short-term PM value (24-hour). Setting a 24-hour value has the advantage that sufficient attention is paid to avoid daily episodes of elevated PM levels. After thorough debate, the majority of the Working Group supported setting a 24-hour limit value for $PM_{2.5}$ based on the 90-percentile. Based on a preliminary evaluation of the frequency distribution of 2001 $PM_{2.5}$ concentrations at a (though limited) number of sites in Germany, Portugal, Sweden and Spain in 2001, an annual average of $20 \mu\text{g}/\text{m}^3$ seems to correspond to a 90-percentile of the 24-hour averages of $35 \mu\text{g}/\text{m}^3$ (see Chapter 4). Hence, this concentration level could be envisaged as a proposal of a 24-hour average, not to be exceeded more than 10% of the days of the year.

The Working Group felt that measuring PM_{10} should not be stopped. Rather, the PM_{10} limit values currently set for Stage 2 of the First Daughter Directive should be maintained as target values.

Monitoring PM₁₀ and PM_{2.5} simultaneously would be consistent with WHO advice that the coarse fraction, i.e. PM_{2.5-10}, the size fraction between PM₁₀ and PM_{2.5} is not innocuous. However, to avoid excessive monitoring costs, the Working Group deemed sufficient that about 20% of the PM_{2.5} monitoring sites be used for parallel measurement of PM_{2.5} and PM₁₀. A reasonable distribution of the respective sites should be ensured, so that the different types of sites (urban background, traffic, industry and rural) are sufficiently well represented. Particular consideration should be given to sites where the coarse fraction may play a significant role.

Monitoring both PM_{2.5} and PM₁₀ would achieve two goals: (1) the results could be used to follow year-long trends of PM₁₀ concentrations and to avoid neglecting sources contributing essentially to PM₁₀, and (2) the results could be used to follow the development of the concentration of the coarse fraction.

11.6 Rationale for the PM concentration levels

The starting point for deriving an appropriate range for a limit value for PM_{2.5} is the recognition that exposure to currently common levels of fine particulate matter is linked to considerable health impacts in EU Member States. The Working Group agreed that a significant reduction of the overall exposure of the population to fine particulate was warranted. It also felt that the choice of the actual limit value should be based – if possible - on quantitative information on health effects. The Working Group felt it reasonable to use integrated assessment modelling in this process.

Based on the advice from WHO, PM_{2.5} was selected as indicator to assess PM related health effects within RAINS, the integrated assessment model to be applied within the framework of CAFE and the UN/ECE Convention on Long Range Transboundary Air Pollution (LRTAP). This proposal was supported by the Task Force on Health and endorsed by the Working Group on Effects under the Convention on LRTAP.

The inclusion of any health endpoint into RAINS requires the selection of an appropriate concentration-response relationship. Such relationships are usually derived from epidemiological studies. Associations between air pollution exposure and mortality have been assessed mainly through two types of epidemiological studies:

- Time-series studies, which investigate the association between daily mortality and variations in recent PM concentrations,
- Cohort studies, which follow large populations for years and relate mortality to exposure to air pollution over extended periods.

Both designs provide estimates of relative risk of mortality that can be associated with exposure to air pollution. It is important to point out that the relative risks derived from time series and cohort studies have different meanings, but refer to similar effects of air pollution. The WHO working group on health impact assessment concluded that both designs could contribute useful, albeit different, information³⁵. Through their design, time series studies yield estimates of deaths due to recent exposure, in all likelihood among those who are frail due to either chronic disease, or to some transient condition. Because such studies cannot quantify chronic effects of long-term exposure, some deaths attributable to air pollution will be missed and the extent to which air pollution advances the time of death cannot be quantified. For this reason, the use of risk estimates from time series studies of daily mortality will in most cases underestimate the impact of pollution exposure on both the attributable numbers of deaths and average lifespan in a given population.

Therefore, the WHO working group on health impact assessment (WHO, 2001) concluded that the most complete estimates of both attributable numbers of death and average reduction in lifespan associated with the exposure to air pollution are those based on cohort studies. Such studies include

³⁵ WHO 2001, Health impact assessment of air pollution in the WHO European Region. Technical Report from WHO/ECEH Project. WHO European Centre for Environment and Health, Bonn, Nov 2001.

not only those whose deaths were advanced by recent exposure to air pollution, but also those who died from chronic disease cause by long-term exposure.

The extended American Cancer Society (ACS) Study (Pope et al., 2002³⁶) is by far the largest cohort study of air pollution and long-term average mortality. This study found for all causes, cardiopulmonary and lung cancer deaths statistically significant increases of relative risks for PM_{2.5}. TSP and coarse particles (PM₁₀ – PM_{2.5}) were not significantly associated with mortality. It was decided to apply the relative risk for PM_{2.5} for all causes of mortality estimated for the average exposure level in the extended ACS cohort study within RAINS. The Task Force on Health endorsed this decision³⁷. It was felt that this risk coefficient was a more appropriate choice than the estimates specific to the PM levels in the initial or final period of the follow-up in the ACS study. The results from this study of long-term exposures to PM_{2.5} suggest that an exposure-response relationship down to the lowest observed levels seems to be appropriate.

The Working Group concludes that a reduction of exposure to fine PM up to the lower end of the concentration range investigated in the ACS study would be desirable from the health point of view. As stated previously, any new regulation should result in a further reduction of total population exposure. However, the Working Group also noted the necessity to take other considerations into account like current levels, natural background concentrations, attainability and cost-benefits when establishing legally binding limit values. A first evaluation of European monitoring data for PM_{2.5} has shown that in 2001 the majority of urban background stations (28 out of 38) have met this level. Clearly, it would be more difficult to meet the level at traffic sites: 15 out of 23 traffic sites did exceed 20 µg/m³. Taking further into account the anticipated continuation of decreasing trends of fine PM, in particular the secondary inorganic PM and that the attainment date for a new PM_{2.5} limit value would be 2010 or later, the Working Group concluded that such a limit value should preferably not exceed 20 µg/m³. The limit value would apply where people are likely to be exposed for a period that is significant in relation to the averaging period of the limit values. Given that in setting a final level, the attainability of any such level would have to be taken into account, the Working Group concludes that no single PM level should be recommended at this time. Rather, values within the range 12 to 20 µg/m³ should be used as an input to the integrated assessment procedure.

³⁶ Pope, C. A. et al. (2002). Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *Journal of the American medical association*, 287: 1132 - 1141.

³⁷ UNECE EB.AIR/WG.1/2003/11

Annex 1 Members of the CAFE Working Group on Particulate Matter

The CAFE Working Group on Particulate Matter had the following Members:

Luc Bertrand	Belgium
Souad Bouallala	France
Peter Bruckmann	Germany
Christian Lange Fogh	Denmark
Hans-Christen Hansson	Sweden
Dick van den Hout	Consultant to European Commission
Stefan Jacobi	for European Commission
Duncan King	for UNICE
Klaas Krijgsheld	Netherlands
Steinar Larssen	for ETC/AAC
Duncan Laxen	for EEB
Martin Meadows (Co-chair)	United Kingdom
Jürgen Schneider	Austria and WHO
Bernd Seifert (Co-chair)	Germany
Peter Straehl	Switzerland
Xavier Querol	Spain
Gabriele Zanini	Italy

Annex 2 Annex to Chapter 3 – PM₁₀ Reference Method

The CEN standard EN 12341 covers basically the following aspects:

- a) Assessment of appropriate and practical European PM₁₀ Reference Method.
- b) Assessment of a workable European PM₁₀ test procedure enabling the Member States to evaluate their own candidate PM₁₀ sampling systems.

For the PM₁₀ field tests conducted by the CEN/TC 264 Working Group 6 at four different European sites in 1994/95 the Wide Range Aerosol Classifier (WRAC)³⁸ was used as primary PM₁₀ Reference Sampler, as from all known samplers the WRAC has the highest capability to sample the (conventional) true particle size distribution in the range from 10 µm up to 100 µm in aerodynamic diameter. However, the practicability of the WRAC presents the major drawback for the use of this system.

The results of the field tests showed that beside the WRAC also the PM₁₀ high volume sampler having a flow rate of 68 m³/h and the PM₁₀ low volume sampler having a flow rate of 2.3 m³/h possess the required performances to sample PM₁₀. The two latter samplers are suitable to be used as PM₁₀ Reference Samplers and PM₁₀ monitoring instruments as well because of their practicability. Thus, the CEN standard EN 12341 produced by the CEN/TC 264 Working Group 6 defines the PM₁₀ Reference Method which are based on the gravimetric evaluation of filters by a balance and the use of three kind of Reference Samplers:

- a) WRAC for the super high volume flow regime with a flow rate of 2340 m³/h.
- b) HVS-PM₁₀ sampler for the high volume flow regime with a flow rate of 68 m³/h.
- c) LVS-PM₁₀ sampler for the low volume flow regime with a flow rate of 2.3 m³/h.

The requirements for proving equivalence of PM₁₀ candidate samplers to one of the Reference Samplers focus mainly on four items:

- a) The test site(s) shall cover PM₁₀ concentrations and particle size distributions being representative for European sites. If already one or two sites fulfil these requirements, it is not necessary to conduct the measurements at further sites.
- b) Limitation of the uncertainty range for candidate methods of $\pm 5 \mu\text{g}/\text{m}^3$ ($<100 \mu\text{g}/\text{m}^3$) respectively $\pm 5\%$ ($>100 \mu\text{g}/\text{m}^3$).
- c) Limitation of the deviation between candidate method and Reference Method of $\pm 10 \mu\text{g}/\text{m}^3$ ($<100 \mu\text{g}/\text{m}^3$) respectively $\pm 10\%$ ($>100 \mu\text{g}/\text{m}^3$).
- d) Correlation between candidate method and Reference Method of $R^2 > 0.95$.

Full details of test and acceptance criteria as well as the statistical data treatment are given in the standard.

The requirements concerning the test location(s) are specified such that only the separation efficiency of PM₁₀ inlets can be judged rather than the sampling efficiency of complete automated instruments. Thus, the EN 12341 explicitly excludes equivalence tests of automated instruments. The reason for that are the experiences made with several PM₁₀ samplers in the framework of the preceding WRAC studies. These studies revealed that the separation efficiency of various PM₁₀ samplers differed from each other by up to 30%, though all these samplers were approved by the US EPA PM₁₀ wind tunnel test procedure. Therefore first of all, establishing an appropriate and reliable European PM₁₀ Reference Samplers was needed. To evaluate additionally e.g. the influence of heated sampling tubes and filter

³⁸ L. Laskus et al. (1989). Measurement of particle size distribution and dust concentration. Comparative measurements with the Wide Range aerosol classifier (WRAC) and other samplers. Staub Reinhaltung der Luft 49, p. 395-400.

units on the sampling efficiency of complete automated instruments would have exceeded the limits of the standard's scope at that time.

The EN 12341 requires, furthermore, to express the measured PM_{10} concentrations with respect to STP conditions (273 k and 101,3 kPa). This requirement is only needed for the purpose of a reliable comparison between candidate methods and the WRAC, as the WRAC is mass-flow controlled in terms of standard m^3 related to STP conditions. This requirement does not touch regular PM_{10} compliance measurements which are to be related to ambient air conditions.

Annex 3 Annex to Chapter 3 – PM_{2.5} test procedure

Unlike the PM₁₀ test procedure, there is no primary PM₁₀ Reference Method like the WRAC for the intercomparison of PM_{2.5} candidate methods available. However, according to the decision of the CEN Working Group the PM_{2.5} Reference Method shall meet the following requirements:

- a) The separation process of the PM_{2.5} Reference Inlet shall base on established first physical principles.
- b) The measurement uncertainty of the Reference Method calculated according to the Guide to the expression of uncertainty in measurement (GUM)³⁹ shall be very low.
- c) The Reference Method shall base on the gravimetric evaluation of filters using a micro-balance.
- d) The Method shall be also suit the practice of PM_{2.5} monitoring in networks preferably with long maintenance intervals and without the expenditure of a daily manual filter exchange.
- e) The Method shall be constructed such that losses of particles by kinetic, thermal and chemical processes are minimised.

Furthermore, also experiences and results derived from other studies shall be considered for establishing an appropriate PM_{2.5} Reference Method.

As the construction of reliable PM_{2.5} (as well as PM₁₀) inlets is no longer a problem nowadays, the equivalence test procedure, the main characteristics of which are already drafted, may be also applied to automated instruments. With respect to that and according to the above-mentioned requirements for a PM_{2.5} Reference Method the Working Group lays emphasis, inter alia on the following test site criteria: High and low relative humidity, high and low ambient temperatures, strong solar radiation in summer as well as high and low amounts of ambient volatile particulate matter (nitrates). To check the feasibility of the test procedure regarding the test of automated instruments, such methods take also part in the field tests beside manual methods. In detail the following samplers and instruments are involved in the field tests:

- 2 Mini WRAC's (special construction of the Fraunhofer Institute for Toxicology and Aerosol Research, Hannover) (0.172 mol/sec ~ ca. 15 m³/h)
- 2 US PM_{2.5} Federal Reference Samplers (WINS) (1.0 m³/h)
- 2 LVS-PM_{2.5} single channel samplers (2.3 m³/h)
- 2 Partisol Plus sequential samplers (1.0 m³/h)
- 2 SEQ 47/50 sequential samplers (2.3m³/h)
- 2 HDI sequential samplers (30 m³/h)
- 1 beta-absorption gauge "ADAM" (1.0 m³/h)
- 1 TEOM SES (filter temperature 30 - 35 °C) (1.0 m³/h)
- 1 beta-absorption gauge FH 62 I - R (1.0 m³/h)
- 1 beta-absorption gauge BAM (1.0 m³/h)

At the test sites, all relevant meteorological parameters as well as the nitrate concentration are measured. For the latter purpose the nitrate analyser described in Section 3.1.5 is used.

In a Decision of the Commission concerning guidance on a preliminary PM_{2.5} Reference Method dated from 16th January 2003⁴⁰ it is outlined that the use of (PM_{2.5}) devices, the sample and/or the filter of which can be heated up by solar radiation during sampling, is to be avoided. To prevent losses of volatile particles as far as possible instruments shall be employed having an inlet system that samples

³⁹ International Organisation for Standardization (1995). Guide to the expression of Uncertainty in Measurement, ISBN 92-67-10188-9, ISO Geneva.

⁴⁰ Commission Decision of 16 January 2003 concerning guidance on a provisional reference method for the sampling and measurement of PM_{2.5} under Directive 1999/30/EC. Official Journal of the European Communities L 12, 17.01.2003, p.31.

as close as possible to the ambient temperature. This recommendation meets the requirements of the Working Group regarding an appropriate PM_{2.5} Reference Sampler.

Annex 4 Annex to Chapter 3 – Modelling of ambient particle concentrations⁴¹

In January 2002 an EMEP-workshop “Dynamic aerosol modelling: from box models to 3D transport models” was held to discuss the possibilities and difficulties with such models and to form recommendations for future use. The general conclusions and recommendations from the workshop are given below. During the workshop the authors of this chapter met together with about 30 scientists representing most groups active on modelling aerosols in Europe (see homepage of EMEP, www.emep.int). To give an orientation on the status of the present modelling work, different specialist gave short presentations on dynamical aerosol modelling describing the fundamentals, implementation and use on urban, regional and global scale of which some is presented in this appendix.

A4.1 Possibilities, needs and suggestions for the future particle modelling work in European policy applications – recommendations from the EMEP-workshop “Dynamic aerosol modelling: from box models to 3D transport models”

The main conclusion of the EMEP Workshop was despite the recognized uncertainties in aerosol modelling; it is possible to get useful data from current models. However considerable and intensified attention is required in order to formulate and test better descriptions giving more reliable results. In this work the most important is validation against measured data, which should be pursued regularly.

The parameters identified as useful from different aspects, but then necessary in testing the models, are shown in Table A4.1.

Table A4.1 Identification of important deliverables (i.e. output variables) from aerosol models

Area of interest	Output variables	Comments
Acidification / Eutrophication	Particulate mass of SO ₄ , NO ₃ and NH ₄	
Health	PM ₁₀ and PM _{1.0} N(Aitken), N(accumulation)	Mass closure needed to accomplish source identification
Climate	Optical properties as scattering and absorption	

Note 1: Validation studies for the above mentioned variables are necessary – Support to level III measurements in EMEP monitoring strategy

Note 2: The workshop recommended the use of PM_{1.0} instead of PM_{2.5} in order to reduce uncertainty as contributions from natural and anthropogenic events of dust are poorly described.

A higher temporal and spatial resolution of measurements as well as the particle chemical composition is needed in order to validate model results. Practical and economical reasons are recognized and supposed to limit the measurement efforts. But some measurements can easily be performed with high time resolution as number and mass and are thus strongly recommended.

In identifying the uncertainties in current aerosol models three main sources were found in current aerosol models. The emission inventories and emission model parameterisations should include sea spray, soil dust and other natural particle emissions, as well as provide characterization of aerosol

⁴¹ The following authors, other than members of the Working Group on PM, have contributed to this annex: R. Berkowicz, DMU, Roskilde, Denmark; M. Kulmala, University of Helsinki, Finland; J. Langner, SMHI, Norrköping, Sweden; B. Schell, Ford Forschungszentrum Aachen, Germany; D. Simpson, S. Tsyro and L. Tarrasón, The Norwegian Meteorological Institute, Oslo, Norway; J. Wilson and E. Vignati, JRC, Ispra, Italy.

composition and size. The description of condensation processes can introduce an important source of error in the models. The partitioning between gas and particle for sulphate, nitrate and ammonium can be model by using equilibrium for daily averages but will have difficulties below that temporal resolution.

Thermodynamics are still not well known and are important for condensation, cloud processes and nucleation. This effects especially the treatment of organics, clouds and nucleation. The composition and characteristics of organic aerosols are largely unknown. This lack of knowledge severely limits the possibilities for applying and evaluating Secondary Organic Aerosols models for policy applications. These areas need considerable and intensified attention to find better descriptions giving more reliable results. Still it is considered possible to have the current models to give useful data. But more research on the current SOA formation descriptions is absolute necessary including continuous test and evaluation by comparing with relevant measurements.

The most common aerosol model approaches are at the moment: sectional, modal and moment based. No clear difference in cpu were agreed on concerning the general approach, while differences exist due to different processes included and the level of detailed with which they are described. The modal and moment based models needs non-physical corrections. Several efficient modal models are available now.

A4.2 Aerosol processes important to Aerosol Dynamics

In aerosol dynamics we study processes like Nucleation, Condensational Growth and Evaporation, Coagulation, Cloud Processes, Deposition, Primary emissions, Re-emissions, Connections with atmospheric chemistry and meteorology.

During the processes of formation and growth of atmospheric aerosols the aerosol dynamics, atmospheric chemistry and meteorology form a coupled system. The importance of atmospheric chemistry (e.g. Pirjola and Kulmala, 1998; Pirjola 1999) as well as meteorological conditions (Nilsson and Kulmala, 1998; Nilsson *et al.*, 2000; Väkevä *et al.*, 2000) on particle formation and growth have been demonstrated under tropospheric conditions. Although ternary nucleation of water-ammonia-sulphuric acid vapours (Korhonen *et al.*, 1999) has shown to be able to explain atmospheric nucleation – i.e. formation of ~1 nm particles – in many cases (Kulmala *et al.*, 2000), the exact routes for formation of 3 nm particles are still unclear, because besides nucleation, also the growth from 1 nm size to 3 nm size is needed.

In order to be able to understand the formation and growth processes of atmospheric aerosols and cloud droplets their thermodynamic properties should be known. For example, in the condensation process, the driving force is the vapour pressure difference between gas phase and surface. However, in the atmosphere where there are multi-component, multiphase mixtures, their thermodynamic state and phase diagrams are typically very complex. It is very important to obtain thermodynamically consistent vapour pressures, chemical activities, surface tensions and densities for organic and inorganic compounds and their water solutions (for the importance see e.g. Korhonen *et al.*, 1999) as a function of temperature and composition.

In future, development of nucleation theories, modelling and nucleation rate parameterisations are needed. So far, conclusions on whether or not certain substances cause nucleation in the atmosphere conditions are usually based on predictions given by the classical nucleation theory (CNT). CNT treats the nucleating molecular clusters as macroscopic droplets, which is a questionable approach since the nucleating clusters often contain less than fifty molecules. Nucleation of various vapours using molecular dynamics (MD) and Monte Carlo (MC) simulation techniques is needed to investigate. So far, some investigations were carried out using *ab initio* calculations on small sulphuric acid-water clusters (Arstila et al 1998), classical MD (Laasonen et al, 2000) and MC (Vehkamäki and Ford, 1999)

simulations of argon nucleation, as well as DFT calculations of nucleation in binary systems imitating water and different organic molecules (Laaksonen *et al.*, 1995, Napari and Laaksonen 2000). Also, a new nucleation mechanism based on stable dimers (Lushnikov and Kulmala, 1998) has been proposed.

In contrast to laboratory conditions, the formation of aerosol in the atmosphere can be kinetically limited by some of the intermediate steps of its formation processes. The equilibrium state is thus not necessarily the aerosol itself but can be, for example, thermodynamically stable clusters (TSC), as we have recently shown (Kulmala *et al.*, 2000). Although there is strong indication that the water-sulphuric acid-ammonia nucleation mechanism (Korhonen *et al.*, 1999, Janson *et al.*, 2001) explains the formation of new atmospheric aerosols (diameter < 3 nm) in many circumstances, the condensation of these vapours does not explain the observed growth rates of the particles (Kulmala *et al.*, 2000), and in atmospheric conditions nucleation and growth are decoupled (Kulmala *et al.*, 2000). The other possible relevant nucleation mechanism is ion-induced nucleation.

Condensation growth increases aerosol mass and changes composition. It is important particularly for nucleation and Aitken modes. The observed growth rate for these modes is 1-20 nm/h (Kulmala *et al.*, 2001). In practise, sulphuric acid cannot explain this alone and other condensable vapours are needed. Different non-volatile or less volatile organics are very good candidates for that extra vapour. When investigating multi-component condensation of sulphuric acid, ammonia, organics and water very complex thermodynamics is involved. Therefore parameterisations are needed also for thermodynamic point of view. The mass accommodation coefficient affects condensation rates, particularly in kinetic and transition regime. The driving force in condensation is the vapour concentration, which is related to vapour sources and to atmospheric chemistry. An open key question is how charged particles with polar condensing molecules will enhance condensation growth.

The chemistry evolving from the ageing by nucleation, coagulation and condensation determines if the particles will activate as cloud droplets. The increased mass transfer from gas to cloud droplets will increase aerosol mass and change composition and will end up with changes in size distribution of the evaporated cloud droplets. The influence of the organic component has been forwards as important in determining what particles activating and further the droplet distribution (Kulmala *et al.*, 1997, Facchini *et al.*, 1999, Li *et al.*). Still it is unclear to what extent organic influence on the atmospheric mix of inorganics and organics influence the activation and other physical properties of the cloud and still needs to be investigated. Precipitation is a major deposition path way determining the residence time of the particles in the atmosphere and thus an accurate description on which particles that actually form cloud droplets is crucial for determining the fraction that is eligible for long rang transport. Subsequently the secondary changes in the atmosphere of the particle chemistry change the chemistry of the particles to more hygroscopic and thus more eligible as cloud condensation nucleus.

Dry and wet deposition investigations including continuous measurements of aerosol fluxes (Buzorius *et al.*, 1998, 2000) are very important from aerosol life cycle point of view. The number of measurements is increasing and new information on the deposition velocity as a function of boundary layer conditions is obtained.

Direct emissions and re-emissions of aerosol particles as well as direct emissions of precursor gases are an absolute requirement to obtain needed initial and boundary conditions for aerosol dynamic models.

A4.3 Urban modelling, connecting street canyon to regional scale

Specifics of the urban pollution modelling are mainly related to the geographical and time scale of the relevant physical and chemical processes considered. For the regional scale models we need to take into consideration transport as well as physical and chemical transformation processes that have a scale of hundreds to thousands kilometres. The scale of an urban domain is normally only few to tens

of kilometres, and when dealing with single streets, the relevant scale is only few metres. The urban environment is characterised by a very short distance to main pollution sources. Actually, when dealing with street level pollution, the modelling domain is practically identical with the source domain. From the modelling point of view this implies that such processes, as dilution by turbulent dispersion are much more important than chemical or physical transformations and deposition, which are crucial for the regional scale modelling. In this respect, the particle pollution modelling is not especially different from modelling of traditional gaseous pollutants on the local scale.

The importance of the meteorological conditions for pollution dispersion within the urban scale implies that modelling techniques based on e.g. box models, are hardly applicable in the current context. Urban scale models must reproduce the vertical dispersion of pollutants on a horizontal scale of one kilometre or less. On this scale, the dispersion and mixing of pollutants is mainly governed by turbulence created by wind. The highest pollution concentrations are always observed in urban regions characterised by low wind speed conditions.

When dealing with street level pollution, the mixing and thus dilution of pollutants can also be inhibited by the presence of buildings along the street, and large concentration gradients can be observed just between two sides of the same street. This is due to the specific wind recirculation conditions that are typical for street canyons.

The special importance of the micro-meteorological conditions, compared with the chemical or physical transformation processes, makes the approach of “down-scaling” of large-scale models to an urban environment scale, quite inefficient. However, coupling between large-scale and local-scale models is inevitable.

The major difference in modelling the particle pollution, compared with the traditional gaseous pollutants, is in the relative importance of the different source contributions. At a highly trafficked street – the Marylebone Road and an urban background site – Bloomsbury the average diurnal variation of NO_x , PM_{10} and $\text{PM}_{2.5}$ shows striking similarities indicating common sources of these pollutants and dispersion conditions. However, at the same time, significant differences are evident too. While the street level pollution, when considering NO_x is much higher than the urban background level, the same is not true neither for PM_{10} nor for $\text{PM}_{2.5}$ pollution concentrations. Remarkable high background level of particulates, compared with NO_x , indicates that the contribution from regional sources is much more important for PM-pollution than for NO_x .

Non-combustion PM complicates the $\text{PM}_{2.5}/\text{PM}_{10}$ pattern in some European cities. In southern Europe there might be a high contribution of Sahara dust, that may raise background levels and also contribute to a large reservoir of road dust, available for vehicle induced resuspension. In northern Europe the use of studded tires and wintertime sand gritting contribute to raised $\text{PM}_{2.5}/\text{PM}_{10}$ levels originating from road/brake/tire wear. In a street canyon in central Stockholm the local contribution of resuspended dust to PM_{10} levels is, as an annual average, almost an order of magnitude higher than PM tail pipe emissions, while for $\text{PM}_{2.5}$ the resuspended dust contributes to about the same amount as the vehicle tail pipe emissions.

A somewhat different situation appears when looking at the particle number concentration, and not the mass. As an example, observing measurements of NO_x and the total particle number concentration at two sites in Copenhagen, Denmark, one at a highly trafficked street Jagtvej and the other site is an urban background site, located on the roof of the Copenhagen University building (ca. 20 m) a similarity between the diurnal variation of NO_x and the ultra-fine particle number concentrations is much better than for PM_{10} or $\text{PM}_{2.5}$ concentrations. This points to the conclusion that contribution from local sources to the particle number concentrations can be treated in a much the same way as the contribution of inert pollutants such as NO_x or CO.

Taking into account that the particle concentrations in an urban environment are usually much higher than in the rural environment (especially considering the particle number) one could expect that the

non-linear aerosol dynamics processes could be of importance here. However, at the same time one should also take into account the relatively short residence time of pollutants in an urban environment. For the street scale, the residence time is normally less than 1 min, while for a city it is of the order of some hours at most. Numerical model calculations show that processes such as e.g. coagulation, are quite negligible when considering aerosol dynamics in a car exhaust plume (Vignati et al., 1999). The rapid dilution and the short travelling time prevent the coagulation process to take place efficiently.

It is still an open question to what extent, and how, the nucleation and condensation processes should be taken into account in urban scale modelling. Rapid formation of nucleation mode particles is most likely a large-scale process. However, particle formation at very short distances from car exhaust pipe in certain conditions cannot be excluded.

Available parameterisations for particle deposition indicate fast deposition of ultra fine particles. Deposition could be of importance for the smallest particles on the street scale. It therefore seems necessary to include deposition when trying to model number concentrations on the urban scale.

Currently the major difficulty in particle modelling can be attributed to estimation of emissions. As has been pointed out earlier, it is well recognised that a significant contribution to particle mass (especially PM_{10}) originates from mechanical particle formation, i.e. by resuspension or abrasion of road and vehicle material. Although, some semi-empirical methods have been elaborated for estimation of these emissions, they are far from being generally applicable. A lot of empirical adjustment must be made before applying these methods to different locations, taking into account the road surface conditions or precipitation amount. From regulatory point of view this is a very crucial question. Modelling the effects of emission reduction scenarios it's important to be able to quantify the contribution from regulated sources, such as exhaust pipe emission, versus other, non-regulated sources, as road abrasion and resuspension. This question must be resolved before applying particle models for local, urban scale air quality estimations.

In cities the highest number concentrations are found in traffic environments, where particles smaller than 50 nm dominate. Those particles consist of semi-volatile organic and sulphur compounds formed through gas-to-particle conversion during the initial dilution and cooling of vehicle exhausts, typically within the first tenths of a second after leaving the pipe (Kittelson, 2001). Laboratory data and also ambient air measurements close to roads show a very large variation in the number concentrations of those small ultrafine particles (Andersson and Wedekind, 2001; Harrison and Collins, 1999). The variation is not only due to different emission factors for different motor, fuel and after treatment technologies, the levels may also be very sensitive to ambient conditions (temperature) as well as driving pattern (velocity, A/F ratio) (Kittelson, 2001; Maricq et al., 1999). Thus for modelling number concentrations, emission factors from laboratory tests may yield erroneous results when applied to "real world" conditions. A number of ongoing street canyon and tunnel studies are working on better estimates of emission factors for particle number to be used in urban dispersion models.

As stated in the beginning a good description of the dispersion processes (turbulence and advection) is necessary in local and urban scale modelling. Models are available which solve this problem for gaseous pollutants on different scales ranging from street canyon models to urban diffusion models based on Gaussian dispersion and urban grid point models. From the above discussion it is also clear that different processes need to be taken into account when modelling particles in an urban environment depending on the spatial scale considered. The processes which need to be considered also differ depending on if aerosol mass or number is considered. If the prime interest is to model aerosol mass and primary PM, available models for gaseous pollutants can be applied more or less directly assuming inert dispersion including deposition. The same may be true for modelling of number concentrations of primary particles outside street canyons. If on the other hand modelling of aerosol number and chemical composition is the aim, aerosol dynamical processes have to be included at least on scales larger than the street scale. Work in this field is ongoing e.g. Lurmann et al. (1997) but is limited by lack of knowledge about emissions, chemical and physical processes and data for validation.

A4.4 Implementation of aerosol dynamics in regional transport models

Representation of the particle size distribution

An overview of different approaches applied in aerosol dynamics modelling to approximate the particle size distribution is given in Plate A4.1 as well as their strengths and limitations (Whitby and McMurry, 1997). Depending on the overall aim of the study, the scales of interest, the dimension of the model, and the detail required, an appropriate approach to represent the particle size distribution should be selected.

On the dynamics of aerosol populations

Dynamic processes affecting an aerosol population are condensational growth, coagulation of particles, formation of new particles (referred to as nucleation), and gas/particle mass transfer. Furthermore, the aerosol population can be altered due to transport processes, i.e. horizontal and vertical advection and diffusion, gravitational settling and deposition, aerosol-cloud interactions and emissions. Here, the processes affecting the aerosol population will be addressed with special emphasis on model implementation aspects rather than describing the processes in detail. The subject of mode merging and numerical issues like time stepping are also briefly touched because they have the potential to influence simulation results.

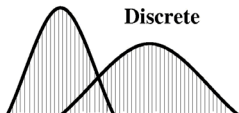
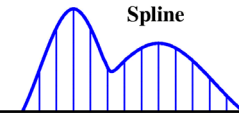
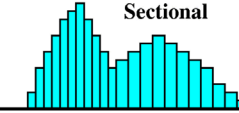
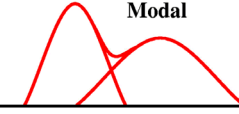
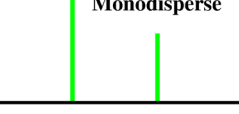
Model Type	Description	# of PDEs	Strengths	Limitations
 Discrete	calculate time dependent concentration of each possible cluster	# PDEs = # cluster sizes		
 Spline	subdivide into series of sections, section functions are splined together	cubic splines: # PDEs = 4 * # sections	accurate	nonphysical distribution function possible, computationally intensive
 Sectional	subdivide into series of sections, distribution function is constant within section	# PDEs = # sections	flexible model structure multicomponent simulations, well developed codes and user interfaces available	significant numerical diffusion for particle growth, computationally intensive, accuracy dependent on number of sections used to discretize size distribution
 Modal	represent concentration by a continuous distribution function for overlapping intervals	# PDEs = 3 * # modes	flexible model structure, computationally fast	accuracy dependent on the form of the mode distribution functions
 Monodisperse	represent concentration at a finite number of discrete sizes	# PDEs = # discrete sizes	flexible model structure, computationally very fast	only useful for rough estimate of aerosol dynamics, no size distribution information

Plate A4.1 Overview of approaches to model aerosol dynamics (Whitby and McMurry, 1997)

Condensational growth: The rate of change of a particle size distribution due to condensation or evaporation is mathematically described by the so-called condensation equation (Seinfeld and Pandis, 1997). Depending on the representation of the particle size distribution the condensation equation has to be converted into an approach specific expression. Sectional approaches can be subject to significant numerical diffusion (Seigneur et al., 1986). However, some efforts have been made to reduce numerical diffusion, e.g. full-moving approach in combination with fixed sections for 3-D processes (hybrid approaches), use of advection algorithms to solve the condensation equation, or the use of characteristics within a section. A comparison of three sectional approaches, all of them currently used in 3-dimensional air quality models, is presented in Zhang et al. (1999).

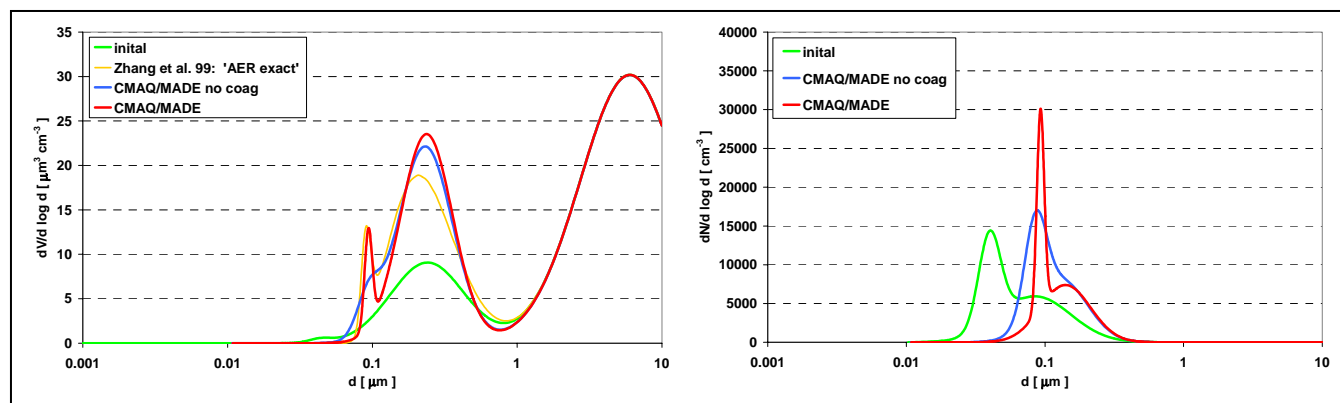


Figure A4.1 Simulation of condensation for hazy conditions (for details see Zhang et al. (1999)) with a condensation rate of $5.5 \text{ m}^3 \text{ cm}^{-3} / 12\text{h}$ applying a recent version of CMAQ (Binkowski and Roselle, 2002), initially (green line) and after 12 h (red line); left: particle volume distributions; right: particle number distribution. Additionally results of a simulation with no coagulation are shown (blue line) after 12 h.

Coagulation: Resulting from the discretisation of the particle size distribution, there are two principle classes of coagulation processes from a model implementation aspect: intramodal coagulation and intermodal coagulation. For intramodal coagulation, there are two possibilities on the whole: collision of particles of the same size mode results in a particle within this size mode or the resulting particle is assigned to a larger size mode. Hybrid strategies involving both possibilities have also been applied in 3-D model approaches (e.g. Ackermann et al., 1998). For intermodal coagulation in contrast, particles formed by collision from two different size modes are usually assigned to the larger size mode involved. Coagulation is an important process and should be included simulating urban conditions. However, the results presented in Figure A4.1 suggest, that coagulation is also important for hazy conditions.

Nucleation: The processes of new particle production results in an increase of the number concentration of ultrafine particles and leads to an increase in particle mass concentration. From a model implementation point of view, the formulation of nucleation rates can be considered as containing two components: (1) the overall formulation that defines whether the nucleation rate is calculated after the condensation rate, before the condensation rate, or together with the condensation rate; and (2) the actual parameterisation of the nucleation rate (Zhang et al., 1999). Further discussion is found above in part by Kulmala and Hansson.

Gas/particle mass transfer: The treatment of mass transfer between the bulk gas phase and the particles has been described by three major approaches: (1) an explicit kinetic approach, which is accurate but computationally demanding; (2) an equilibrium approach, assuming that the bulk gas phase is in equilibrium with the particles, which is computationally efficient but may be less accurate in the presence of sea salt; (3) hybrid approaches, using equilibrium assumptions for the bulk gas phase and the particle phase, but considering some aspects of mass transfer processes, e.g. using diffusion limited assumptions. Zhang et al. (1999) conclude, that the explicit treatment of gas/particle mass transfer is only important for areas, where compounds containing chloride and carbonate comprise a significant fraction of the particulate matter.

Mode merging: As particles age in the atmosphere, they may continuously grow by condensation processes and coagulation from smaller size ranges to larger sizes, i.e. particles from the nucleation size mode grow into the Aitken size mode and finally to the accumulation size mode. Eventually, a new nucleation mode may appear at smaller diameters. While this is probably true in nature, it usually violates the modelling paradigm, that a fixed number of well defined distinct size areas, i.e. sections, modes, or bins, exist to represent the particle size distribution. Therefore, algorithms are required to transfer characteristics like number, surface, and volume between the model particle size modes. Currently, there are two common criteria used to determine if renaming by mode merging has to be done: fixed borders or dynamic mode overlap criteria. Some effort has been started to develop a general framework and algorithms for managing the modes in modal aerosol dynamics models (Whitby et al., 2002).

Numeric solutions and time stepping: Several numerical methods are used to solve the general dynamic equations, describing mathematically the effect of aerosol dynamic processes. Usually analytical solutions or iterative solutions to the problem are used depending on the specific model formulation. Analytical solutions are by nature computationally very effective, but cannot always be derived. However, it has to be noted, that analytical solutions are often derived on the basis of specific assumptions, e.g. that terms or coefficients constant in time. Therefore, adjustable time steps may be needed even for analytical solutions.

Implementation into a regional model framework

Implementation of an aerosol dynamic model, which includes the processes very briefly touched in the previous section, into a regional air quality model framework involves additional processes like gas phase production of condensable material, transport, gravitational settling, deposition, aerosol-cloud interaction and primary particle emissions. These processes also alter the particle size distribution. Since the coupling of a particular aerosol dynamics model and a particular regional air quality model can be very specific, here these aspects are only addressed very generally.

In general, horizontal and vertical advection as well as horizontal and vertical diffusion are considered as transport processes in regional air quality models. The quantities, which have to be transported in order to describe the particle size distribution, can usually be coupled to the transport algorithms available. However, this has to be done with some attention, since large concentration gradients in the order of several magnitudes can occur, e.g. during nucleation events. In order to be able to cope with the mixing of different particle size distributions due to transport processes computationally efficient, the same basic representation of the particle size distribution, i.e. the same sectional discretisation or an equal number of modal modes, is usually applied in each grid cell as a matter of principle.

Important loss processes for aerosol particles are sedimentation and dry deposition (e.g. Jacobson, 1999). The two main aspects usually considered in regional models are Brownian diffusion and gravitational settling of particles. Both processes are dependent on the actual particle size distribution and require online calculation for that reason. Ackermann et al. (2001) found in their simulations, that coarse particle concentrations are considerably higher and that long-range transport is enhanced, if gravitational settling is neglected.

Aerosol-cloud interactions are very complex in detail. Since cloud processes itself and therefore also aerosol-cloud interactions, e.g. activation of aerosol particles, cloud droplet formation, aqueous-phase chemistry, interstitial aerosol, vertical mixing, scavenging, and wet removal, occur at subgrid scale in regional air quality models, these processes are highly parameterised. As an example, Binkowski and Roselle (2002) may be mentioned at this point, how aerosol-cloud interactions can be implemented into a regional air quality model framework.

Although primary particle emission can influence the particle size distribution considerably, particulate matter emission inventories available for regional air quality modelling do currently not contain sufficient information about neither size distribution nor chemical speciation. Some information on anthropogenic particle emission is available, but European inventories of natural particle sources are lacking.

A4.5 Particle models within EMEP

An aerosol dynamics model has been recently implemented in the EMEP MSC-W modelling system. This new model, UNI-AERO, takes into account aerosol dynamics, i.e. nucleation, condensation and coagulation, and allows for size dependent description of removal processes. An overview of the emission, transformation and removal processes included in UNI-AERO is schematically presented in Figure A4.2.

UNI-AERO distinguishes 7 aerosol components in 4 different size modes, sulphate, nitrate, ammonium, EC, OC and seasalt. The size ranges considered are nucleation, Aitken, Accumulation and Coarse mode. All particles within each mode are assumed to have the same size (monodisperse) and chemical composition (internally mixed).

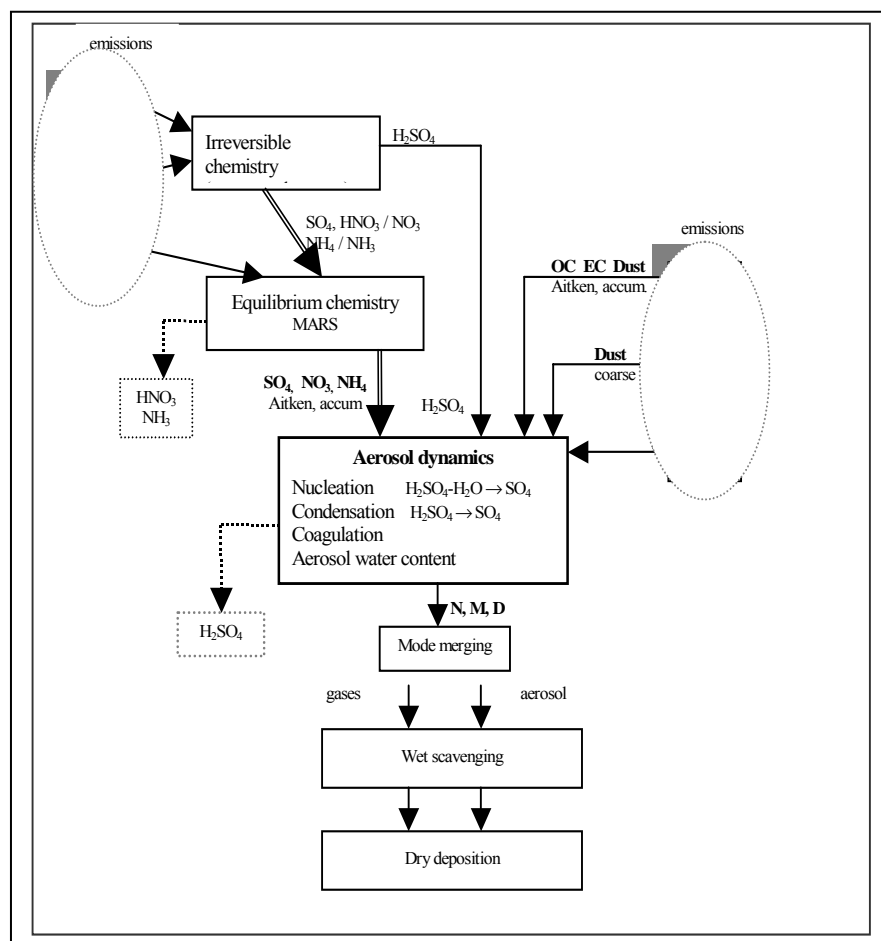


Figure A4.2 Schematic computational structure of the Unified EMEP aerosol model version (UNI-AERO).

Aerosol water is treated as a diagnostic parameter, following Binkowski & Shankar (1995). Sea salt particle generation rate is calculated as a function of size, based upon Monahan *et al.* (1986). The chemical scheme is based upon that of the acid deposition version of the EMEP model (e.g. Olendrzynski *et al.*, 2000), but with two differences. Firstly, SO_2 gas-phase and heterogeneous oxidation reactions are described separately, thus allowing the explicit description of sulphate formation by nucleation and condensation of gaseous H_2SO_4 . Secondly, equilibrium reactions for NO_3 and NH_4 are now performed with the thermodynamic model MARS (Saxena *et al.*, 1986, Binkowski & Shankar, 1995). Aerosol dynamic processes are coupled to the chemistry and parameterised following the MM32 version of MULTINOMO (Pirjola *et al.*, 2002, Tsyro *et al.*, 2002; Pirjola and Kulmala, 2000) except for nucleation where rates from the MADMAcS model from the Institute of Tropospheric Research in Leipzig are used. Finally, removal by dry and wet deposition is considered to be a size dependent process.

UNI-AERO is flexibly designed so that the model can be extended as necessary to include more chemical components and size modes. The present version solves 27 prognostic equations (7 for gases and 20 for particle number and masses) and thus is relatively cost-efficient in terms of CPU usage.

Initial testing of UNI-AERO has focused on the analysis of aerosol dynamic processes and on the effect of size-resolved dry and wet deposition on primary PM concentrations. A significant conclusion so far is that accounting for size dependent emissions and dry and wet deposition of aerosols in the model has a larger impact on the calculated concentrations of primary PM than introducing coagulation processes. These results underline the need for accurate descriptions of size dependent emissions and dry and wet removal processes. Current testing aims to establish the effect of aerosol dynamics on the calculated secondary inorganic aerosol (SIA) concentrations.

Model evaluation is of crucial importance to establish the soundness of a model system and to aid further development. Initial comparison of UNI-AERO with available observations at EMEP sites indicates quite a reasonable agreement between model results and measured PM₁₀ concentrations in Norway, Switzerland and Germany, although differences in composition are found. In some regions, e.g. in the Mediterranean area, UNI-AERO considerably underestimates ambient PM₁₀ levels, presumably partly because natural and re-suspended anthropogenic mineral dust and biogenic aerosols are currently not incorporated in the model.

Model validation requires sufficient measurements in terms of site density, data quality and chemical/physical parameters. The EMEP monitoring programme still provides insufficient data for model evaluation, particularly regarding chemical composition. Measurements of the size distribution of PM mass and number, extremely important for model verification and development, are presently being compiled under different programs associated with EMEP, and will form the basis for the next stage of model evaluation.

Secondary Organic Aerosols (SOA) are created from the condensed phase of semi-volatile organic compounds (SOCs). Such SOCs are either emitted directly (for example, heavy alkanes or aromatic species have very low vapour pressures and thus belong to this class), or are formed in chemical reactions (e.g. from oxidation of terpenes). Two different approaches have been taken in the EMEP model (Andersson-Sköld and Simpson, 2001) to deal with SOA formation. In one method, the SOCs created following oxidation of a primary hydrocarbon are parameterised in terms of a two-species system (e.g. Odum et al., 1997, Griffin et al., 1999), with one species typically being much less volatile than the other. In a second approach (following Kamens et al., 1999), a more realistic chemical treatment is attempted, in which products of oxidation are involved in gas-phase chemistry as specific species.

However, the organic aerosol systems observed in atmospheric samples display features which are hard to explain with current approaches, such as strong contributions of long chain (>C₁₀-C₃₀) compounds (e.g. Seinfeld and Pandis, 1998). Reactions in the aerosol may be important sources of such long-chain compounds. SOC sources could be from anthropogenic or foliar emissions, but other more speculative sources (e.g. arising from organic debris in soils, Gelencser et al., 2002) are possible. Further, the complexity of organic aerosols precludes proper characterisation of thermodynamic properties (vapour pressure, activity coefficients). Unfortunately, we do not currently know what proportion of atmospheric organic carbon is due to primary emissions, secondary sources, anthropogenic or natural sources. In such conditions, Andersson-Sköld and Simpson (2001) concluded that real progress can only be made when observational data can provide more information on the composition and sources of organic carbon in the atmosphere. For this reason, we do not currently include SOA modelling as part of policy calculations within EMEP.

A4.6 Global Aerosol Transport Models

Modelling global aerosol distributions in global 3-dimensional chemical transport models and general circulation models is primarily driven by the need to evaluate the impact of aerosols on climate. In this section, we review progress in global aerosol modelling and identify remaining problems.

Mass based estimates of the direct and indirect effect

The direct effect of aerosols on the earth's radiative balance is a function of their mass concentration, size distribution and composition. First estimates of the direct effect considered the sulphate fraction alone, and estimated the radiative forcing from mass concentration fields and observed mass scattering coefficients, e.g. Charlson et al., (1991), which used the mass distribution of *Langner and Rodhe* (1991). A refinement of this assumed a size distribution, and a state of mixing for the sulphate aerosol, and calculated the mass scattering coefficient and radiative forcing accordingly (e.g., Kiehl and Briegleb, 1993).

This approach has been extended to mixed aerosols by superimposing mass distributions of different aerosol components, e.g. Sulphate & Black Carbon (*Hayward & Shine, 1995*). Again size distributions and mixing states for each component are assumed, from which the mass scattering and absorption coefficients and radiative forcings can be calculated. Several models now exist that have extended this approach to multi-component mixtures including Sulphate BC/OC, Dust & Seasalt, and their results have been compared with satellite derived aerosol optical fields (*Penner et al., 2002*).

The indirect effect is similarly a function of the aerosol mass concentration, size distribution and composition. Specifically it is a function of the fraction of the size distribution that is capable of acting as cloud condensation nuclei. Accordingly the indirect effect has been estimated, both from empirical relationships between sulphate mass and cloud droplet number concentrations (CDNC), (e.g., *Boucher and Lohmann, 1995*), and by assuming a fixed size distribution and deriving the CDNC from the mass concentration (e.g., *Jones et al., 1994*).

All these models assume that the aerosol is externally mixed, and has a constant size distribution. In reality however aerosols are normally internally mixed (e.g., *Bates et al., 1998*) and the size distributions vary spatially and temporally.

Representing aerosols as an internal mixture

The challenge in global aerosol modelling, as in regional scale modelling is to resolve mass concentration, size distribution and composition. This creates two problems:

- An aerosol microphysical model is needed to describe the development of the aerosol mixture, which is computationally intensive.
- The aerosol mixtures contain compounds of differing volatility, the equilibrium between the aerosol and the vapour phases needs to be calculated, which is also computationally intensive.

Models have therefore tended to emphasise either the microphysics or the calculation of the equilibrium composition, not both, frequently adopting the same techniques as described in the above sections.

Aerosol microphysics

Resolving aerosol microphysics in a global model requires a multi-component approach. A microphysical model of a single component will not produce realistic size distributions, as it cannot represent interactions with the rest of the aerosol population. One of the first attempts to include aerosol microphysics in a global model of mixed aerosols is *Wilson et al. (2001)*. In this study a sulphate, black carbon, organic carbon & sea salt microphysical model is coupled to an offline chemical transport model, TM2. Mixed modes develop by condensation of sulphate onto pure black carbon modes, and thence by coagulation with the nucleation and Aitken mode sulphate. The accumulation mode population is the sum of a 'pure' sulphate accumulation mode, mixed black carbon modes, and a sea salt mode. The 'pure' sulphate' modes indicate 'clean' atmosphere processes, while the mixed biomass burning, and fossil fuel black carbon modes are indicative of anthropogenic influences. The spatial distribution of the dominant contributors to the accumulation mode is shown in Figure A4.3.

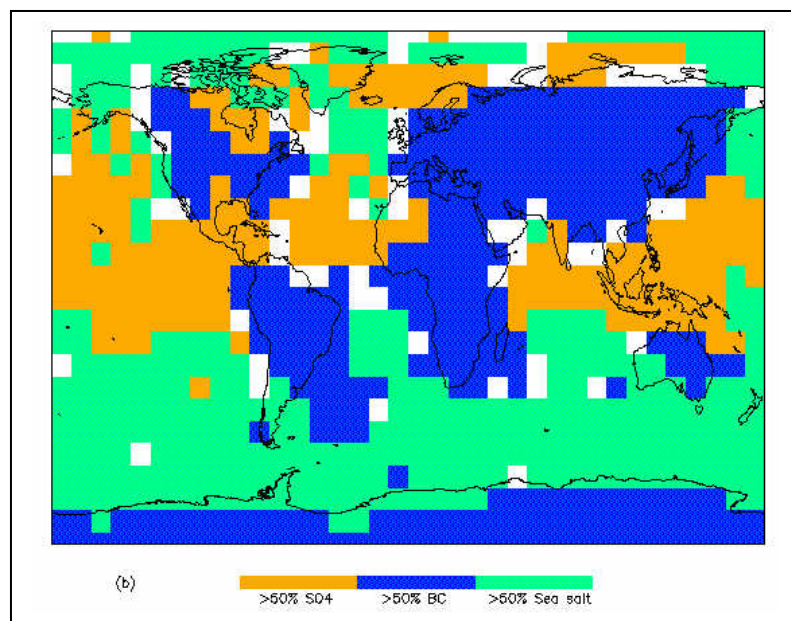


Figure A4.3 The distribution of areas where the accumulation mode number concentration is more than 50% one type (Wilson et al., 2001).

The model agrees qualitatively with surface observations, but has a tendency to predict unrealistically high concentrations of nucleation and Aiken mode sulphate particles in the upper free troposphere.

Aerosol equilibrium models

More studies have been conducted with aerosol equilibrium models coupled to Chemical Transport Models and General Circulation models e.g. Jacobson (2001), Jeuken et al. (2001). Typically in these studies, the aerosol physical properties are assumed constant. Even though there may be, some explicit description of the aerosol size distribution in the models, e.g. Jacobson (2001) where it is represented by 17 size bins, this is assumed to be constant, so that there is no internal aerosol dynamics processing in the model. The study of Jeuken et al. is interesting in it uses a parameterisation scheme that is sufficiently fast for it to be coupled to a microphysical model.

Future developments

An important requirement for size resolving global aerosol models is the characterization of primary emission size distributions. The development of size resolving models capable of evaluating equilibrium dimensions is also a key improvement. Several parameterisation schemes for example for nucleation, and equilibrium dimensions are not appropriate for the entire range of conditions found in global models, in particular in the upper free troposphere, and these schemes need to be extended into these areas. Where models resolve both number and mass, the impact of differential advection of these properties on the size distribution needs also to be investigated. Finally the coupling of size resolving aerosol schemes to cloud activation schemes also needs further development.

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Annex 5 Annex to Chapter 6 – Selected examples of source apportionment of PM in Member States

In addition to the local and regional anthropogenic particulate emissions, both the levels and composition of ambient air PM depend on the climatology (mainly temperature, humidity, photochemistry, resuspension of soil particles, rain scavenging potential, re-circulation of air masses vs. dispersive atmospheric conditions) and on the geography (mainly proximity to the coast, soil cover and proximity to arid zones) of a given region. Therefore, wide variations on PM levels and characteristics may be expected when considering different EU regions such as Southern Mediterranean and the Scandinavian countries with very different climatologic and geographical patterns. Consequently, due to the difficulty of reporting general PM compositional characteristics, in this chapter examples of source apportionment analysis performed in Germany, Spain, the United Kingdom, the Netherlands and Sweden are given to illustrate different scenarios on PM in the EU.

The main objective of this report is not to cover all possible scenarios of PM from different EU sites, but to compare and interpret PM characteristics and sources of 5 selected regions, having very different climatic patterns, within EU. Consequently, PM₁₀ levels and contributions in other countries and cities may well fall outside the general picture presented here. Putaud et al. (2002) recently compiled a very complete report on PM phenomenology in the EU, and detailed studies on PM speciation and source apportionment analysis may be found in APEG (1999), Visser et al. (2002), Quass and Kuhlbusch (2002) and Querol et al. (2003), for the United Kingdom, the Netherlands, Germany and Spain, respectively.

The data selected here were obtained from the following sources:

- PM data from Berlin in 1998, Germany from Lenschow et al. (2001). Data for 2000-2001 the data were obtained from Abraham (2001) and Abraham H.J. (Senate Department of Urban Development, Berlin, Germany, personal communication).
- PM data from rural and urban background and kerbside sites in Spain in the period 1999-2001 were obtained from Querol et al. (2001 and 2003), Artíñano et al. (2003) and Rodriguez et al. (2002) and from EMEP for 2001.
- PM data from Sweden for 199-2001 were extracted from Areskoug et al. (2004).
- PM data from the Netherlands were obtained from different sources but compiled by Dr E. Buringh from RIVM, National Institute of Public Health and the Environment in the Netherlands.
- PM data from the United Kingdom were derived from four paired urban background and roadside sites in London and Birmingham operated by the University of Birmingham (Harrison et al., 2003).

In most of these examples, source apportionment analysis has been carried out by considering the scenario of an urban background as an "island" of elevated PM levels arising from a regional background and on top definite peak concentrations due to traffic hot spots at road side sites (Lenschow et al., 2001). Consequently, PM₁₀ and PM_{2.5} measurements performed in these three typical locations (regional background, urban background and roadside sites), including speciation, has been used as a basic information for a source apportionment analysis.

A5.1 Berlin

The conurbation of Berlin (3.4 million inhabitants) is situated in a flat area with low population density. A summary of the results from PM₁₀ and PM_{2.5} measurements performed in 1998 at three typical locations (regional background, urban background and traffic exposed), including speciation, are shown in Table A5.1. Lenschow et al. (2001) for the source apportionment analysis, complemented the information supplied from these data with the emission inventories for PM and gaseous (NO_x, SO_x, NH₃, VOC) emissions for Germany (regional background) and Berlin (urban

background). The natural background such as wind blown dust, pollen or volcanic eruptions was assumed to be $5.9 \mu\text{g}/\text{m}^3$.

Table A5.1 shows that, comparing the PM_{10} urban background and regional background levels (38 and $19 \mu\text{g}/\text{m}^3$ in 1998), 50% of the urban background pollution was caused by emissions within the agglomeration, and about 50% by long range transport and natural sources.

PM_{10} levels near busy streets ($52 \mu\text{g}/\text{m}^3$ in 1998) were about 40% higher than in the urban background. From the chemical speciation, it can be concluded that 55% of this additional pollution consisted of carbonaceous material (exhaust emissions, tyre abrasion) and 45% could be attributed to road dust, the other species being roughly the same (Figure A5.1). The comparison of the simultaneously measured PM_{10} and $\text{PM}_{2.5}$ concentrations and speciation showed that one third of the resuspension was in the fine mode ($\text{PM}_{2.5}$) and two thirds in the coarse mode.

Table A5.1 Average concentration of PM_{10} and $\text{PM}_{2.5}$ and main chemical components in the city of Berlin and the regional background ($\mu\text{g}/\text{m}^3$)

	Traffic ¹⁾ (Frankfurter Allee)	Urban back. ¹⁾ (Nansens tr)	Regional back. ¹⁾ (Neuglowsow)	Berlin ²⁾ Kerbside	Berlin ²⁾ Kerbside	Berlin ²⁾ Urban back	Berlin ²⁾ Urban back
	1998	1998	1998	2000	2001	2000	2001
PM_{10}	51.5	37.7	18.6	37	42	28	29
OM	11.0	7.5	3.0				
EC	9.0	5.1	1.8				
OM+EC	20.0	12.6	4.8	11.2	12.7	7.1	8.1
SO_4^{2-}	5.7	6.1	3.9	5.4	4.6	4.9	4.9
NO_3^-	4.6	4.4	2.4	5.8	5.2	5.3	5.2
NH_4^+	3.3	3.4	2.0	3.0	2.7	2.9	3.0
Secondary inorg.	13.6	13.9	8.3	14.2	12.5	13.1	13.1
Cl^-	0.8	0.7	--				
Marine				2.3	2.2	2.1	2.0
Unaccounted ³⁾	17.9	11.2	5.5	9.3	14.7	5.8	5.8
$\text{PM}_{2.5}$	38.6	30.2	--	25	27	22	22.5
OM	9.4	7.0	--				
EC	7.7	4.7	--				
OM+EC	17.1	11.7		9.5	10.4	6.0	6.8
SO_4^{2-}	5.6	5.5	--	4.6	4.2	4.6	4.6
NO_3^-	4.3	3.7	--	4.6	4.1	4.6	4.0
NH_4^+	3.8	3.7	--	2.9	3.1	2.8	2.8
Secondary inorg.	13.7	12.9		12.1	11.4	12.0	11.4
Cl^-	0.7	0.5	--				
Marine				1.0	1.1	0.9	1.1
Unaccounted ³⁾	7.8	5.6	--	2.4	4.2	3.1	3.1

¹⁾ from Lenschow et al. (2001)

²⁾ Abraham (2001) and Abraham H.J. (personal communication)

³⁾ Unaccounted, mainly metal oxides (soil) and water

Levels of $\text{PM}_{2.5}$ measured in 1998 reached $38 \mu\text{g}/\text{m}^3$ near busy streets and $30 \mu\text{g}/\text{m}^3$ at urban background sites.

Allocating these levels according to the contributions that the main source groups have in the corresponding emission inventories (adjusted by scaling factors due to different dispersion conditions) lead to the source apportionment for traffic hot spots shown in Figure A5.1. About 36% of this pollution burden stems from sources outside of the agglomeration and cannot be influenced by local measures.

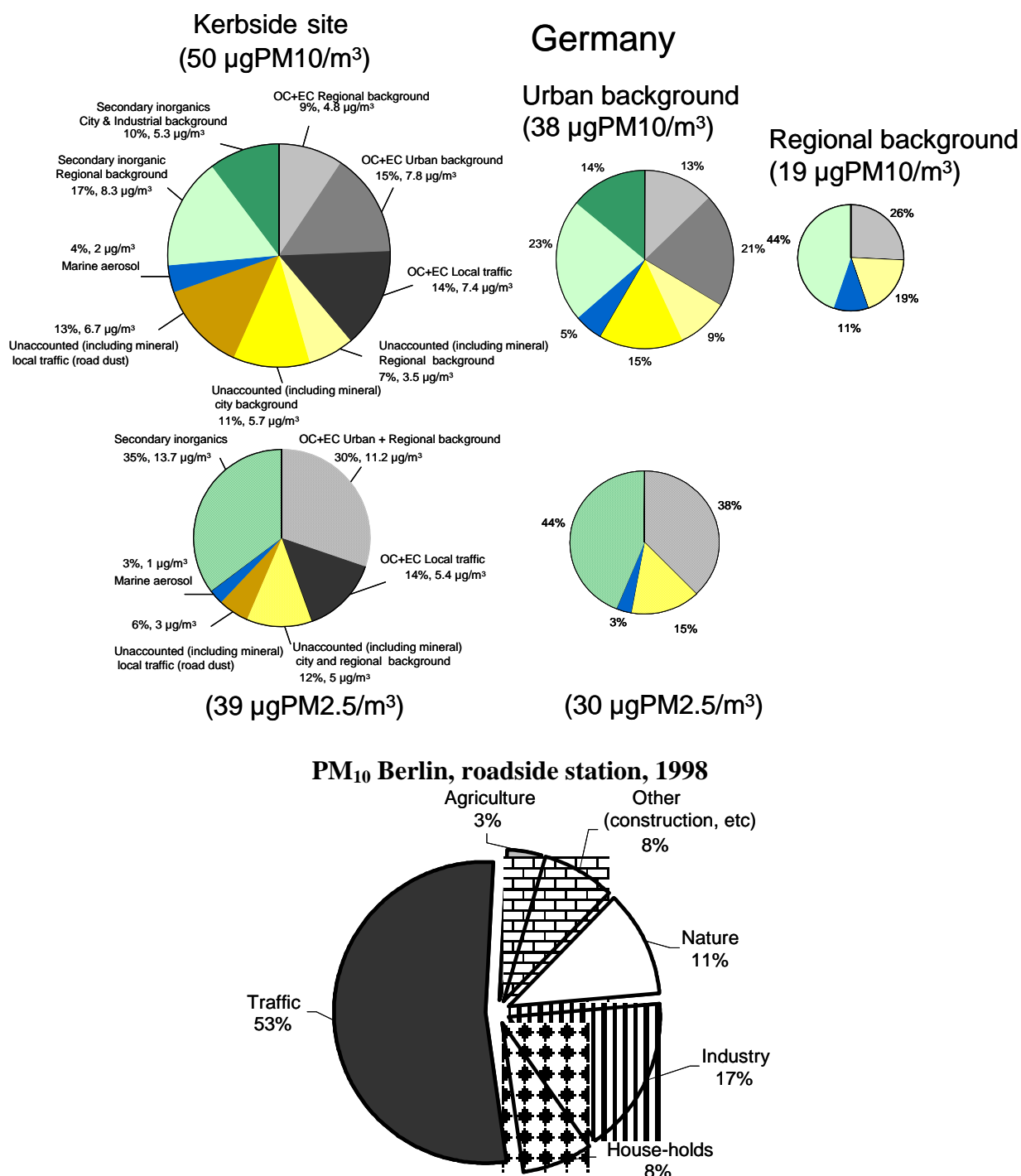


Figure A5.1 Top: Results of the source apportionment analysis of PM₁₀ and PM_{2.5} performed in regional background, urban background and kerbside sites from Berlin, Germany. Based on data from Table A5.1. Bottom: Source contribution analysis of PM₁₀ in a busy traffic spot in Berlin in 1998.

As already mentioned, sources within the conurbation were responsible for 50% of the particulate burden at urban background locations. Half of this local contribution was due to traffic emissions, 15% to domestic heating and households, 19% to other sources, such as construction, and about 9% to industrial sources.

In a similar manner, the regional background could be allocated to source categories as follows: 37% industrial sources, 22% from traffic, 22% from natural sources and 10% from agriculture.

Subsequent studies performed during 2000 and 2001 reported the PM_{10} and $PM_{2.5}$ concentrations at a kerbside site and an urban background station from Berlin determined by field campaigns. A total of 228 samples (114 $PM_{2.5}$ and 114 PM_{10}) were collected using manual gravimetric instruments. The average PM_{10} level over the whole periods of the measurement campaigns was 42 and 37 $\mu g/m^3$ at traffic site for 2001 and 2000, respectively. The $PM_{2.5}$ concentrations was on average 35% lower than PM_{10} (mean of 27 and 25 $\mu g/m^3$). Average PM levels at the urban background station reached 29 $\mu g/m^3$ for PM_{10} and 22 $\mu g/m^3$ for $PM_{2.5}$. In both cases levels are significantly lower than in the measurements performed in 1998. The mean $PM_{2.5}/PM_{10}$ ratios reached 0.66 and 0.78 at the urban traffic and background sites, respectively for the data 2000-2001 and 0.75 and 0.80 for the 1998 data.

A5.2 Spain

Table A5.2 shows the results of studies on source apportionment analyses of PM_{10} and $PM_{2.5}$ carried out at rural and urban background and kerbside sites in Spain in the period 1999-2001. The data discussed here is the product of research projects sponsored by the Spanish Ministries of Environment and Science and Technology. Results have been published by Artiñano et al. (2003) for a roadside site of Madrid-1999, Querol et al. (2001) for a roadside site of Barcelona-1999, Rodriguez et al. (2002) for a rural background site (Monagrega) and Querol et al. (2003) for 2001 data from Tarragona, Huelva, Bemantes, Llodio, Alcobendas, Barcelona and Gran Canaria. The following conclusions may be drawn from the comparison of these studies with the PM_{10} and $PM_{2.5}$ EMEP data for 2001:

- The regional background levels of PM are 15 $\mu g PM_{10}/m^3$ in the Atlantic regions, 20 $\mu g PM_{10}/m^3$ in the other Spanish regions, and 9-14 $\mu g PM_{2.5}/m^3$ for the whole territory.
- The most common urban background concentrations of PM_{10} and $PM_{2.5}$ in Spain fall in the range 30-40 and 17-25 $\mu g/m^3$, respectively.
- The most common kerbside concentrations of PM_{10} and $PM_{2.5}$ in Spain fall in the range 45-50 and 25-35 $\mu g/m^3$, respectively.

It is important to note that the source apportionment studies shown here are carried out by determining the levels of more than 50 parameters in each sample, but a fraction of around 4 to 9 $\mu g/m^3$ in PM_{10} and 3 to 6 $\mu g/m^3$ in $PM_{2.5}$ is still unaccounted for at regional and urban sites, respectively. A fraction of this unaccounted mass may be attributed to water since the filter samples can not be heated or dried to avoid loss of volatile material. This water may be an important fraction of PM_{10} and $PM_{2.5}$ levels.

Regional and rural background sites

As shown in Figure A5.2, the fraction of background secondary inorganic phases (sulphate ammonium and nitrate) accounts for only 20% of PM_{10} (5 $\mu g/m^3$) in the Canary Islands, 30% (5 $\mu g/m^3$) in the Atlantic regions of the Iberian Peninsula and 35% (7.5 $\mu g/m^3$) in the rest of the Peninsula. Conversely, the mineral background fraction increases from only 13% (2.5 $\mu g/m^3$) in the Atlantic regions, to 25-30% (5-7 $\mu g/m^3$) in the rest of the Iberian Peninsula and the Canary Islands. The marine aerosol contribution to PM_{10} may be estimated to range from around 5% (1 $\mu g/m^3$) in the Central areas to 15% (2.5 $\mu g/m^3$) in the coastal areas of the Iberian Peninsula, but it reaches around 35-40% (11 $\mu g/m^3$) in the Canary Islands. Finally, the organic and elemental carbon concentrations account for 10 to 25% (2.5 to 4.5 $\mu g/m^3$) of the PM_{10} levels from the Mediterranean to the Atlantic regions.

Given that the marine aerosol and mineral elements mainly occur in the 2.5-10 μm fraction, the range of variations of their concentrations measured in $\text{PM}_{2.5}$ at the different sites is narrower when compared with PM_{10} . Thus, as shown in Figure A5.3, the fraction of background secondary inorganic phases accounts for 35-45% of $\text{PM}_{2.5}$ (4-6 $\mu\text{g}/\text{m}^3$), the mineral background for 10-20% (1.5-3 $\mu\text{g}/\text{m}^3$), the marine aerosol contribution for 5-10% (0.5-1.5 $\mu\text{g}/\text{m}^3$) and the organic and elemental carbon for 20% (2-3.5 $\mu\text{g}/\text{m}^3$).

Urban background sites

As shown in Figure A5.2, in urban background areas of Spain, on the average around 60% (20 $\mu\text{g}/\text{m}^3$) of the PM_{10} mass is accounted for by regional background particulate matter (with the above composition), the additional 40% (12-14 $\mu\text{g}/\text{m}^3$) is made up of the city background contribution of which one third (4-5 $\mu\text{g}/\text{m}^3$) is mineral dust (mainly from erosion of pavement in roads and demolition dust). Another third is carbonaceous material, mainly arising from traffic emission (both exhaust emission and tyre erosion), and the remaining third is unaccounted mass (an important fraction of which is water).

Only 20-35% of the PM_{10} city background mineral dust input is present in $\text{PM}_{2.5}$, whereas a fraction 80-90% of carbonaceous particles input is also present in $\text{PM}_{2.5}$.

In the case of $\text{PM}_{2.5}$ (Figure A5.3), the regional background accounts for 60-70% (11-13 $\mu\text{g}/\text{m}^3$) of the urban background levels of $\text{PM}_{2.5}$. The additional 30-40% (8-10 $\mu\text{g}/\text{m}^3$) is accounted by the city background input from which only 20% (1.5 $\mu\text{g}/\text{m}^3$) is made of city mineral dust, 50% (4 $\mu\text{g}/\text{m}^3$) of carbonaceous material and the remaining 30% (3 $\mu\text{g}/\text{m}^3$) by the unaccounted mass.

Table A5.2 PM_{10} and $\text{PM}_{2.5}$ composition in different environments from Spain.

	Monagrega Bemantes		Tarragona Huelva Alcobendas Llodio Canary					Madrid L'Hospitalet Barcelona		
	Rural background		Urban background stations.					Kerbside sites		
	1999-2000	2001	2001	2001	2001	2001	2001	1999-2000	1999-2000	2001
PM_{10} ($\mu\text{g}/\text{m}^3$)	20.7	18.9	37.4	37.5	32.2	31.7	42.9	47.7	49.8	46.2
N	132	87	88	91	84	85	90	69	115	90
OC+EC	2.3	4.4	8.0	5.8	8.3	6.7	6.7	16.5	12.1	11.9
Mineral	5.2	2.5	9.2	11.8	8.6	7.3	11.5	15.2	12.9	15.2
Marine	0.7	2.6	2.3	2.8	1.2	2.3	11.5	0.8	2.3	2.4
Unaccounted	5.0	3.5	6.6	8.0	8.4	4.7	6.7	7.5	7.2	6.1
Secondary Inorganic	7.5	5.2	10.2	7.9	6.4	8.7	4.9	7.7	15.3	10.8
nmSO_4^{2-}	3.7	3.0	4.9	4.7	2.9	5.4	2.7	4.4	6.8	4.9
NO_3^-	2.5	0.9	3.9	1.8	2.0	1.5	1.6	2.1	5.8	3.9
NH_4^+	1.3	1.3	1.5	1.5	1.5	1.6	0.8	1.2	2.7	2.0
$\text{PM}_{2.5}$ ($\mu\text{g}/\text{m}^3$)	Nd	13.5	21.8	19.3	24.9	23.9	18.6	34.1	34.5	27.7
N		45	43	49	34	44	49	38	63	45
OC+EC	Nd	4.0	6.4	5.5	7.7	6.4	6.4	15.7	11.2	10.1
Mineral	Nd	1.5	1.9	2.7	2.7	2.3	3.2	5.6	4.2	4.2
Marine	Nd	1.0	1.0	0.8	0.6	1.2	1.6	0.5	0.9	0.7
Unaccounted	Nd	2.0	3.8	4.7	5.0	3.4	2.8	5.8	4.2	2.7
Secondary Inorganic	Nd	4.4	7.2	5.5	5.3	7.4	3.0	6.5	13.0	8.3
nmSO_4^{2-}	Nd	2.9	3.9	3.6	2.7	4.9	1.9	3.8	5.8	4.2
NO_3^-	Nd	0.4	1.9	0.5	1.3	0.8	0.4	1.3	4.0	2.3
NH_4^+	Nd	1.2	1.4	1.4	1.4	1.7	0.7	1.4	3.2	2.0

Kerbside sites

At kerbside sites around 40 and 30% of the PM_{10} mass is accounted by the regional and urban background particulate inputs, respectively. The additional 30% (13-15 $\mu\text{g}/\text{m}^3$) is supplied by local traffic from the monitored street. As shown in Figure A5.2, at least 85% of this kerb contribution is

made up of road dust (45%, $6.5 \mu\text{g}/\text{m}^3$) and carbonaceous particles from exhaust emissions and tyre erosion (40%, $6 \mu\text{g}/\text{m}^3$).

As for the urban background sites, only 30% of the PM_{10} local road dust input at kerbside sites is present in $\text{PM}_{2.5}$ (around $4\text{--}5 \mu\text{g}/\text{m}^3$ of mineral dust in the annual $\text{PM}_{2.5}$ mean is accounted by mineral dust), whereas a fraction $>90\%$ of the kerb PM_{10} carbonaceous particles input at these sites is also present in $\text{PM}_{2.5}$.

In the case of $\text{PM}_{2.5}$ (Figure A5.3) the same contributions of regional and urban background and local kerb emissions described for PM_{10} are measured (40/30/30%, $13/9/9 \mu\text{g}/\text{m}^3$). In this case at least 60% ($5.5 \mu\text{g}/\text{m}^3$) of this kerb contribution is accounted by carbonaceous particles from exhaust emissions and tyre erosion and only 20% ($2 \mu\text{g}/\text{m}^3$) by road dust.

Industrial contribution

From the data shown in Table A5.2 it is evident that some urban background and kerbside sites have also a proportion of PM_{10} and $\text{PM}_{2.5}$ made of secondary inorganic species much higher than the regional and most urban background sites. This is probably an artefact introduced by the fact that two of the three kerbside sites and one urban background site investigated are located in highly industrialised areas from the Mediterranean region (Barcelona and Tarragona), where higher levels of sulphate, and specially of nitrate, with respect other Spanish regions, are usually recorded. To attribute higher levels of inorganic secondary species at urban and kerbside sites with respect regional background areas, regional background sites for source apportionment studies should be located very close to the kerbside and urban background sites. In any case, bearing in mind the differences in the levels of secondary inorganic species, it may be concluded that the mass attributable to industrial inorganic secondary material may range from 3 to $8 \mu\text{gPM}_{10}/\text{m}^3$ and 1 to $6 \mu\text{gPM}_{2.5}$ in highly industrialised areas. The carbonaceous fraction is only slightly influenced by most of the industrial emissions, since at Madrid, Alcobendas and Canary islands the ratio organic and elemental carbon vs. inorganic secondary species ranges from 1.2 to 1.9 in PM_{10} and 1.3-2.2 in $\text{PM}_{2.5}$, whereas in the above industrialised urban sites this ratio is constantly maintained in the range 0.7-0.8 in PM_{10} and 0.7-1.1 in $\text{PM}_{2.5}$.

Source apportionment

Given all the above PM_{10} and $\text{PM}_{2.5}$ contributions reported for regional background, urban background and kerbside sites, Figures A5.2 and A5.3 summarise the source apportionment analysis for PM_{10} and $\text{PM}_{2.5}$ at different sites of Spain.

From these data it becomes evident that the natural contribution accounts for 15-20, 20-25 and 30-40% of PM_{10} ($7\text{--}9 \mu\text{g}/\text{m}^3$) measured at different kerbside sites, urban and regional background stations of the Iberian Peninsula, respectively (Figure A5.4). At the Canary Islands this natural fraction increases markedly up to 45-65% ($18\text{--}19 \mu\text{g}/\text{m}^3$, Figure A5.4) of the PM_{10} mass measured at urban and regional background sites. The natural input in $\text{PM}_{2.5}$ is markedly reduced down to a around 11-25% of the bulk $\text{PM}_{2.5}$ mass ($3\text{--}4 \mu\text{g}/\text{m}^3$), depending of the type of site.

As shown in Figure A5.4, a fraction of 30-50% of the PM_{10} (and higher for $\text{PM}_{2.5}$) at kerbside sites are attributable to traffic emissions, whereas this contribution reach only 20-35% in urban background stations. At the urban background sites the industrial contribution may be estimated in 12-35% of the PM_{10} levels (higher in $\text{PM}_{2.5}$) depending on the site of Spain.

As a result of the previously described PM compositional differences, the relative proportions of $\text{PM}_{10}/\text{PM}_{2.5}/\text{PM}_{1.0}$ measured at different Spanish stations vary widely. Thus, the proportion of the coarse fraction ($\text{PM}_{2.5-10}$) range from $> 55\%$ of the bulk PM_{10} mass in the Canary Islands, 41-47% in Southern and Eastern Spain, and 26-36% in Central and Northern Spain. This resulting in $\text{PM}_{2.5}/\text{PM}_{10}$ ratios ranging from 0.4 in the Canary Islands, 0.5-0.6 in most of the Iberian Peninsula, and 0.7-0.8 in the Central and Northern regions. Conversely, the ratio $\text{PM}_{1.0}/\text{PM}_{10}$ increase from 0.5-0.6 for the

Central and Northern Iberian Peninsula, 0.4 for the Southern and Eastern Regions down to 0.2 in the Canary Islands. Thus, the proportion of PM₁₀ mass made by <1µm particles in Northern Spain is increased by a 50% with respect the Southern and Eastern regions, and up to near 3 times with respect the Canary Islands.

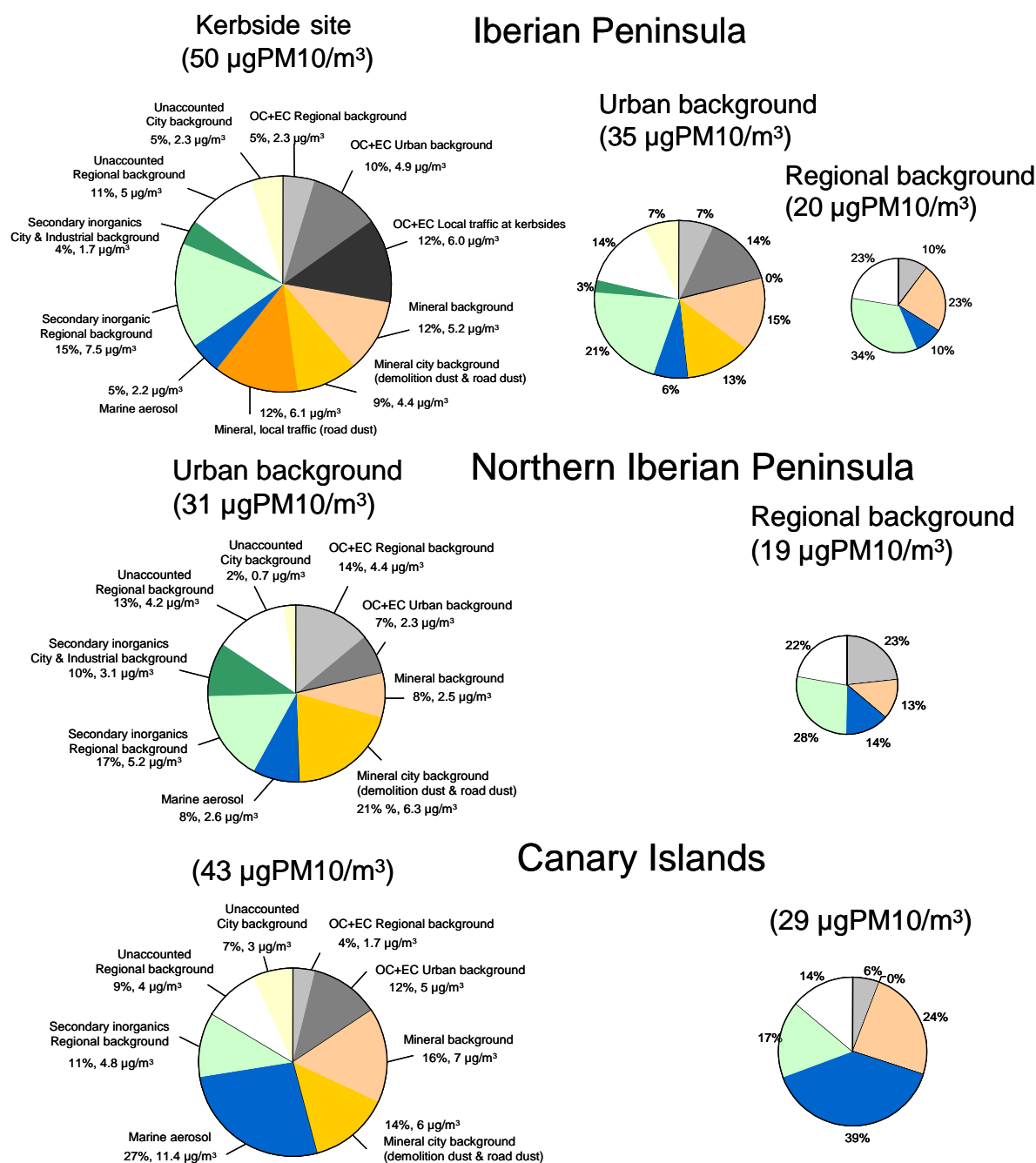


Figure A5.2 Results of the source apportionment analysis of PM₁₀ performed in regional background, urban background and kerbside sites from Eastern and Northern areas of the Iberian Peninsula and the Canary Islands in 1999-2001.

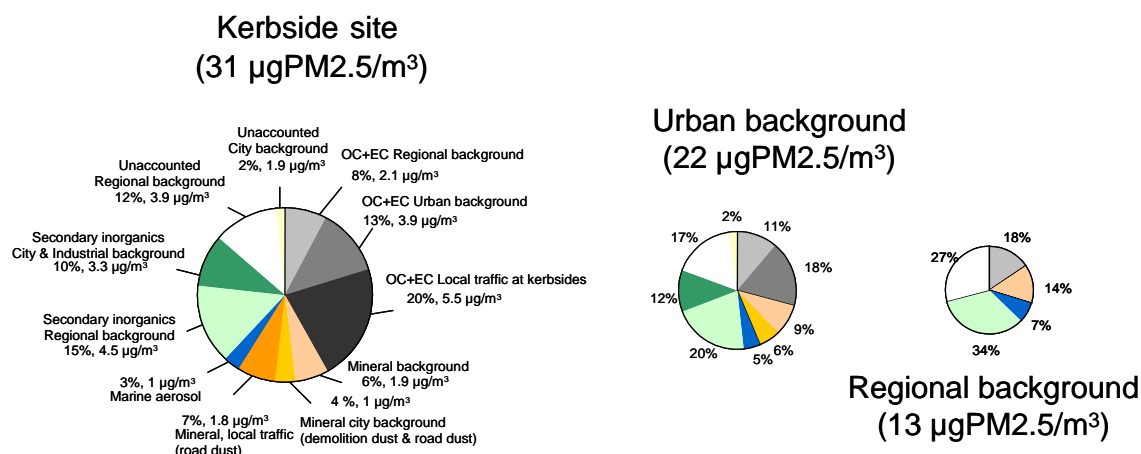


Figure A5.3 Results of the source apportionment analysis of $\text{PM}_{2.5}$ performed in regional background, urban background and kerbside sites from Spain in 1999-2001.

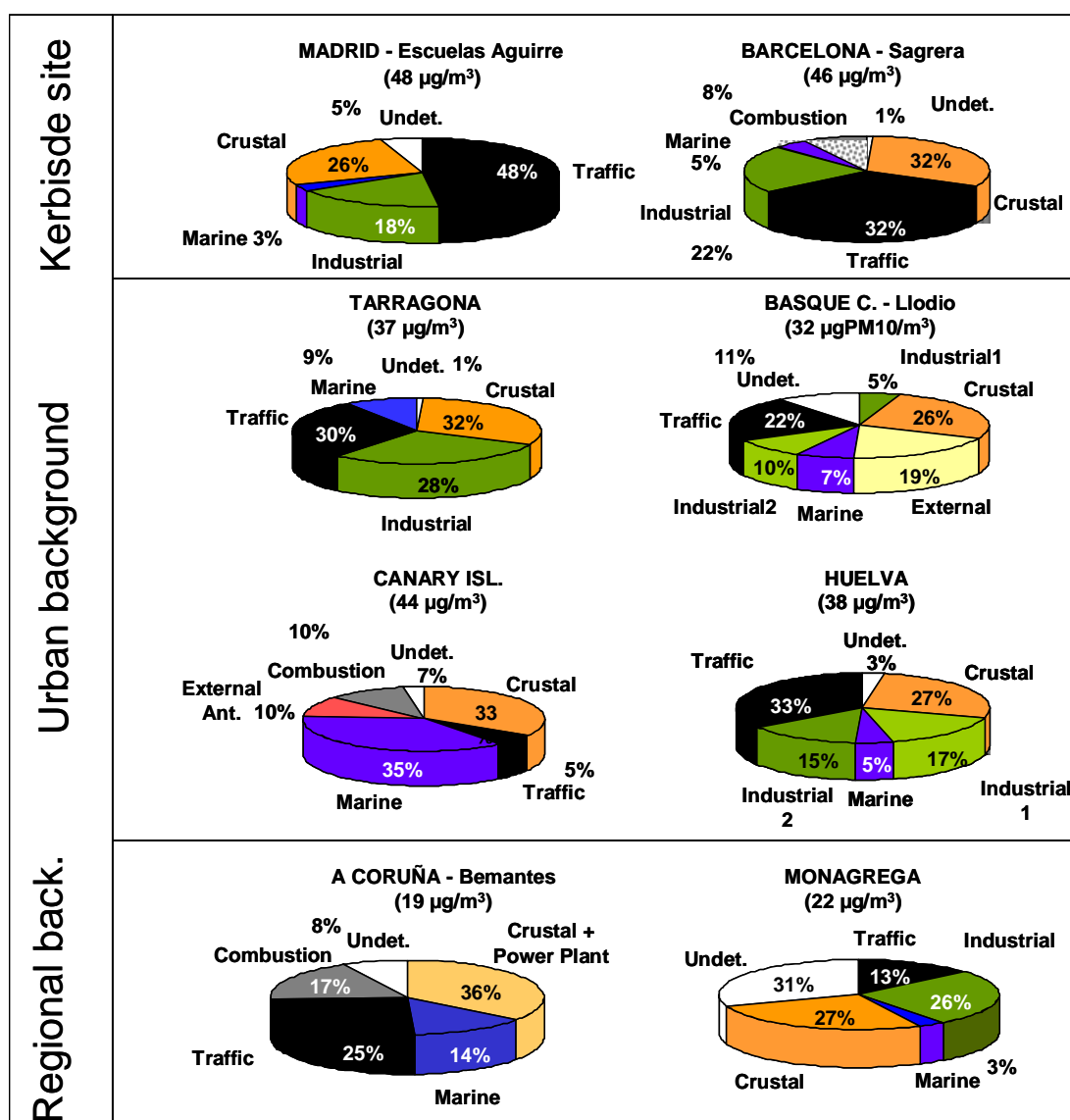


Figure A5.4 Results of the source apportionment analysis of PM_{10} performed in regional background, urban background and kerbside sites from Spain in 1999-2001.

Chemical speciation of the exceedances of the PM₁₀ 24-hour limit value

Figure A5.5 shows the number of exceedances (n) of the 24-hour PM₁₀ limit value (50 µg/m³) recorded in 2001 at 6 monitoring stations. As shown in this figure the exceedances recorded in days with African dust outbreaks may account for a high proportion of the total exceedances, and may be decisive in meeting the requirements of the First Daughter Directive for 2005 (n <35). Thus, the number of annual exceedances recorded simultaneously with these natural episodes range from 63 at traffic influenced sites from Las Palmas to 12 at the urban and background sites from Central and Northern Spain. Anthropogenic exceedances reached n= 9 at rural background sites from Northern Spain, 17 at the traffic influenced sites of Gran Canaria, 40 at urban background stations in Northern, Central and Southern Spain and 74 at a kerbside site in Barcelona.

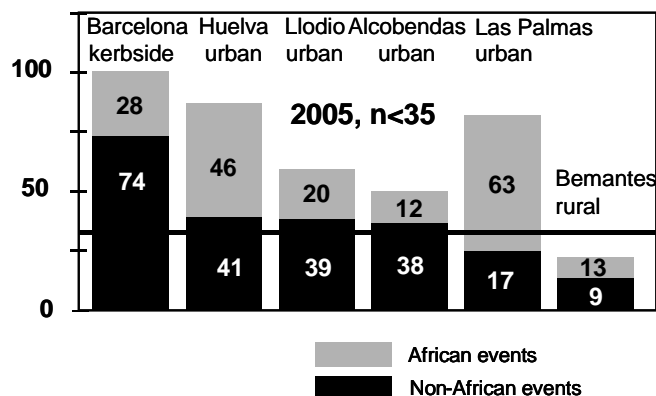


Figure A5.5 Number of exceedances of the 24-hour PM₁₀ limit value (50 µg/m³) recorded at the 6 monitoring stations in Spain 2001. The 24-hour exceedances occurred in days with African dust outbreaks are highlighted.

In addition to the African episodes, high PM₁₀ levels (18 to 25 µg/m³ as daily means in regional background and EMEP stations) are also recorded in the Mediterranean basin during the episodes defined as 'regional re-circulation events' (Millán et al., 1997 and Rodríguez et al., 2003). These episodes occur mainly in June to October under meteorological scenarios characterised by low pressure horizontal gradients over the Mediterranean. Under this scenario, the advection is inhibited and the atmospheric circulation is mainly controlled by local processes. The high insolation associated with these events favours the formation of secondary PM, as well as the turbulence and convective dynamics that cause the resuspension of soil particles from semiarid zones. Taking this into consideration, as well as the low air mass renovation, the persistence of these scenarios gives rise to an increase in PM₁₀ levels with a regional (both natural and anthropogenic) origin. In this context, these PM₁₀ events take place simultaneously with ozone episodes, and although the origin of the particles may be anthropogenic and/or natural, the causes for the atmospheric accumulation of PM have a natural origin related to the persistent re-circulation of the air masses. These events are usually limited in time by the development of advection episodes which renovate the air masses.

In order to determine the major causes of the exceedances of the PM₁₀ limit value (50 µgPM₁₀/m³), Figure A5.6 shows the mean levels of PM₁₀ components for days with PM₁₀ > 50 µg/m³ measured at Barcelona (kerbside site with industrial influence, data from 2001), Madrid (kerbside site, data from 1999-2000), Llodio (industrial urban background station, data from 2001) and Canary Islands (urban background station, data from 2001). The results show the prevalence of three PM₁₀ fractions at the different types of sites studied: mineral dust, secondary inorganic phases and carbonaceous components. At the Canary islands, the mineral fraction and the marine aerosol are clearly dominant during days exceeding the limit value, with mean levels of 36 µg/m³ and 20 µg/m³ (42 and 24% of the bulk PM₁₀ levels), respectively. The proximity of the African continent and the Atlantic island features accounted for the very high natural contribution measured during days exceeding the limit values in this site. At the industrial sites (Llodio and Barcelona) the fraction of secondary inorganic species accounts for 14-25 µg/m³ (22-41%) of the bulk PM₁₀ load for days exceeding the limit value. The high

traffic influence in Barcelona also accounts for a large contribution of mineral dust ($24 \mu\text{g}/\text{m}^3$, 38% of PM_{10} mass) and carbonaceous matter ($12 \mu\text{g}/\text{m}^3$, 19%) in days exceeding the limit value. Finally, it is the carbonaceous ($22 \mu\text{g}/\text{m}^3$) and mineral ($22 \mu\text{g}/\text{m}^3$) fractions that constitute 62% of PM_{10} in days exceeding the limit values at a kerbside site of Madrid, given that traffic is the main contributor to PM for those days. Sporadically, African dust outbreaks over Southern EU may induce very high PM_{10} and $\text{PM}_{2.5}$ levels. Up to 400 and $80 \mu\text{g}/\text{m}^3$ daily PM_{10} and $\text{PM}_{2.5}$ levels with >80% being mineral dust have been measured for some of those events over Spain.

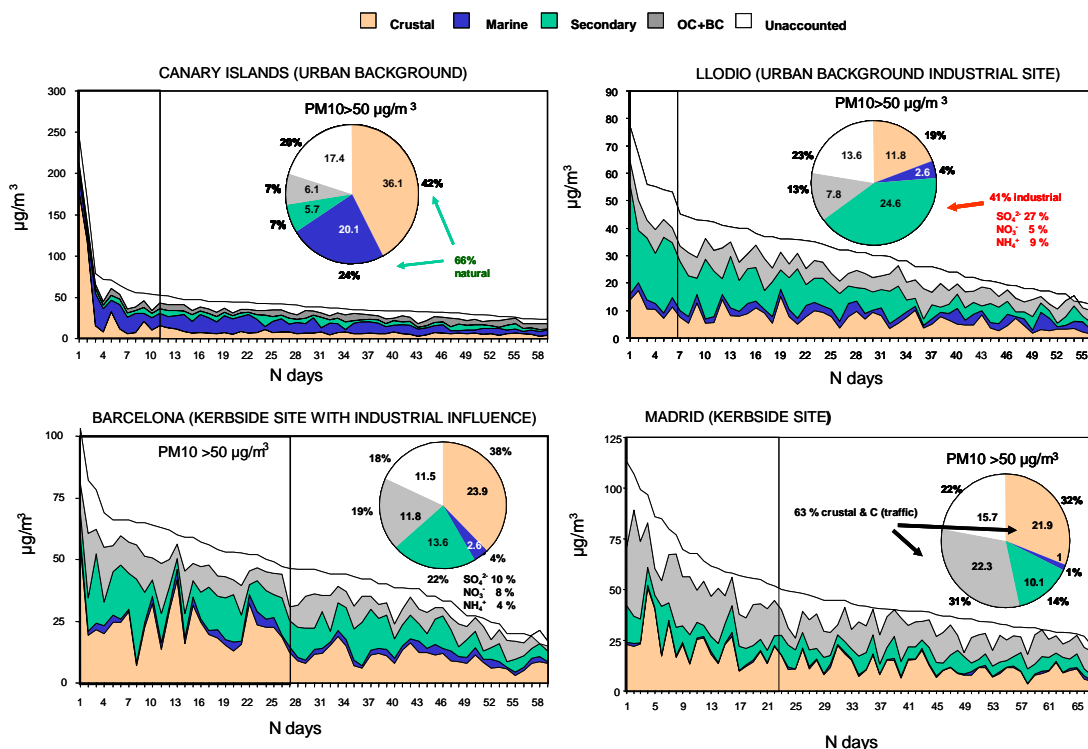


Figure A5.6 Chemical speciation of days exceeding the PM_{10} 24-hour limit value ($50 \mu\text{g}/\text{m}^3$) at urban background, kerbside and industrial sites.

A5.3 Sweden

The $\text{PM}_{2.5}$ and PM_{10} data evaluated here was obtained from 13 Swedish sites operated from September 1, 1999 to August 31, 2001 in Sweden. The measurement sites were selected to be representative for the major cities in Sweden, for areas in cities where biomass combustion is one of the major domestic heating procedures and for background rural areas without important local sources of PM emissions. Table A5.3 summarises the PM_{10} and $\text{PM}_{2.5}$ data reported by Areskoug et al. (2003).

The results show two important factors with special incidence on PM concentrations, long distance transport of particles and resuspension of road dust. Strong low level inversion events, especially in the inland of Northern Sweden during lock out the long distant transport, may also induce sporadically very high PM concentrations, especially in winter.

As shown in Table A5.3, PM_{10} annual levels in rural background areas of Sweden are close to $16 \mu\text{g}/\text{m}^3$. These concentrations rise to 17 – 24 and 25 – $50 \mu\text{g}/\text{m}^3$ for urban background and kerbside sites, respectively. Consequently, in Sweden the regional background levels account on the average for 80 and 50% of the PM_{10} levels recorded at urban background and kerbside sites, respectively. In other words at a kerbside site the regional background, the city background and the local traffic contributions account for 40, 10 and 50% of the PM_{10} levels. In most of the other EU examples these proportions reach around 40, 30 and 30%, with the exception of the Netherlands (70, 10, 20%).

Levels of $PM_{2.5}$ in rural background areas of Sweden are close to $12 \mu\text{g}/\text{m}^3$. These concentrations rise to 13 – 16 and 13 – 18 $\mu\text{g}/\text{m}^3$ for urban background and kerbside sites, respectively. The $PM_{2.5}$ levels recorded at urban and traffic oriented sites are markedly lower (by a factor of 1.3 to 2) than the other EU examples given here. Consequently, the regional background levels are in Sweden 75-90 and 65-90% of the $PM_{2.5}$ levels recorded at urban background and kerbside sites, respectively.

The ratios $PM_{2.5}/PM_{10}$ measured at rural background, urban background and kerbside sites are 0.8, 0.6–0.7 and 0.4–0.6, respectively. The strongly decreasing ratio implies the strength of road dust emissions and other mechanical wearing processes giving coarse particle emissions.

Local sources are effectively revealed by the diurnal variations at different types of sites. The sites have been classified as rural, urban background and kerbside sites. The rural sites are in forested areas with very limited traffic, several km from larger roads. Urban background stations are placed on roofs, parks or calm residential areas.

Figure A5.7 shows that daytime hourly PM_{10} levels are markedly higher, by a factor 2.5, at kerbside sites and by only 1.5 at urban background sites, when compared with rural concentrations. At nighttimes the city concentrations seems to be fully in line with rural background concentrations.

Daytime hourly $PM_{2.5}$ levels show a lot less pronounced increases at kerbside and urban background sites, by factors of 1.6 and 1.3 respectively. This is quite striking and shows a strong influence of sources contributing to the PM_{10} but not to $PM_{2.5}$.

Using NO_x as a tracer for traffic intensity, the emission factors for PM_{10} and $PM_{2.5}$ were calculated. As shown in Figure A5.8, the calculated total emission factors over 2 years at a kerbside site show large variations. When using a common exhaust emission factor of 25 mg/veh.km the non-exhaust fraction an average of 45% and 95% of the total emissions factors for $PM_{2.5}$ and PM_{10} are estimated, respectively. In total, the non-exhaust emission factor for PM_{10} is 9 times larger than the PM_{10} exhaust emission factor but for $PM_{2.5}$ the non-exhaust emission factor is equal to the exhaust emission factor. Consequently traffic non-exhaust emission strongly contributes to $PM_{2.5}$ and totally dominates the PM_{10} at a kerbside station in Sweden. This is most likely valid for all the Nordic countries, though it depends on use of road salt and sand together with tyre abrasion products. However, during the summer months the contribution to PM_{10} is still very high.

Table A5.3 Statistical summary of 24-hour average concentrations of PM_{10} and $PM_{2.5}$ at thirteen sites in Sweden from September 1, 1999 to August 31, 2001. n = Number of days with data (i.e. 18 or more validated one hour averages).

	PM_{10} ($\mu\text{g}/\text{m}^3$)				$PM_{2.5}$ ($\mu\text{g}/\text{m}^3$)			
	n	Mean	90 perc	98 perc	n	Mean	90 perc	98 perc
Rural								
Aspvreten	615	15,9	23,2	34,5	628	12,5	18,4	26,6
Vavihill	455	16,4	25,8	44,6	450	12,5	20,2	38,2
Urban background								
Lycksele	523	16,9	29,5	67,0	535	11,8	18,9	32,9
Umeå, Town Library	655	14,7	24,2	39,5	656	8,4	12,9	18,1
Stockholm, Rosenlund	703	18,6	30,8	43,5	663	11,9	18,7	29,2
Göteborg, Femman	472	20,1	33,9	49,4	406	12,2	21,1	34,3
Växjö	566	17,2	29,1	47,3	572	11,9	21,1	33,7
Malmö, Lernacken	630	20,0	30,2	46,5	625	13,1	22,8	37,1
Malmö, Town Hall	647	22,8	35,5	53,4	567	14,8	24,6	39,5
Kerb side								
Umeå, Storgatan	303	25,0	40,1	84,7	304	13,4	21,9	27,0
Stockholm	722	51,0	116,2	183,2	722	18,5	28,2	38,8
Göteborg, Gårda	497	33,7	58,1	104,0	497	16,9	25,3	38,4
Göteborg, Järntorget	177	35,8	56,3	85,7	173	15,6	22,7	32,8
Malmö, Mobile station	641	22,7	34,4	52,8	639	13,5	22,6	38,8

The meteorology determines the dilution of the emissions and thus is strongly affecting observed concentrations. In northern Sweden, with a snow cover and quite cold temperatures, ground inversions can be frequent. In Figure A5.9 the $PM_{2.5}$ and PM_{10} concentrations are shown for Lycksele, a small town with about 9000 inhabitants in the northern inland of Sweden. Very high concentrations are measured when the temperature is falling, which is indicative of a thermal inversion period. In spite of low emissions, the number of limit value exceedances depends on the number of inversion events.

Overall estimate

The collected information on the different sources allows an estimate of mean annual $PM_{2.5}$ and PM_{10} concentrations in different environments all over Sweden (see Table A5.4). Background, i.e. influence of long distant transport decrease roughly with a factor of 2 comparing northern and southern areas. Subtracting the background from the concentrations found and using the above found relation between the coarse and fine emissions, it is possible to assess the contribution from resuspension in urban environment where traffic is the main local source. It should be noted that Table A5.4 only reports annual mean concentrations, thus concentrations at occasions from the different sources may undergo a wider variation.

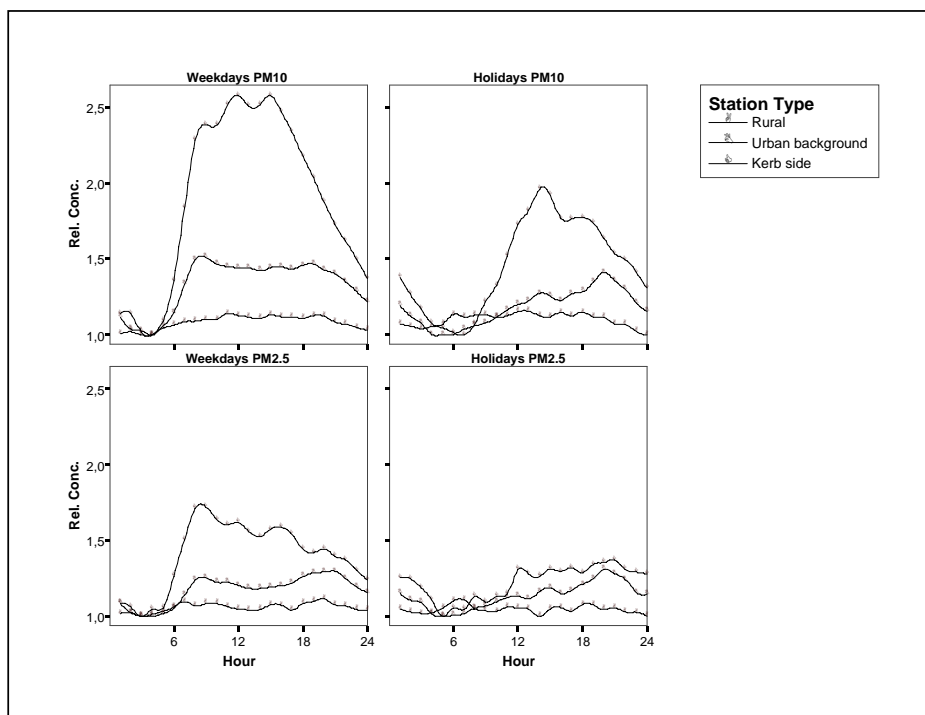


Figure A5.7 Hourly PM_{10} and $PM_{2.5}$ levels measured at rural and urban background stations and kerbside sites from Sweden during weekdays and holidays.

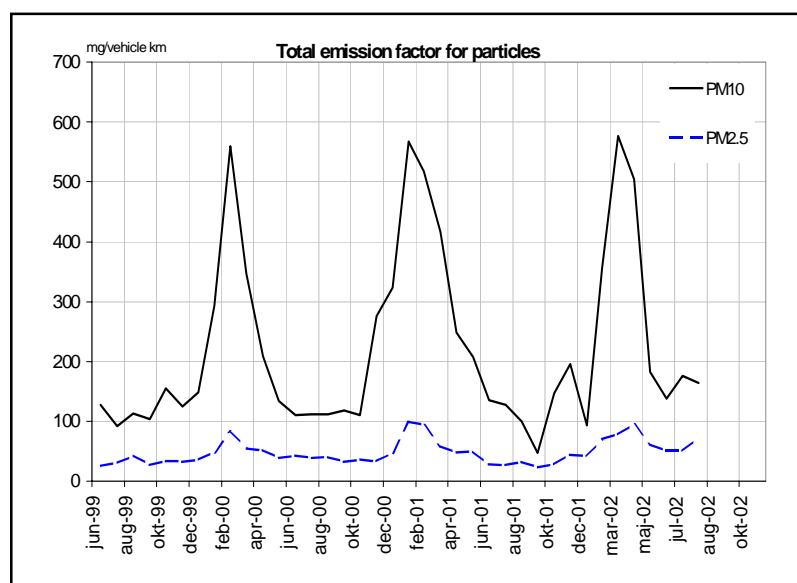


Figure A5.8. Total traffic emissions for PM_{10} and $PM_{2.5}$ estimations (mg/vehicle.km) from June 1999 to October 2002.

Other sources included both exhaust from traffic and heating (which in the north could be an important source as wood fires are used).

Road dust emerges as the major local source responsible for exceedance of limit values in Sweden. However it is superimposed on strongly enhanced background concentrations due to long distance transport.

Under thermal inversion scenarios, the local sources will totally dominate the local PM levels and very high PM concentrations may be reached due to the impact of small local sources. Basically the meteorology determines the concentrations. The number of exceedances of the 24-hour PM_{10} limit

value in Lycksele, a small town situated in the inland of northern Sweden, the region where inversions are most frequent, will give an idea of the importance of this phenomenon. During the two year period, 10 days with higher concentrations than $50 \mu\text{g}/\text{m}^3$ were detected in Lycksele. It is likely that a major part of the inland is affected by such events. Consequently local inversion and then local sources can have considerable effects in spite of small emissions.

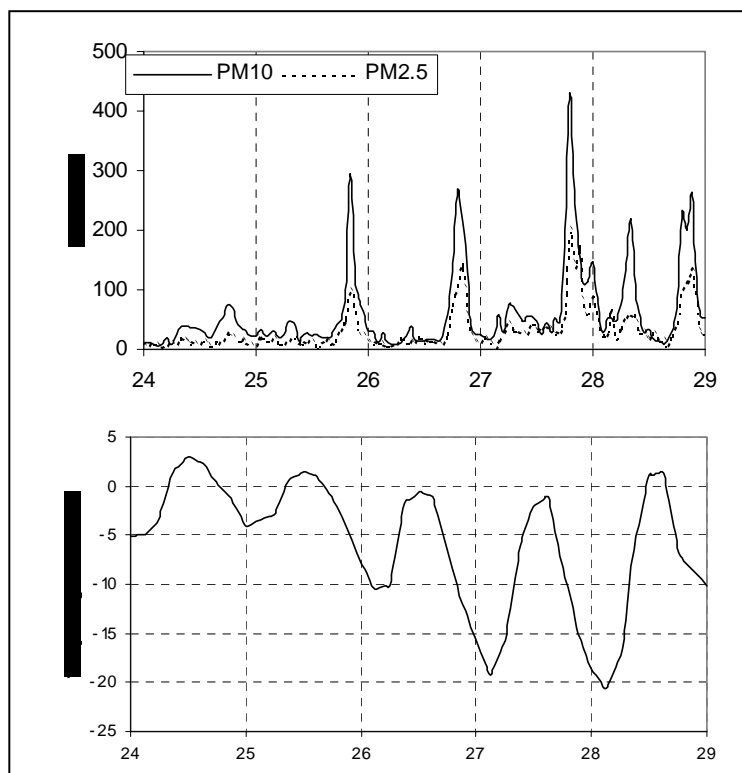


Figure A5.9 Hourly $\text{PM}_{2.5}$ and PM_{10} concentrations recorded in January 24th to 29th, 2000 at Lycksele, a small town with about 9000 inhabitants in the northern inland of Sweden, showing peak events with temperature falls.

Table A5.4 Estimates of annual mean source contribution at different type of sites ($\mu\text{g}/\text{m}^3$). The range given for the long range contribution depends on location in Sweden, with the lowest values in the northern regions.

Source Site	Long range	Resuspension	Other local sources	Sum
Rural				
PM_{10}	8 – 17	-	-	8 – 17
$\text{PM}_{2.5}$	7 – 13	-	-	7 – 13
Urban background				
PM_{10}	8 – 17	5	1	14 – 23
$\text{PM}_{2.5}$	7 – 13	1	1	9 – 15
Kerbside				
PM_{10}	8 – 17	23	4	23 – 51
$\text{PM}_{2.5}$	7 – 13	3	4	14 – 21

A5.4 Netherlands

In the Netherlands Aerosol Programme (NAP, Buringh et al., 2002), the ambient PM concentrations in the Netherlands have been the subject of research. Important in this respect were measurements and model calculations for PM_{10} and $\text{PM}_{2.5}$ at rural, urban and industrial locations during a one-year period

in 1998-1999 (Visser et al., 2001). Relevant emission sources were identified using source receptor modelling in combination with dispersion modelling and national/European emission inventories for PM and the precursor gases SO₂ and NO_x for Secondary Inorganic Aerosol (SIA). In addition, special studies were directed to estimate contributions to PM concentrations in the Netherlands by emission sources outside Europe (Weijers et al., 2000), wind-blown soil particles and resuspension of road-dust (Keuken et al., 1999) and background information on EC/OC and ultra fine particles (Ten Brink and Keuken, 2000). Part of the research in the framework of NAP is still ongoing.

The average results of PM research in the Netherlands during the period August 1998 to September 1999 at three spatial scales (street, urban background and rural background) and six locations have been presented by Visser et al. (2001). From this project and the previous references an estimate for the annual average of PM₁₀ in the Netherlands has been compiled (Table A5.5). A default value of 10% for unknown, not measured components and/or water has been assumed.

The range of 27 – 42 µg/m³ in annual average PM₁₀ reflects the uncertainties and spatial variation in the estimated contribution by various sources to PM₁₀ in the Netherlands.

Table A5.5 Estimate of the annual average PM₁₀ contribution of various sources to PM levels in the Netherlands at the end of the nineties. Natural secondary organic aerosol is not included as it represents only a small fraction (<< 10%) of the anthropogenic contribution

	PM ₁₀ µg/m ³	Source
Primary natural		
Sea salt	4-7	Visser et al., 2001
Crustal material	2	Visser et al., 2001
<i>Subtotal natural</i>	6-9	
Primary anthropogenic		
EU and NL contribution (non-traffic)	4-7	Visser et al., 2001)
Traffic (EU and NL)	2-5	Visser et al., 2001
Netherlands road dust	1-2	Keuken et al., 1999
<i>Subtotal primary anthropogenic</i>	7-14	
Secondary Inorganic Aerosol (SIA)		
European contribution	7-9	Visser et al., 2001
Dutch contribution	3-5	Visser et al., 2001
<i>Subtotal SIA</i>	7-14	
Other		
Northern Hemisphere background	1	Weijers et al., 2000
Unknown 10%	3-4	Default
<i>Subtotal Other</i>	4-5	
Total PM₁₀	27-42	

The emission database for PM contains information on precursor gases for SIA and the primary Dutch and EU contribution of other sources including tailpipe emissions from traffic. Therefore these fractions can be modelled. According to Table A5.5 this fraction had a spatial variability of 16 – 25 µg/m³ on an annual average basis. The emission database does not contain information on natural contributions as sea salt, plant material and crustal material. Neither does it contain information on the Northern Hemisphere background and Secondary Organic Aerosols (SOA). Therefore the contribution to the annual average of these fractions cannot be modelled in dispersion models. This non-modelled fraction accounts for approximately 18 µg/m³ in the Netherlands.

Measurements in the air quality monitoring network for 1998 and 1999 resulted in annual PM₁₀ levels in the non-urban (regional) parts of the Netherlands of respectively 33 and 32 µg/m³ (Hammingh, 2001). In the large cities assessed urban background concentrations still were slightly higher than 40 µg/m³ in the nineties.

For PM monitoring the National Air Quality Monitoring Network (NAQMN) considers the following spatial scales: regional, urban background (number of passing vehicles within a radius of 35 m from the station <2750 per 24 hours) and street canyon (the number of passing vehicles within a radius of 35 m from the station > 10,000 per 24 hours).

Table A5.6 Yearly average PM₁₀ levels in µg/m³ measured in the National Air Quality Monitoring Network of the Netherlands

The Netherlands									
Year	1993	1994	1995	1996	1997	1998	1999	2000	2001
Regional stations									
131 Vredepeel	50	43	38	43	40	37	35	35	36
133 Wijnandsrade	39	37	42	39	38	34	29	30	31
230 BiestHoutakker	46	48	48	47	45	40	37	34	36
318 Braakman	44	41	42	46	45	42	33	34	33
437 Westmaas	42	36	39	42	41	37	35	34	32
444 DeZilk		31	37	41	38	34	30	31	29
538 Wieringerwerf	32	34	34	43	33	30	33	31	25
722 Eibergen	36	37	37	44	40	33	29	28	28
724 Wageningen	43	42	39	43	44	36	34	33	32
928 Witteveen	36	33	32	35	32	27	27		
929 Valthermond								29	30
Urban background station									
404 Den Haag	41	41	42	47	41	44	43	34	36
418 Rotterdam		41	42	47	42	42	38	36	37
441 Dordrecht		37	40	43	39	33	32	32	29
520 Amsterdam	41	37	41	49	39	38	35	33	29
Street station									
236 Eindhoven	41	42	45	49	42	39	36	35	35
433 Vlaardingen	40	39	41	49	43	39	36	32	32
641 Breukelen		42	41	46	43	38	40	37	31
639 Utrecht	42	40	47	54	49	36	37	35	37
728 Apeldoorn	39	39	42	46	42	36	34	33	37

Large differences were found compared with results in other Member States, which is related to the similarity of levels measured at regional, urban and roadside sites in the Netherlands, with ranges of 25-36, 39-37 and 31-37 µg PM₁₀/m³, respectively, in 2001 (Table A5.6). The ratios PM_{2.5}/PM₁₀ were also rather similar in all types of site (from regional to kerbside: 0.6-0.7). It is also important to notice that, as shown by Figure A5.10, there is a fraction of around 18 µg/m³ of PM₁₀ in all sites which cannot be influenced by measures within the Netherlands.

Another important difference becomes evident when comparing the contributions of carbonaceous particles and mineral matter at the regional and city background and local traffic inputs in the roadside sites in the Netherlands with those obtained elsewhere. According to Visser et al. (2001), the levels of carbonaceous (OC+EC) particulate matter at kerbside sites in the Netherlands are estimated at around 6-7 µgOC+EC/m³ and around 13-18 µgOC+EC/m³ at kerbside sites from Germany, Spain and the United Kingdom. Similarly to the United Kingdom, the levels of mineral particulate matter reach 6-7 µg/m³ at kerbside sites in the Netherlands, much lower values than in Spain and Berlin: 12-15 µg/m³. However, it has to be pointed out that the above levels of mineral elements in Berlin must be lower given that unaccounted mass (mostly water) was also included when this fraction was estimated. The Dutch levels of secondary inorganic particulate matter are in the usual range found in other EU studies (8-12 µg/m³). The levels of the marine aerosol contribution measured ranged from 4 to 7 µg/m³, twice the values measured in Berlin, the United Kingdom and Spain. These peculiarities described for PM₁₀

measured in the Netherlands may account for the lower differences between the levels of PM₁₀ at regional, urban and roadside sites. However, it may be also possible that the criteria to define a roadside site station may be different in the Netherlands than in the above countries, given that the levels and composition of the Dutch roadside sites fall in the usual ranges defined for urban background stations in the other examples described in this report.

As for Spain and Berlin, in rural areas distant from industrial areas, the levels of secondary inorganic phases in PM₁₀ are around 4-5 µg/m³ lower than in urban areas with industrial influence.

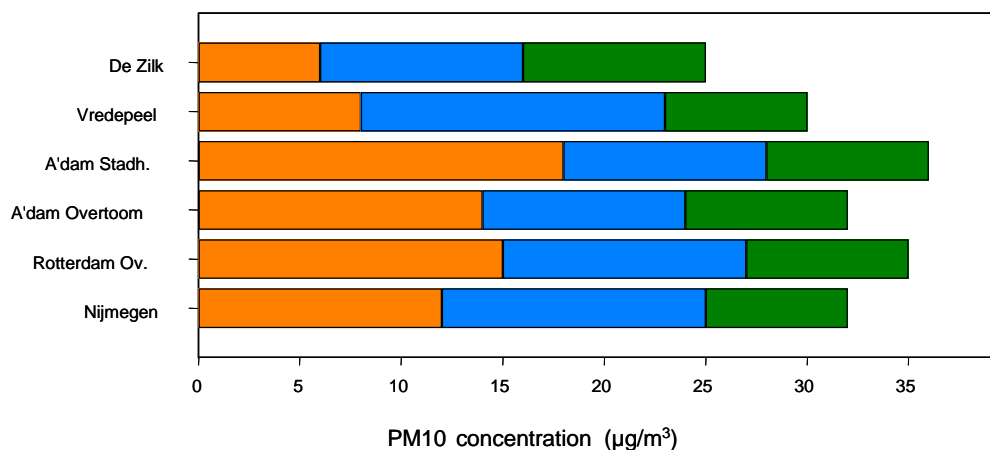


Figure A5.10 PM₁₀ concentrations at six sampling sites, divided into (i) the maximal part which can be influenced by measures within the Netherlands (orange), (ii) the maximal part which can be influenced by measures outside the Netherlands (blue), and (iii) the contribution of natural sources (green). Data are taken from Visser et al. (2001).

Results from modelling based on the PM₁₀ emission databases from anthropogenic sources and precursor gases of secondary inorganic aerosols in the Netherlands and EU, account for only around half of the yearly average PM₁₀ levels. The other half (18 µg/m³) is composed of PM that is not contained in the emission databases. Part of it is probably sea salt (4 to 7 µg/m³) and (anthropogenic) crustal or natural material (3 - 4 µg/m³). Approximately 1 µg/m³ is a non-modelled contribution from the northern hemisphere. From measurements a reliable non-modelled value for PM_{2.5} could not yet be drawn up. Table A5.7 summarises the source apportionment of this modelled concentration for 1995, with only 50% of the measured PM₁₀ levels accounted for.

The modelled contribution for emissions from the Netherlands was 5.5 µg/m³ and that of foreign countries 10.9 µg/m³. Transport (including shipping) contributes the major part (5.9 µg/m³), of which 2.6 µg/m³ is the result of emissions in the Netherlands. It is estimated that 6 µg/m³ of the anthropogenic PM is from primary emissions, whereas 10.5 µg/m³ are secondary inorganic aerosols.

Table A5.7 Annually averaged primary and secondary inorganic concentrations of PM₁₀ averaged over the Netherlands by anthropogenic source (µg/m³). Calculated for the year 1995, based on emissions for the Netherlands and the CEPMEIP inventory for European countries

	Primary PM ₁₀	NH _x	NO _y	SO _x	Total
Dutch sources					
Industry	0.4	0.0	0.1	0.1	0.6
Energy	0.0	0.0	0.1	0.1	0.2
Transport ¹⁾	1.5	0.0	1.0	0.1	2.6
Agriculture	0.5	0.9	0.0	0.0	1.4
Others	0.5	0.1	0.1	0.0	0.7
Sum	2.9	1.0	1.4	0.2	5.5
Other countries					
Industry	0.9	0.0	0.1	0.1	1.0
Energy	0.4	0.0	0.7	1.9	3.0
Transport ¹⁾	0.9	0.0	2.1	0.3	3.3
Agriculture	0.1	1.2	0.0	0.0	1.4
Others	0.7	0.0	0.5	1.1	2.3
Sum	3.0	1.2	3.3	3.4	10.9
All sources					
Sum	6.0	2.2	4.6	3.6	16.5
Measured PM₁₀					36

¹⁾ Including international shipping

A5.5 United Kingdom

Intensive research on the chemical characterisation of PM₁₀ has been summarised by APEG (1999). The data on the United Kingdom discussed here (Table A5.8) was supplied by Harrison et al. (2003). These are the mean values of samples collected at four roadside sites and four urban background sites. In both cases, three sites were in London and one site in Birmingham. The data are presented as follows:

- Ammonium sulphate calculated from the measured sulphate concentration.
- Ammonium nitrate calculated from the measured nitrate concentration.
- Elemental carbon determined by combustion.
- Organic carbon determined by combustion. This is multiplied by a factor of 1.6 at the background site and 1.4 at the roadside sites.
- Resuspended particles estimated from the measured iron and calcium concentrations.
- Sodium chloride estimated from the measured chloride concentration.
- The small mass of unaccounted material which is described as unmeasured.

From this data, the following conclusions on the composition of PM₁₀ and PM_{2.5} from urban background and roadside sites from the United Kingdom can be drawn:

- The marine contribution is estimated at around 2 µg/m³ in PM₁₀ (8 and 6% in urban background and roadside sites) and only 0.3 µg/m³ in the PM_{2.5} fraction (1-2% of PM_{2.5} mass). These values are very similar to those reported for Berlin and Spain, and lower than the Dutch levels.

- Road dust levels reach 5.5 and 6.5 $\mu\text{g}/\text{m}^3$ in PM_{10} (22 and 18% at urban background and roadside sites) and only 1.6 $\mu\text{g}/\text{m}^3$ in the $\text{PM}_{2.5}$ fraction (11-6% of $\text{PM}_{2.5}$ mass). These values are relatively low when compared with data obtained from Spain, and similar to the data reported for the Netherlands.
- The levels of carbonaceous components reach 9.3 and 18.1 $\mu\text{g}/\text{m}^3$ in PM_{10} (38 and 50% at urban background and roadside sites). These values are in the range of data reported for Berlin and Spain, and higher than the values reported for the Netherlands.
- Organic C predominates over elemental C at urban background sites (26 versus 12% in PM_{10}) whereas in roadside sites similar levels are determined for both type of carbonaceous (25 versus 25%) in PM_{10} .
- The carbonaceous components are mostly present in $\text{PM}_{2.5}$. Thus, around 90 and 80% of the OC and EC mass, respectively, are present in fine fractions at both background and roadside sites. Around 50 and 65% of the $\text{PM}_{2.5}$ mass at urban background and roadside sites is made up by carbonaceous compounds.
- Levels of secondary inorganic are close to 8 $\mu\text{g}/\text{m}^3$ in both urban background and roadside sites, accounting for 32 and 23% of the PM_{10} mass, respectively, in the range of levels found for the other EU examples reported here.
- The ratio ammonium sulphate/ammonium nitrate in PM_{10} is also constantly 1:1 at both type of sites, and around 30% of nitrate in PM_{10} is present in the coarse mode ($\text{PM}_{2.5-10}$, probably as sodium nitrate), whereas this proportion is around 12% in the case of ammonium sulphate.

Table A5.8 Mean composition of PM_{10} , $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ at 4 urban background and 4 roadside monitoring stations in the UK (6 in London and 2 in Birmingham). Source: Harrison, 2003.

Urban background (calculated concentration)									
In $\mu\text{g}/\text{m}^3$									
	Ammonium sulphate	Ammonium nitrate	Secondary inorg.	Elemental carbon	Organic matter ¹⁾	Resuspended	Sodium chloride	Unmeasured	Total
$\text{PM}_{2.5}$	3.41	2.65	6.06	2.55	4.90	1.77	0.32	0.13	15.73
$\text{PM}_{2.5-10}$	0.53	1.21	1.74	0.36	1.51	3.64	1.61	0.06	8.92
PM_{10}	3.94	3.85	7.79	2.91	6.41	5.42	1.93	0.19	24.65
¹⁾ Using factor $\text{OC} \times 1.6$									
Percentage									
$\text{PM}_{2.5}$	22	17	39	16	31	11	2	1	
$\text{PM}_{2.5-10}$	6	14	19	4	17	41	18	1	
PM_{10}	16	16	32	12	26	22	8	1	

Urban roadside (calculated concentration)									
In $\mu\text{g}/\text{m}^3$									
	Ammonium sulphate	Ammonium nitrate	Secondary inorg.	Elemental carbon	Organic matter ²⁾	Resuspended	Sodium chloride	Unmeasured	Total
$\text{PM}_{2.5}$	3.43	2.71	6.14	8.42	7.41	1.53	0.34	0.20	24.03
$\text{PM}_{2.5-10}$	0.59	1.29	1.88	0.64	1.60	4.93	1.65	0.90	11.60
PM_{10}	4.02	4.00	8.02	9.06	9.01	6.46	2.00	1.09	35.63
²⁾ Using factor $\text{OC} \times 1.4$									
Percentage									
$\text{PM}_{2.5}$	14	11	26	35	31	6	1	1	
$\text{PM}_{2.5-10}$	5	11	16	6	14	43	14	8	
PM_{10}	11	11	23	25	25	18	6	3	

When subtracting the urban background to the roadside site levels for each component (Figure A5.11), it becomes evident that around 8 (35%) and 11 $\mu\text{g}/\text{m}^3$ (30%) of $\text{PM}_{2.5}$ and PM_{10} are from local roadside emissions. The remaining 65-70% arise from the city background input including the regional PM contribution. These values are in the same range that those obtained in the other EU examples

reported here, with the exception of the Netherlands, where the local contribution from the monitored street in a roadside site is lower (by a factor of $\frac{1}{2}$).

In the case of $PM_{2.5}$ this local road contribution is totally made of carbonaceous compounds, mainly from vehicle exhaust. For PM_{10} the carbonaceous fraction accounts for around 80% of the local road site contribution, with an additional 12% of road dust and around 8% of unmeasured components + secondary inorganic phases. These contributions to PM_{10} are different from the Spanish and German data where the carbonaceous and the mineral PM_{10} contributions to the local road input are around 1:1.

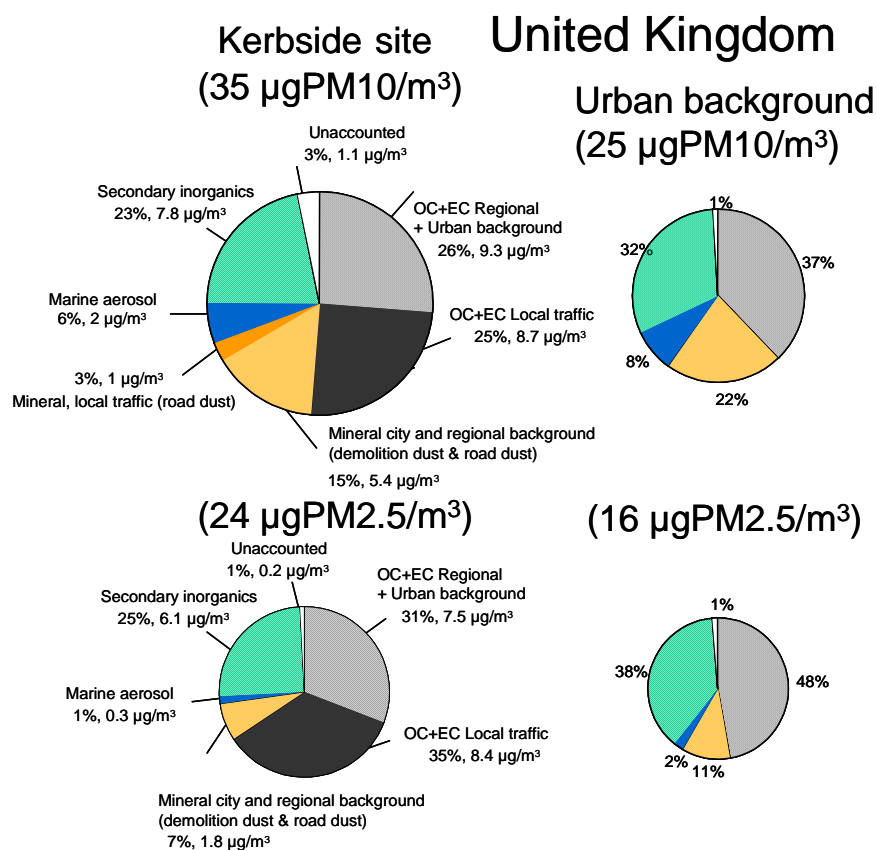


Figure A5.11 Results of the source apportionment analysis of PM_{10} and $PM_{2.5}$ performed at urban background and kerbside sites from UK. Based on data from Table A5.8.

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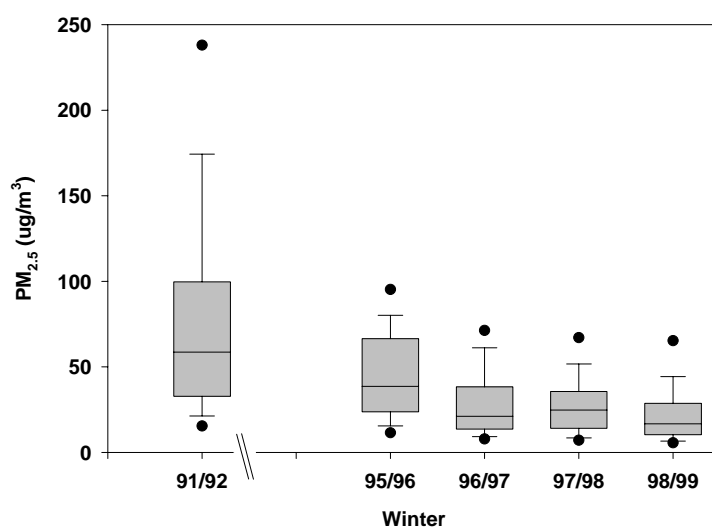
Annex 6 Annex to Chapter 7 – Trend data from individual countries

This annex to Chapter 7 shows further data provided by some countries and extracted from the EMEP database, provide additional examples of trends for PM₁₀ concentrations in Europe.

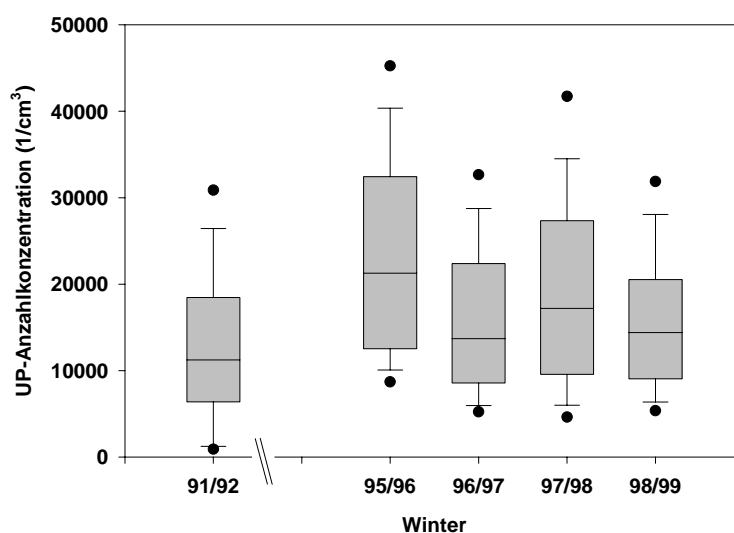
Erfurt, Germany

In Erfurt, Germany (see Figure A6.1), PM_{2.5} concentrations have decreased substantially since 1991/92 (winter averages). Ultrafine particles show no trend.

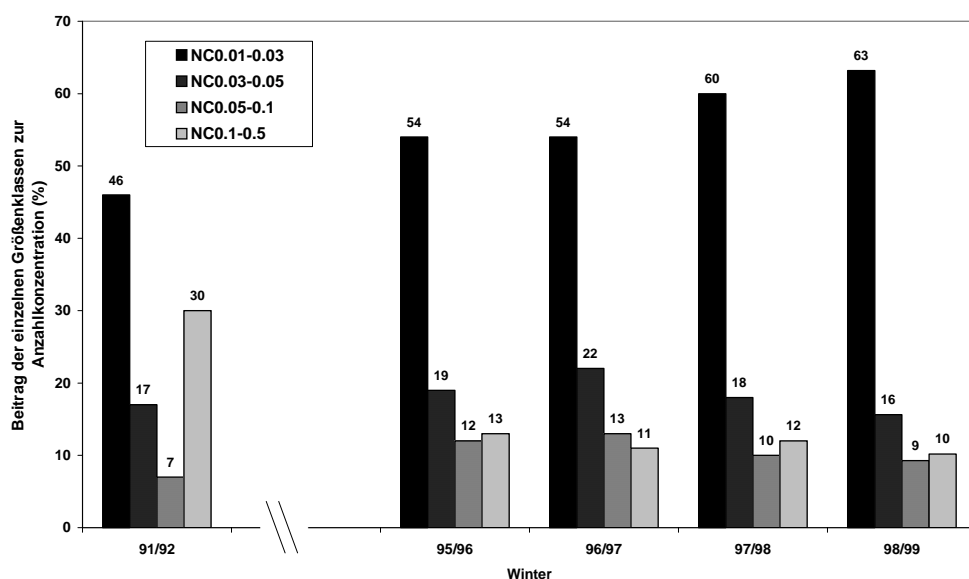
Figure A6.1 PM trends in Erfurt.



a) Trend in PM_{2.5} in Erfurt, winter 1991/92 to 1998/99. Box-Whisker plots (mean, 25/75-percentiles, 10/90-percentiles, min/max value)



b) Trend in ultrafine PM concentration (UP-Anzahlkonzentration) in Erfurt, winter 1991/92 to 1998/99



c) 7-year trend in relative number concentration (in %) – for various size classes (0.01-0.03, 0.03-0.05, 0.05-0.1, 0.1-0.5 µm) in Erfurt, winter 1991/91 to 1998/99. The number in the smallest size class increases steadily. Vertical axis: Contribution of the size classes to the total number concentration (%).

Germany, EMEP stations

PM₁₀ at EMEP stations at rural sites in Germany (Figure A6.2) show a substantially decreasing trend at all stations included, since 1977. Concentrations are now between 10 and 20 µg/m³ (annual average), while they were before 1980 20-70 µg/m³. Note that before 1999, it was TSP that was measured at these stations, and PM₁₀ has been calculated from TSP, using a constant factor of 0.85.

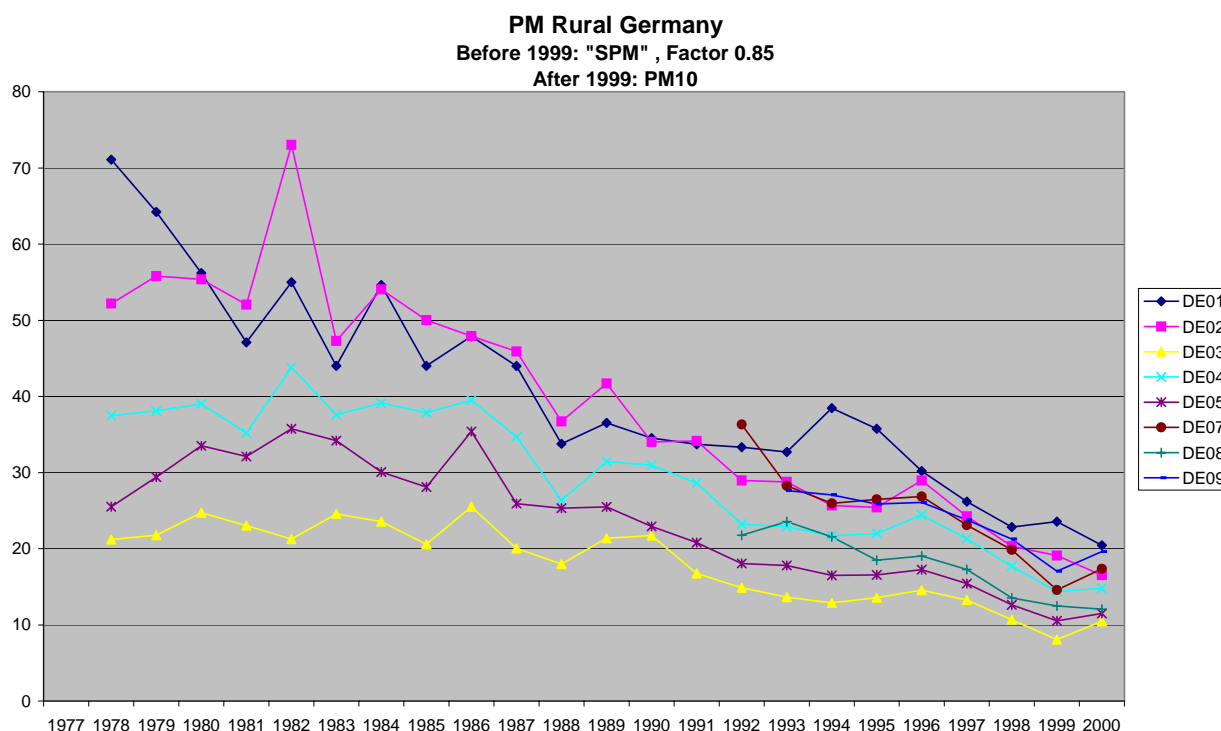
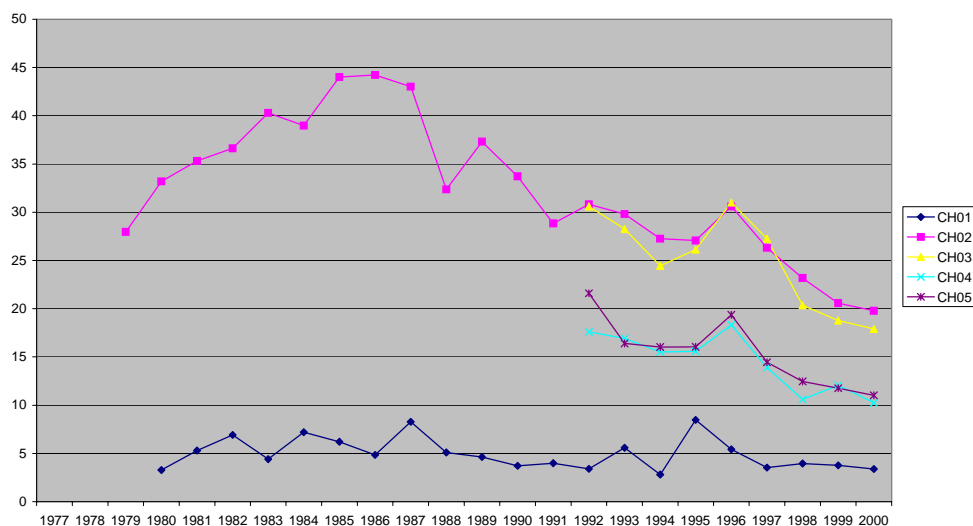


Figure A6.2 PM Rural Germany. Source: EMEP/CCC.

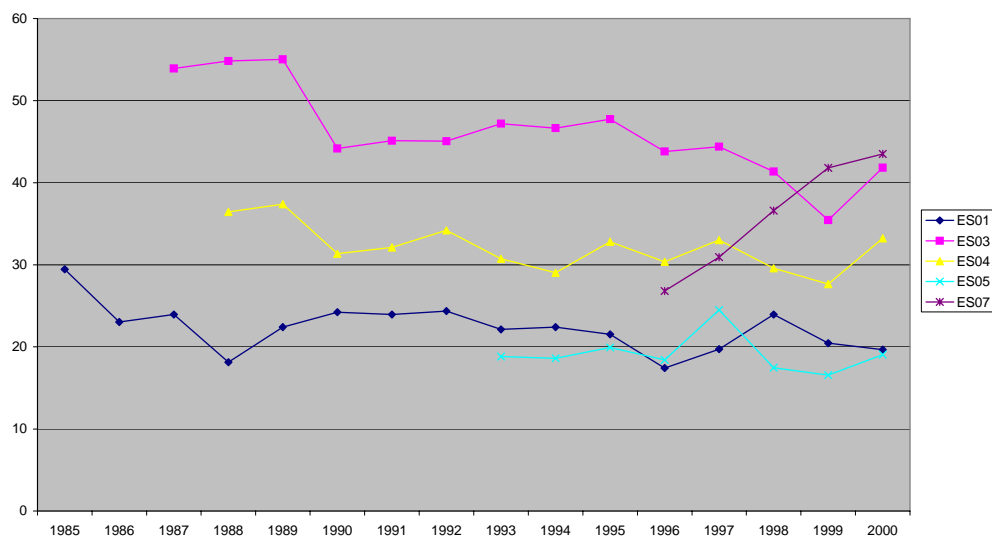
Switzerland, Spain, Ispra: EMEP stations

PM at EMEP stations at rural sites in Switzerland (Figure A6.3) showed decreasing concentrations at four stations, while no change at station CH5 (Rigi). In rural Spain, the concentrations remains rather unchanged at four EMEP stations, while a substantial increase was observed at station ES07 since 1996. The EMEP station at JRC in Ispra, Italy PM, showed a slightly decreasing tendency.

PM Rural Switzerland



PM Rural Spain



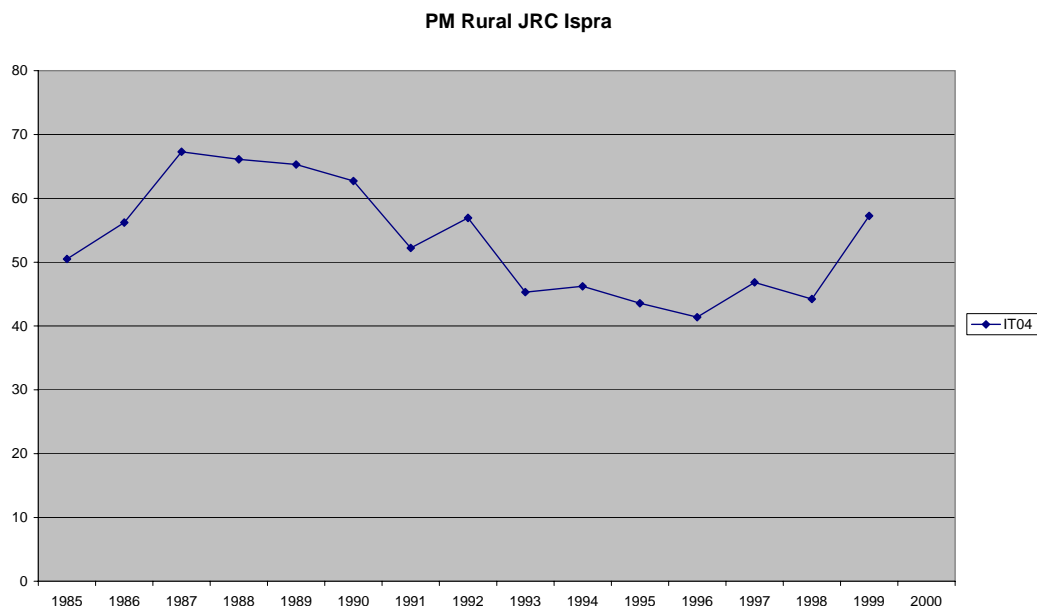
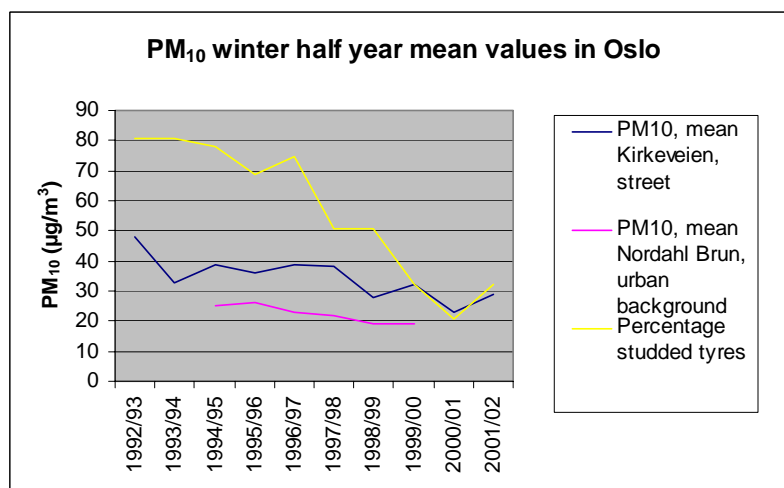


Figure A6.3 Rural (EMEP) sites in a) Switzerland, b) Spain, and c) JRC Ispra station. Source: EMEP/CCC.

Urban sites in Oslo, Norway, PM_{10} and $PM_{2.5}$

Figure A6.4 shows that at urban sites in Oslo, PM_{10} has decreased steadily since 1992/93, both at a traffic site (Kirkeveien) and at an urban background site (Nordahl Brun), both for winter average and number of days above $50 \mu\text{g}/\text{m}^3$. During the same period, the usage of studded tyres on cars has decreased from about 80% of the cars in 1992-94 to about 20% in the winter of 2000/01. The figure also shows that the $PM_{2.5}$ concentration (winter average) at the traffic site has decreased somewhat during the same period.



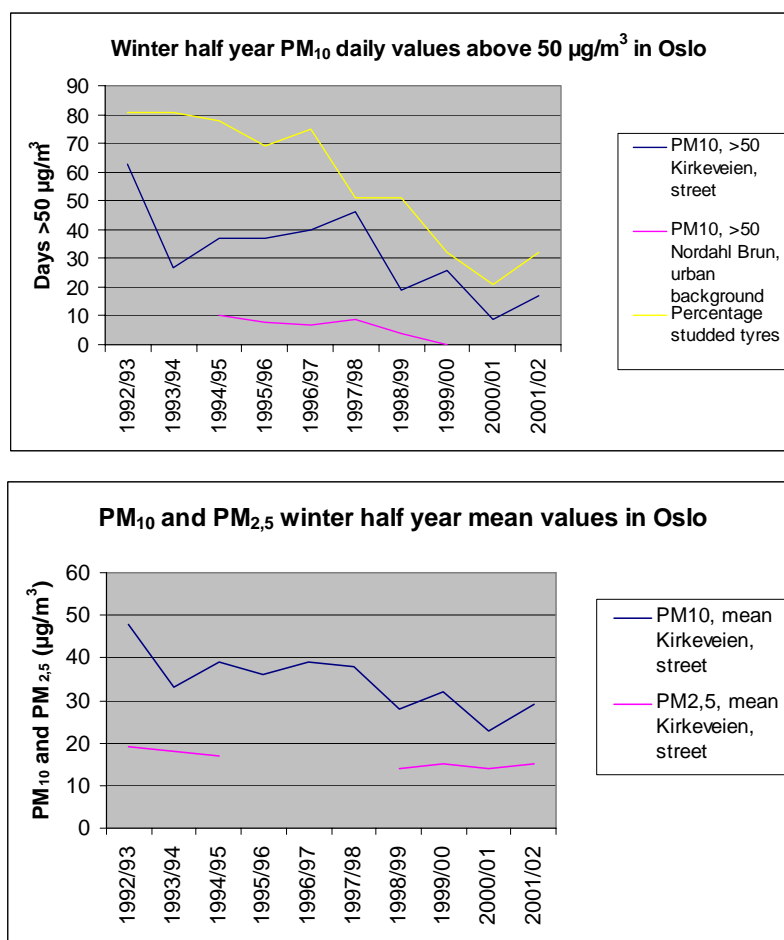


Figure A6.4 PM trends in Oslo, Norway. Source: NILU.

Urban sites in the United Kingdom

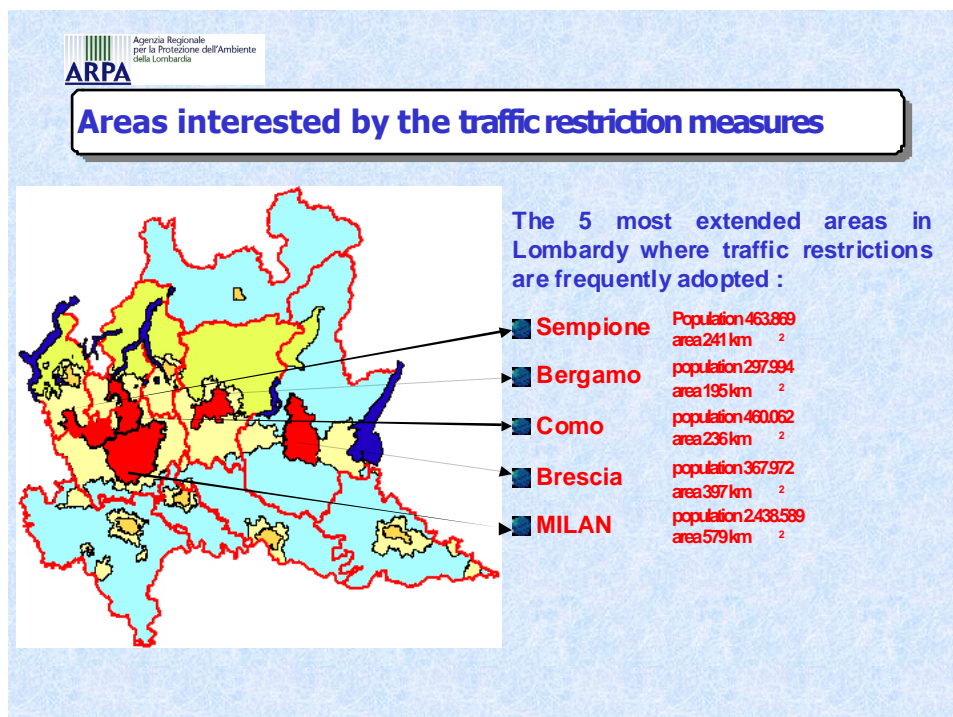
Section 7.4 of Chapter 7 also shows examples of recent trends at some urban sites in the United Kingdom. These also include projections and so are located in the projections section.

Annex 7 Annex to Chapter 8 – Short term actions for reducing PM₁₀ in Italy

In North Italy along the Po river valley, the most industrialized and populated area of Italy, many exceedances of the PM₁₀ 24-hour limit value occur during the winter, typically lasting several days. In the last few years some local Authorities have imposed traffic restrictions during those episodes. Two Regions, Lombardy and Emilia Romagna, adopted significant measures in view of the extent of the areas covered and the temporal extension.



In Lombardy Region during high PM₁₀ levels traffic restrictions were enforced in the agglomerations of Milan (2,438,000 inhabitants; 579 km²); Sempione (463,000 inhabitants; 241 km²); Como (260,000 inhabitants; 236 km²); Bergamo (298,000 inhabitants; 195 km²) and Brescia (368,000 inhabitants; 397 km²).



The Milan, Como and Sempione agglomerations are contiguous; the circulation limitations in the three areas were generally simultaneous. The traffic limitations were of three types:

- ban of pre-EURO vehicles from 8 am to 8 pm (adopted when PM_{10} concentrations exceeded $50 \mu g/m^3$ for more than seven days);
- “alternate odd-even license plates”, a complete stop of vehicles from 8 am to 8 pm with even plates on even days and vice versa (experimental campaign);
- ban of all the vehicles on Sundays (adopted on Sundays when PM_{10} concentrations exceeded $50 \mu g/m^3$ for more than 9 days).

During the 2002-2003 winter in the Milan agglomeration, there were 2 days of complete stop of vehicles circulation on Sunday; 6 days of odd-even plates stop and 15 days of stop for pre-EURO vehicles.

In Emilia Romagna Region, 81 municipality (accounting 60% of the total regional population) adopted coordinated measures for reducing traffic emissions from October 2002 and March 2003, when many exceedances are likely to occur. The actions scheme was quite different from the Lombardy's one:

- ban of the private cars according to the “alternate odd-even license plates” rule on Thursday and on Sunday from 8.30 to 12.30 and from 14.30 to 19.30;
- during the same days, all not-catalysed and diesel vehicles (except Direct Injection (DI) diesel and common rail) must stop as well as two-stroke not-catalysed mopeds and motorcycles;
- further limitations in specific urban areas from Monday to Sunday (7.30-9.30 and 17.30-19.30) for all the diesel vehicles, except the DI and common rail ones.

Both in Lombardy and in Emilia Romagna, the effectiveness of the measures has been recently assessed by the Regional Environmental Protection Agencies (ARPA Lombardia 2003 and ARPAER 2003), paying particular attention to the confounding effect of the meteorological changes which can obviously lead to misinterpretation.

In Lombardy the analysis showed that the measures reduce the concentrations of the primary pollutants, but have only limited influence on the PM_{10} concentrations. The small size of the effects may be explained considering that the agglomerations affected by the traffic limitations were surrounded by a territory with more than 2,200,000 of inhabitants within an area of about 4500 km², characterized by important sources of PM_{10} and its precursors such as NO_x , SO_x and NH_3 .

A more detailed comparison among the provisions lead to the conclusion that a pre-EURO vehicles ban is not very effective: the average reduction of the PM_{10} concentrations was around 9% during the ban period, with a decrease in kilometres driven below 5%.

An alternate odd-even license plate ban appears a slightly more efficient, with a reduction of PM_{10} concentration of about 12-14% and kilometres driven reduced by the same extent. During a complete ban of the vehicles circulation – with an evident decrease of primary pollutants concentration – the reduction of PM_{10} concentration still depends on meteorological conditions and may reach 35-45%. The PM_{10} composition changes during alternate odd-even plates bans are still under evaluation.

In Emilia Romagna, the analysis of the trend in PM concentrations at the monitoring stations of the regional network showed an average reduction of PM_{10} concentrations on the Thursdays between October 2002 and March 2003 of about 10%. In contrast, the concentrations on Sundays are not far from those of a normal Sunday with typical weekend traffic levels. On a regional basis, the average reduction of the circulating vehicles during the regulated days has been nearly 15%, while the number of passengers on the public transportation system increased even more (30%).

Both the experiences reveal that the effectiveness of the measures acting on the mobile emissions increases as the restrictions become more and more rigorous and extended in space and time. The “alternate odd-even license plates” measure, which is the only measure reasonably applicable for long time and over large areas (though the risk to induce people to buy more than one car in order to have always the “right” number on the license plate is not negligible), may contribute to slightly reduce the average long term concentrations, while the ability in reducing the number of the exceedances or the peak concentration values during severe episodes is questionable since the processes are dominated by the meteorological conditions. In that case only the strongest measure (complete ban of the traffic) might bring to a detectable effect.