

PM_{2.5} in the Netherlands

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Consequences of the new European air quality standards

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ECN Energy Research Centre of the Netherlands

MNP Netherlands Environmental Assessment Agency

TNO Institute for Applied and Scientific Research

RIVM National Institute for Public Health and the Environment

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FOREWORD

Thus far, air pollution policy in the Netherlands has focused on PM₁₀ as the target fraction for particulate matter. Based on new findings of the World Health Organization (WHO) policy attention in the EU has shifted towards the finer fraction (PM_{2.5}). This means that policy, monitoring and models in the Netherlands will have to be adapted. The new EU Air Quality Directive is in the final stages of decision making and will probably go into force in 2008. This Directive results from the Thematic Strategy on air pollution that was adopted in 2005. The new Directive combines four existing EU directives and establishes new air quality standards for fine particulate matter (PM_{2.5}).

The two key elements in the Directive with respect to PM_{2.5} constitute the following:

1. a limit value for PM_{2.5} that applies in the entire Member State.
2. a reduction of the PM_{2.5} average exposure levels in major urban agglomerations.

The exposure reduction aims to lower average levels in urban agglomerations, whereas the limit value has been introduced to also control concentrations at local hot spots.

The underlying report addresses these two issues and provides insight into the standards and associated assessment requirements. The report is meant as an aid for policy-makers and scientists involved with PM_{2.5}. The report focuses specifically on issues that are pertinent to the Netherlands. It begins with an inventory of the data on PM_{2.5} levels and the methods used for measuring these levels. Next, the report describes how future levels or levels at locations other than measurement sites are derived from model calculations and emissions in combination with measurements. Finally, an inventory is made of the current knowledge on sources of PM_{2.5}, policy measures to reduce emissions and the instruments regarding measurements, models and emissions which are used to support policy development.

In the following five chapters, this report addresses the state of knowledge on the following questions related to PM_{2.5}.

1. *Why is legislation on PM_{2.5} being introduced and what will be the new guidelines?*
2. *What are the current levels and composition of PM_{2.5}, and how should PM_{2.5} be measured?*
3. *How can the levels of particulate matter be calculated using models?*
4. *How large is the emission?*
5. *What policy instruments are available?*

Director of the Energy research Centre of the Netherlands (ECN)

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Director of the Netherlands Environmental Assessment Agency (MNP)

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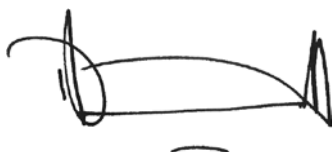
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Ir. D.Ph. Schmidt

SUMMARY FOR POLICY MAKERS

Introduction

Thus far, air pollution policy in the Netherlands has focused on PM_{10} as the target fraction for particulate matter. Based on new findings of the World Health Organization (WHO) policy attention in the EU has shifted towards the finer fraction ($PM_{2.5}$). This requires that policy, monitoring and models in the Netherlands have to be adapted. A new EU Air Quality Directive is in the final stages of decision making and will probably go into force in 2008. This Directive results from the Thematic Strategy on air pollution that was adopted in 2005. The new Directive combines four existing EU directives that include legislation on PM_{10} , and it establishes new air quality standards for fine particulate matter ($PM_{2.5}$).

This report takes advantage of the lessons drawn from the policy concerning PM_{10} and aims to anticipate possible policy actions concerning $PM_{2.5}$. This is approached by summarizing the proposed particulate matter standards and projecting whether or not the Netherlands can meet these standards. The report makes an inventory of the national knowledge on which the conclusions given below are based, taking into account that some aspects of the new Directive are still undecided.

What is $PM_{2.5}$ and where does it come from?

Particulate matter, or PM, is the term for particles found in the air. PM_{10} and $PM_{2.5}$ are good approximations of particles smaller than 10 and 2.5 micrometers in diameter, respectively. $PM_{2.5}$ is the finer fraction in PM_{10} . Many man-made and natural sources emit PM directly. Man-made sources include industrial processes and all types of combustion activities such as motor vehicles, power plants and wood burning. Other particles may be formed in the air due to chemical processes. They are formed indirectly when gases, from burning fuels and ammonia from manure, react in the atmosphere. Natural sources of $PM_{2.5}$ include, for instance, sea salt.

Why has $PM_{2.5}$ been chosen as the new metric in the EU?

There are two main reasons:

1. In general $PM_{2.5}$ is considered to be more hazardous to human health than PM_{10} (WHO, 2006a; Brunekreef and Forsberg, 2005), and because it penetrates more deeply into the lungs.
2. $PM_{2.5}$ originates more from man-made sources than PM_{10} and is therefore in principle more manageable.

Health studies have shown that there is a significant association between both short-term and long-term exposure to fine particles and premature death. Other important effects include aggravation of respiratory and lung disease, asthma attacks, heart attacks and irregular heartbeat. PM_{2.5} consists of many compounds originating from many sources. PM_{2.5} mass is regulated instead of individual components, because up to now all PM_{2.5} has been considered as equally harmful - although some components are believed to be more harmful than others - and because the health benefits of reducing the components individually are unknown.

What are the standards for PM_{2.5} in the new EU Directive on air quality?

The common position adopted by the Council (CS, 2007a) has been used as the basis for the present report. The main thrust with respect to PM_{2.5} is that two standards are proposed, a limit value and an exposure reduction target value; the latter is a new policy approach.

- Applied throughout the EU, the proposed *limit value* must be attained at every public location by 1 January 2015; it has been set at 25 µg/m³. This value has been set as an annual average target value for the period 2010-2015. The recent draft recommendation for amendments by the European Parliament (CS, 2007b) refers to a value of 20 µg/m³ instead of 25 µg/m³.
- For the Netherlands, the proposed *exposure reduction target value* implies a 20% decrease in exposure to PM_{2.5} in urban agglomerations in 2020 compared to the exposure in 2010. In 2013, the provisions for PM_{2.5} will be reviewed and the reduction target value could then become legally binding.

The main question addressed in this report is whether these standards can be attained in the Netherlands. This is discussed below.

Attainability of the standards in the Netherlands

Limit value

- The available data on the current levels of PM_{2.5} suggest that the proposed limit value of 25 µg/m³ can probably be attained in 2015, apart from a very limited number of hot spots (see Figure 1). Under current legislation, the value of 20 µg/m³ will probably still be exceeded in busy streets in urban agglomerations and at several locations in agricultural areas. Due to the recently outlined additional measures for the Netherlands, the number of exceedances of the value of 20 µg/m³ will decline towards 2015, but even then the number of exceedances is expected to become very limited only by 2020.
- The Directive aims at regulating both PM₁₀ and PM_{2.5}. Since these parameters are strongly interrelated, it makes sense to review the stringency of the limit values

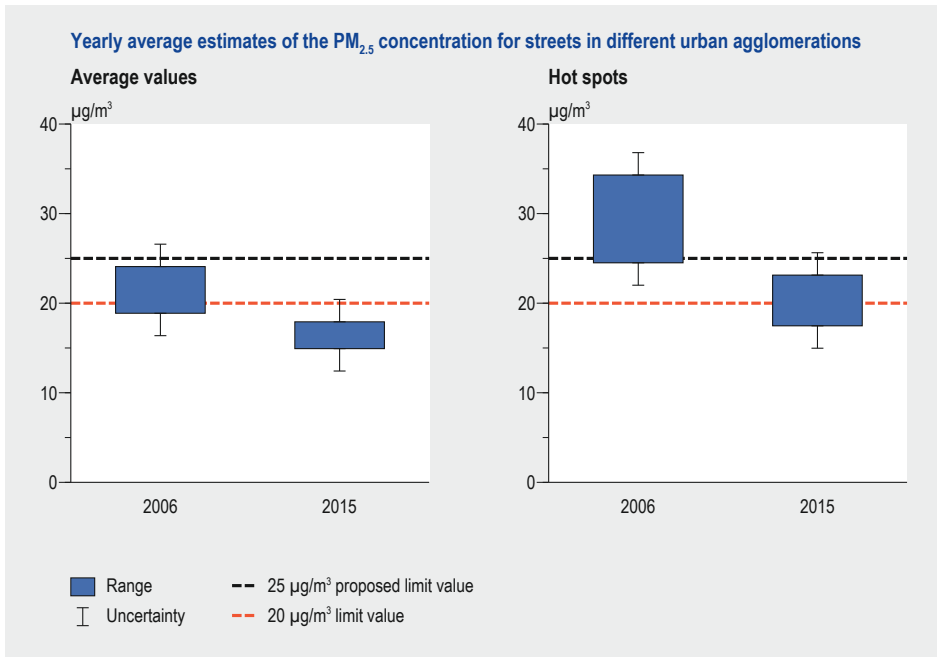


Figure 1 Estimates of the PM_{2.5} concentration ranges for streets in urban agglomerations in the Netherlands in 2006 and 2015. On the left, ranges for average values and, on the right, ranges for hot spots. The estimates are based on a combination of model calculations and measurements. The emission projections are based on current legislation (see chapter 4). The uncertainties shown represent a lower limit, because not all sources of uncertainty have been quantified (e.g. the effect of uncommon meteorological conditions on yearly average concentrations).

proposed. The current status in the Netherlands is the following. The strictest limit value for PM₁₀ concerns 24-hour concentrations, which are not to exceed 50 µg/m³ more than 35 times in a calendar year. This limit value appears to be more stringent than the proposed PM_{2.5} limit value of 25 µg/m³. However, a value of 20 µg/m³ for PM_{2.5} could be more stringent than the PM₁₀ limit value for 24-hour concentrations. This implies that in 2015 the PM_{2.5} limit value of 20 µg/m³ might lead to more exceedances than the PM₁₀ limit value for 24-hour concentrations. Note that the stringency is viewed with respect to PM₁₀ and PM_{2.5} levels per year. The fact that the limit values go into force in different years has not been taken into account.

National exposure reduction target value

- A reduction of 20% in the exposure between 2010 and 2020 will almost certainly not be reached under current legislation (see Figure 2). Moreover, it is likely that the recently outlined additional measures for the Netherlands will also be insufficient to reach the reduction target value. Therefore, extra national and local measures will probably be necessary.
- Average urban background concentrations in the Netherlands are largely due to sources abroad. Consequently, the effect of national policies is limited and the

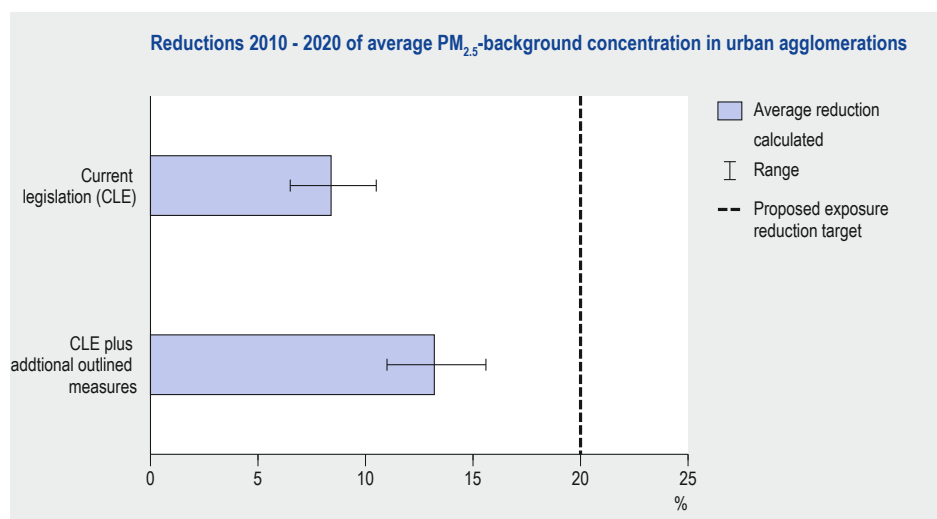


Figure 2 $PM_{2.5}$ reductions (%) between 2010 and 2020 of the average background concentration in urban agglomerations in the Netherlands calculated for emissions based on current legislation and if additional measures (recently outlined) are also taken. The margins show the highest and lowest calculated reductions.

attainability in the Netherlands of the proposed exposure reduction target value depends for an important part on the implementation of policy measures abroad.

- Future levels will be affected by reductions in the EU and especially the neighboring countries. The present analysis is based on the assumption that countries will meet their emission goals as set by existing agreements: for 2010 by the National Emission Ceilings Directive and for 2020 by the Thematic Strategy on Air Pollution. Therefore, larger reductions than those shown in Figure 2 can only be expected if Member States apply measures that go beyond their national emission goals. At present, not all Member States expect to be able to meet their emission goals in time.
- Other Member States probably also need to make plans for further emissions reductions in order to meet the new $PM_{2.5}$ standards. At what time this may lead to substantial extra emission reductions is yet unclear. Therefore, further concentration reductions should only be expected from extra national and local measures in the Netherlands, at least until 2015.

Attainability of the standards in the rest of Europe

An assessment of the attainability in the EU was published by the Institute for European Environmental Policy (IEEP, 2006). It was based on an extrapolation of $PM_{2.5}$ levels in Europe for 2004. It concluded that only a limited number of exceedances of the proposed limit value of $25 \mu\text{g}/\text{m}^3$ can be expected after 2010. However, under current legislation – and even including the emission reduction called for in the Thematic Strategy – the same information suggests that the value of $20 \mu\text{g}/\text{m}^3$ will very probably

not be attained everywhere in Europe in 2015. Specifically, urban background concentrations may then be close to, or exceed, the annual average concentration of $20 \mu\text{g}/\text{m}^3$ in densely populated and industrialized areas (such as the Po Valley), certain areas in Central European countries, the Ruhr area, the Benelux countries, and certain large cities in Europe. At the street level, the attainability problems will probably be more severe.

Attainability problems regarding the $\text{PM}_{2.5}$ limit value in the Netherlands thus appear similar to those in other densely populated and industrialized regions in Europe. However, it is unclear whether the relevant Member States will face similar problems meeting the proposed exposure reduction target value of 20%, because the level of implementation of technical and non-technical reduction measures differs throughout Europe.

How to proceed?

A number of steps will be taken. Some follow directly from the new Directive as a legal obligation, and others are necessary to reduce the uncertainties about $\text{PM}_{2.5}$ in order to provide more robust support to policy decisions regarding $\text{PM}_{2.5}$.

The underlying $\text{PM}_{2.5}$ data of this report are rather uncertain. This is because the instruments that are used to support $\text{PM}_{2.5}$ policy development (measurements, models and emissions, etc.) are still at an initial or research phase. The conclusions in this report are often based on preliminary data and extrapolation of information on PM_{10} .

- The most urgent step is to prepare for and comply with the measurement obligations in the guidelines of the new Directive. National preparations are being made to start measurements of $\text{PM}_{2.5}$ by 2008, in accordance with the Directive guidelines.
- Such measurements form the basis for further assessments of $\text{PM}_{2.5}$ levels and the associated health effects. In addition, focused measurements seem necessary, especially concerning 1) the contribution of sources to exceedances that occur at the local level and 2) the association of health effects with specific local sources such as traffic.
- Additional steps include several actions to improve models and emissions, especially regarding primary and secondary organic particles. Model performance in urban agglomerations and streets should be improved; this can lead to a better assessments of measured or modeled values compared to limit values and to a better understanding of source contributions.
- A more accurate inventory of primary $\text{PM}_{2.5}$ emissions is also necessary in the process of defining and monitoring a national emission ceiling for $\text{PM}_{2.5}$.
- At the same time it is important to assess which additional national, local and possibly European policy measures would be necessary to attain the proposed standards in the Netherlands in time. Planned national and European legislation to

combat climate change can affect particulate matter levels, and should therefore be integrated.

Instruments for policy support

Given the uncertainties, the quality of the measurements and models currently used for policy support is insufficient to assess the levels of $PM_{2.5}$ with an accuracy required by the Directive guidelines. Especially in urban agglomerations, which have large emission dynamics and complex terrain, the uncertainties are large. However, the current models may be adequate to assess the relative effect of given emission reductions on the concentrations.

Better information will become available when the steps indicated above are taken. This information will reduce the currently large uncertainties about $PM_{2.5}$. Even then, however, the uncertainties will probably remain substantial. If these uncertainties can be taken into account in the implementation of the new Directive, the enforcement in practice could become more effective.

The following Technical Summary discusses the main conclusions in more detail and makes suggestions about how to bridge the present knowledge gap regarding $PM_{2.5}$.

TECHNICAL SUMMARY

The main conclusions and suggestions about how to bridge the knowledge gap with regard to $PM_{2.5}$ are based on the detailed work in the present report, which is summarized here. For the actual approach used in this inventory, the reader is referred to the main text of the report.

Attainability of the standards in the Netherlands

Exceedances of the proposed limit value of $25 \mu\text{g}/\text{m}^3$ will be localized and are expected to be limited in number. Exceedances of the value of $20 \mu\text{g}/\text{m}^3$ are expected in busy streets and local hot spots, even if the recently outlined additional national measures are taken. Concentration reductions of average urban background levels between 2010 and 2020 which result from current legislation and additional outlined measures in the Netherlands will be too small to meet the proposed exposure reduction target.

Current concentrations and projections

Estimates of the concentrations in the Netherlands are based on a combination of measurements and model calculations. Most measured data do not have an official status, because they were not obtained according to the guidelines for the measurements. In the present report, the measured levels have been corrected with a provisional calibration factor; these measurements are mostly biased low.

- Current annual regional background concentrations range between 12 and $16 \mu\text{g}/\text{m}^3$. Urban background concentrations are found in the range of 16 to $18 \mu\text{g}/\text{m}^3$. Street increments – the added concentration due to traffic in streets – range between 2 and $6 \mu\text{g}/\text{m}^3$ for average traffic conditions and between 7 and $14 \mu\text{g}/\text{m}^3$ for very busy streets.
- With current legislation, projections for 2015 range between 12 and $14 \mu\text{g}/\text{m}^3$ for the regional background concentrations and between 14 and $15 \mu\text{g}/\text{m}^3$ for the urban background. Street increments could range between 1 and $3 \mu\text{g}/\text{m}^3$ for average traffic conditions and between 3 and $6 \mu\text{g}/\text{m}^3$ for very busy streets.
- In neighboring regions in Belgium and Germany, measurements of the annual concentrations appear to be somewhat higher than the current levels in the Netherlands, mainly at urban background and street sites.
- In the EU, data on $PM_{2.5}$ are generally scarce. Levels that have been reported range from insignificant in coastal Ireland to concentrations greatly exceeding the proposed limit value of $25 \mu\text{g}/\text{m}^3$ in Poland and in cities in southern Europe.

Components of PM_{2.5} in the Netherlands

Particulate matter is not a simple parameter in chemical terms. PM_{2.5} is a complex blend of the following chemical compounds:

- Ammonium nitrate and sulfate particles comprise on average about $8 \mu\text{g}/\text{m}^3 \pm 10\%$ of the PM_{2.5} concentration in the Netherlands (approximately equal amounts of ammonium nitrate and ammonium sulfate). Ammonium nitrate and sulfate are not directly emitted, but are formed in the air from sulfur dioxide, nitrogen oxides and ammonia. Ammonium nitrate is semi-volatile and therefore complicates measurements.
- Carbon is the other main PM_{2.5} component, with a contribution of about $5\text{-}6 \mu\text{g}/\text{m}^3$. The concentration of carbon is the most uncertain of the major components ($\pm 35\%$). There are strong indications that the elevated PM_{2.5} concentrations in the urban background and streets are associated with increased levels of carbon.
- Ammonium nitrate and sulfate and carbon originate almost completely from anthropogenic sources.
- Sea salt is the most important natural component, with an estimated average contribution in the Netherlands of about $1 \mu\text{g}/\text{m}^3 \pm 20\%$.
- Mineral dust is a component with an estimated average contribution in the Netherlands of $0.6 \mu\text{g}/\text{m}^3 \pm 60\%$.
- Water forms a normal part of particulate matter. The amount depends for a great deal on the measurement method.

Current emissions and projections

- Primary emissions of PM_{2.5} have been included in the national Pollutant Emission Register in 2007. In 2005, primary PM_{2.5} emissions amounted to 22 kt in the Netherlands. Projections for 2010 and 2020 are 19 and 17 kt, respectively, with current legislation. Recently outlined additional measures can lower the 2020 projections for the Netherlands by an extra 10%. If all technical measures are applied, larger reductions will be possible.
- Primary emissions due to ocean shipping – not included in the national totals above – amounted to an extra 8 kt in 2005. Projections for the ocean shipping sector indicate an increase to 11 kt in 2010 and 13 kt in 2020.
- For the EU-27, current legislation will probably lead to a 50% emissions reduction of primary PM_{2.5} between 2000 and 2020. For the EU-27, if all technical potential is used, an extra 40% emissions reduction of primary PM_{2.5} could be achieved in 2020.
- Sources of the gases sulfur dioxide, nitrogen oxides, ammonia and volatile organic compounds, which have a large indirect contribution to PM_{2.5}, are regulated by the National Emission Ceilings Directive. National emission projections for 2010 indicate that the emission ceilings in the Netherlands can be reached or closely approached with current legislation. Emission targets for 2020, which also include relative targets for PM_{2.5}, are presently under negotiation in a revision of the National Emission Ceilings Directive.
- The present discussion in the EU on burden sharing of the reduction of greenhouse gases could lead to a shift in the emissions relevant for PM_{2.5} within the EU. Generally speaking, measures to combat climate change will provide significant ancillary benefits for air pollution abatement by 2030, although the use of biofuels and biomass also can cause increased concentrations of particulate matter. Transition towards a more intensive use of biofuels and biomass will require additional

Source contributions to PM_{2.5} in the Netherlands

- About 20% on average of current PM_{2.5} concentrations in the Netherlands originate from nationally registered sources, 50% from registered sources abroad and on the sea and 30% from natural and other sources. In urban agglomerations, the contribution from national registered sources to the annual average concentration is 30% or less. In busy streets, however, the contribution from national registered sources is larger (up to 60%).
- The main contributor to the average PM_{2.5} concentration in the Netherlands is the sector Industry/Energy/Refineries abroad. The sectors traffic and agriculture, both national and abroad, are the next largest contributors.
- Secondary inorganic aerosols contribute about half of the total PM_{2.5} concentration. These aerosols are chemically produced in the air from sulfur dioxide, nitrogen oxides and ammonia. The present emission projections do not lead to significant changes in the amount of primary PM_{2.5} relative to secondary inorganic PM_{2.5}.
- The other half of the total PM_{2.5} concentration is due to primary registered anthropogenic sources, sea salt, mineral dust and other sources of volatile organic compounds that lead to the formation of secondary organic aerosol.

regulation to limit particulate matter emissions from, for instance, cars, biofuel refineries and small woodstoves.

Main uncertainties

Current assessments of concentrations and sources of particulate matter contain significant uncertainties.

- Measurements of PM_{2.5} are still in the initial phase; absolute levels have at least 25% uncertainty, which is the maximum value allowed by the Data Quality Objectives of the new Directive. In addition, measured levels in EU countries may be difficult to compare due to the use of different, but officially allowed, technical measurement conditions.
- There are major uncertainties about primary PM_{2.5} emissions. Estimates of the uncertainty vary from around 30% up to a factor of two.
- Models underestimate PM_{2.5} because not all sources are included or because sources and formation pathways are not well represented. An average difference of $5 \mu\text{g}/\text{m}^3 \pm 2.5 \mu\text{g}/\text{m}^3$ was found between the measurements and the model used for the present assessment.
- The uncertainty about the health effects associated with different fractions in PM_{2.5} hampers policy development to reduce the adverse health effects. There are indications that particle emissions from traffic and, in general, combustion-related particles like soot, play a more important role in the associated health effects than secondary inorganic aerosols (WHO, 2006b). Consequently, reducing the emissions of the precursor gases sulfur dioxide, nitrogen oxides and ammonia may reduce the contribution to background concentration on a large scale, but not necessarily the potentially higher health risk from primary combustion-related sources (local and low-level) in urban agglomerations.

How to proceed and reduce the main uncertainties

Measurements are to be conducted according to the guideline in the new Directive. An assessment of the necessary policy measures to attain the PM_{2.5} standards has to take place. Improved emission inventory, modeling and monitoring methods, together with more widespread measurements, would help reduce the main uncertainties.

The measurement requirements in the guidelines lead to the following actions and recommendations:

- PM_{2.5} measurements should begin in 2008 or 2009, at the latest, in order to determine the starting point for assessing the 20% reduction target value in 2020.
- The national measuring locations must be chosen according to the guidelines. A specific type of monitor must also be chosen.
- The new Directive aims at establishing a measurement network which integrates both PM₁₀ and PM_{2.5} measurements. Due to the limited understanding of PM_{2.5}, the measurements for PM₁₀ and PM_{2.5} can not be merged at this time.
- If the Netherlands decides to use an automated monitor, its equivalence with the reference method will have to be determined.
- Member States are required to measure a series of components of PM_{2.5} at a regional site, starting two years after the Directive goes into force. The regional background site *Cabauw*, which is already a focus of PM-related and climate research, could be a good location for this purpose.

Determine policy measures needed to attain the PM_{2.5} standards while taking into account the co-benefits and/or trade-offs of other EU legislation.

- Assess the national and local measures, such as regulation of traffic volume, that are necessary to attain the new PM_{2.5} standards in relation to the effect of possible future reduction measures in the EU that go beyond existing legislation.
- Assess the effects on particulate matter levels of policy measures to combat climate change, such as the effects of a substantially increased use of biofuels and/or biomass.

Improve emission inventories and models and conduct more widespread measurements.

- Improve the quality of the inventory of primary emissions. There is also a need for chemical classification of the officially reported primary PM_{2.5} emissions, both nationally and abroad, because modeled primary PM_{2.5} levels can not as yet be verified with measured data.
- Improve assessments of PM_{2.5}, with emphasis on the local and urban scales, especially regarding primary PM_{2.5} and secondary organic aerosol.
- Improve model assessments of PM_{2.5} by describing the sources and formation pathways of PM_{2.5} that have not yet been modeled.
- Determine and minimize the magnitude of inevitable measurement uncertainties, such as those caused by the presence of semi-volatile components in PM_{2.5}, and assess the consequences for national policies.

- Develop an optimal measurement strategy for monitoring the compliance with both PM_{2.5} standards.
- Reduce the remaining uncertainties about the formation pathways of ammonium nitrate and the emission estimates of ammonia. Even though the uncertainties have been significantly reduced in recent years, this still may be worthwhile; this is because the contribution of ammonium nitrate to current regional and urban background levels of PM_{2.5} in the Netherlands is substantial, especially during episodes.

Contents

FOREWORD 5

SUMMARY FOR POLICY MAKERS 7

TECHNICAL SUMMARY 13

- 1 PM_{2.5} in the new EU Directive for Air Quality 21
 - 1.1 Why is PM_{2.5} being introduced and what will be the new standards? 21
 - 1.2 Implications of the new PM_{2.5} standards for the Netherlands 24
 - 1.3 Reader's guide 26

 - 2 Levels and measurements 29
 - 2.1 Current levels of PM_{2.5} 29
 - 2.2 How can we measure PM_{2.5}? 33
 - 2.3 Chemical composition of PM_{2.5} 36

 - 3 Calculating PM_{2.5} levels using models 41
 - 3.1 Introduction 41
 - 3.2 The role of model calculations 42
 - 3.3 Background concentrations of PM_{2.5} 42
 - 3.4 Composition and sources 44
 - 3.5 Concentrations in urban agglomerations 46
 - 3.6 Contribution of local sources 48
 - 3.7 Uncertainty in the calculated PM_{2.5} concentrations 51

 - 4 How large is the emission? 55
 - 4.1 Sources of PM_{2.5} 55
 - 4.2 Anthropogenic primary sources 56
 - 4.3 Anthropogenic sources of SO₂, NO_x and NH₃ 59
 - 4.4 Natural and other sources 60
 - 4.5 Emission uncertainties 60

 - 5 PM_{2.5} policy instruments and measures 63
 - 5.1 Current legislation 63
 - 5.2 Additional policy instruments 65
 - 5.3 Policy measures in addition to current legislation 65
 - 5.4 Current status of PM_{2.5} assessment instruments 68
- References and further reading 73

1 PM_{2.5} IN THE NEW EU DIRECTIVE FOR AIR QUALITY

This chapter summarizes the main requirements for PM_{2.5} in the new EU Air Quality Directive and discusses the background to this legislation. Implications for the Netherlands are highlighted with respect to the requirements, thereby focusing on the question of attainability. The role of PM_{2.5} in other environmental issues is briefly addressed.

The Directive is presently at the stage of a common position adopted by the Council of Ministers (CS, 2007a). After the second reading by the European Parliament, the final version is scheduled to appear around the end of 2007.

1.1 Why is PM_{2.5} being introduced and what will be the new standards?

The introduction of PM_{2.5} as an air quality parameter is part of a novel approach to air quality in the framework of the recent 6th Environment Action Plan of the EU; this Action Plan stems from the Thematic Strategy on air pollution (CEC, 2005).

The basic argument for the choice of PM_{2.5} as an air quality parameter stems from conclusions and recommendations in reports by the World Health Organization (WHO, 2000; WHO, 2003; WHO, 2006a; WHO, 2006b). The report from 2003 was specifically intended to support the development of EU policy on clean air for Europe. It states that fine particulate matter (commonly measured as PM_{2.5}) is strongly associated with mortality and other effects such as hospitalization for cardio-pulmonary disease; it therefore recommends that air quality guidelines for PM_{2.5} be developed further. Both WHO reports also indicate that a smaller body of evidence suggests that coarse mass (particles between 2.5 and 10 µm) also has some effects on health and that a separate guideline for coarse mass may be warranted.

There is as yet no identifiable threshold below which PM_{2.5} does not pose a health risk. This component of air pollution should therefore not be regulated in the same way as other air pollutants. The approach should focus on an overall reduction of concentrations in the urban background so that large sections of the population benefit from improved air quality. However, to ensure a minimum degree of health protection everywhere, there is also a limit value.

Another reason for the choice is that PM_{2.5} is more often of anthropogenic (man-made) origin and therefore is theoretically more manageable. Essentially, this is the scientific basis for focusing on the regulation of PM_{2.5}, while retaining the existing air quality limit values for PM₁₀.

Reduction target for exposure to PM_{2.5}

A 20% reduction is proposed as a target value. This focuses on the average urban background concentrations and must be achieved between 2010 and 2020. Member States are not obliged to incur 'disproportionate' costs to realize this reduction target. Moreover, there will be a review of the exposure reduction target in 2013 with the aim of making this legislation legally binding.

The average annual urban background concentrations must be assessed as a three-year average. The reason is that this approach moderates the impact of the year-to-year meteorological variability.

Limit value

The Directive presently calls for a limit value of 25 µg/m³, expressed as an annual average that must be attained by 2015. The level of the limit was initially intended to be consistent with the existing limit values for PM₁₀, so that Member States would not be confronted with extra measures for PM_{2.5}. However, there is also discussion of a more stringent strategy for PM_{2.5}, with an annual value of 20 µg/m³. The consequences of both values for the Dutch situation are explored in this report.

Requirements for assessment of the levels

The Directive provides detailed requirements for assessment and specifically the measuring strategy:

- Regarding the reference method for measuring PM_{2.5}, see section 2.2.
- Measurements should start in 2008 or at the latest by 1 January 2009 to establish a reference point for the exposure reduction target.
- For assessment of compliance with the proposed limit value, the Directive requires measurements for levels above the upper threshold (corresponding to 70% of the limit value). At lower levels a combination of measurements and modeling is allowed, which reduces the number of costly measurements. Measurements to assess compliance with the limit value should start in 2010.

The monitoring requirements as they are pertinent to the Netherlands can be summarized as follows:

- Assessment of compliance with the proposed **limit value**

There is a combined requirement for the number of locations for measuring PM_{2.5} and PM₁₀. The key issue is that the number of PM_{2.5} sampling points depends on the number for PM₁₀ (see text box below: **Summary of measuring requirement to assess the limit values for PM in agglomerations**).

Summary of measuring requirement to assess the limit values for PM in agglomerations in the Netherlands

| Population in thousands | Sum of stations for PM ₁₀ and PM _{2.5} at levels higher than 70% of limit value | Sum of stations for PM ₁₀ and PM _{2.5} At levels less than 70% of limit value |
|-------------------------|--|--|
| 0-249 | 2 | 1 |
| 250-499 | 3 | 2 |
| 500-749 | 3 | 2 |
| 750-999 | 4 | 2 |
| 1000-1499 | 6 | 3 |

- The total number of PM_{2.5} and PM₁₀ sampling points in a Member State must not differ by more than a factor of 2.

- Assessment of compliance with the proposed **exposure reduction target**
The reduction target is associated with the Average Exposure Indicator (AEI). This AEI is based on measurements in urban background locations. It should be assessed as a 3-year continuous annual mean concentration, averaged over all sampling points. The guidelines to assess the AEI (e.g. number of sampling points) are ambiguous: either one sampling point per million inhabitants summed over agglomerations and additional urban areas, or a sufficient number of urban background stations for the calculation of the AEI. The AEI for the reference year 2010 will be the mean concentration of the years 2008, 2009 and 2010. The mean concentration over the years 2009 and 2010 may be used or the mean concentration over the years 2009, 2010 and 2011 if a Member State is unable to start monitoring by 1 January 2008. The Commission must be informed about this before 1 January 2008.
- The Directive refers to: 'detailed measurements should be made of fine particulate matter and components at rural background locations in order to understand better the impacts of this pollutant and to develop appropriate policies'. This involves the measurement of a number of chemical compounds.

The deadline for starting the measurements to establish a reference point for the exposure reduction target is 1 January 2009. The national decision about the technical measuring tool to be used has not been made as yet. More details on measurements and associated measuring requirements are given in chapter 2.

1.2 Implications of the new PM_{2.5} standards for the Netherlands

Attaining the PM_{2.5} standards

The annually averaged PM_{2.5} regional and urban background concentration in the Netherlands is, at present, between 14 and 21 µg/m³. Due to the contribution of traffic, concentration increments between 2 and 14 µg/m³ may be expected at the street level. The continued effect of current policies will lead to a considerable reduction of the PM_{2.5} concentrations in 2015. By then, the regional and urban background concentrations are expected to range between 12 and 17 µg/m³ and street increments between 1 to 6 µg/m³. Therefore, it can be concluded that large-scale exceedances of the proposed 25 µg/m³ limit value in 2015 will not take place. However, due to the present lack of data on PM_{2.5} concentrations and also considering the ambiguities in the current measurement of PM_{2.5} as described in chapter 2, the possibility of some hot spot exceedances should not be ruled out.

Attaining a value of 20 µg/m³ – as discussed by the European Parliament (CS, 2007b) – is more challenging. This value could be exceeded at several locations, for instance, along motorways and in cities in 2015. Measures on a local scale, such as regulations which reduce the local traffic volume, can help to attain a limit value at specific hot spots.

As for the national exposure reduction target of 20% between the reference year 2010 and 2020, achieving this goal will require more stringent emission reductions than those currently foreseen. The reduction target translates into a 3-4 µg/m³ downward concentration trend in urban air in the period 2010-2020, whereas the most ambitious policy package of those currently considered would reduce average urban PM_{2.5} levels by no more than 2.3 µg/m³ between 2010 and 2020. Current legislation will lead to relative concentration reductions of 6 to 10%, on average for urban agglomerations between 2010 and 2020. Additional recently outlined policy measures, discussed in chapter 5, may lead to a further reduction of urban concentrations by as much as 16% in some urban agglomerations.

The reduction percentages are sensitive to the level of implementation of reduction measures in 2010 and the absolute concentrations in 2010. Altogether the 20% reduction target would require drastic measures in the Netherlands. Additional national policy measures are necessary. If the currently available technical reduction measures in the EU are implemented, this could, roughly estimated, result in an extra reduction in PM_{2.5} concentrations of up to 2 µg/m³ on average in the Netherlands (see chapter 5). Such an additional reduction would be sufficient to attain the exposure reduction target value of 20%.

Relationship between the limit values for PM_{2.5} and PM₁₀

The proposed Directive aims at regulating both PM_{2.5} and PM₁₀. Since PM_{2.5} is contained in PM₁₀, the parameters are strongly interrelated and it therefore makes sense to review the stringency of the targets and limit values for PM_{2.5} in the context of the existing limit values for PM₁₀. Another issue is that the national measurement strategy will focus on that fraction for which the standards are the most stringent.

The stringency of one PM standard with respect to the other can be assessed by converting PM_{2.5} standards to PM₁₀ standards using the fraction of PM_{2.5} in PM₁₀ concentrations. This approach to assess the stringency of PM standards is, however, sensitive to the actual levels, which are not yet known with sufficient accuracy for PM_{2.5} in the Netherlands. Data from the literature indicate that PM_{2.5}/PM₁₀ ratios in Northwest Europe lie between 0.6 and 0.8. At present, the general picture is that the share of PM_{2.5} in total PM₁₀ is low (0.6-0.75) along roadways – where the highest concentrations will occur (see chapter 2). The low fraction of PM_{2.5} in PM₁₀ is partly explained by re-suspended road dust which generally contributes much more to PM₁₀ than to PM_{2.5}.

The yearly average proposed PM_{2.5} limit value of 25 µg/m³ corresponds, given PM_{2.5}/PM₁₀ ratios of 0.6 to 0.8, with PM₁₀ standards of 42 µg/m³ and 31 µg/m³, respectively. This is less or about the same as the current PM₁₀ limit value for 24-hour concentrations (equivalent to a yearly average concentration of 31 µg/m³). The PM₁₀ limit value is thus more stringent than or about equivalent to the proposed 25 µg/m³ limit value. The PM₁₀ limit value is also more stringent in a temporal sense, because the maximum delay in meeting this limit value expires 3 years after the Directive goes into force (i.e. possibly in 2011), whereas the PM_{2.5} limit value should be achieved in 2015. We conclude that in the Council's proposal the PM₁₀ limit value for 24-hour concentrations remains the more stringent limit value.

The yearly average PM_{2.5} value of 20 µg/m³ would correspond to a yearly average PM₁₀ concentration of between 33 µg/m³ and 24 µg/m³. Given that the limit value for 24-hour PM₁₀ concentrations corresponds to an annual limit value for PM₁₀ of 31 µg/m³, a standard of 20 µg/m³ PM_{2.5} could be more stringent than the limit value for 24-hour PM₁₀ concentrations. See also Figure 2.2 in the next chapter.

Exceptions/derogations

The Commission is expected to publish guidelines for the demonstration and subtraction of exceedances attributable to natural sources. The Council's common position includes the possibility to subtract the contribution of natural sources of air pollutants when exceedances of air quality limit values occur. While there is a possibility to extend the deadline for attaining limit values, it is important to note that such a postponement is not allowed for PM_{2.5}.

1.3 Reader's guide

The national issues discussed above are the key ones in the following chapters. We will begin in chapter 2 by addressing the levels and the measurement methods. Chapter 3 describes the path from emission via transport to levels at exposure. In chapter 4 the sources are specified. Chapter 5 sketches out the status of the policy instruments that affect the national levels of air pollution and the reduction potential.

In summary, the report is divided into four chapters addressing the knowledge on:

- Levels/measurements
- Calculation of the levels
- Emissions
- Policy instruments

PM_{2.5} is contained in PM₁₀ and is a major fraction of PM₁₀. The national knowledge on PM₁₀ was used in an interpolation to address the previously indicated issues concerning PM_{2.5}. Most of the expertise and knowledge on PM₁₀ was, and still is, acquired from the implementation of the current Daughter Directive for PM₁₀ (EU, 2001).

If you would like to explore specific aspects of this new parameter in more detail, a list of suggested reading has been included at the end of the report. Internet sites are also listed and are recommended as an easily accessible source of information; links to these sites have been included.

The present chapter, chapter 1, summarizes the key elements of the proposed Directive. It should be mentioned that although the Directive is based on the recommendations of the World Health Organization (WHO, 2003), it did not follow these recommendations literally. The WHO recommended giving further consideration to black carbon (black

The influence of aerosols on climate

Particulate matter plays a role in the enhanced greenhouse effect (IPCC, 2007). Particulate matter is usually indicated in this context with the term aerosols. Aerosols reflect sunlight that would otherwise warm the earth. There are natural aerosols (see Section 2.4), and their reflection and associated cooling is a natural climate phenomenon. However, as in the case of the enhanced greenhouse effect, the additional manmade aerosols lead to enhanced cooling. One component, elemental carbon (soot), absorbs sunlight much more effectively and thus acts as a true greenhouse component. This suggests that reduction of soot could be beneficial. Such a policy has both a positive health effect and a positive climatic effect by partly neutralizing the enhanced greenhouse

effect, especially at the regional scale. However, a decrease in the other aerosol components would lead to less cooling.

Aerosols and carbon dioxide frequently originate from the same sources. For example, they are simultaneously emitted during combustion processes. To calculate the total effect of source measures on climate, both products must be taken into account. One example is traffic. Modern diesel vehicles are 20% to 30% more efficient than comparable petrol vehicles and therefore emit 10% to 20% less carbon dioxide for each kilometer traveled. This is beneficial for counteracting the enhanced greenhouse effect. On the other hand, diesel cars emit soot that acts as a greenhouse component.

smoke) or other measures of traffic soot and to the effects of ultrafine particles (particles with a diameter smaller than 100 nm), which are also related to traffic emissions.

Addendum: Relationship with other environmental issues

PM_{2.5} is related to other environmental issues besides human health; a reduction in PM_{2.5} levels would also affect those issues. The new Air Quality Directive mentions proposed new national emission ceilings (NEC) for trace gases, which could greatly affect PM_{2.5} levels. This is because half or even more of PM_{2.5} derives from gases like SO₂, NO_x, NH₃ and VOCs. This issue is addressed in detail in chapters 3 and 4. PM_{2.5} affects climate in the sense that it reflects and absorbs solar radiation/energy (see text box **The influence of aerosols on climate**).

2 LEVELS AND MEASUREMENTS

This chapter addresses the current knowledge about $PM_{2.5}$ levels in the EU in general, but focuses primarily on the Netherlands and its neighboring regions. The $PM_{2.5}$ measurement requirements, as they apply to the Netherlands, are discussed. In a final section, the composition of $PM_{2.5}$ is briefly addressed.

2.1 Current levels of $PM_{2.5}$

Levels in the EU

Member States have started measuring $PM_{2.5}$ because of requirements in the current Daughter Directive for PM_{10} (EU, 2001). In this context, data have been reported to the European Environmental Agency, which has placed the data in the publicly accessible database Airbase (Airbase, 2007).

For 2005, the number of $PM_{2.5}$ measurement series reported to Airbase is about a factor of 10 smaller than the number available for PM_{10} (see the maps for $PM_{2.5}$ and PM_{10} in Figure 2.1). While the basis for an assessment of $PM_{2.5}$ levels Europe-wide is still rather small, the AirBase data show that in several countries the current concentrations are higher than the proposed limit value of $25 \mu\text{g}/\text{m}^3$.

Exceedances of the $25 \mu\text{g}/\text{m}^3$ level occur specifically in highly industrialized regions in Central Europe and at urban sites in Southern Europe. It is obvious that there are even more sites at which the concentration level of $20 \mu\text{g}/\text{m}^3$ is exceeded. In Germany, for instance, current levels are close to the $25 \mu\text{g}/\text{m}^3$ level. Low levels are typically found in the less populated countries in Northern Europe. Data from the Netherlands are not yet available in Airbase, but are presented below.

The value of the $PM_{2.5}$ data is questionable, because most of these were not obtained according to the official measuring guideline, but come from automated monitors; in general, these monitors systematically underestimate the levels (see section 2.2)¹.

Levels in the Netherlands and neighboring regions

As an indication of the differences at the various scales, the data have been subdivided according to regional, urban and street locations. Due to the limited national

¹ While the Airbase data-set contains data for sites for which the data availability for a given year is more than 75%, the Directive only mentions that the capture should be more than 90%, excluding servicing of instrumentation. It does not give a standard for data availability.

information, data from two neighboring regions, Flanders/Brussels and North Rhine-Westphalia, have also been analyzed and are shown in Figure 2.2.

PM_{2.5} networks and data

In 2004, monitoring of PM_{2.5} started in the Dutch national air quality network (LML, 2007). In addition to the national network, PM_{2.5} is also monitored in the Netherlands by the local networks in Amsterdam and the Rotterdam area. Since 2002, they have reported the yearly average PM_{2.5} concentrations (GGD Amsterdam, 2007; DCMR, 2007). The data capture is on average better than the required 90%. All measurements provided by the networks concern raw data, in the sense that an equivalence factor has not been used. This factor, which translates raw data into values which would be obtained with the reference method, is discussed in detail in section 2.2.

The data from the Netherlands (RIVM, 2008) and Belgium (IRCEL, 2007) were obtained with automated monitors and have been corrected with a provisional equivalence factor (Velders et al., 2007b).

The data from North Rhine-Westphalia were obtained with manual gravimetric samplers (LANUV NRW, 2007) that approached the official reference samplers (section 2.2). Data capture of the gravimetric samplers in North Rhine-Westphalia was 20% on average. These data have been averaged per station over the period of measurement (2002-2006) to arrive at a single representative value; this value can be compared with the more extended annual data from Belgium and the Netherlands that derive from a high data capture of over 80%.

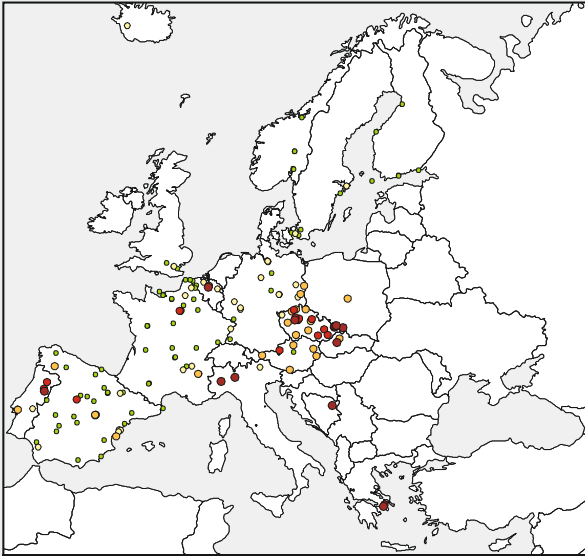
Levels at regional and urban background locations

As can be seen in Figure 2.2, the PM_{2.5} concentrations at the regional background sites range between 12 and 18 µg/m³. In urban agglomerations, the ranges are larger and fluctuate around an average concentration of about 18 µg/m³. During the period October 2002 to March 2004, concentrations of 19 to 21 µg/m³ were measured at sites in the Amsterdam metropolitan area (Puustinen et al., 2007). These are at the higher limit for an urban agglomeration in the Netherlands (Figure 2.2, left).

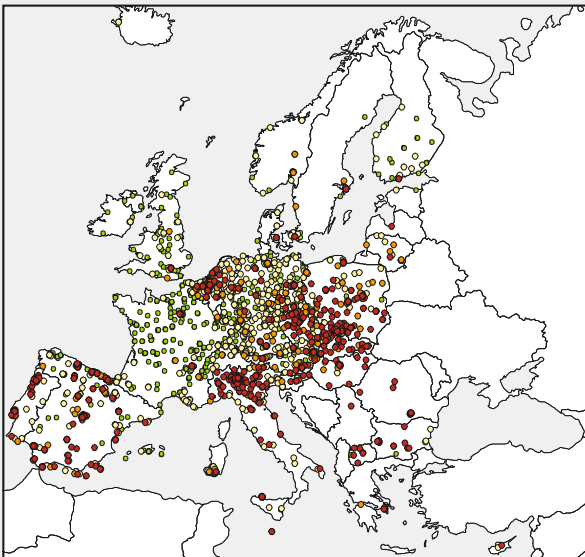
The data suggest a concentration increment in urban background areas with respect to the regional background (urban increment) of around 4 to 5 µg/m³ in all three regions. Model studies, however, suggest a somewhat smaller urban increment range (see chapter 3).

Levels at street locations

PM_{2.5} concentrations measured at the two street sites in North Rhine-Westphalia are on average 25 µg/m³. The three street sites in Belgian urban agglomerations range between 18 and 28 µg/m³. In the Netherlands, PM_{2.5} concentrations in streets (2 sites) are at the lower end of the range of the levels at street locations in the other regions.

Annual mean $PM_{2.5}$ concentration in 2005

- > 30 $\mu\text{g}/\text{m}^3$
- 25 - 30
- 20 - 25
- 15 - 20
- 0 - 15

 PM_{10} , number of exceedance days (concentration higher than 50 $\mu\text{g}/\text{m}^3$) in 2005

- > 50 days
- 35 - 50
- 7 - 35
- 0 - 7

Figure 2.1 Upper panel: $PM_{2.5}$ concentrations (annual mean) in 2005. All stations with data coverage of more than 75% are included. Lower panel: PM_{10} , number of exceedance days (with concentrations above 50 $\mu\text{g}/\text{m}^3$) in 2005 – exceedances of the most stringent limit value for PM_{10} . Source: Airbase.

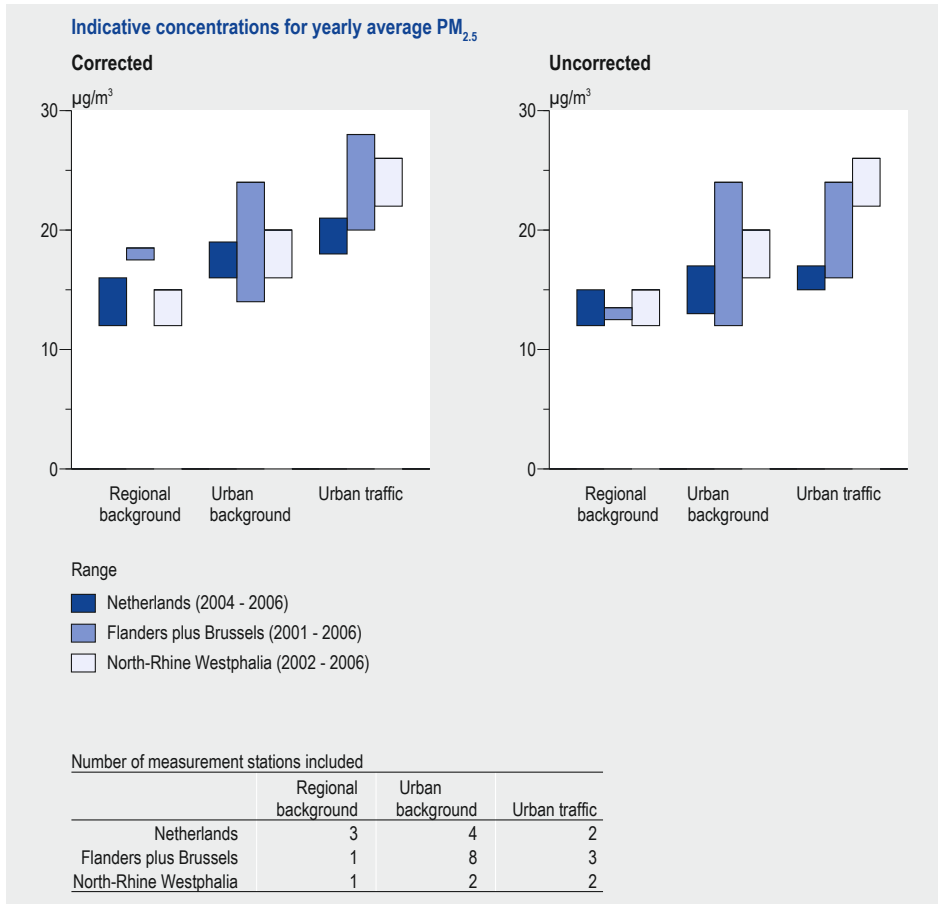


Figure 2.2 Indicative concentrations ranges for yearly average $PM_{2.5}$ ($\mu\text{g}/\text{m}^3$) based on measurements in the Netherlands and neighboring regions in Germany and Belgium. On the right, the original, uncorrected concentration ranges. On the left, the corrected data ranges. Urban background stations include suburban locations.

The variability of $PM_{2.5}$ concentrations in streets in the Netherlands has only been preliminarily explored by means of model calculations (see chapter 3). Literature data show that the additional $PM_{2.5}$ burden in streets, compared to urban background air, can vary considerably, i.e. from $1 \mu\text{g}/\text{m}^3$ to more than $20 \mu\text{g}/\text{m}^3$. The available measurements for street increments in the Netherlands and adjacent regions indicate a range of 2 to $7 \mu\text{g}/\text{m}^3$ (Table 2.1).

Measured street increments are unlikely to be comparable for cities across Europe (UN-ECE, 2007) or even within the same city, because these increments strongly depend on the location of the monitoring site with respect to the local traffic emissions. Climate differences may also account for large variations in $PM_{2.5}$ levels and composition for similar traffic flow and siting criteria (e.g. Harrison et al., 2004).

Table 2.1 Indicative values for the traffic increments of $PM_{2.5}$ ($\mu\text{g}/\text{m}^3$) in the Netherlands, Flanders, Brussels and North Rhine-Westphalia based on measurements (see also Figure 2.2). The street increment is defined here as the difference between $PM_{2.5}$ concentrations measured at urban street sites and urban/suburban background locations. The street increment is an indication of the contribution of local traffic emissions to $PM_{2.5}$. It should be noted that the number of stations used for this comparison is very limited.

| | Street increment |
|------------------------|------------------|
| Netherlands | 5-6 |
| Flanders, Brussels | 2-6 |
| North Rhine-Westphalia | 5-7 |

The data acquired so far are clearly limited. Consequently, they should be regarded as indicative and are only suitable for an initial analysis of the $PM_{2.5}$ concentrations. In this respect, it should also be mentioned that the German and Belgium regional background levels are based on measurements from only one station.

The ratio of $PM_{2.5}$ and PM_{10} concentrations

The ratio of $PM_{2.5}$ to PM_{10} is important due to the stringency of legislation on the two parameters. The networks in our country provide data for both $PM_{2.5}$ and PM_{10} . The data from North Rhine-Westphalia on the ratio of $PM_{2.5}/PM_{10}$ are also from previous measurements (Kuhlbusch et al., 2000). This information suggests that the ratio of $PM_{2.5}$ to PM_{10} is 0.7, with a range between 0.6 at urban/traffic sites and 0.8 at the regional sites. Given the scarcity of data and the absence of a reference method and equivalence data, the estimates of the ratio of $PM_{2.5}$ to PM_{10} are device-dependent. Taking all the data together, we have arrived at an average ratio between 0.7 and 0.75. At the traffic sites, the ratio varies between 0.6 and 0.8. The rather large amount of network data from Belgium has not yet been analyzed, but it could help to reduce the uncertainty.

Airbase data and scientific studies (Putaud et al., 2004; Querol et al., 2004) indicate that the ratio of $PM_{2.5}$ to PM_{10} in Southern Europe is often less than 50%. This is due to the dry soil in this region, which leads to increased re-suspension of dust in the PM fraction between 2.5 to 10 μm . Similarly, in the Nordic countries the ratio can be low, with a larger fraction of coarse PM. This is caused by the re-suspension of coarse road salt particles. In coastal areas, high sea salt concentrations lower the $PM_{2.5}/PM_{10}$ ratio.

2.2 How can we measure $PM_{2.5}$?

The technical sections in the common position adopted by the Council state that the measuring procedure will follow, and very likely not deviate from, the existing EN guideline, which is also valid in the Netherlands as a NEN standard (NEN-14907, 2005).

Uncertainty requirements

European regulations allow a maximum uncertainty of 25% in the data, but the national reduction target for the Netherlands is 20%. Since the uncertainty allowed in the measurements is larger than the target reduction, it would appear that a reduction of this magnitude will not be easily measurable with these means. *Note:* As part of the review in 2013, the experience with monitoring $PM_{2.5}$ will be reported. If appropriate, new reference methods for measuring $PM_{2.5}$ will be proposed that could offer more precision.

Reference method

The reference measurement method for $PM_{2.5}$ is comparable with that for PM_{10} , in the sense that it involves collection (of $PM_{2.5}$) by drawing air through a filter and weighing the amount of PM captured. In case of $PM_{2.5}$, a device will be placed in the inlet that transmits 50% of the particles with a diameter of 2.5 μm , with a sharp cut-off of larger particulate matter. Improved quality assurance is central to the new guideline for $PM_{2.5}$; excessive heating of samples and sampling air, which are associated with evaporation of particulate matter, will be avoided. This follows the sampling guidelines used in the United States.

From a technical perspective, however, there could be a complication. This concerns the material of the filter. Three types of material are allowed, for which significant differences were found in the amount of material sampled (also by the scientific community in Europe). The use of different types of filters could introduce a bias in the data per Member State and complicate the comparability of levels between Member States. For more information, see the text box **Measurement problems due to semi-volatile components in particulate matter**.

Automated measuring methods and equivalence

Most networks in the EU use automated monitors for measurement. The monitors available are very similar to those for PM_{10} , although with a different inlet. The national network and the regional authorities also use such automated instruments. For that matter, all data for $PM_{2.5}$ mentioned in this report are from automated monitors. A central question is the comparability of the data that are obtained with such automated monitors with data from the manual reference method. The major complication is that the air has to be dried. In most monitors, drying occurs by heating. This leads to evaporation of semi-volatile compounds (see text box **Measurement problems due to semi-volatile components in particulate matter**). Monitoring instruments thus provide lower values.

The commission that prepared the guideline has stated that the equivalence factor for $PM_{2.5}$ is very likely higher than that for PM_{10} . This is based on the fact that $PM_{2.5}$ contains relatively more volatile material than PM_{10} .

Remote sensing

Satellites that observe the earth are also able to detect PM. An advantage of satellite measurements is that a broad area can be viewed. Information from satellites can thus be useful to complement measurements from ground-based networks (e.g. Koelemeijer et al., 2006). Two drawbacks are that current satellite observations are 1) limited to a few per day per location and 2) not easily linked to $PM_{2.5}$ concentrations at ground level.

Consequences for the Netherlands

The Netherlands has yet to decide on a monitor for $PM_{2.5}$. Preparations for this decision are ongoing in the Netherlands and a choice harmonized with local networks is anticipated. The $PM_{2.5}$ concentration, measured with automatic samplers, must be compared with the reference method according to the European standard (NEN-14907, 2005).

Measurement problems due to semi-volatile components in particulate matter

Automated monitors and equivalence

Most national air quality networks use automated monitors for PM_{10} . For $PM_{2.5}$ measurements it is likely that the same monitor types will be used, but with a different inlet size. For automated monitoring of particulate matter, there are two monitoring methods that are named after the method used to determine the on-line accumulation of mass on a filter:

- the Beta-Attenuation Monitor (BAM, also known as beta-gauge and FAG)
- the Tapered Element Oscillating Microbalance (TEOM).

There is a major complication with the data from these automated monitors: filtered mass must be dried. Drying is usually accomplished by heating. This is a source of error, because the values obtained with the monitors are usually substantially lower than those obtained with the reference filter method. This is because semi-volatile compounds evaporate due to the heating. These losses of semi-volatile compounds are corrected to the 'actual' concentration as it would be measured with the reference method. The accepted overall average value of the difference (loss) for PM_{10} in automated monitors in the EU is 30%.

However, there has been a recent development. The amount of semi-volatile material that disappears during measurement with a TEOM can be determined and added back to the measurement. This modification is known as the Filter Dynamics Measurement System (TEOM-FDMS). Some of the data presented here (in Figure 2.2) are from this advanced instrument.

It should be noted that the chemical composition of $PM_{2.5}$ is significantly different from that of PM_{10} . In particular, the relative amount of semi-volatile particulate matter (e.g. ammonium nitrate) is higher in the $PM_{2.5}$ fraction. The particulate matter in the size range between PM_{10} and $PM_{2.5}$ mainly consists of inert components such as silica and metal oxides. Consequently, the problems with losses of semi-volatile matter already observed when sampling PM_{10} will probably be more pronounced for $PM_{2.5}$ measurements. The calibration factor for $PM_{2.5}$ is therefore expected to be substantially higher than that for PM_{10} . Detailed information on evaporation, as well as adsorption artefacts and their dependence on filter-type, can be found in Chow (1995) and Ten Brink et al. (2004).

2.3 Chemical composition of PM_{2.5}

Besides requiring PM_{2.5} monitoring, the Directive also calls for measurements of the chemical composition of PM_{2.5} at a regional site. At the central site at Cabauw, intermittent compositional measurements of particulate matter are being performed as part of air quality and climate-change research in Europe (e.g. CESAR, 2007). Cabauw would be a good candidate for such a regional site.

Required composition measurements

Measurements are required of the following minimum list of chemical species (which are part of PM_{2.5}): sulfate (SO₄²⁻), sodium (Na⁺), ammonium (NH₄⁺), calcium (Ca²⁺), elemental carbon (EC), nitrate (NO₃⁻), potassium (K⁺), chloride (Cl⁻), magnesium (Mg²⁺) and organic carbon (OC). The list shows that PM_{2.5} is composed of a variety of chemical substances. Compositional analysis allows the assessment of sources and is also a check on the mass measurement, i.e. the sum of the mass of the chemical compounds should be equal to the directly measured mass.

Regarding data from the Netherlands on the composition of PM_{2.5}, there is information dating back to the mid-1990s (Erisman et al., 1996). The most comprehensive database on the composition of PM_{2.5} in the Netherlands is provided by Visser et al. (2001) for the period 1998-1999. Measurements were performed on the same day at six different sites in the Netherlands, thereby providing information on the spatial gradient in the composition.

SIA and carbon

The current knowledge on the composition is summarized in Figure 2.3. A general conclusion is that more than 75% of the regional PM_{2.5} is composed of SIA and carbon. SIA is the dominating component. SIA stands for Secondary Inorganic Aerosol and comprises the combination of sulfate, nitrate and ammonium. The SIA compounds are present as ammonium sulfate and ammonium nitrate. The latter compound is semi-volatile and may evaporate during measurement.

The term *carbon* is used here for a very complex mixture of compounds that is often reduced to two main categories: elemental carbon and organic carbon. Elemental carbon contributes less to the carbon (approximately 20%). It derives mostly from diesel traffic emissions. A useful first assessment of the carbon deriving from fossil fuel combustion versus that from so-called biogenic sources, specifically wood-burning, is made using isotope (¹⁴C) analysis (Szidat et al., 2004).

Recent data on the carbon component can be found in Sillanpää et al. (2005). The data were collected at the same site and within the period of the previously mentioned campaign of Puustinen et al. (2007). Of specific interest are the measurements of carbon

at Kollumerwaard, a rural background site (EMEP, 2006a). A very recent measurement campaign at a traffic site along the ring-road, A10-south, in Amsterdam, is described by Viana et al. (2007).

At street sites, the concentration of the carbon fraction is higher than the regional concentrations, but insufficient data are available for assessing the absolute magnitude of the increment.

The contribution of carbon is the most uncertain in an absolute sense, due to the small data base and complexity of the measurement. Its estimated uncertainty is 35%. New information on the composition will become available in the near future from studies in the BOP program (BOP, 2007) that will commence in the near future.

Sea salt and mineral dust

Sea salt and mineral dust are of lesser importance. The other substances mentioned in the Directive are so-called tracers for source categories. Potassium is a possible tracer for wood combustion. Magnesium, in combination with calcium, is a tracer for sea salt.

Water

Water forms a normal part of particulate matter, but it is not included in Figure 2.3 because the amount is very uncertain and depends on the measurement method. Water binds to hydrophilic components in particulate matter like sulfate, ammonium, nitrate and sea salt. Abating SO_2 , NO_x and NH_3 lowers the concentration of their secondary particulate components and therefore reduces $\text{PM}_{2.5}$. Lower secondary levels may also reduce the uptake of water by fine particles. This leads, in turn, to a further reduction in $\text{PM}_{2.5}$ concentration. In this way water can magnify trends in secondary particulate matter. The amount of water associated with SIA is, however, highly uncertain; for details, see the text box **On the natural component in $\text{PM}_{2.5}$** .

The overall composition of $\text{PM}_{2.5}$ in the neighboring regions appears to be quite similar to that in the Netherlands (Flanders: Maenhaut, 2006; North Rhine-Westphalia: Quass and Kuhlbusch, 2004). However, the concentration of sea salt is lower than in the Netherlands.

Natural components of $\text{PM}_{2.5}$

The contribution of natural components to $\text{PM}_{2.5}$ is much smaller than to PM_{10} . Only the contribution of sea salt is substantial. The average concentration of sea salt in $\text{PM}_{2.5}$ in the Netherlands is around $1 \mu\text{g}/\text{m}^3$, with a contribution of material deriving from marine organisms of less than 10%. There are insufficient data as yet to assess a gradient in sea salt over the Netherlands. The contribution of natural mineral dust and

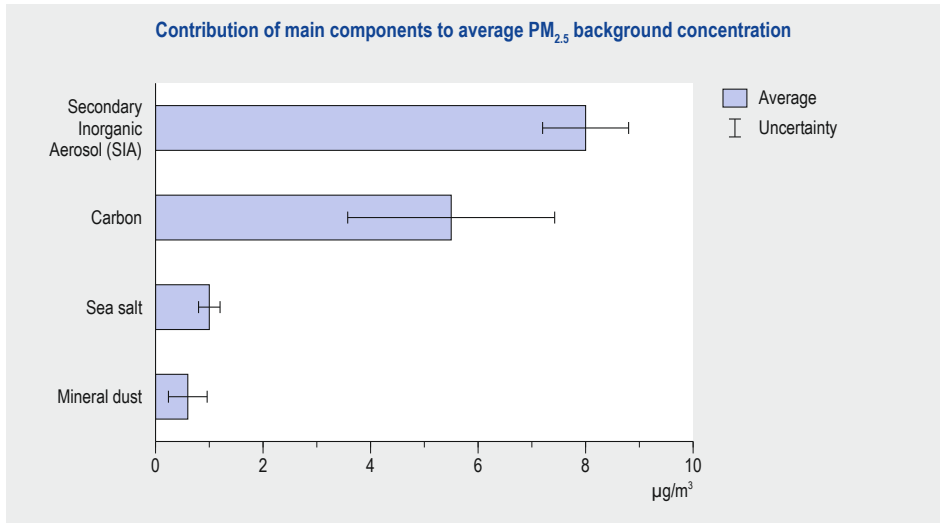


Figure 2.3 Best estimate of the contribution ($\mu\text{g}/\text{m}^3$) of the main components to the $PM_{2.5}$ background concentration in the centre of the Netherlands. The upper and lower margins are uncertainty ranges. Estimates and uncertainty ranges are based on all available information in the last decade, including that from neighboring regions, extrapolated to the year 2005.

carbon to the $PM_{2.5}$ levels is very likely negligible. Details for these analyses can be found in the text box **On the natural component in $PM_{2.5}$** .

In the text box, the major natural component of PM – water – is also described. It is not considered as being health-relevant and can therefore be excluded when the mass is determined. Nevertheless, water appears to be measured and this is a major complicating factor in assessing the mass concentration of $PM_{2.5}$ as described in section 2.2.

On the natural components of $PM_{2.5}$

Sea salt

The major natural source of PM (in the Netherlands) is sea salt. It usually appears in the form of large particles. However, a fraction also resides in $PM_{2.5}$. An initial estimate, which is based on data from the Netherlands, neighboring countries and model studies, indicates that the average ratio of sea salt in $PM_{2.5}$ and that in PM_{10} is between 1:2 and 1:4. Sea salt is mainly composed of sodium chloride. In the sea salt in $PM_{2.5}$, chloride is partly replaced by manmade nitrate and sulfate. The concentration of the natural material is thus less than that based on the concentration of sodium, which is the standard procedure. The average contribution of sea salt to $PM_{2.5}$ is about $1 \mu\text{g}/\text{m}^3 \pm 20\%$ in the Netherlands. Some natural marine material derives from gases that are produced by algae. The contribution of this marine material is estimated to be less than $0.1 \mu\text{g}/\text{m}^3$.

Mineral dust

Another possibly natural PM component is mineral dust. This component is present mostly in PM_{10} . In addition, it is impossible to distinguish natural dust from dust that is suspended by human activities, vehicles and agricultural activities. The total contribution of mineral dust to $PM_{2.5}$ is small ($0.6 \mu\text{g}/\text{m}^3 \pm 60\%$); the natural contribution is therefore even smaller.

Carbon

Compounds that originate in the biosphere (plants/trees) form a third class of theoretically natural components. When stirred under very dry conditions, plant debris is a source of particulate matter. However, the debris is almost completely contained in the size fraction between $PM_{2.5}$ and PM_{10} , and therefore not in $PM_{2.5}$. Natural carbon in the $PM_{2.5}$

fraction is also a result of reactions of gases, emitted by trees, forming secondary particles. This fraction is known as biogenic secondary organic aerosol. Carbon can also derive from forest fires; this has been shown to be case in other European countries.

According to the definitions of the International Panel for Climate Change, emissions that derive from agriculture and forestry should be considered as anthropogenic rather than natural. For that reason, we estimate that the contribution of truly natural carbon in the Netherlands is probably negligibly small.

Water

Water is the major component of atmospheric $PM_{2.5}$ in the Netherlands. However, this water has to be removed before the mass of $PM_{2.5}$ is determined. There is a complication, because some water is tightly bound to components like ammonium nitrate. The amount of water in collected $PM_{2.5}$ is therefore not measured directly, but estimated as follows. The mass of all chemical compounds is added and compared to the directly measured mass. The difference in mass is attributed to water. In this way we arrive at a water content of up to 20% in $PM_{2.5}$ in the Netherlands, based on an extrapolation of data from the neighboring regions. However, recent information shows that this water may not be part of $PM_{2.5}$ (De Jonge, GGD Amsterdam, report in preparation). This study demonstrated that quartz-fiber filters adsorb water vapor during collection of $PM_{2.5}$ and part of this water is retained during drying. This adsorbed water vapor is then erroneously counted as water associated with $PM_{2.5}$. In summary, the contribution of water to $PM_{2.5}$ is highly uncertain, but potentially large.

3 CALCULATING PM_{2.5} LEVELS USING MODELS

This chapter discusses the model results of yearly average PM_{2.5} concentrations in the Netherlands for the years 2006, 2010, 2015 and 2020. For this purpose, the OPS model (Van Jaarsveld, 2004) was used.

This chapter addresses the following aspects:

- The contribution to PM_{2.5} of present and future anthropogenic primary PM_{2.5} emissions and their origin.
- The origin of the secondary particulates sulfate nitrate and ammonium, and their contribution to PM_{2.5}.
- The effect of current legislation on future PM_{2.5} levels.
- PM_{2.5} concentrations in urban agglomerations.
- PM_{2.5} concentrations due to local sources.
- The uncertainty in the PM_{2.5} concentration maps which have been derived for the Netherlands.

3.1 Introduction

The dispersion of PM_{2.5} is in many ways similar to the dispersion of PM₁₀. However, because PM_{2.5} excludes the heavier coarse particles, it can be transported over longer distances than PM₁₀. Typical transport distances for PM_{2.5} are about 2500 km, whereas characteristic transport scales for coarse particles are 500 to 1000 km. Therefore, distant sources which contribute to PM_{2.5} are relatively more important for PM_{2.5} than for PM₁₀. Under unusual conditions, particulate matter can be transported over even larger distances, such as the transport of Saharan dust into Europe. Removal from the air of particulate matter takes place by dry deposition and precipitation (rain, etc.).

The OPS model calculates regional and urban background PM_{2.5} concentrations using registered anthropogenic emissions for Europe. The model estimate is calibrated using measurements. For more details on the calculation methodology of the OPS model, see the text box **Methodology for calculating PM_{2.5} concentrations**.

The model results allow a source apportionment of the PM_{2.5} concentrations resulting from the emissions. Model results for both present emission levels and projections show how PM_{2.5} concentrations are believed to be brought about now and how they may evolve in the future. It should be noted that model results are very uncertain due to uncertainties in the model itself with regard to PM_{2.5} and due to important shortcomings of PM_{2.5} emission and monitoring data.

Models for the assessment of particulate matter

OPS model. The Netherlands Environmental Assessment agency (MNP) uses this model (Van Jaarsveld, 2004) to generate annual maps showing the large-scale concentrations of several air quality components in the Netherlands that are subject to European regulations (e.g. Velders et al., 2007a). Local, provincial and other authorities use these maps for reporting exceedances as part of the EU Air Quality Directives and for planning. The OPS model provides a much higher resolution than the EMEP model, which is used in preparing policy for Europe. The results of the OPS model are limited to the air quality in the country itself. This aspect plays a role, for example, in determining the Dutch standpoint in Brussels. OPS model results have been compared with the EMEP results for PM_{2.5}.

EMEP model. The unified EMEP model (EMEP, 2003) is a chemical transport model for the European domain. For European policy development, the EMEP model results are used as input for the integrated assessment model GAINS. Yearly assessments for different air pollutants are derived from EMEP model calculations for the European domain and for each Member State (EMEP, 2006a, 2006b and 2006c). The EMEP model therefore plays an important role in preparing policy for the Euro-

pean Union. PM_{2.5} is part of the model output on a resolution of 50x50 km².

RAINS/GAINS models. The RAINS/GAINS models (RAINS/GAINS, 2007) are integrated assessment models that calculate air quality and climate forcing (GAINS) for the entire European land area (Wagner et al., 2006 and 2007). The models generate integrated evaluations of emissions across the entire chain, from source to effect and the reverse, as well as generating mitigation scenarios. For national use, a RAINS/GAINS version which focuses on the Netherlands has been made available (RAINS-NL and GAINS-NL; Aben et al., 2005).

LOTOS-EUROS model. The LOTOS-EUROS model (Schaap et al., 2005) is a chemical transport model for the European domain. It is generally used in the Netherlands for research purposes and policy support (e.g. Schaap and Denier van der Gon, 2007). EUROS-LOTOS is used in the Netherlands research program on PM₁₀ and PM_{2.5} (BOP, 2007). A recent evaluation of long-term PM simulations from seven regional air quality models for Europe includes results from LOTOS-EUROS and the EMEP model (Schaap et al., 2007).

3.2 The role of model calculations

Model calculations are used to evaluate and explore environmental policy, and they are essential to the interpretation of measurement data and understanding physical and chemical processes that determine the PM_{2.5} levels. Moreover, the Netherlands previously chose to use models, in addition to measured data, to ascertain air quality in order to report this to the European Commission. The OPS model is the operational model for the background concentrations which are used in these reports. For the Netherlands, other models are also relevant for the support of national policy measures regarding particulate matter (for more details, see text box **Models for the assessment of particulate matter**).

3.3 Background concentrations of PM_{2.5}

The national average PM_{2.5} concentration in 2006 has been calculated to be 15-16 µg/m³. Figure 3.1 shows a breakdown into contributions per sector. The spatial distribution of PM_{2.5} background concentrations in the Netherlands (Figure 3.2) has a pattern similar to that of the PM₁₀ background concentrations. Background concentrations vary between 11 and 20 µg/m³. Large areas in the middle and south

of the Netherlands have background concentrations above 16 $\mu\text{g}/\text{m}^3$. In parts of the main urban agglomeration, *de Randstad*, background concentrations are higher than 18 $\mu\text{g}/\text{m}^3$. The contribution of local emissions is additional and can reach 14 $\mu\text{g}/\text{m}^3$ in street canyons. Future traffic contribution to PM_{2.5} may be considerably reduced due to the expected effects of increased emission restrictions for cars and heavy duty vehicles.

Source contribution

About 70% of the PM_{2.5} background concentrations can be attributed to registered anthropogenic sources in Europe; for PM₁₀ this is only 50%. Consequently the ability to abate PM_{2.5} concentrations on a European scale is larger than for PM₁₀. About 20% of the PM_{2.5} concentration can be attributed to Dutch anthropogenic sources (3 $\mu\text{g}/\text{m}^3$), while 50% percent originates from anthropogenic sources abroad and from ocean shipping (7.5 $\mu\text{g}/\text{m}^3$). The ocean shipping sector comprises emissions

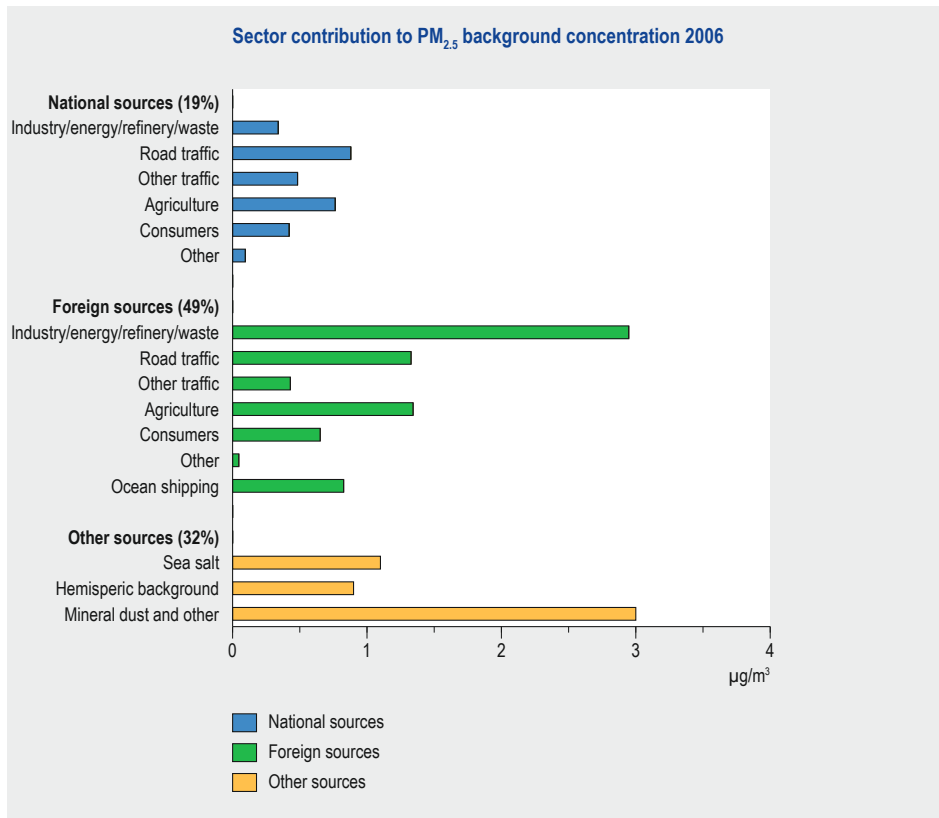


Figure 3.1 Average contributions of anthropogenic and natural sources to background PM_{2.5} concentrations in the Netherlands in 2006. The ocean shipping sector comprises emissions from shipping on Dutch territory, seagoing ships and ships moored in harbors. Emissions from ocean shipping are not included in the EU directives. Sea salt estimates are based on the sea salt contribution to PM₁₀ and a fixed contribution to PM_{2.5} of 25%.

from shipping on Dutch territory, seagoing ships and ships moored in harbors. The rest represents natural and other sources ($5 \mu\text{g}/\text{m}^3$), and is about 30% of the average PM_{2.5} concentration in the Netherlands. The contribution from sea salt and the northern hemispheric background (together $2 \mu\text{g}/\text{m}^3$) cannot be abated.

Similar to PM₁₀, the Industry, Energy and Refineries sector provides the largest contribution from abroad to PM_{2.5} ($3 \mu\text{g}/\text{m}^3$). Other important sources are the road transport and agriculture sectors, both in the Netherlands and abroad. The contribution of ocean shipping to PM_{2.5} in 2006 was about $1 \mu\text{g}/\text{m}^3$ (5%).

Concentration developments between 2010 and 2020

If based on current legislation, emission scenarios for 2010 to 2020 lead to a gradual reduction of PM_{2.5} concentrations during 10 years of about $1 \mu\text{g}/\text{m}^3$ on average (Figure 3.2). The modeled effects of relative emission changes are believed to be fairly reliable, which is not the case with the absolute modeled concentration levels. In 2010, PM_{2.5} concentration levels are already expected to be $2 \mu\text{g}/\text{m}^3$ lower, on average, than in 2006. The average background concentrations in 2010, 2015 and 2020 (Figure 3.2) are expected to be 13.5, 13 and $12.5 \mu\text{g}/\text{m}^3$, respectively. Under current legislation the largest reduction ($1 \mu\text{g}/\text{m}^3$) of PM_{2.5} concentrations between 2010 and 2020 will be related to the transport sector. The second largest reduction ($0.3 \mu\text{g}/\text{m}^3$) will take place due to emission reductions in the Industry/Energy/Refineries abroad sector. Increased contributions are expected from ocean shipping ($0.2 \mu\text{g}/\text{m}^3$) and Dutch agricultural emissions ($0.1 \mu\text{g}/\text{m}^3$).

3.4 Composition and sources

In 2006, about $10\text{--}11 \mu\text{g}/\text{m}^3$ of PM_{2.5} background concentrations were attributed to registered anthropogenic emissions in Europe (Figure 3.3). This component is composed of $7.5 \mu\text{g}/\text{m}^3$ secondary particulates (nitrate, sulfate and ammonium) and $3 \mu\text{g}/\text{m}^3$ primary PM_{2.5}. Consequently, to reduce the levels of fine particulate matter in ambient air, the abatement of the emissions of the gaseous precursors is a key factor.

The majority of the secondary particulates ($6 \mu\text{g}/\text{m}^3$) originates abroad or on the sea. Of the secondary particulates in PM_{2.5}, nitrate is the most abundant ($3.2 \mu\text{g}/\text{m}^3$). About 40% ($1.3 \mu\text{g}/\text{m}^3$) of primary PM_{2.5} originates from land-based sources in the Netherlands, while 60% ($1.8 \mu\text{g}/\text{m}^3$) is from sources abroad or on the sea. At the local scale, for instance in streets, PM_{2.5} concentrations may be elevated mainly due to local emissions of primary particles.

Like PM₁₀, PM_{2.5} contains contributions of sea salt, hemispheric background, mineral dust, and other substances such as water and secondary organic aerosol (see chapter 2). An estimate of the total contribution of these fractions is $5 \mu\text{g}/\text{m}^3$, with a range of $2.5\text{--}7.5 \mu\text{g}/\text{m}^3$ (see the text box **Methodology for calculating PM_{2.5} concentrations**).

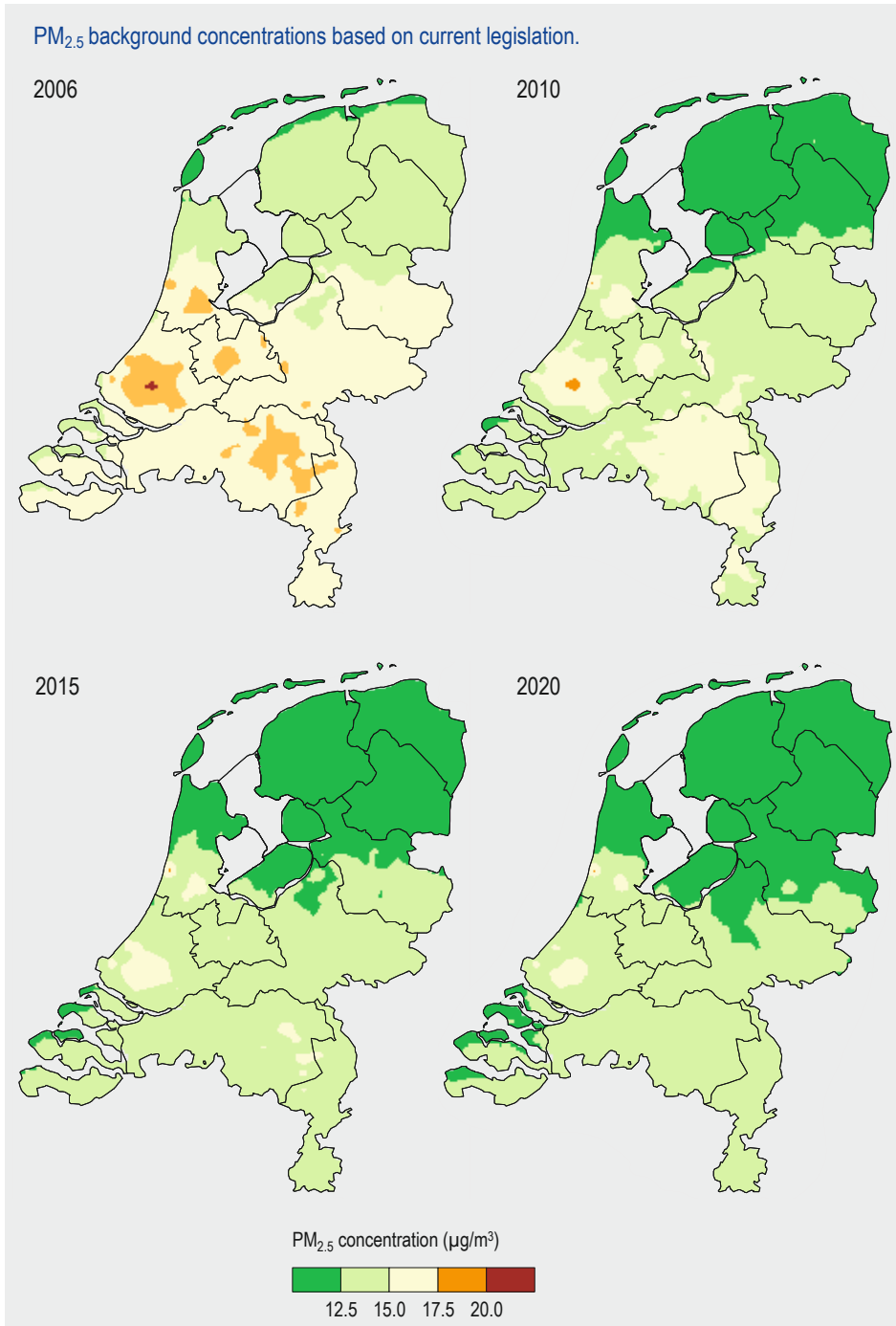


Figure 3.2 Maps of the PM_{2.5} background concentrations in 2006, 2010, 2015 and 2020 based on current legislation. To represent the contribution of natural and other sources, the modeled contribution of anthropogenic PM_{2.5} is augmented with $5 \mu\text{g}/\text{m}^3$ ($\pm 2.5 \mu\text{g}/\text{m}^3$). This means that a concentration of $17.5 \mu\text{g}/\text{m}^3$ could be, with a certain likelihood, as high as $20 \mu\text{g}/\text{m}^3$ or as low as $15 \mu\text{g}/\text{m}^3$. Note that legend colors change with the same amount as the uncertainty.

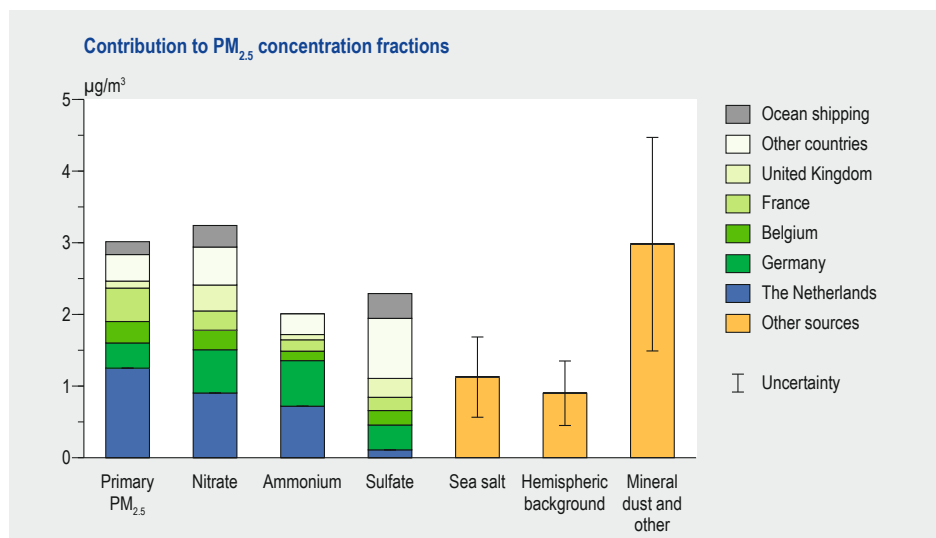


Figure 3.3 Contribution to different fractions of PM_{2.5} in the Netherlands in 2006 of various EU countries and ocean shipping (Velders et al., 2007b). The ocean shipping sector comprises emissions from shipping on Dutch territory, seagoing ships and ships moored in harbors. Emissions from ocean shipping are not included in the EU directives. Sea salt estimates are based on the sea salt contribution to PM₁₀ and a fixed contribution to PM_{2.5} of 25%.

The sea salt fraction in PM_{2.5} is on average about 1 µg/m³ in the Netherlands, with higher concentrations along the coast and lower concentrations inland. The hemispheric background concentration is due to sources outside Europe and is believed to be about 1 µg/m³ or less in the Netherlands. The contribution of mineral dust to PM_{2.5} is believed to be small (< 1 µg/m³). Secondary organic aerosol in the Netherlands is partly anthropogenic, but is not included in the model calculations. The contribution of the secondary organic aerosol to PM_{2.5} concentrations is expected to be less than 1 µg/m³.

3.5 Concentrations in urban agglomerations

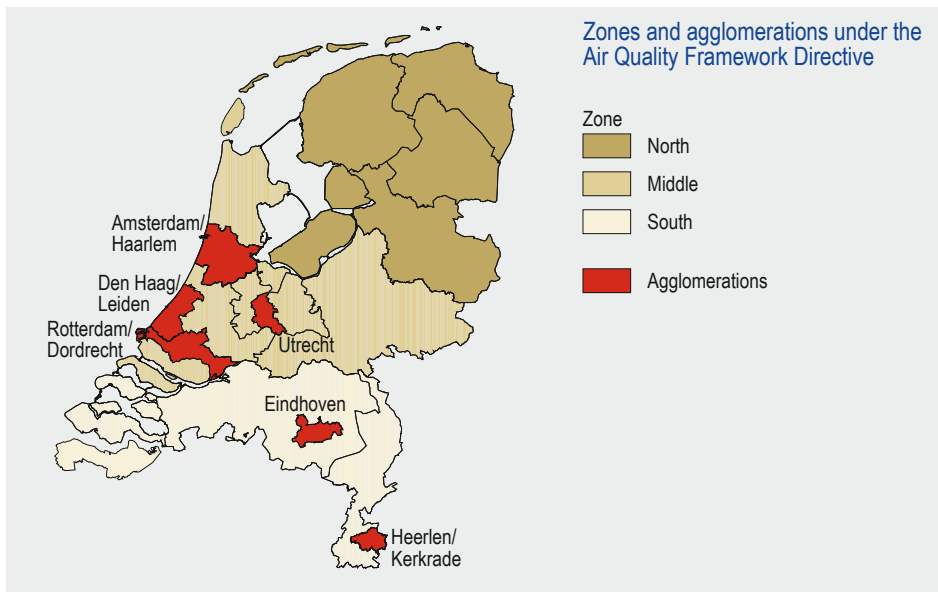
The calculated PM_{2.5} concentrations for the standard urban air quality agglomerations (Figure 3.4) should be seen as the average background concentration in an urban agglomeration and can therefore be associated with the exposure reduction target. PM_{2.5} concentrations in urban agglomerations tend to be higher than in the surrounding region. This leads to a difficult situation because PM_{2.5} levels tend to be highest at densely-populated locations. Unfortunately, emission and measurement data from urban agglomerations are rather uncertain, and the performance of models for urban agglomerations with large emission dynamics and complex terrain is still very weak. This section provides some insight into the urban background concentrations of PM_{2.5} (see also Figure 3.5). Detailed information on calculated sector contributions per urban agglomeration can be found in Velders et al. (2007b).

Estimated concentration reduction between 2010 and 2020

The estimated average concentration reduction between 2010 and 2020 in urban agglomerations ranges between 6 and 10% with current legislation (see Figure 2). Recently outlined additional measures lead to a concentration reduction ranging between 11 and 16%. These reductions do not meet the proposed exposure reduction target of 20%. The percentages are sensitive to the level of implementation of reduction measures in 2010 and the absolute concentrations in 2010. Both are uncertain. However, it seems unlikely that the 20% reduction target will be attained in the Netherlands even if the intended and additional measures, which are outlined in Velders et al. (2007a), are carried out. Consequently, extra emission reduction measures are necessary.

The present calculations show that 30% or less of average urban background concentrations are due to registered national sources. This limits the effect of national policies aimed at attaining the proposed exposure reduction target value and makes attainability largely dependent on the implementation level of policy measures abroad. At the same time, additional measures that go beyond the national goals set by existing European legislation are now not envisioned abroad. However, other Member States probably also need to make plans for further emissions reductions in order to meet the new PM_{2.5} standards. At what time this may lead to substantial extra emission reductions is yet unclear. Therefore, further concentration reductions should only be expected from extra national and local measures in the Netherlands, at least until 2015.

Figure 3.4 Division of the Netherlands into zones and agglomerations in accordance with the Air Quality Framework Directive (Van Breugel and Buijsman, 2001).



Urban increment

The OPS model calculates PM_{2.5} concentrations for the standard air quality agglomerations (Figure 3.4). The modeled concentrations in these agglomerations are on average 1-3 µg/m³ higher than the surrounding regional background concentrations; this is the urban increment. The urban increment varies and depends on factors like city size, topography and meteorology.

Results from an extensive model study of European cities show a similar range of urban increments (Cuvelier et al., 2006; Amann et al., 2007c). The urban increments found for 27 cities in the Netherlands (0.3-3.1 µg/m³) were among the lowest of the 473 cities included in the study. The reason for this is unclear. Urban areas in Belgium and Germany have ranges with somewhat higher increments, 1.2-5.0 µg/m³ and 0.3-4.2 µg/m³, respectively. Concentrations of PM_{2.5} in urban agglomerations, Europe-wide, can be 10 µg/m³ or higher than in regional background locations (Amann et al., 2007c). Urban increments are believed to be mainly due to primary sources. In the Netherlands, important sectors in this respect are road traffic and consumers. Measured urban increments for the Netherlands and adjacent regions (about 4 to 5 µg/m³, see Figure 2.2) seem to be somewhat higher than modeled increments. However, when the uncertainties are taken into account, the modeled urban increments are in agreement with the measurements (which are still limited).

3.6 Contribution of local sources

In addition to the regional and urban background concentrations (Figure 3.2), contributions of local sources like traffic and large animal housings can be substantial. The contribution of low-level primary sources like traffic in densely-populated areas is also believed to be potentially of higher risk.

Local contribution of traffic in streets

Little is known about PM_{2.5} concentrations in streets in the Netherlands. The street increment, additional to the urban background concentration, depends on street type and traffic volume and composition. The available measurements in the Netherlands and adjacent regions give a range from 2 to 7 µg/m³ (see chapter 2).

Model estimates have been derived for the current and future traffic contributions to PM_{2.5} in the Netherlands, see text box **Methodology for calculating PM_{2.5} concentrations**. Current PM_{2.5} street increments range between 2 and 6 µg/m³ for average traffic conditions and between 7 and 14 µg/m³ in very busy streets (98-percentile). In 2015, PM_{2.5} street increments are expected to be about 50% smaller than the current increments: contributions range between 1 and 3 µg/m³ for average traffic conditions and between 3 and 6 µg/m³ in very busy streets. Similar street

increments have been reported in a model study of narrow street canyons conducted by the European Environmental Agency (EEA, 2006a).

Urban background concentrations plus the contribution of traffic

The highest PM_{2.5} concentrations in the Netherlands are probably found in streets in urban agglomerations. Therefore, current and future estimates of PM_{2.5} concentrations in streets are believed to be the best indicator for assessing whether the proposed PM_{2.5} limit value of 25 µg/m³ or the more stringent value of 20 µg/m³ will be attained in the Netherlands. Figure 3.5 shows estimates for calculated PM_{2.5} concentrations in urban agglomerations augmented with a calculated approximation for the street increment due to traffic emission in 2006 and 2015. The concentrations shown are average concentrations and *hot spot* concentrations (98-percentile), calculated for each of the six urban agglomerations (see Figure 3.4). The street contributions are the calculated ranges for an average street and a very busy street (98-percentile), as mentioned above.

Therefore, the upper two graphs indicate possible concentrations for average conditions in 2006 and 2015, while the lower two graphs indicate possible concentrations for very busy streets with high urban background concentrations. The number of locations with *hot spot* concentrations is always much lower than the number of locations with average concentrations.

Limit values and attainability

The data in Figure 3.5 suggest that the proposed limit value of 25 µg/m³ can probably be attained in 2015, except for a very limited number of hot spots. Average concentrations in streets are expected to be even below 20 µg/m³ in 2015. However, under current legislation the more stringent limit value of 20 µg/m³ will probably still be exceeded in busy streets in urban agglomerations. Due to the recently outlined additional measures, the urban background concentrations will decline by an extra 1 µg/m³ in 2015. This will lead to a further decline in the number of exceedances, but they will not be completely eliminated. Even if the additional measures are implemented, the number of exceedances of the 20 µg/m³ limit value is expected to become very limited only by 2020.

Local contribution of animal housings to PM_{2.5}

Bleeker et al. (2006) have estimated the current and future PM₁₀ contribution of large animal housings in a number of provinces in the Netherlands. They concluded that exceedances of the PM₁₀ limit value can take place due to agriculture-related emissions. The exceedances are generally limited to very large animal housings. This study was used to make a preliminary estimate of the effect of local agricultural

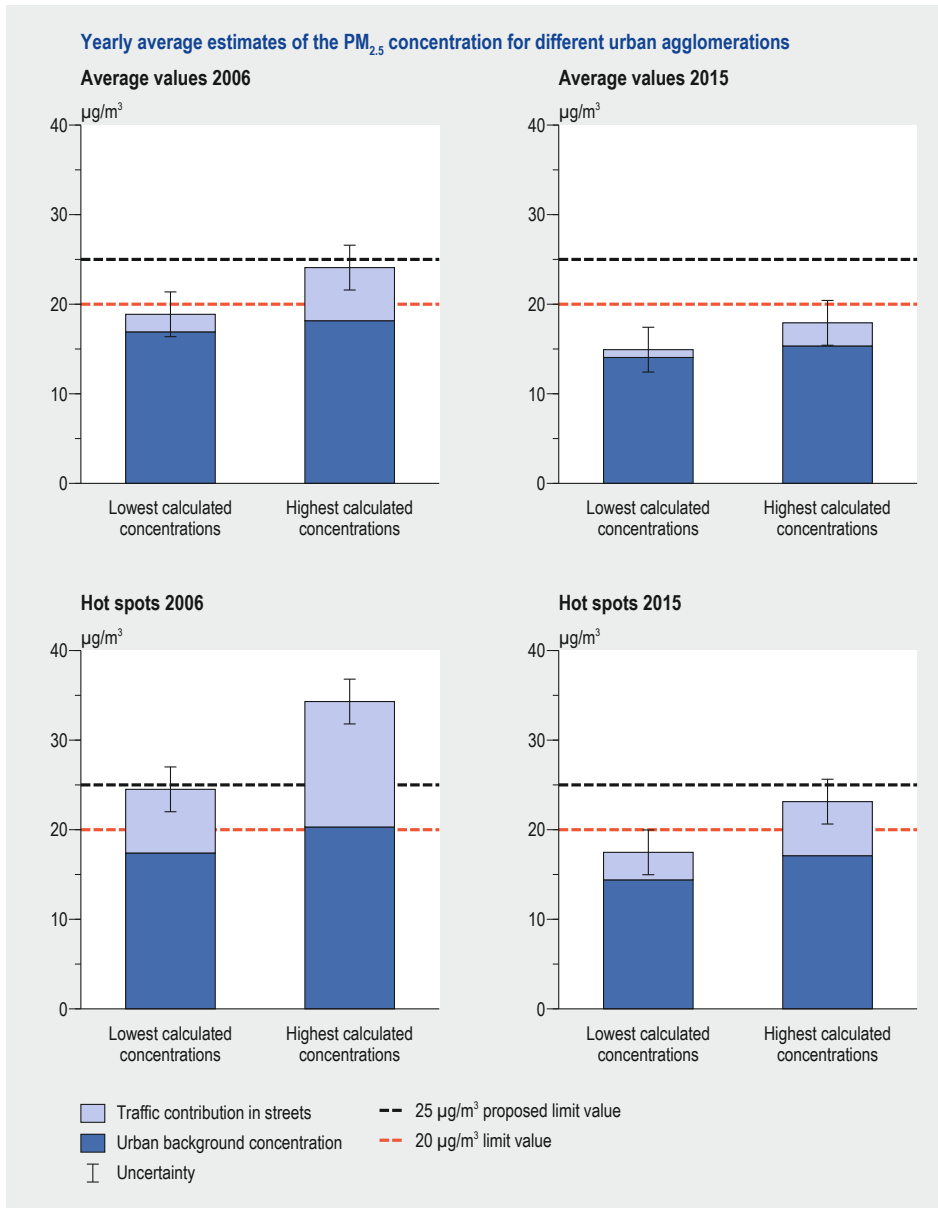


Figure 3.5 Estimates of the yearly averaged PM_{2.5} concentrations for streets in different agglomerations in 2006 and 2015. The upper two graphs show the lowest and highest calculated average concentration for the different agglomerations and streets. The lower two graphs show the lowest and highest calculated hot spot concentration (98-percentile) for the different agglomerations and streets. The estimates are based on a combination of measurements and model calculations, based on current legislation. The uncertainties shown represent a lower limit, because not all sources of uncertainty have been quantified (e.g. the effect of unusual meteorological conditions on yearly average concentrations).

PM_{2.5} emissions on the attainability of PM_{2.5} standards, now and in the future, in provinces with regional background. When compliance with the proposed PM_{2.5} limit value of 25 µg/m³ is tested, the number of exceedances that are associated with the PM_{2.5} emissions of large animal housings will be very limited by 2015 (approximately 1 out of 1000 cases). Testing compliance with a PM_{2.5} value of 20 µg/m³ does increase the number of exceedances in 2015 by a factor of 4, but the number will still be rather limited. The agricultural emission projections took account of recent requirements for more humane animal treatment. These policies generally lead to increased emissions of particulate matter and precursors.

3.7 Uncertainty in the calculated PM_{2.5} concentrations

The PM_{2.5} levels presented here are based on a combination of model calculations and measurements of PM_{2.5}. The PM_{2.5} levels that resulted have an uncertainty of ±2.5 µg/m³. This uncertainty includes the net effect of errors in the model and the measurements and results from the calibration of the modeled concentrations (see text box **Methodology for calculating PM_{2.5} concentrations**). However, the uncertainty range of ±2.5 µg/m³ is a minimum and could be larger for a number of reasons:

- Calibration is based on a very limited number of measurements.
- The uncertainty of the different PM_{2.5} measurements has not been quantified.
- Uncertainties in the modeled street contribution to PM_{2.5} have not been quantified.
- Projections of future PM_{2.5} concentrations have been derived for average meteorological conditions. More extreme meteorological conditions can cause annual PM_{2.5} concentrations to be several µg/m³ higher than those under average conditions. This phenomenon leads in practice to a larger uncertainty range for projections of future PM_{2.5} concentrations.

Comparison with EMEP model results

The source contributions of PM_{2.5} reported above have been compared with results from the EMEP model. The EMEP model plays an important role in preparing policy for Europe (see text box **Models for the assessment of particulate matter**). The discrepancies between the PM_{2.5} results from different model approaches are an indication of the uncertainty in the calculated levels (see Schaap et al., 2007).

The conclusions from a previous comparison between OPS model results and the EMEP model still apply (Velders et al., 2003):

- Large discrepancies are found between the models for oxidized nitrogen (NO_x and particulate nitrate). For instance, when compared with measurements, the NO_x and particulate nitrate concentrations from the EMEP model were lower and higher, respectively. The results of the OPS model for the Netherlands were in reasonable agreement with measurements (see Buijsman et al., 2005).

- The source-receptor matrices are in reasonable agreement for reduced nitrogen (NH₃ and particulate ammonium).
- The source-receptor matrices are in good agreement for oxidized sulfur (SO₂ and particulate sulfate).

The two models also produced different results concerning the contributions of other countries to the deposition of acidifying compounds in the Netherlands. A comparison of the PM_{2.5} concentrations modeled with the EMEP also shows distinct differences between the primary PM_{2.5} and nitrate contributions of countries to the PM_{2.5} concentration in the Netherlands (Figure 3.6). The models agree on the fact that about 80% of the anthropogenic PM_{2.5} levels in the Netherlands are determined by emissions from the Netherlands itself and nearby regions in Germany, France, the United Kingdom, Belgium and the North Sea.

The relative contributions to primary PM_{2.5}, nitrate, ammonium and sulfate from the EMEP and OPS models are in reasonable agreement, except for the contribution of Germany and the Netherlands to primary PM_{2.5} and nitrate. The OPS model attributes about 40% of primary PM_{2.5} to Dutch sources and about 10% to German sources, whereas the EMEP model shows the share from both countries to be about equal (between 25 and 30%). The Dutch and German contribution to nitrate is also different in both

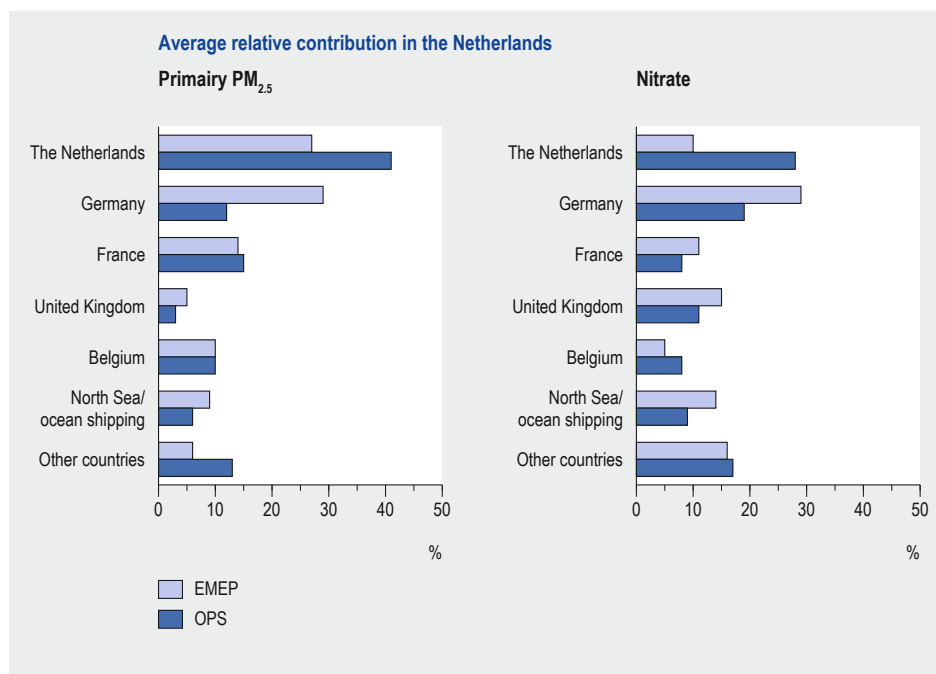


Figure 3.6 Average relative contributions (%) by country to primary PM_{2.5} and nitrate in PM_{2.5} in the Netherlands resulting from the EMEP model (EMEP, 2006a) and OPS model for 2004 and 2006, respectively.

models: the OPS model calculates values of about 30 and 20%, respectively, whereas the EMEP model determines values of about 10 and 30%. The reason for these differences may be related to resolution differences between the models. An explanation is not obvious and needs to be worked out.

Methodology for calculating PM_{2.5} concentrations

The methodology for calculating the background concentration at any arbitrarily-chosen location in the Netherlands can be divided into two steps. These are briefly described below.

Step 1. Calculating the background concentration.

This concerns the calculation of the background concentration (the regional concentration and the urban concentration) with the OPS calculation model (Van Jaarsveld, 2004). This model includes source contributions in all of Europe. Primary and secondary (sulfate, nitrate, ammonium) fractions are calculated separately and then added together to obtain the total calculated particulate matter concentration. The secondary fraction mainly consists of fine particles. Based on literature and limited measurements, the model assumes that the following fractions of secondary particles are part of PM_{2.5}: 1.0 (ammonium), 0.9 (sulfate) and 0.8 (nitrate). The model results are sensitive to this approach, since about half of PM_{2.5} consists of secondary inorganic aerosol. The secondary contribution to PM_{2.5} from VOC (secondary organic aerosol) is still at the research phase and has not been included in the OPS model. The calculation resolution is 5x5 km. The inputs for the model include data on emissions, such as the strength of emissions and spatial and temporal distribution of the sources, both for the Netherlands and for other European countries. Meteorological data are also required. For calculations involving years from the past, the emission data for the Netherlands from the national Pollutant Emission Register (MNP, 2007; MB 2007) and meteorological data for the relevant year are used. For calculations involving future years, the future emissions are estimated based on assumptions about developments of economic activities and emission factors, along with many-year average meteorological input (1990-1999). In the future projections, the effect of established and/or future national and international policy is taken into account. Countries are assumed to meet their emission goals set by existing agreements: for 2010 these goals are set by the National Emission Ceilings Directive (EU, 2001b) and for 2020 by the Thematic

Strategy on Air Pollution (CEC, 2005).

Step 2. Calibration to measurements. Calibration is required for PM_{2.5}, which was also the case for PM₁₀. This is because the model does not include all sources. Only about two-thirds of the measured concentrations can be explained by the model. Due to the large uncertainties in the model, the emissions and the measurements, the calculated PM_{2.5} background concentrations are calibrated simply by adding a fixed amount of 5 µg/m³. This value of 5 µg/m³ is the mean difference between the model results and the available PM_{2.5} measurements in the Netherlands. This amount represents not only the contribution to PM_{2.5} by sea salt, mineral dust, water, secondary organic aerosol and other sources, but also includes the effect on other sources which may be misrepresented by the model. The value of 5 µg/m³ is a rough estimate with an uncertainty of ± 2.5 µg/m³. The estimate certainly needs improvement and revision when more data become available. More information on the model uncertainties and methodology can be found in Velders et al. (2007b) and Matthijsen and Visser (2006).

Calculation of the contribution of local sources.

The contribution of local sources on top of the background concentration, such as the contribution of urban traffic in streets, can be calculated with separate models. For this assessment, preliminary estimates have been made for current and future traffic contributions to PM_{2.5} for different sets of streets (total 1000) in the cities of Amsterdam, Utrecht and Rotterdam by using the CAR II street model (Eerens et al., 1993; Jonker en Teeuwisse, 2006). In the Netherlands, CAR II is a widely used tool to assess the traffic contribution of air-pollutants in streets. The model has recently been updated (Wesseling and Sauter, 2007). Vehicle emission factors for PM_{2.5} are still rather uncertain compared to the PM₁₀ emission factors. The uncertainties concern the non-tailpipe emissions of PM_{2.5} which are due to tire, brake and road wear.

4 HOW LARGE IS THE EMISSION?

This chapter summarizes the present knowledge on the various anthropogenic (man-made) and natural sources in Netherlands and Europe that are believed contribute to $PM_{2.5}$.

The following aspects are addressed:

- Present and future anthropogenic primary $PM_{2.5}$ emissions in the Netherlands and nearby regions.
- Present and future anthropogenic emissions of precursor gases of secondary particulate matter in the Netherlands and Europe.
- Natural sources which can contribute to $PM_{2.5}$ in the Netherlands.
- Uncertainties in the $PM_{2.5}$ emissions.

4.1 Sources of $PM_{2.5}$

Emissions of different substances lead to concentration levels of $PM_{2.5}$ after transport in air, possible transformation and removal. $PM_{2.5}$ is usually classified as primary or secondary, depending to the formation mechanism. Primary particles are emitted directly into the atmosphere and are either anthropogenic or the result of natural emissions like sea salt. Secondary particles are those formed in the air by chemical reactions of gases like sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOC). Such secondary particles can also occur in the fine fraction.

In Europe, anthropogenic emissions are the main contributors to $PM_{2.5}$, although natural sources are important and may be dominant during certain episodes. Emission data serve as input for models to assess $PM_{2.5}$ levels, origin and composition. In general, most models use anthropogenic primary emissions and SO_2 , NO_x and NH_3 emissions. The other sources, including natural emissions, are still very uncertain, as is their effect on levels of particulate matter. Model estimates of $PM_{2.5}$ from natural sources and elemental carbon (EC) have been included in the EMEP model only very recently (EMEP, 2006b).

The $PM_{2.5}$ model exercises, presented in the previous chapter, follow the general approach and are therefore based on anthropogenic emissions of primary $PM_{2.5}$ and the gases SO_2 , NO_x and NH_3 . In the section below, the available emission data for these substances are presented and discussed. The other sources which contribute, in model terms, to $PM_{2.5}$ are summarized thereafter.

4.2 Anthropogenic primary sources

PM_{2.5} has recently been added to the Dutch national Pollutant Emission Registration. Because PM_{2.5} is part of PM₁₀, the sources of PM_{2.5} are implicitly known in those cases where PM₁₀ sources are known. The national database contains PM_{2.5} emission data, which are inferred from PM₁₀ emissions by applying fixed fractions of PM_{2.5} to PM₁₀ emissions per activity sector (Visschedijk et al., 2007; Velders et al., 2007b). All combustion processes generate particulate matter emissions, of which 95 to 100% consist of PM_{2.5}. However, sources such as tire abrasion and road wear due to road traffic do not contribute to PM_{2.5} emissions, or only very slightly. PM₁₀ emissions from agriculture, storage, loading/unloading and building activities contain between 10 and 20% PM_{2.5}. From other sectors, PM₁₀ emissions contain about 20 to 80% PM_{2.5}.

Anthropogenic primary PM_{2.5} emission data in the national database are given as a total sum of different primary substances per sector. For example, combustion emissions consist mainly of elemental carbon (soot) and particulate organic matter. Consequently, with speciated carbon measurements such as elemental carbon and organic carbon, it is impossible to verify the contribution of specific primary PM_{2.5} sources to the PM_{2.5} concentrations.

Primary PM_{2.5} emissions in the Netherlands

Figure 4.1 shows the development in time of the national emission estimates for primary PM_{2.5} and PM₁₀ – the latter is provided for comparison purposes (MNP, 2007b; Velders et al., 2007a, 2007b). The source strengths of several anthropogenic sources of PM_{2.5} are uncertain. For instance, re-suspension due to road traffic, which is not included in the national database, is probably a small but relevant source of PM_{2.5} that could be on the order of 1 kt (van Harmelen et al., 2004). Note that the emissions from ocean shipping are not included in the EU directives and are therefore not included in the national total. The ocean shipping sector produces emissions due to shipping on Dutch territory, from seagoing ships and emissions from ships moored in harbors. Without ocean shipping, the emissions of primary PM_{2.5} in 2005 have declined by more than 50% since 1990. Road traffic and ocean shipping comprised almost 70% of all registered PM_{2.5} emissions in 2005. Other sectors contributed 10% or less.

Under current legislation, primary PM_{2.5} emissions from most sectors are expected to decline between 2000 and 2020 (total reduction of 40%). Transport is expected to show the largest emission reduction (70%). A small increase of 10% is foreseen by the agriculture sector. Ocean shipping emissions are also expected to increase between 2000 and 2020 by about 60%. In 2020, the national PM_{2.5} emission total will be on the same order as the PM_{2.5} emissions due to ocean shipping on Dutch territory. For 2020, the calculated contribution to the PM_{2.5} concentrations of ocean shipping in the Netherlands is around 1 µg/m³ (see chapter 3), which is small considering the emitted quantity. One of the reasons for this is that most of the sources are off-shore and emissions are greatly diluted before they reach land.

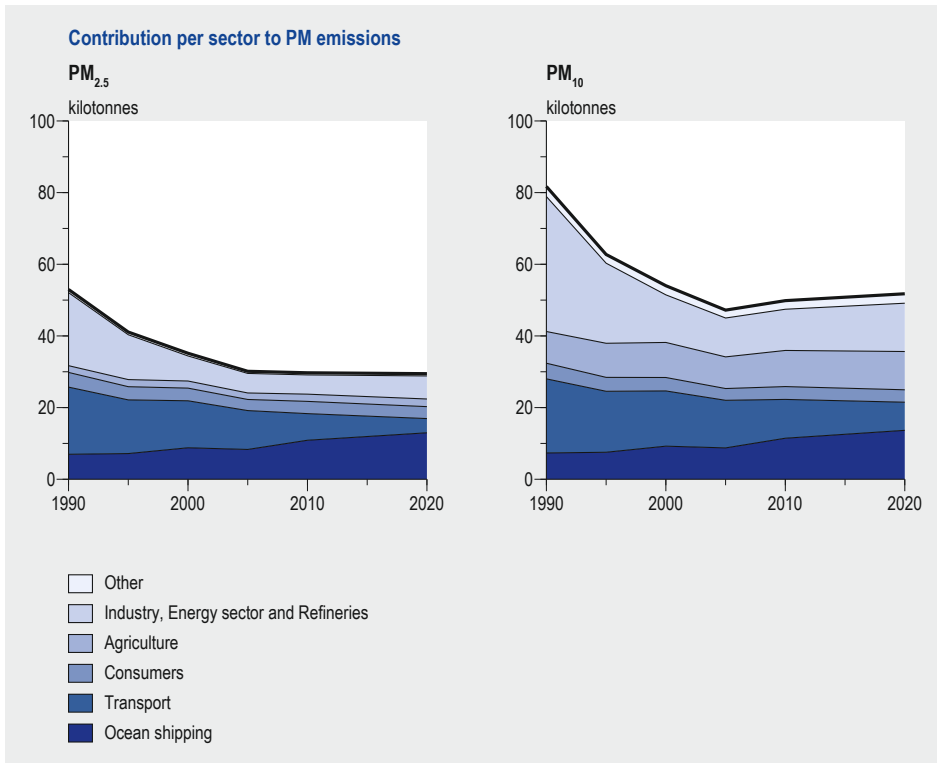


Figure 4.1 $PM_{2.5}$ and PM_{10} emissions for 1990, 1995, 2000 and 2005 and projections for 2010 and 2020; based on current legislation as reported by the Dutch National Pollutant Emission Registration (Visschedijk et al., 2007; Velders et al., 2007b). See VROM (2003) for the composition of the various sectors. The ocean shipping sector consists of emissions due to shipping on Dutch territory, from seagoing ships and emissions from ships moored in harbors. Emissions of ocean shipping are not included in the EU directives.

Primary $PM_{2.5}$ emissions in other European countries

As part of the Convention on Long-range Transboundary Air Pollution, data about the emissions of primary particulate matter and SO_2 , NO_x and NH_3 in other European countries must be reported annually to the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). Not all countries have met this obligation. Nevertheless, 27 of about 40 countries, among these the Netherlands, met their EMEP obligation in 2006 to report on 2004 $PM_{2.5}$ emissions. The officially reported emissions of SO_2 , NO_x and NH_3 and of primary PM_{10} and $PM_{2.5}$ have recently undergone a thorough revision and quality control regarding their consistency and comparability (EMEP, 2006a). For the other countries, the EMEP estimates the magnitude of their annual emissions.

The established emissions were obtained from the officially reported emissions (EMEP, 2007). Emission projections per country are according to Amann et al. (2007a). For some of these countries the national projections differ by up to 20% from the

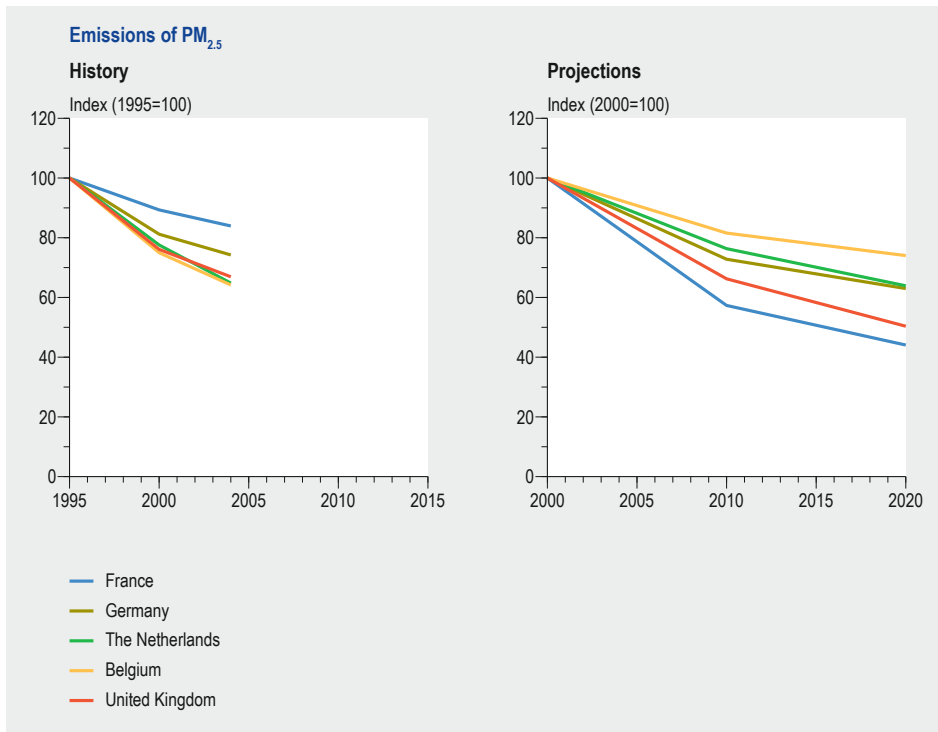


Figure 4.2 Indexed national emissions of primary PM_{2.5} in the Netherlands and neighboring countries (left panel) for 1995-2004 and (right panel) for 2000, 2010 and 2020.

integrated projections on energy use calculated with the PRIMES model (PRIMES, 2007). Nevertheless, for 2020 the total of the national projections and the PRIMES projections for these seven countries are about equal.

Non-industrial combustion is one of the major sources of primary PM_{2.5} emissions in Europe. Other important sources are production processes and road transport. According to calculations with the RAINS/GAINS model (Amann et al., 2007b), in the rest of Europe (EU-27) the future anthropogenic emissions of primary PM_{2.5} will also decline by 36% to 38% in 2020, depending on the volume of coal consumption (Amann et al., 2006). Major contributions to these reductions are made through the ongoing replacement of solid fuels for home heating and for electricity generation, pollution control equipment in the power sector and emission control measures for mobile sources.

Between 1990 and 2005, the trend of primary PM_{2.5} emission in the Netherlands and neighboring countries showed the largest declines in Belgium and the Netherlands. However, future emissions in other countries will decline more rapidly than those in the Netherlands (Figure 4.2 - right panel). This is because other countries will begin to catch up with the Netherlands, which has already implemented a relatively large number of control technologies. In 2020, total primary PM_{2.5} emissions from the EU-27 are expected to be half of the 2000 total due to implementation of current

EU legislation, along with the Gothenburg and the Heavy Metals Protocols under UN-ECE (UN-ECE, 2007).

Emissions outside Europe

Emissions outside Europe can contribute to background hemispheric levels of $PM_{2.5}$, but their average share is believed to be small (less than $1 \mu\text{g}/\text{m}^3$). Data on global emissions are collected in the EDGAR database (Olivier and Berdowski, 2001). EDGAR provides global annual emissions for 1990 and 1995 for the greenhouse gases CO_2 , CH_4 and N_2O and the precursor gases CO , NO_x , NMVOC and SO_2 . Particulate black and organic carbon for all anthropogenic sources are also included.

4.3 Anthropogenic sources of SO_2 , NO_x and NH_3

The EU has set national emission targets (ceilings) for SO_2 , NO_x , NH_3 and VOC that are to be attained by EU countries before 2010 (EU, 2001). All these substances contribute to the $PM_{2.5}$ concentration when they are chemically transformed into secondary particulate matter.

Emissions in the Netherlands

The contribution of SO_2 , NO_x , NH_3 and VOC to $PM_{2.5}$ concentrations in the Netherlands is 50% or more on average. For the Netherlands, the ceilings are 50 (SO_2), 260 (NO_x), 128 (NH_3) and 185 (VOC) kt per year. In 2004, the emissions were 65, 379, 181 and 134 kt, respectively. The emissions of these gases have been monitored for about two decades and the uncertainty has been reduced over this period. In the Netherlands, the registered emissions of SO_2 , NO_x , NH_3 and VOC have declined by 65%, 38%, 62% and 47%, respectively, between 1990 and 2005. National emission projections for 2010 indicate that the emission ceilings in the Netherlands can be reached or closely approached with current legislation (MNP, 2007a). Emission targets for 2020, which include targets for $PM_{2.5}$, are presently under negotiation as part of a revision of the EU legislation on national emission ceilings.

Emissions in other European countries

Europe wide, emission targets set for 2010 – compared to 2000 – would lead to reductions of about 45% for SO_2 , 25% for NO_x , 10% for NH_3 and 35% for VOC. Projections for 2010 indicate that the emission targets for NO_x and SO_2 of the nearby countries of Belgium, France, Germany and the United Kingdom are, similar to the Netherlands, the least likely to be attained (Amann et al., 2007b). The revision of the national emission ceilings in the EU foresees more stringent national ceilings in 2020 for NO_x and SO_2 , whereas the NH_3 and VOC targets will remain about the same or will be even less stringent. The emission trends per sector and country since 1990 are discussed in the Annual Emission Inventory by the European Environmental Agency (EEA, 2006b).

4.4 Natural and other sources

For the Netherlands, the average contribution of natural and other sources to $PM_{2.5}$ concentrations is about $5 \mu\text{g}/\text{m}^3$. This estimate is uncertain and ranges between 2.5 and $7.5 \mu\text{g}/\text{m}^3$. The value of $5 \mu\text{g}/\text{m}^3$ is derived from the difference between measured and modeled $PM_{2.5}$ concentrations. This amount therefore represents more than the contribution of natural sources only. For instance, the model calculations do not include sources from outside Europe. These sources can be of natural or anthropogenic origin and also contribute to $PM_{2.5}$ in the Netherlands due to long-range transport. The average annual contribution from these sources, known as the northern hemispherical background concentration, is believed to be $1 \mu\text{g}/\text{m}^3$ or less in the Netherlands. The value of $5 \mu\text{g}/\text{m}^3$ is supported by rough estimates of the average contribution to $PM_{2.5}$ from separate sources: sea salt ($1 \mu\text{g}/\text{m}^3$), mineral dust ($0.6 \mu\text{g}/\text{m}^3$), water ($1\text{-}2 \mu\text{g}/\text{m}^3$), secondary organic aerosol ($<1 \mu\text{g}/\text{m}^3$), sources outside Europe ($1 \mu\text{g}/\text{m}^3$) and other sources (see also chapter 2).

4.5 Emission uncertainties

Complete and consistent emissions inventories and reliable air quality modeling are needed for assessing compliance with target values. Emission inventories of primary particulate matter are not yet considered to be complete and consistent for all sectors, although ongoing work should allow significant improvements in the accuracy and consistency of inventories over time.

Uncertainty in primary emissions

The guidebook for $PM_{2.5}$ from EIONET states that the uncertainty in the emission factors of primary $PM_{2.5}$ ranges from -20% to +30%. However, model studies suggest that the uncertainties are larger by up to a factor of two (e.g. Schaap and Denier van der Gon, 2007). The uncertainty of the $PM_{2.5}$ emission factors is determined by a number of factors such as source height, profile and sector. Few uncertainty estimates of the primary $PM_{2.5}$ emissions are available. The uncertainty in the particulate matter emission data is not well understood. The TNO institute has reported on various sources of inaccuracy and uncertainty (Van Harmelen et al., 2004). Measurements of the emission amounts are uncertain due to several highly dynamic processes in the plume of the source. Especially for large sources of primary $PM_{2.5}$, the quality of emission measurements is difficult to ascertain (Corio and Sherwell, 2000).

Additional uncertainties in $PM_{2.5}$ emissions can be due to the uncertainties in particle size distribution when $PM_{2.5}$ emissions are derived as a fraction of the corresponding PM_{10} emissions. The overall uncertainties of $PM_{2.5}$ emissions may be smaller than those for PM_{10} emission inventories, since much of $PM_{2.5}$ is emitted from fuel combustion sources which are relatively well known. Emissions of coarse particulate matter, which are not included in $PM_{2.5}$, are associated with rather significant uncertainties. The

large uncertainties in primary PM_{2.5} emissions estimates make it more difficult to use emission trading and emission ceilings as policy instruments to reduce emission.

Uncertainty in emissions of secondary precursor gases

Emission data for the gases which lead to the formation of secondary particulate matter are much more certain than those for primary PM_{2.5}. However, uncertainties in the registered emissions of VOC have not been quantified. National emission estimates for SO₂, NO_x and NH₃ have an uncertainty of ±6%, ±15% and ±17%, respectively (MNP, 2007b). The emissions of these gases abroad can be more uncertain.

5 PM_{2.5} POLICY INSTRUMENTS AND MEASURES

This chapter addresses policy instruments that impact on the levels of air pollution in the Netherlands, specifically PM_{2.5}. Current legislation is discussed and possible additional measures are outlined in the light of the fact that extra measures are probably necessary to attain the new PM_{2.5} standards. The chapter also provides an overview of the measurements, emissions and models that are used to assess the situation regarding PM_{2.5} and to support policy development on PM_{2.5} in the Netherlands. The status of current instruments and measures is discussed, and recommendations for research are provided.

5.1 Current legislation

Current legislation that affects particulate matter levels is a conglomeration of different policy measures established on the European, national and local scales (Figure 5.1). The measures are all driven by European policy instruments.

At present, both national and European air quality policy measures are mostly linked to European policy instruments. These policy instruments essentially take a three-track approach to reducing the negative effects of air pollution.

They do this by:

1. establishing National Emission Ceilings (NEC),
2. establishing air quality standards,
3. achieving emission reductions for vehicles and in production processes.

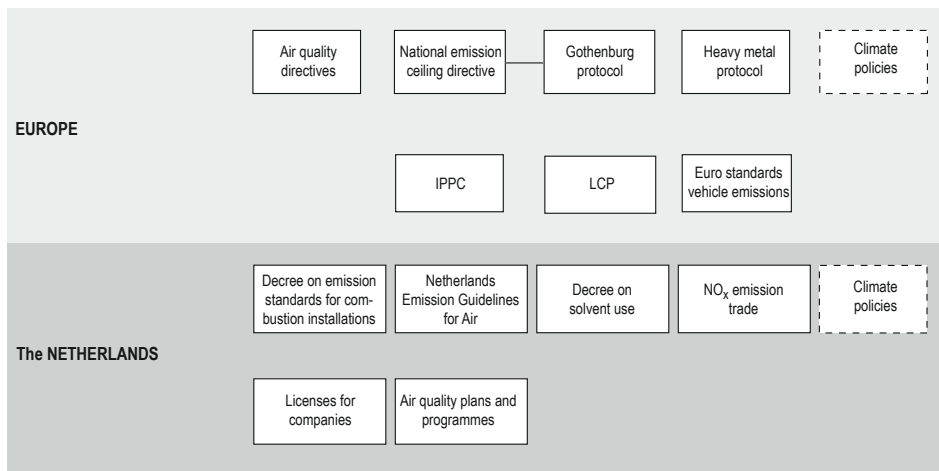


Figure 5.1 Overview of the European and national policy instruments which comprise the current legislation affecting particulate matter levels. New climate policy instruments are being developed.

National Emission Ceilings

National Emission Ceilings are established by the NEC Directive (EU, 2001b). The Directive stipulates emissions maximums for each Member State for the air pollutants, SO₂, NO_x, NH₃ and VOC. Ceilings for particulate matter were not included in the Directive because at that time knowledge on particulate matter was insufficient. Nevertheless, particulate matter levels have decreased substantially due to the reduction of emissions of the PM precursors SO₂, NO_x, NH₃ and VOC. The national emission ceilings are based on a Europe-wide optimization of costs of air pollution abatement and benefits to human health and natural habitats.

The NEC Directive was preceded by the Gothenburg Protocol and the Heavy Metal Protocol, which were established under the auspices of the UN-ECE (UN-ECE, 1999; UN-ECE, 1998). The UN-ECE involves a larger number of countries (41) than the EU. For instance, Russia is also a party to the UN-ECE. Consequently, the UN-ECE protocols have led to an early decline of air pollution levels in Europe. Even now, the UN-ECE establishes emission reductions for the present EU-27 that are additional to those brought about by EU directives.

EU air quality standards

The aim of the EU air quality standards is to ensure that EU citizens enjoy a minimum level of protection against air pollution. This is realized by means of EU-wide standards for 13 air pollutants, which are regulated by several air quality Directives (EU, 1996b, 1999, 2000, 2002 and 2004). For instance, for PM₁₀ there are legally binding limit values for 24-hour and annual averaged concentrations.

Emission reductions for vehicles and production processes

Several source policies have been issued which aim to achieve emission reductions in production processes. These policies are established by the Integrated Pollution Prevention and Control (IPPC) Directive (EU, 1996a) and the Large Combustions Plants (LCP) Directive (EU, 2001a) and other directives that define product requirements, such as those for cars (the so-called Euro-standards; emission requirements) and fuels. For example the IPPC Directive prescribes best available techniques (BAT) to reduce fugitive emissions during the storage and handling of goods. The LCP Directive has established explicit emissions requirements. The EU strives for a level playing field for enterprises with regard to abatement costs that result from these source policies. This means that enterprises are not obligated to take prescribed actions if this negatively affects their competitiveness within Europe.

The European legislation on air quality has been implemented in the Netherlands. The relevant laws at the national level are the following: the Environmental Management Act (*Wet Milieubeheer*), the Emission Requirements for Combustion Installations Decree (*BEES*), the Netherlands Emission Guidelines for Air (*NeR*), the Solvents Decree (*Oplosmiddelenbesluit*) and the Air Quality Decree (*Besluit Luchtkwaliteit*). In addition,

there is policy concerning NO_x emissions trading and there are national incentives to use cars with lower emissions. On a more local scale, provinces and municipalities can control air quality by making air quality plans and by stipulating emissions when issuing operating licenses to companies.

In Europe, the Netherlands is an air quality *hot spot*. Therefore, some Dutch policy measures to reduce emissions are more stringent than is strictly required by the relevant EU directives. For instance, the Netherlands has passed additional legislation (*BEES B*) concerning existing, small-scale energy installations that are not regulated by European legislation. As a result, the most important actions to reduce emissions have already been carried out. For example, the Industry/Energy/Refineries sector in the Netherlands now ranks among the most eco-efficient in Europe.

5.2 Additional policy instruments

At present, a new EU Air Quality Directive is being developed, and tighter emission ceilings are being established. A revision of the NEC Directive has been foreseen to ensure further emission reductions in 2020 for sulfur dioxide, nitrogen oxides, ammonia and non-methane volatile organic compounds, but this time including particulate matter (PM_{2.5}). The European Commission will probably propose a relative emission ceiling for PM_{2.5}, since the quality of the PM_{2.5} emission inventories in the EU-27 is not yet satisfactory.

The aim for the revision of the NEC Directive was formulated in the Thematic Strategy on Air Pollution (CEC, 2005) of the European Commission. The aim is expressed in terms of environmental and health targets for 2020. For instance, as an EU-wide goal, the health impacts of PM_{2.5} – expressed in the number of years of life lost – must be reduced by 47% in 2020 with respect to the year 2000.

The preparatory track for a proposal of the European Commission for a new NEC Directive began in September 2006 and is expected to be completed in the first half of 2008. The process was postponed for six months due to the current discussion in the EU on a possible proposal for burden sharing of the greenhouse gas reductions in the EU-27. This discussion could have important consequences for additional air pollution abatement in countries due to synergy between climate change and air pollution abatement measures (see text box **Climate policy influences air quality policy**).

5.3 Policy measures in addition to current legislation

Additional national and European policy measures may be necessary to attain the deadlines for the PM_{2.5} standards in the Netherlands. The target of a 20% reduction in PM_{2.5} exposure appears to be the most difficult to attain. The proposed PM_{2.5} limit value of 25 µg/m³ can probably be attained in 2015, apart from a limited number of

exceedances in busy streets and other hot spots (for instance in agricultural areas). But if the PM_{2.5} limit value is set at 20 µg/m³, such exceedances are expected to occur much more frequently in 2015, although for average conditions the value of 20 µg/m³ will probably not be exceeded.

Several national and European policy measures in addition to current legislation are described by Velders et al. (2007a and 2007b). Intended and additional national measures defined in 2007 will lead to an extra emission reduction of primary PM_{2.5} of between 1.3 and 3.3 kt, depending on the scenario conditions. Similarly, extra measures are foreseen to reduce precursor gases of secondary inorganic aerosol. Altogether, an additional reduction in average PM_{2.5} concentrations of up to about 1 µg/m³ will be realized with respect to current legislation. The extra reduction may be enough to prevent exceedances of the limit value of 25 µg/m³, but probably not exceedances of the 20 µg/m³ limit value. Even more national policy measures are probably required, especially to attain the target of a 20% reduction to PM_{2.5} exposure.

Further national measures

The cost-effectiveness for the Netherlands of specific measures has been explored for PM₁₀. The measures are additional to current national and European legislation using currently available technology. The results have been extrapolated to PM_{2.5} (see text box **Cost-effectiveness of additional measures in the Netherlands**). For PM₁₀ the studied measures will lead in 2020 to an estimated reduction of 1 to 3 µg/m³, depending on whether the specific measures are applied in the Netherlands only or on a European level. Similarly, the reduction of PM_{2.5} concentrations in 2020 could range from less than 1 µg/m³ to about 2 µg/m³, depending on whether the specific measures are applied in the Netherlands only or Europe-wide.

Climate policy influences air quality policy

The European ambition to reduce greenhouse gases in 2020 by 30% (compared to the base year 1990) also has consequences for the emissions of air pollutants like SO₂, NO_x and PM_{2.5}. Measures such as increasing energy efficiency, stimulating wind and solar energy and hydropower, and promoting carbon capture and storage at coal-fired power plants also have positive ancillary benefits for the emissions of air pollutants like SO₂ and PM_{2.5}. Depending on the level of climate policy in a country, part of the task to reduce air pollution has already been accomplished by climate measures. As a result, climate policies will reduce the overall cost of the air pollution abatement measures needed to meet the objectives of the Thematic Strategy

on air pollution by 2020. However, the costs for climate measures will increase at the same time. Ancillary benefits will probably be greater in 2030 than in 2020 (EEA, 2006a).

Beside positive ancillary benefits, negative side effects can also occur: for example this may be the case if biomass and biofuels are used in small-scale combustion installations with emission standards that are less stringent than those at large combustion plants. Moreover, it is still unclear to what extent the application of biofuels in cars and trucks will influence their emissions. Further research on this topic is needed.

Further local measures

Measures on a local scale, such as regulations which reduce the traffic volume locally, can help to attain a limit value at specific hot spots. Traffic volume regulations should be conducted on a much larger scale in order to help attain the exposure reduction target, which aims at a reduction of average urban background concentrations.

Further European measures

Under UN-ECE mandate, the Expert Group on Particulate Matter (EGPM) has investigated the scope for additional measures at the EU and UN-ECE level with currently available technical measures. The main conclusions are the following:

- Additional measures for primary PM_{2.5} emissions: With currently available technical measures (i.e. maximum reduction according to Amann et al., 2007a) there is potential for a further reduction of 40% of the projected 2020 total within the EU-27, and of 70% in non-EU countries. Of that 70%, one-third could be achieved by full implementation of current regulations and two-thirds by applying further measures.
- Additional measures for PM precursor emissions: The potential for further reduction with currently available technical measures is estimated to be 40% for SO₂, 15% for NO_x and 10% for NH₃ (of the projected 2020 total within the EU-27). Figures for European non-EU countries are not available to the Expert Group, but are expected to be generally higher.

The possible effect of these reductions on future PM_{2.5} concentrations in the Netherlands has not yet been assessed. A rough estimate is that this could result in an extra reduction in PM_{2.5} concentrations up to 2 µg/m³ on average in the Netherlands. Note that further national and European measures have a certain overlap, but the extent of the overlap is still unclear.

Ocean shipping measures

Hammingh et al. (2007) reported on international emission control measures for ocean shipping particulate matter levels in the Netherlands. This study showed that under current legislation, a high ambition scenario for ocean shipping would reduce the average urban particulate matter concentrations in the Netherlands by an additional 1-2% (i.e. 0.2-0.4 µg/m³) in 2020, compared to the emission projections. Although the absolute reductions in urban particulate matter concentrations resulting from shipping measures may seem small, they constitute a substantial proportion of air pollution that can be reduced by policy.

Cost-effectiveness of additional measures in the Netherlands

Smeets et al. (2007a and 2007b) have evaluated the cost-effectiveness of technical measures for improving air quality for PM₁₀ in the Netherlands. They looked at measures that go beyond the measures set down in current national and European legislation. In addition to technical measures, they also studied the effects and costs of introducing road pricing schemes for light and heavy-duty vehicles.

The results for PM₁₀ were extrapolated to PM_{2.5}. They assumed that primary PM₁₀ and PM_{2.5} emissions are being removed with equal efficiency by the additional abatement technologies. For most industrial sources, this assumption seems reasonable.

The analysis for PM_{2.5} shows that road pricing is the most cost-effective policy option to improve the exposure of the Dutch population to PM_{2.5}. The second-best option is an additional retrofit of soot filters and NO_x catalysts on inland ships (5-7 million euros per 0.01 µg/m³ decline in PM_{2.5} concentration). The third-best option consists of a large group of very different measures with a cost-effectiveness ranging between 9 and 15 million euros per 0.01 µg/m³. This group is composed of measures to reduce

emissions of primary particulates as well as measures aimed at the further reduction of the secondary precursors SO₂, NO_x and NH₃. More stringent (EuroVI) emission standards for particulates for heavy-duty vehicles (at the level of soot filters) are relatively costly compared to other measures to improve PM_{2.5} exposure in the Netherlands (25-45 million euros per 0.01 µg/m³). The placement of advanced dust abatement technologies in specific industries (with a low PM_{2.5} fraction) such as the food industry is also relatively costly.

The abovementioned third group of measures consists of concrete measures such as the prevention of diffuse emissions from the storage and handling of bulk goods, the placement of improved dust abatement technologies in specific industries (with a large PM_{2.5} fraction) and the placement of air scrubbers in the larger pig and poultry housing systems. Measures directed at secondary precursors include additional NH₃ control measures in agriculture, sharpening the national NO_x performance standard for heavy industry, additional SO₂ measures in industry and more stringent NO_x emission standards for road vehicles.

5.4 Current status of PM_{2.5} assessment instruments

This section views the quality of the various instruments: measurements, emissions and models necessary for compliance monitoring and to support assessments and national policy development with regard to PM_{2.5}. The main question addressed is the following: are these instruments adequate and of sufficient quality to assess the attainability of PM_{2.5} targets and to develop the correct policy measures for PM_{2.5}?

The new Air Quality Directive, which includes targets for PM_{2.5} and regulations for measurement and models, has not yet been finalized. Therefore, it is theoretically somewhat premature to answer the above question. The present analysis concerns the regulations outlined in the common position of the Council (CS, 2007a). As an initial analysis of PM_{2.5} in relation to the proposed Directive, the present report collected the available data and sketched the outlines of the PM_{2.5} situation for the Netherlands. Clearly, many figures are uncertain and need improvement. The following section sketches out the current status and probable actions regarding the instruments.

Measurements

Preparations are being made to meet the national requirements for measurements of PM_{2.5} and some of the prescribed components. In accordance with the First Daughter Directive (EU, 1999), routine measurements of PM_{2.5} began at a limited number of locations in the Dutch air quality network in 2004. The following actions, which are in preparation, must still be carried out:

- Make a preliminary assessment in order to ascertain the required number of PM_{2.5} measurement sites and their locations.
- Decide on the measurement strategy for PM_{2.5} and the prescribed components.
- Make technical choices such as the PM_{2.5} monitor type and filter type to be used for reference instruments.
- Harmonize the national and local networks for air quality measurements.
- Install monitors and start routine measurements according to the regulations in the Directive.
- Measure natural fractions in PM_{2.5}: this is required in case the natural fractions of particulate matter must be subtracted when determining compliance with limit values.

The present uncertainty in the particulate matter concentrations and the levels of the PM components hinders the above developments. For instance, concentration levels in the Netherlands are too uncertain to determine whether PM₁₀ limit values are more stringent than the PM_{2.5} limit value, or the reverse. The most stringent indicator for particulate matter will become the focus of the measurement strategy. Moreover, if the natural fractions are subtracted when determining compliance with limit values, this could also change the stringency of the particulate matter limit values. Consequently, official measurements of PM₁₀ and PM_{2.5} can only be integrated when the relationship between the fractions is determined with satisfactory accuracy.

If relative improvement is used for the reduction target value, compliance assessment requires a different strategy than if the absolute limit value is used.

When the Netherlands decides to use an automated monitor for PM_{2.5} measurements, an official equivalence method must be applied. Such a method regulates how automated measurements can be coupled to the results of PM_{2.5} reference instruments. This is needed to obtain officially recognized PM_{2.5} concentrations from automated PM_{2.5} monitors. There are still large uncertainties about measurements with regard to semi-volatile components in PM_{2.5} such as ammonium nitrate and water. The relative amount of semi-volatile components in PM_{2.5} is larger than in PM₁₀. Consequently, systematic differences due to evaporation of these fractions will also be relatively larger.

The knowledge on the levels, composition and representativity of both PM_{2.5} and PM₁₀ is small. The absence of a downward trend during the last seven years in the urban

particulate matter concentrations is not understood, given a downward trend in the estimates of all relevant emissions during that same period. Therefore, additional data on the composition of PM_{2.5} in cities and at street sites is necessary in order to better assess and understand the urban background levels and the increment in streets.

Some components are poorly understood because they are difficult to measure, like carbon and particle-bound water, and some because they are not widely measured, particularly mineral dust. Much additional work is required to characterize carbon as being either natural or man-made and primary or secondary in origin.

Emission inventory

In 2007, primary PM_{2.5} was included in the national Pollutant Emission Register. The emissions are inferred from primary PM₁₀ emissions. The uncertainty is still very large and needs improvement. A number of international initiatives are presently underway with the aim of improving inventory quality and assisting inventory experts in the compilation of robust national inventories. In this regard, EMEP (2006) lists the following actions to be taken:

- Revise the EMEP CORINAIR Guidebook to update information on PM methodologies and emission factors for inventory compilers.
- Develop an EU-27 PM_{2.5} emission inventory for the year 2000, focusing on completeness and comparability to ensure that consistent emission estimates are available for each country.
- Revise the Reporting Guidelines, to be done by UN-ECE, EMEP and the Task Force on Emission Inventories and Projections.
- Complete the annual review of inventory quality under the auspices of the Task Force on Emission Inventories and Projections Expert Panel on Review.
- Quantify the emissions of air pollutants from natural sources as part of the European NATAIR project. Results are expected in 2008 and 2009.

Anthropogenic emissions of primary particulate matter consist largely of carbonaceous material. Verification of these emissions with measurements is limited by the present uncertainties and difficulties regarding the measurements of these organic species. Consequently, model results of primary PM_{2.5} are also uncertain and difficult to verify with measurements. So there is a need for chemical classification of the officially reported primary PM_{2.5} emissions, both in the Netherlands and abroad.

There are indications that particle emissions from traffic and, in general, combustion related particles like soot play a more important role in the associated health effect than components such as secondary inorganic aerosols. Consequently, reducing the emissions of the precursor gases sulfur dioxide, nitrogen oxides and ammonia may reduce the contribution to background concentration on a large scale, but not necessarily the potentially higher risk from local, low-level primary, combustion related sources in urban areas. There is a need for information which helps policy makers to develop *best strategies* for emission reduction. Such strategies should be effective from

a health point of view and at the same time help to attain the particulate matter standards.

Extra measures to reduce emissions in addition to the presently outlined emission scenarios (see Velders et al., 2007a) are necessary in order to attain the PM_{2.5} standards. The effect of extra policy measures has only been examined preliminarily in this assessment. The extra national, international and local measures that will be necessary to meet the new standards must still be assessed.

Models

The task force on measurements and modeling, which operates under the mandate of the UN-ECE, recently prepared an assessment on particulate matter in Europe (EMEP, 2007b). In their report they stated: ‘There is little confidence that the PM models currently used for policy development are ready to accurately describe the urban and street increments in PM_{2.5} which are required for integrated assessment modeling and for policy assessments of urban health effects. Current models for particulate matter used in policy development may thus be adequate for the assessment of the relative magnitudes of emission reductions of some PM components and their precursors in PM mass concentrations, but their quantitative assessment against target and limit values for PM is limited by large uncertainties.’

The PM models used for policy assessment in the Netherlands are limited by the uncertainty of the emission input and of the measurements used for calibration. The OPS model, similar to most models, does not operationally include the contribution to PM_{2.5} of sea salt, re-suspension, secondary organic aerosol, mineral dust and water. Although the direct contribution of mineral dust to PM_{2.5} is limited, it drives the contribution of re-suspension in streets to PM_{2.5} by traffic and wind erosion. Therefore it is considered to be important for the assessment of PM_{2.5} levels.

The contribution of these primary sources to exposure in densely populated areas is probably underestimated by the present models due to a limited model resolution and large uncertainties in the primary emissions. A more accurate calculation of the urban and street increments in PM_{2.5} is required for integrated assessments and for policy assessments of urban health effects.

Uncertainties and how to proceed

Clearly, the particulate matter dossier still has many uncertainties. At present the knowledge base for the assessment of PM_{2.5} is very small. Several actions are necessary to improve this situation. The main and most urgent actions are listed in the Technical Summary and are covered by the following three main categories.

1. Define the measuring requirements from the guidelines.
2. Determine policy measures needed to attain the PM_{2.5} standards and the co-benefits and/or trade-offs of other EU legislation.

3. Improve emission inventories and models and perform more widespread measurements.

Some of the actions are being addressed in the national program on PM₁₀ and PM_{2.5} (BOP, 2007). The present report is part of this program, which is funded by the Dutch Ministry of Housing, Spatial Planning and the Environment (VROM). The aim of the program is to reduce the uncertainties and the number of policy dilemmas that complicate the development and implementation of adequate policy measures. The program focuses on the dispersion and composition of PM₁₀ and PM_{2.5}. It does not address the uncertainties in the health aspects of PM.

The uncertainties in the PM dossier addressed by the BOP program concern the following aspects:

- **Measurements:** the composition of PM₁₀ and PM_{2.5}; spatial and temporal variability of PM and fractions; indicative measurements of PM from wood burning.
- **Emissions:** description of poorly understood sources like mineral dust, non-tailpipe traffic emissions and re-suspension by road-traffic, inland and ocean shipping.
- **Models:** include description of sea salt, mineral dust, and secondary organic aerosol; coupling with global air quality model for boundary conditions.

Measurements, emissions and model results of PM_{2.5} are now more uncertain than those for PM₁₀. This situation will continue in the near future. Although better information will improve the situation and reduce the uncertainty, the uncertainty margins for PM_{2.5} will probably remain relatively large. If these uncertainty margins can be taken into account during the implementation of the Directive, the enforcement in practice will benefit and become more effective.

REFERENCES AND FURTHER READING

- Aben, J.M.M., Hettelingh, J-P and Schöpp, W. (2005). RAINS-NL: An Integrated Assessment model to support Dutch air quality policy making, In: Proc. 19th Intern. Conf. Informatics for Environmental Protection, pp 513-517, Brno. See also http://www.mnp.nl/images/rainsnl_background_tcm60-21210.pdf.
- AIRBASE (2007). The air quality database of the European Topic Centre on Air and Climate Change. See <http://air-climate.eionet.eu.int/databases/airbase>.
- Amann, M., Bertok, I., Cofala, J., Heyes, C., Klimont, Z., Posch, M., Schöpp, W. and Wagner, F. (2006). Baseline scenarios for the revision of the NEC Emission Ceilings Directive Part 1: Emission projections, NEC Scenario Analysis Report Nr. 1, Laxenburg, Austria. See <http://www.iiasa.ac.at/rains/reports.html>.
- Amann, M., Asman, W., Bertok, I., Cofala, J., Heyes, C., Klimont, Z., Schöpp, W. and Wagner, F. (2007a). Updated Baseline projections for the revision of the emissions ceilings directive of the European Union. NEC scenario Analysis Report Nr. 4. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria. See <http://www.iiasa.ac.at/rains/reports.html>.
- Amann, M., Asman, W., Bertok, I., Cofala, J., Heyes, C., Klimont, Z., Schöpp, W. and Wagner, F. (2007b). Cost-effective Emission Reductions to meet the Environmental Targets of the Thematic Strategy on Air Pollution under Different Greenhouse Gas Constraints. NEC scenario Analysis Report Nr. 5. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria. See <http://www.iiasa.ac.at/rains/reports.html>.
- Amann, M., Cofala, J., Gzella, A., Heyes, C., Klimont, Z. and Schöpp, W. (2007c). Estimating concentrations of fine particulate matter in urban background air of European cities, IIASA interim Report Nr. IR/07/001. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria. See <http://www.iiasa.ac.at/rains/reports.html>.
- Bleeker, A., Gies, E. and Kraai, A. (2006). Fijn stof uit stallen, berekeningen in het kader van het NSL. ECN report (in Dutch) ECN-E-06-045, Petten, The Netherlands.
- BOP (2007). A Dutch national research program on PM₁₀ and PM_{2.5}. (Beleidsgeoriënteerd Onderzoeksprogramma PM). See http://www.mnp.nl/nl/dossiers/fijn_stof/nieuws/index.html.
- Brink, H. M. ten, Müller, K., Spindler, G., Maenhaut, W., Wieprecht, W., Hitztenberger, R. and Baltensperger, U. (2004). Artefacts in measuring (the composition of) Particulate Matter in Europe; INTERCOMP 2000, a campaign to assess the comparability of methods in use in Europe for measuring aerosol composition. *Atmos. Environ.* 38, 6457-6466.
- Brunekreef, B. and Forsberg, B. (2005). Epidemiological evidence of effects of coarse airborne particles on health. *Eur Respir J*, 26, 309-318.
- Bruegel, P. van and Buijsman, E., (2001). Preliminary assessment of air quality for sulfur dioxide, nitrogen dioxide, nitrogen oxides, particulate matter, and lead, in the Netherlands under European Union legislation. RIVM report 725601005, Bilthoven, The Netherlands.
- Buijsman, E., Beck, J.P., Bree, L. van, Cassee, F.R., Koelemeijer, R.B.A., Matthijsen, J., Thomas, R. and Wieringa, K. (2005). Particulate matter: a closer look. MNP-report 500037011, Bilthoven, The Netherlands. (<http://www.rivm.nl/bibliotheek/rapporten/500037011.pdf>)
- CEC (2005). Communication from the Commission to the Council and the European Parliament. Thematic Strategy on Air Pollution. COM (2005) 446 final. Brussels.
- CESAR (2007). Cabauw Experimental Site for Atmospheric Research. See <http://www.cesar-observatory.nl>.
- Chow, J.C. (1995). Measurement Methods to Determine Compliance with Ambient Air Quality Standards for Suspended Particles; a critical review *J. Air & Waste Manage. Assoc.*, 45, 320-382
- Corio, L. A. and Sherwell, J. (2000). In-Stack Condensable Particulate Matter Measurements and Issues, *J. Air & Waste Manage. Assoc.* 50, 207-218.
- CS (2007a). Interinstitutional File, 2005/0183 (COD). Common Position adopted by the Council with a view to the adoption of a Directive of the European Parliament and of the Council on ambient air quality and cleaner air for Europe (16477/06), 30 March

2007. (<http://register.consilium.europa.eu/pdf/en/06/st16/st16477.en06.pdf>)
- CS (2007b). Draft recommendation for second reading on the Council common position for adopting a directive of the European Parliament and of the Council on ambient air quality and cleaner air for Europe (16477/2006 – C6 0000/2007 – 2005/0183(COD)), 31 July 2007. ([http://www.europarl.eu.int/registre/commissions/envi/projet_rapport/2007/392253/ENVI_PR\(2007\)392253_EN.doc](http://www.europarl.eu.int/registre/commissions/envi/projet_rapport/2007/392253/ENVI_PR(2007)392253_EN.doc))
- Cuvelier, C., Thunis, P., Vautard, R., Amann, M., Bessagnet, B., Bedogni, M., Berkowicz, R., Brocheton, F., Builtjes, P., Denby, B., Douros, G., Graf, A., Hellmuth, O., Honore, C., Jonson, J., Kerschbaumer, A., de Leeuw, F., Moussiopoulos, N., Philippe, C., Pirovano, G., Rouil, L., Schaap, M., Stern, R., Tarrason, L., Vignati, E., Volta, L., White, L., Wind, P. and Zuber, A. (2006). CityDelta: A model intercomparison study to explore the impact of emission reductions in European cities in 2010. *Atmos. Environ.*, 41, 189-207.
- DCMR (2007). Annual data reported by the DCMR, the Rotterdam Environmental Protection Agency. See <http://www.dcmr.nl/en/index.html>.
- EEA (2006a). Air quality and ancillary benefits of climate change policies EEA Technical report No 4/2006. See <http://reports.eea.europa.eu>.
- EEA (2006b). Annual European Community LRTAP Convention Emission Inventory 1990-2004 EEA Technical report No 8/2006. See <http://reports.eea.europa.eu>.
- EC (2007). Communication from the Commission to the Council, the European Parliament, the European Economic and Social Committee and the Committee of the Regions. Limiting Global Climate Change to 2 degrees Celsius. The way ahead for 2020 and beyond. COM(2007) 2 final. Brussels.
- Eerens, H.C., Sliggers, C.J. and Hout, K.D. van den (1993). The CAR model: the Dutch method to determine city street air quality. *Atmos. Environ.* 27B, 389-399.
- EMEP (2003). Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe, PART I, Unified EMEP Model Description. EMEP Report 1/2003, Nilu, Oslo. See http://www.emep.int/index_model.html.
- EMEP (2005). EMEP EC/OC campaign and results, in: Transboundary Particulate Matter in Europe EMEP Status Report 4/2005. See http://www.emep.int/publ/common_publications.html.
- EMEP (2006a). Transboundary acidification, eutrophication and ground level ozone in Europe since 1990 to 2004. The Norwegian Meteorological Institute, Oslo (EMEP Status Report 1/2006).
- EMEP (2006b). Transboundary particulate matter in Europe. The Norwegian Meteorological Institute, Oslo, EMEP Status Report 4/2006. See http://www.emep.int/publ/common_publications.html.
- EMEP (2006c). H. Klein and A. Benedictow: MSC-W Data Note 1/06, Transboundary data by main pollutants (S, N, O3) and PM, The Netherlands. ISSN 1890-0003. See http://www.emep.int/publ/common_publications.html.
- EMEP (2007a). See <http://webdb.emep.int>.
- EMEP (2007b) EMEP Particulate Matter Assessment Report, Editors: Wenche Aas, Peter Bruckmann, Richard Derwent, Nathalie Poisson, Jean-Philippe Putaud, Laurence Rouil, Sonja Vidic, Karl-Espen Yttri. EMEP/CCC-Report 8/2007, O-7726, Nilu, Oslo. See <http://www.nilu.no/projects/ccc/reports.html>.
- Erisman, J.W. ed. (1996). Towards development of a deposition monitoring network for air pollution in Europe. RIVM report 722108014.
- EU (1996a). Council Directive 96/61/EC of 24 September 1996 concerning integrated pollution prevention and control. OJ L 257 , 10.10.1996, page26.
- EU (1996b). Directive 96/62/EC of the European Parliament and the Council on ambient air quality assessment and management, OJ L 296, 21.11.1996, page 55.
- EU (1999). Directive 1999/30/EC of the European Parliament and the Council relating to limit values for sulphur dioxide and the oxides of nitrogen, particulate matter and lead in ambient air, OJ L 163, 29.6.1999, page 41. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/1999/l_163/l_16319990629en00410060.pdf)
- EU (2000). Directive 2000/69/EC of the European Parliament and the Council relating to limit values for benzene and carbon monoxide in ambient air, OJ L 313, 13.12.2000, page 12. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/2000/l_313/l_31320001213en00120021.pdf)
- EU (2001a). Directive 2001/80/EC of the European Parliament and the Council of 23 October 2001 on the limitation of emissions of certain pollutants into the air from large combustion plants, OJ L 309, 27.11.2001, page 1. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/2001/l_309/l_30920011127en00120011.pdf)

- europa.eu/LexUriServ/site/en/oj/2001/L_309/L_30920011127en00010021.pdf)
- EU (2001b) Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on national emission ceilings for certain atmospheric pollutants, OJ L 309 , 27.11.2001, page 22. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/2001/L_309/L_30920011127en00220030.pdf)
- EU (2002). Directive 2002/3/EC of the European Parliament and the Council relating to ozone in ambient air, OJ L 67, 9.3.2002, page 14. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/2002/L_067/L_06720020309en00140030.pdf)
- EU (2004) Directive 2004/107/EC of the European Parliament and the Council relating to arsenic, cadmium, nickel, mercury and polycyclic aromatic hydrocarbons in ambient air, OJ L 23, 26.1.2005, page 3. (http://eur-lex.europa.eu/LexUriServ/site/en/oj/2005/L_023/L_02320050126en00030016.pdf)
- Folkert, R.J.M., Buringh, E., Hammingh, P., Aben, J., Beck, J., Blom, W., Bree, L. van, Brink, R. van den, Buijsman, E., Esbroek, M. van, Hettelingh, J.P., Jaarsveld, H. van, Jimmink, B., Matthijsen, J., Hinsberg, A. van, Koelemeijer, R., Peters, J., Ruiter, J. de, Smeets, W., Tol, S. van, Thomas, R., Velders, G., Velze, K. van, Vries, W. de, Wieringa, K. and Zeijts, H. van (2005). Consequences for the Netherlands of the EU thematic strategy on air pollution, MNP report 500034002, Bilthoven. (<http://www.rivm.nl/bibliotheek/rapporten/500034002.pdf>)
- GGD Amsterdam (2007). Annual data reported by the Health Service Amsterdam. See <http://www.ggd.amsterdam.nl>.
- Hammingh, P., Aben, J.M.M., Blom, W.F., Jimmink, B.A., Vries, W.J. de and Visser, M. (2007). Effectiveness of international emission control measures for North Sea shipping on Dutch air quality, MNP report 500092004/2007, Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500092004.pdf>)
- Harrison, R.M. , A.M. Jones and Barrowcliffe, R. (2004). Field study of the influence of meteorological factors and traffic volumes upon suspended particle mass at urban roadside sites of differing geometries, *Atmos. Environ.*, 38, 6361-6369.
- Harmelen, A.K. van, Denier van der Gon, H.A.C., Kok, H.J.G., Appelman, W.J., Visschedijk, A.J.H. and Hulskotte, J.H. (2004). Particulate Matter in the Dutch Pollutant Emission Register: State of Affairs, TNO-report, R2004/428, Apeldoorn, The Netherlands.
- IEEP (2006). Proposed air quality Directive: Assessment of the Environmental impact of Parliament's amended proposal, Policy Brief for the EP Environmental Committee IP/A/ENVI/FWC/2005-35. (See http://www.mnp.nl/en/publications/2006/Assessment_new_limit_values_Particiulate_Matter.html)
- IPCC (2007). Intergovernmental Panel on Climate Change 4th Assessment Report, 2007; WG 1. See <http://ipcc-wg1.ucar.edu/wg1/wg1-report.html>.
- IRCEL (2007). Annual data reported by the Belgian Interregional Environment Agency. See <http://www.irceline.be>.
- Jaarsveld, J.A. van, 2004. The Operational Priority Substances model, description and validation of OPS-Pro 4.1, MNP-rapport 500045001/2004, Bilthoven, Nederland. (<http://www.rivm.nl/bibliotheek/rapporten/500045001.pdf>) (see also: <http://www.mnp.nl/ops>)
- Jonker, S. and Teeuwisse, S. (2006). Handleiding CARII, versie 5.0. TNO-report (in Dutch) 2006-A-R00078/B, Apeldoorn, the Netherlands.
- Kassteede, J. van de, Koelemeijer, R.B.A., Dekkers, A.L.M., Schaap, M., Homan, C.D. and Stein, A. (2006). Statistical mapping of PM₁₀ concentrations over Western Europe using secondary information from dispersion modeling and MODIS satellite observation. In: Stochastic environmental research and risk assessment (SERRA), 21, 2, 183-194.
- Koelemeijer, R.B.A., Homan, C.D. and Matthijsen, J. (2006). Comparison of spatial and temporal variations of aerosol optical thickness and particulate matter over Europe, *Atmos. Environ.*, 40, 5304-5315.
- Kuhlbusch, T. and A. John (2000). Korngrößenabhängige Untersuchungen von Schwebstaub und Inhaltsstoffen, Abschlußbericht für das Ministerium für Umwelt, Raumordnung und Landwirtschaft (in German), NordRein-Westfalen, Germany. See http://www.iuta.de/Verfahrenstechnik/Luftreinhaltung/luftpub_download.htm.
- LANUV NRW (2007). Annual data reported by the Landesamt für Natur, Umwelt und Verbraucherschutz NordRein-Westfalen, Germany. See <http://www.lanuv.nrw.de/luft/immissionen/aktluftqual/einzel.htm>.
- LML (2007). Data from the Dutch national air quality network. See <http://www.rivm.lml.nl>.
- Maenhaut, W., Chi, X. and Wang W. (2006). Analyses voor EC/OC en ionen in fijn

- stof, 2005-2006, UGent, eindverslag voor studie in opdracht van de Vlaamse Milieumaatschappij (in Flemish), Belgium. See <http://www.milieurapport.be>.
- Matthijssen, J. and Visser, H. (2006) PM₁₀ in Nederland, Rekenmethodiek, concentraties en onzekerheden. MNP report 500093005 (in Dutch), Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500093005.pdf>)
- MNP (2007a). Realisatie milieu doelen, voortgangsrapport 2007, Milieu- en Natuurplanbureau, rapport 500081002 (in Dutch), Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500081002.pdf>)
- MNP (2007b). Mileubalans 2007, Milieu- en Natuurplanbureau, rapport 500081004 (in Dutch), Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500081004.pdf>)
- NEN-14907 (2005). Nederlandse norm NEN-EN 14907; 2005 Ambient air quality - Standard gravimetric measurement method for the determination of the PM_{2,5} mass fraction of suspended particulate matter Nederlands Normalisatie-instituut, The Netherlands. See <http://www.nen.nl>.
- Olivier, J.G.J., Berdowski, J.J.M., Peters, J.A.H.W., Bakker, J., Visschedijk, A.J.H. and Bloos, J.-P.J. (2001). Applications of EDGAR. Including a description of EDGAR 3.0: reference database with trend data for 1970-1995. RIVM report 773301001 NOP report 410200051, Bilthoven, The Netherlands.
- PRIMES (2007) A modeling system that simulates a market equilibrium solution for energy supply and demand in the European Union. See <http://www.e3mlab.ntua.gr/>.
- Putaud, J.-P., Raes, F., Van Dingenen, R., Brüggemann, E., Facchini, M.C., Decesari, S., Fuzzi, S., Gehrig, R., Hüglin, C., Laj, P., Lorbeer, G., Maenhaut, W., Mihalopoulos, N., Müller, K., Querol, X., Rodriguez, S., Schneider, J., Spindler, G., ten Brink, H.M., Tørseth, K. and Wiedensohler, A. (2004). A European aerosol phenomenology-2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmos. Environ.*, 38, 2579-2595.
- Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., ten Brink, H., Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S., Harrison, R., Ayres, J., van der Zee, S. and Hoek, G. (2007). Spatial variation of particle number and mass over four European cities, *Atmos. Environ.*, in press.
- Quass, U. and Kuhlbusch, T. (2004). Identifizierung von Quellgruppen für die Feinstaubfraktion, IUTA-Report Nr. LP 15/2004 (in German). See http://www.iuta.de/Verfahrenstechnik/Luftreinhaltung/luftpub_download.htm.
- Querol, X., Alastuey, A., Ruiz, C.R., Artinano, B., Hansson, H.C., Harrison, R.M., Buringh, E., Brink, H.M. ten, Lutz, M., Bruckmann, P., Straehl, P. and Schneider, J. (2004). Speciation and origin of PM₁₀ and PM_{2,5} in selected European cities, *Atmos. Environ.* 38, 6547-6555.
- RAINS/GAINS (2007). Regional Air Pollution Information and Simulation/Greenhouse Gas and Air Pollution Interactions and Synergies. See <http://www.iiasa.ac.at/rains/gains/documentation.html>.
- RIVM (2008). Measurement data on PM_{2,5} mass-concentrations, to be published.
- Schaap, M., Timmermans, R.M.A., Sauter, F.J., Roemer, M., Velders, G.J.M., Boersen, G.A.C., Beck, J.P. and Bultjes, P.J.H. (2007). The LOTOS-EUROS model: description, validation and latest developments, *Int J. of Environment and Pollution*, in press.
- Schaap, M., Spindler, G., Schulz, M., Acker, K., Maenhaut, W., Berner, A., Wierprecht, W., Streit, N., Mueller, K., Brüggemann, E., Putaud, J.-P., Puxbaum, H., Baltensperger, U. and Brink, H.M. ten (2004). Artefacts in the sampling of nitrate studied in the INTERCOMP campaigns of EUROTRAC-AEROSOL. *Atmos. Environ.*, 38, 6487-6496.
- Schaap, M. and Denier van der Gon, H.A.C. (2007). On the variability of Black Smoke and carbonaceous aerosols in the Netherlands, *Atmos. Environ.* 41, 5908-5920.
- Schaap et al. (2007). Evaluation of long term aerosol simulations from seven regional air quality models and their ensemble in the EURODELTA study (submitted).
- Sillanpää, M., Frey, A., Hillamo, R., Pennanen, A.S. and Salonen, R.O. (2005). Organic, elemental and inorganic carbon in particulate matter of six urban environments in Europe, *Atmos. Chem. Phys.*, 5, 2869-2879.
- Smeets, W.L.M., Blom, W.F., Hoen, A., Jimmink, B.A., Koelemeijer, R.B.A., Peters, J.A.H.W., Thomas, R. and Vries, W.J. de (2007a). Kosteneffectiviteit van aanvullende maatregelen voor een schonere lucht, MNP report 500091001 (in Dutch), Bilthoven,

- The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500091001.pdf>)
- Smeets, W.L.M., Blom, W.F., Hoen, A., Jimmink, B.A., Koelemeijer, R.B.A., Peters, J.A.H.W. and Vries, W.J. de (2007b). Cost-effective abatement options for improving air quality in the Netherlands, Dustconference, 23-24 April, 2007, Maastricht. (<http://www.mnp.nl/bibliotheek/digitaaldepot/Costeffectiveabatementoptions.pdf>)
- Szidat, S., Jenk, T.M., Gäggeler, H.W., Synal, H.-A., Fisseha, R., Baltensperger, U., Kalberer, M., Samburova, V., Reimann, S. and Hajdas, I. (2004). Radiocarbon (¹⁴C)-deduced biogenic and anthropogenic contributions to organic carbon (OC) of urban aerosols from Zürich, Switzerland. *Atmos. Environ.* 38, 4035-4044.
- Tsigaridis, K. and Kanakidou, M. (2003). Global modelling of secondary organic aerosol in the troposphere: a sensitivity analysis. *Atmos. Chem. Phys.* 3, 1849-1869.
- UN-ECE (1998). The 1998 Protocol on Heavy Metals; 28 Parties. Entered into force on 29 December 2003. See http://www.unece.org/env/lrtap/status/lrtap_s.htm.
- UN-ECE (1999). The 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone; 23 Parties. Entered into force on 17 May 2005. See http://www.unece.org/env/lrtap/status/lrtap_s.htm.
- UN-ECE (2007). Report of the fourth and fifth meetings of the Expert Group on Particulate Matter by the Co-Chairs in collaboration with the secretariat. ECE/EB.AIR/WG.5/2007/18, 26 June 2007, Geneva.
- Velders, G.J.M., Waal, E.S. de, Jaarsveld, J.A. van and Ruiters, J.F. de (2003). The RIVM-MNP contribution to the evaluation of the EMEP Unified (Eulerian) model, RIVM report 500037002, Bilthoven, The Netherlands. (<http://www.rivm.nl/bibliotheek/rapporten/500037002.pdf>)
- Velders, G.J.M., Aben, J.M.M., Beck, J.P., Blom, W.F., Dam, J.D. van, Elzenga, H.E., Geilenkirchen, G.P., Hoen, A., Jimmink, B.A., Matthijsen, J., Peek, C.J., Velze, K. van, Visser, H., Vries W.J. de and Velders, G.J.M. (eds) (2007a). Concentratiekaarten voor grootschalige luchtverontreiniging in Nederland, Rapportage 2007, MNP report 500088001 (in Dutch), Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500088001.pdf>)
- Velders, G.J.M., Matthijsen, J., Aben, J.M.M. and Vries, W.J. de (2007b). Grootschalige PM_{2.5} concentratiekaarten van Nederland, Een eerste analyse, MNP report 500088003 (in Dutch), Bilthoven, The Netherlands. (<http://www.mnp.nl/bibliotheek/rapporten/500088003.pdf>)
- Viana, M., Maenhaut, W., Brink, H.M. ten, Chi, X., Weijers, E., Querol, X., Alastuey, A., Mikuška, P. and Večeřa, Z. (2007). Comparative analysis of Organic and Elemental Carbon concentrations in carbonaceous aerosols in three European cities *Atmos. Environ.*, in press
- Visser, H., Buringh, E. and Breugel, P.B. van (2001). Composition and origin of airborne particulate matter in the Netherlands, RIVM report 650010029, Bilthoven, The Netherlands. (<http://www.rivm.nl/bibliotheek/rapporten/650010029.pdf>)
- Visschedijk, A.J.H., Appelman, W., Hulskotte, J., Coenen, P. (2007). Onderhoud van methodieken Emissieregistratie 2006-2007, TNO report (in Dutch) 2007-A-R0865/B, TNO B&O, Apeldoorn.
- VROM (2003). Erop of eronder, Uitvoeringsnotitie emissieplafonds verzuring en grootschalige luchtverontreiniging 2003 (in Dutch). See <http://www.vrom.nl>.
- Wagner, F., Schöpp, W. and Heyes, C. (2006). The RAINS optimization module for the Clean Air For Europe (CAFE) Programme. IASA Interim Report IR-06-029. (<http://www.iasa.ac.at/Admin/PUB/Documents/IR-06-029.pdf>)
- Wagner, F., Amann, M. and Schöpp, W. (2007). The GAINS Optimization Module as of 1 February 2007. IASA Interim Report IR-07-004. (<http://www.iasa.ac.at/Admin/PUB/Documents/IR-07-004.pdf>)
- Wesseling, J.P. and Sauter F.J. (2007). Kalibratie van het programma CAR II aan de hand van metingen van het Landelijk Meetnet Luchtkwaliteit van het RIVM, RIVM report 680705004 (in Dutch), Bilthoven, The Netherlands. (<http://www.rivm.nl/bibliotheek/rapporten/680705004.pdf>)
- WHO (2000). Air Quality Guidelines for Europe, Second edition. Copenhagen, WHO Regional Office for Europe, 2000 (WHO Regional Publications, European Series, No 91).
- WHO (2003). Health Aspects of Air Pollution with Particulate Matter, Ozone and Nitrogen Dioxide. WHO, Bonn, Germany. (<http://www.euro.who.int/document/e79097.pdf>)
- WHO (2006a). WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: global update 2005: summary of risk assessment. document

number: WHO/SDE/PHE/OEH/06.02., World Health Organization, Geneva. (http://whqlibdoc.who.int/hq/2006/WHO_SDE_PHE_OEH_06.02_eng.pdf)

WHO (2006b). Health risks of particulate matter from long-range transboundary air pollution, document number: EUR/05/5046028, WHO Regional Office for Europe, Copenhagen. (<http://www.euro.who.int/document/e88189.pdf>)