# Policy research programme on particulate matter Main results and policy consequences

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

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## Main results and policy consequences

Jan Matthijsen (PBL) Robert Koelemeijer (PBL)

PBL Netherlands Environmental Assessment Agency

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Contact: jan.matthijsen@pbl.nl

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PBL Netherlands Environmental Assessment Agency

Location The Hague
P.O. Box 30314
2500 GH Den Haag
T 070 3288700
F 070 3288799
E: info@pbl.nl
www.pbl.nl

Location Bilthoven
P.O. Box 303
3720 AH Bilthoven
T 030-2742745
F 030-2744479

# Foreword

Elevated atmospheric concentrations of particulate matter are associated with adverse health effects and are the most important environmental disease burden the Netherlands and Europe. To counteract these effects, standards have been established at the European level for aspects such as the overall concentration of particulate matter ($PM_{10}$) and the finer fraction ($PM_{2.5}$) of particulate matter. These standards are the maximum allowable concentrations that European countries must comply with beginning in 2011 (for $PM_{10}$) and 2015 (for $PM_{2.5}$). In supplementation to European policy, generic national policy has been developed so that the Netherlands can comply on time with these standards everywhere in the country. In addition, location-specific measures are being taken that focus on eliminating the particulate matter 'hotspots'.

Scientific uncertainties in this dossier make it difficult to formulate effective and efficient policy. How do the natural and anthropogenic contributions to particulate matter vary in space and time? How much do various sources contribute and to which fractions? What are the policy targets for reducing particulate matter concentrations and what is the potential health impact of measures to control particulate matter? To answer these questions, the Ministry of Housing, Spatial Planning and the Environment (VROM) financed a policy research programme to improve the knowledge about particulate matter. The Policy Research Programme on Particulate Matter (BOP) was implemented by four cooperating research institutes in the Netherlands: the Energy Research Centre of the Netherlands (ECN), the PBL Netherlands Environmental Assessment Agency, the National Institute for Public Health and the Environment (RIVM) and TNO. The programme ran from 2007 to 2009. The research led to various new insights into the composition and sources of particulate matter and into the progress in particulate matter policy. These insights have in turn led to policy recommendations. The results have been summarised in 15 reports.

This report summarises the main results and the policy implications of these results.

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

Dr A.B.M. Hoff

Director of the PBL Netherlands Environmental Assessment Agency

Prof. M.A, Hajer

Director of the Department of Environment and Chemicals of the National Institute for Public Health and the Environment (RIVM/MEV)

Dr *Ir.* R.D. Woittiez

Director TNO Built Environment and Geosciences

*Ir.* D.Ph. Schmidt

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# Samenvatting

## Chemische samenstelling van fijn stof

- De totale bijdrage door menselijk handelen aan fijnstofconcentraties ($PM_{10}$) en van de fijnere fractie hiervan ($PM_{2,5}$) blijkt groter te zijn dan waar tot nu toe op basis van de beschikbare kennis van was uitgegaan: 25 procent meer voor $PM_{10}$ en 20 procent voor $PM_{2,5}$.
- Atmosferische omzettingsproducten van zwaveldioxide, stikstofoxiden en ammoniak dragen 50 procent meer bij aan de fijnstofconcentraties dan tot nu toe werd gemeten en berekend.
- Natuurlijke bronnen, waaronder zeezout, dragen minder bij dan volgens eerdere schattingen. Bij verhoogde $PM_{10}$-concentraties is de bijdrage van zeezout geringer.
- Nederlandse bronnen dragen meer bij aan de fijnstofconcentraties dan gedacht. Bij fijnstofconcentraties boven 30 microgram per kubieke meter (µg/m³) is de bijdrage van fijn stof uit stikstofoxiden en ammoniak extra hoog.
- Houtverbranding in kachels en open haarden dragen hooguit enkele procenten bij aan de gemiddelde $PM_{10}$-concentratie. Dit kan echter oplopen tot 30 procent voor $PM_{10}$ en 40 procent voor $PM_{2,5}$ in de wintermaanden op locaties waar veel hout wordt gestookt.
- Bovenstaande bevindingen over de samenstelling van fijn stof hebben geen invloed op de hoogte van de gemeten totale $PM_{10}$ en $PM_{2,5}$ concentraties. Het verandert dus niets aan het huidige aantal gemeten overschrijdingen van de $PM_{10}$- and $PM_{2,5}$-normen.

## Trends in concentraties

- De $PM_{10}$-concentraties zijn tussen 1993 en 2007 met 24 tot 32 procent gedaald. Deze daling is in lijn met de ontwikkelingen van de relevante antropogene - door menselijk handelen veroorzaakte - emissies.
- Alle gemeten concentraties van antropogene bestanddelen van fijn stof zijn in de afgelopen 20 jaar in Nederland gedaald. Het gaat om zware metalen, zwarte rook en fijn stof uit zwaveldioxide, stikstofoxiden en ammoniak. De mate waarin verschilde echter wel per bestanddeel. De grootste dalingen vonden plaats tussen 1990 en 2000.
- De concentraties van zwarte rook namen tussen 1990 en 2007 in buitenstedelijke gebieden af met 50 procent. Wegverkeer is waarschijnlijk de belangrijkste bron van dit type fijn stof uit verbrandingsprocessen. In steden zijn de zwarterooktrends echter niet zo eenduidig.

## Verwachte ontwikkelingen

- Projecties voor de emissies van $PM_{10}$ en $PM_{2,5}$ leren dat alle Europese normen voor $PM_{2,5}$ waarschijnlijk haalbaar zijn op basis van het huidige en het voorgenomen nationale en Europese emissiebeleid. Een onzekere factor hierbij is het percentage waarmee de gemiddelde $PM_{2,5}$ concentratie in steden moet afnemen tussen 2010 en 2020. De hoogte van deze doelstelling is afhankelijk van de concentraties in 2009, 2010 en 2011 en zal daarom niet eerder dan 2012 met zekerheid bekend zijn. Nu wordt een doelstelling van 15 procent verwacht, maar deze kan 20 procent worden. Om een afname van 20 procent te realiseren is waarschijnlijk additioneel nationaal en Europees beleid nodig.
- Voorziene maatregelen zullen mogelijk meer effect sorteren dan met de huidige projecties aanvankelijk was becijferd. De verwachte $PM_{10}$ concentratiedaling tussen 2010 en 2020 zal maximaal met ongeveer 50 procent extra dalen, ongeveer 3 µg/m³ in plaats van circa 2 µg/m³.

## Gezondheidseffecten

- De gemiddelde gezondheidswinst is grofweg 30 procent geweest in Nederland tussen 1993 en 2007 als deze aan de blootstelling aan $PM_{10}$ zou worden afgemeten. Niet alle bestanddelen van fijn stof zijn even relevant voor de gezondheid. Als het gezondheidseffect wordt afgemeten aan de blootstelling aan fijn stof uit verbrandingsprocessen, een bestanddeel van fijn stof dat juist als gezondheidsrelevant geldt, dan is de gezondheidswinst mogelijk nog groter geweest.
- $PM_{10}$- of $PM_{2,5}$-concentraties zijn minder geschikt om op lokale schaal het effect te volgen van maatregelen op emissies die vanuit gezondheidsoogpunt als relevant gelden. Uit nader onderzoek moet blijken of zwarte rook of elementair koolstof kunnen dienen als een aanvullende indicator voor het fijn stof uit verbrandingsprocessen.

## Beleidsconsequenties

- Vermindering van de emissies van stikstofoxiden en ammoniak in Nederland en Europa is het effectiefste middel om te voldoen aan de normen voor fijn stof.
- Herziening van de regeling voor de aftrek van zeezout bij overschrijding van de grenswaarden van $PM_{10}$ is gewenst.

Want de bijdrage van zeezout aan de fijnstofconcentraties is minder dan tot nu toe werd aangenomen.

- De grotere antropogene bijdrage aan de fijnstofconcentraties betekent in principe dat beleidsmaatregelen om de concentraties van fijn stof te verlagen effectiever kunnen zijn. De vraag in welke mate dit het geval is zal worden behandeld in een vervolg op het beleidsgericht onderzoeksprogramma fijn stof.

# Findings

# Policy Research Programme on Particulate Matter

## Main results and policy consequences

### Summary

#### Chemical composition of particulate matter

- The total anthropogenic contribution to concentrations of particulate matter ($PM_{10}$) and to the finer fraction of particulate matter ($PM_{2.5}$) is higher – by 25 and 20 per cent, respectively – than was previously assumed on the basis of available data.
- Contributions to particulate matter from the atmospheric transformation products of sulphur dioxide, nitrogen oxides and ammonia appear to be 50 per cent larger than were measured and calculated in the past..
- Contributions from natural sources, such as sea salt, appear to be smaller than earlier estimations. In elevated $PM_{10}$ concentrations, the contribution of sea salt is smaller.
- Contributions from Dutch sources to PM concentrations are larger than previously thought. When PM concentrations rise above beyond 30 µg/m³, there is an extra increase in the contribution of particulate matter from nitrogen oxides and ammonia.
- The relative share from wood-burning heaters and fireplaces to average $PM_{10}$ concentrations is no more than a few per cent. During winter months, however, this share can increase up to 30 per cent for $PM_{10}$ and 40 per cent for $PM_{2.5}$ in locations with many wood burners.
- The above findings on the composition of particulate matter have no influence on the measured total $PM_{10}$ and $PM_{2.5}$ concentration levels. It therefore does not change the current number of measured exceedances of the $PM_{10}$ and $PM_{2.5}$ standards.

#### Trends in concentrations

- Between 1993 and 2007, $PM_{10}$ concentrations have decreased by 24 to 32 per cent. This decrease is consistent with the developments of the relevant anthropogenic emissions – those resulting from human activities.
- In the Netherlands, all measured concentrations of anthropogenic constituents of particulate matter have decreased over the past 20 years. This decrease involved heavy metals, black smoke, and secondary particulate matter from sulphur dioxide, nitrogen oxides and ammonia. However, the observed decreases varied between constituents. The largest decreases occurred between 1990 and 2000.
- Between 1990 and 2007, concentration levels of black smoke decreased by 50 per cent in outer urban areas. Traffic-related combustion processes are likely to be the dominant source of this type of particulate matter. Inside the urban area, however, the trend is not as clear.

#### Expected developments

- According to projections of $PM_{10}$ and $PM_{2.5}$ emissions based on present and proposed national and European emission policies, all European standards for $PM_{2.5}$ can probably be attained. One uncertainty regarding this attainability is the required average decrease in $PM_{2.5}$ concentrations in the urban area between 2010 and 2020. This target level depends on the concentration levels in 2009, 2010 and 2011, and will therefore will not be known for certain until 2012. The target level is now expected to be around 15 per cent, but could be as high as 20 per cent. Attaining a 20 per cent decrease would probably require additional national and European policy.

- The proposed measures could be more effective than initially projected. Between 2010 and 2020, the maximum additional decrease in $PM_{10}$ concentrations is expected to be 50 per cent (about 3 µg/m³ instead of 2 µg/m³).

### Health effects

- Between 1993 and 2007, the average positive health impact in the Netherlands, if measured according to exposure to $PM_{10}$, would have been roughly 30 per cent. But not all constituents of particulate matter are equally relevant to health. If the health impact was measured according to exposure to particulate matter from combustion processes – a constituent that is considered to be especially health-relevant – the positive effect might have been even greater.
- On the local scale, $PM_{10}$ and $PM_{2.5}$ concentrations are less suitable for monitoring the effect of health-relevant emission reduction measures. Research is currently being done to determine if black smoke or elementary carbon could be used as an additional indicator of particulate matter from combustion processes.

### Policy consequences

- The most effective means of meeting the standards for particulate matter would be to decrease emissions of nitrogen oxides and ammonia in the Netherlands and in Europe.
- Revision of the sea salt deduction during exceedances of the $PM_{10}$ limit values is advisable, because the sea salt contribution is less than previously assumed.
- The larger anthropogenic contribution to PM concentrations theoretically means that there is a greater potential for policy measures to decrease concentration levels of particulate matter. To what extent this is actually the case is a question that will be addressed in a follow-up to the BOP research.

## Introduction

Elevated atmospheric concentrations of particulate matter are associated with adverse health effects; in fact, this is the most important environment-related disease burden in the Netherlands and Europe. To counteract these adverse health effects, standards have been established at the European level for aspects such as the concentration of particulate matter ($PM_{10}$) and the finer fraction of particulate matter ($PM_{2.5}$). This concerns maximum allowable concentrations of the two fractions; the limit value for $PM_{10}$ will be in force beginning in 2011 and that for $PM_{2.5}$ in 2015.

Particulate matter is an important policy concern especially in the Netherlands because the limit values for $PM_{10}$ were widely exceeded here. A complication in the Dutch situation is that the options for reducing these exceedances appear to be limited. Moreover, failure to comply on time with European legislation has turned out to have serious economic consequences; exceedances of air quality standards can result in planned development projects being stopped or delayed. In supplementation to European policy, generic national policy has been developed so that the Netherlands can comply on time with these standards everywhere in the country. In addition, location-specific measures are being taken that focus on eliminating particulate matter 'hotspots' (local exceedances).

Due to the scientific uncertainties about particulate matter, it has not always been easy to formulate effective and efficient policy. This uncertainty concerns questions such as the following:
How do the natural and anthropogenic contributions to particulate matter vary temporally and spatially? How much do various sources contribute, and to which fractions?
What are the policy targets for reducing particulate matter concentrations?

In addition, there are many questions about the trend in particulate matter concentrations, both in the Netherlands and elsewhere in Europe. However, understanding the trend is a precondition to evaluating the effectiveness of the policy. To answer these questions, the Ministry of Housing, Spatial Planning and the Environment (VROM) financed a policy research programme on particulate matter, which ran from 2007 to 2009.

### Research aims and mode of operation

The Policy Research Programme on Particulate Matter (BOP) aimed to improve the knowledge about particulate matter so that policy making could be supported more adequately in the future. The most important research aims of the programme were:

- Improving the knowledge about $PM_{10}$ and $PM_{2.5}$ concentrations and about the composition and sources of particulate matter.
- Improving the understanding of the behaviour of particulate matter in the urban area.
- Determining the trends in particulate matter concentrations and in the individual constituents of particulate matter.
- Clarifying the effects of past and the future policy measures on the $PM_{10}$ and $PM_{2.5}$ concentrations.

The research focussed on situations where limit values were exceeded. Exceedances of the European limit values for $PM_{10}$ still occur in the urban area or near motorways and at locations with high local emissions. The constituents of particulate matter and the contributions from various anthropogenic and natural sources are still insufficiently known, especially in situations where limit values are exceeded. The knowledge about the spatial differences in particulate matter concentrations is also limited. This concerns not only the spatial variability of particulate matter in the urban area, but also the differences between urban and rural areas.

The Policy Research Programme on Particulate Matter (BOP) was implemented by four cooperating research institutes in the Netherlands: the Energy Research Centre of the Netherlands (ECN), the PBL Netherlands Environmental Assessment Agency, the National Institute for Public Health and the Environment (RIVM) and TNO Built Environment and Geosciences.

In addition, the DCMR Environmental Protection Agency, the municipality of Breda, the Municipal Health Service of Amsterdam and Wageningen University and Research Centre contributed to the research programme by providing measurements, measurement facilities and/or research capacity.

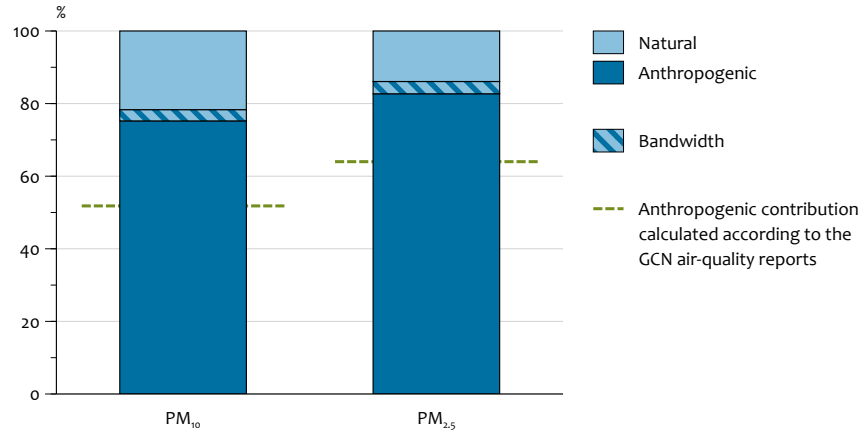
BOP was completed in 2009 and has provided much new information. The results have been published in a separate series of 15 reports. Some of these reports concerned the composition and sources of particulate matter, with specific attention to constituents such as sea salt, secondary inorganic aerosol, elementary carbon (EC) and organic carbon (OC). Other BOP reports addressed topics such as particulate matter concentrations in the urban area, trends in particulate matter concentrations, particulate matter emissions from shipping, EC and OC emissions from traffic, particulate matter emissions from wood combustion and the feasibility of new norms for $PM_{2.5}$. Technical details about the research programme were addressed in two background documents: one about measurements and one about model developments.

Even before the programme was completed, results from the BOP research were used to support national and European air-quality policy. For example, the initial study on the finer fraction ($PM_{2.5}$) of particulate matter (Matthijsen & ten Brink 2007) played an important role in determining the European standard for $PM_{2.5}$. Results from the BOP research were also used in the National Air Quality Cooperation Programme (NSL) and in the future outlook studies on particulate matter (Matthijsen et al. 2009; Velders et al. 2009; PBL 2009a).

### Reader's guide

The first part of this study, the Findings, presents the most important results of the research. The following topics are discussed: the physical and chemical composition of particulate matter, trends in concentrations, expected

**Anthropogenic and natural contributions to particulate matter, 2007 – 2008** **Figure 1**




*Approximately 50 per cent of $PM_{10}$ and 65 percent of $PM_{2.5}$ are calculated to be anthropogenic in origin (based on registered sources). However, BOP measurement data indicate that the anthropogenic contribution to both $PM_{10}$ and $PM_{2.5}$ is significantly greater.*

developments, adverse health effects, policy consequences and suggestions for continuing the particulate matter dossier after the completion of the programme. In the second part of the study, the Deepening, the findings are accounted for and the underlying analyses are explained.

## Chemical composition of particulate matter

### Larger anthropogenic contribution to particulate matter

Atmospheric particulate matter consists of various constituents; some of these constituents are of natural origin and some of them enter the atmosphere due to human activities (the anthropogenic contribution). On average, particulate matter ($PM_{10}$) consists of 75 to 80 per cent anthropogenic constituents; for the finer fraction of particulate matter ($PM_{2.5}$) this is 85 to 90 per cent (see Figures 1 and 2). At heavily affected locations, such as urban roads, the anthropogenic contribution to $PM_{10}$ is more than 80 per cent. Figure 1 shows the distribution between anthropogenic and natural contributions to $PM_{10}$ and $PM_{2.5}$ based on data from BOP and on air quality reports. The bandwidth indicates the uncertainty about the anthropogenic constituent in carbonaceous particulate matter. Figure 2 shows the average composition of $PM_{10}$ and $PM_{2.5}$. Anthropogenic constituents that were previously underestimated consist of secondary particulate matter formed in the air from volatile organic compounds, sulphur dioxide, nitrogen oxides and ammonia, and the anthropogenic mineral dust contribution to primary particulate matter.

### Secondary inorganic aerosol is more important than previously assumed

The largest modification of the anthropogenic component resulted from improved understanding of the particulate matter that is formed from sulphur dioxide, oxides of nitrogen and ammonia. This component turned out to be approximately 50% larger than previously measured and calculated; this result is also in accordance with measurements taken in Belgium and Germany. Secondary aerosol is particulate matter that is formed in the air and is not emitted directly from a source, which is the case with primary particulates.

### New insights could lead to more effective policy measures

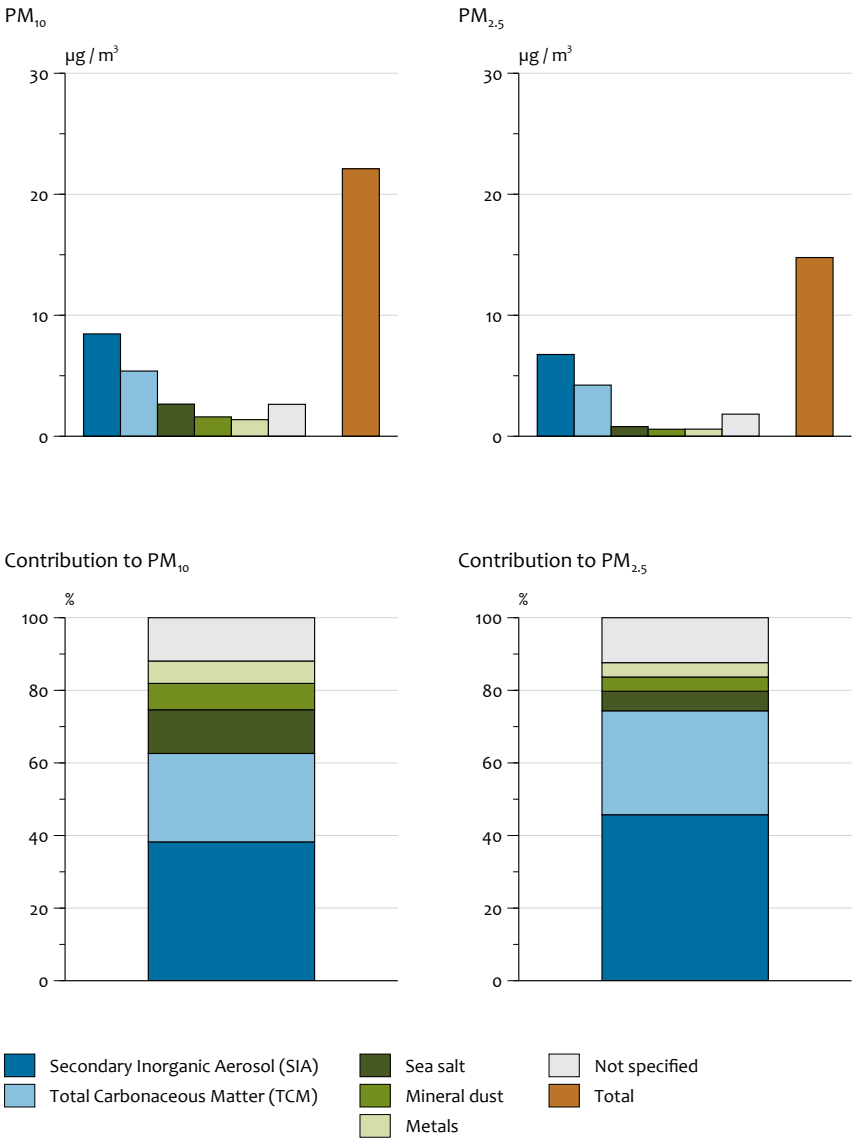
These findings about the composition of particulate matter have no effect on the total particulate matter concentration as it is measured. However, a larger anthropogenic component means that government policy theoretically has a greater potential to reduce the $PM_{10}$ and $PM_{2.5}$ concentrations than was previously thought. The extent to which this is actually the case cannot be determined precisely at this time. Policy measures to reduce emissions of sulphur dioxide, nitrogen oxides and ammonia could possibly be more effective in reducing total particulate matter concentrations than indicated in the current projections. If the maximum effect is achieved, the expected $PM_{10}$ concentration could be reduced by an additional 50 per cent between 2010 and 2020.

### Continuing uncertainties about mineral dust and carbonaceous particulate matter

Part of the anthropogenic particulate matter consists of dust that is resuspended by traffic and by agricultural activities. This source of particulate matter is very uncertain and is not included in the Netherlands Pollutant Release & Transfer Register. The contribution of anthropogenic mineral dust to the particulate matter concentrations it is therefore not calculated separately for the air quality reports. Moreover, measures to reduce the particulate matter contribution from road dust appear to have little or no effect, as shown from previous outlook studies in the Netherlands and Germany.

Carbonaceous particulate matter is partly anthropogenic and partly natural in origin. The ratio of anthropogenic to natural carbonaceous constituents is uncertain. The anthropogenic constituent is estimated at 50 to 75 per cent of the total quantity of carbonaceous particulate matter. Part of the carbonaceous particulate matter of anthropogenic origin

**Composition of particulate matter concentration, 2007 – 2008** **Figure 2**




*Secondary inorganic aerosol and carbonaceous particulate matter supply the largest contribution to both $PM_{10}$ and $PM_{2.5}$.*

is emitted directly, and part is formed in the atmosphere from volatile organic compounds. Although the emissions of both constituents are included in the Netherlands Pollutant Release & Transfer Register, little is known about the effects of measures to reduce volatile organic compounds on the particulate matter concentration. However, these measures are thought to be relevant for reducing the adverse health effects of particulate matter.

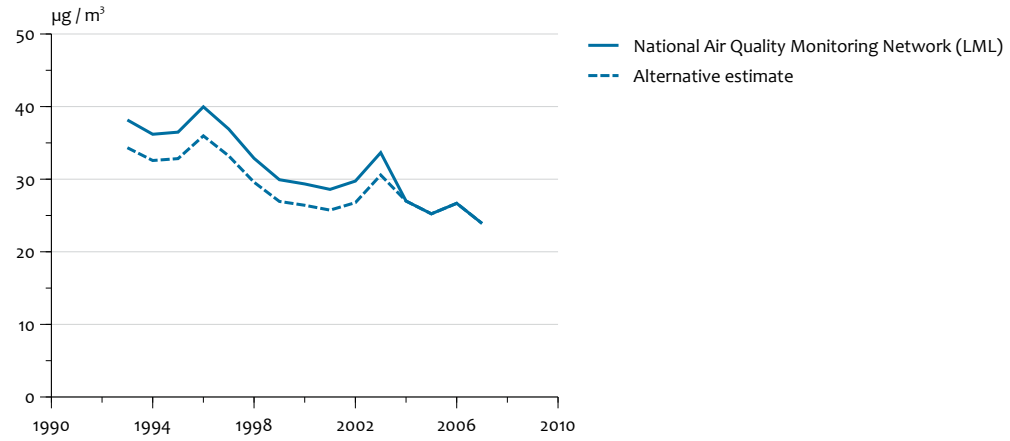
### Contribution of sea salt is significantly lower

Sea salt aerosol is of natural origin and in the Netherlands contributes 12 per cent on average to the $PM_{10}$ concentrations and 5 per cent to $PM_{2.5}$ concentrations. However, this contribution is greatly affected by weather conditions and varies both temporally and spatially. In 2007 and 2008, the concentration of sea salt aerosol ranged from 4 µg/m³ in Rotterdam, which is near the coast, to 2 µg/m³ in Vredepeel, which is further inland. For 24-hour average concentrations, maximums were measured ranging between 16 µg/m³ in Rotterdam and 10 µg/m³ in Vredepeel. The contribution of sea salt to $PM_{2.5}$ turned out to be one-third on average of its contribution to $PM_{10}$. On days with elevated particulate matter concentrations, the contribution of sea salt is actually lower than average (less than 1 µg/m³), because on such days there is usually a land breeze.

### Sea salt deduction

Sea salt particles are not harmful to health. European rules allow the contribution of sea salt to particulate matter to be disregarded when determining compliance with particulate matter legislation. This is known as the 'sea salt deduction'. The intention of the sea salt deduction is to create a level playing field for European Member States when tackling their air quality problems. As a result, Member States are not required to make additional efforts if there happens to be a high contribution from natural sources. The disadvantage of this

**$PM_{10}$ concentration** **Figure 3**




*Between 1993 and 2007, the $PM_{10}$ concentrations fell by 0.7 to 1.0 µg/m³ per year on average. This is equivalent to a decline of 24 to 32 per cent since the beginning of the measurements.*

policy is that the limit values for particulate matter, as a result of deducting the natural contribution, provide a somewhat lower level of health protection. This is because the deduction leads de facto to a relaxation of the standard. The sea salt deduction allows additional anthropogenic contributions, while still complying with the standard. Unlike sea salt, however, the anthropogenic contribution is probably hazardous. Essentially, the sea salt contribution can then be replaced with additional anthropogenic contributions up to the limit value.

### Revision of the sea salt deduction

The Netherlands deducts the contribution of sea salt in accordance with the Air Quality Assessment Scheme 2007. However, BOP research indicates that the scheme for deducting sea salt when the limit values of $PM_{10}$ are exceeded results in a too high sea salt deduction. Revision of the sea salt deduction is therefore desirable from a health perspective. If the sea salt deduction was eliminated, the most critical limit value for particulate matter – the 24-hour average particulate matter concentration – would become somewhat more stringent. It is still unclear to how a smaller sea salt deduction would affect the number of hotspots in the National Air Quality Cooperation Programme (hotspots are locations where the limit value for 24-hour average $PM_{10}$ concentrations is exceeded).

## The trend in particulate matter concentrations

### Declining concentrations

Between 1993 and 2007, the $PM_{10}$ concentrations fell by 0.7 to 1.0 µg/m³ per year on average (Figure 3). Approximately two-thirds of this decline was the result of reduced emissions, especially of sulphur dioxide, and to a lesser extent nitrogen oxides and ammonia. The remainder of the decline (one-third) was due to reduced emissions of primary particles, secondary carbonaceous particulate matter and water on particulates (assuming that the quantity of water on particulates is proportional to the quantity of particulate matter from sulphur dioxide, nitrogen oxides and ammonia).

The annual progression of the decline is difficult to interpret due to a combination of the large measurement uncertainty and the year-to-year variations in particulate matter caused by the weather, which are on the order of 2.5 µg/m³. Since 2000, the rate of decline in emissions and concentrations has flattened out. Consequently, the trend since 2000 is not significant. A slightly declining trend cannot be distinguished from a non-declining trend, even if the effects of weather on particulate matter concentrations are taken into account.

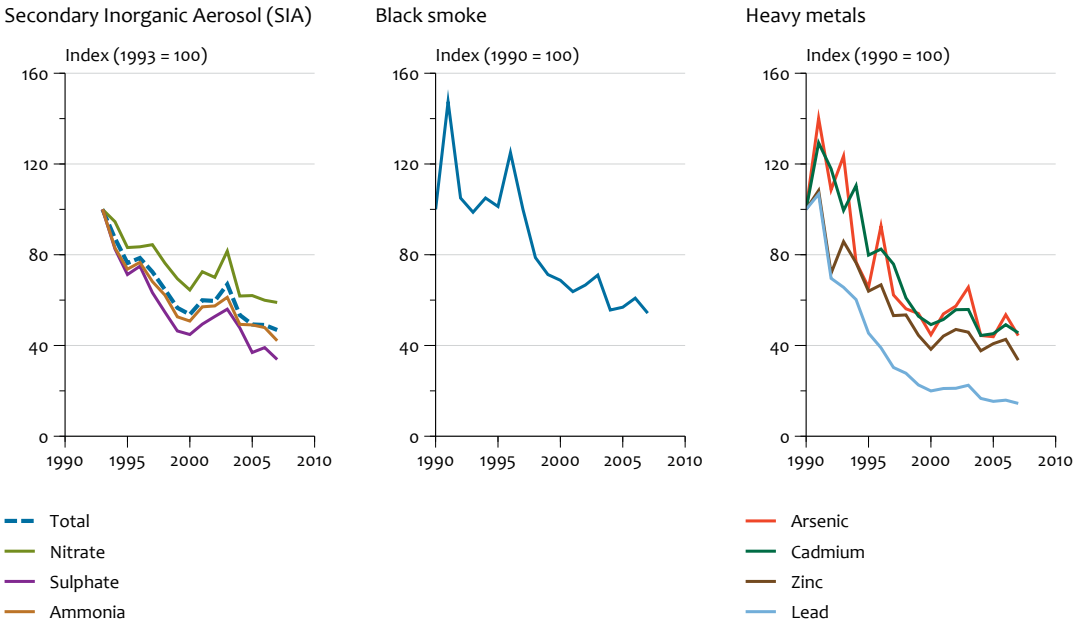
In the Netherlands, all measured concentrations of anthropogenic constituents of particulate matter have decreased over the past 20 years (see Figure 4). This decrease concerned particulates from sulphur dioxide, nitrogen oxides and ammonia, and those from black smoke and heavy metals. However, the observed decreases varied between the constituents. The largest decreases occurred between 1990 and 2000. Between 1990 and 2007, concentrations of black smoke decreased by 50 per cent outside the urban areas. Traffic-related combustion processes are likely to be the dominant source of this type of particulate matter. In urban areas, however, the black smoke trends are not as clear; stagnating concentrations and declining trends have both been observed.

In addition, there are still many questions about the trend in particulate matter concentrations, both in the Netherlands and elsewhere in Europe. According to recently published research, the concentrations in Europe no longer appear to be declining, even though the relevant emissions still are declining. In the Netherlands, however, there is no discrepancy between the trend in measured concentrations and emissions.

### Composition of particulate matter is location-dependent

The composition of particulate matter can differ according to location, and there appear to be location-dependent variations in the trend. If such local differences have a significant health impact, then $PM_{10}$ and $PM_{2.5}$ by themselves are inadequate indicators of the health effects of policy measures.

**Regional background concentration of particulate matter components in the Netherlands** **Figure 4**




*In the outer urban areas of the Netherlands, all measured concentrations of anthropogenic particulate matter constituents have declined on average. The largest decline occurred between 1990 and 2000.*

An average of 30 to 40 per cent of particulate matter is formed from nitrogen oxides, ammonia and sulphur dioxide in gaseous form. On days with elevated concentrations (above 30 µg/m³), this percentage is even higher (Figure 5). On such days, other particulate matter constituents do not appear to increase disproportionally, with the exception of constituents that have not yet been chemically specified. In contrast, the contribution of sea salt on such days appears to be especially low. The combination of low-velocity winds from the east or south and the high emission density in Northwest Europe, especially of ammonia and nitrogen oxides, plays an important role in causing exceedances of the limit value for 24-hour average particulate matter concentrations. Consequently, elevated particulate matter concentrations in the Netherlands are a pre-eminently anthropogenic phenomenon.

Policy that aims to reduce emissions of nitrogen oxides and ammonia would therefore be the most effective way of meeting the standards for particulate matter. This reduction is also important for alleviating the excessive nitrogen deposition on natural habitats. But it is probably of limited importance for protecting human health. For this purpose, large-scale control of combustion aerosol – especially soot and metals – appears to be more relevant.

### Elementary carbon as an indicator

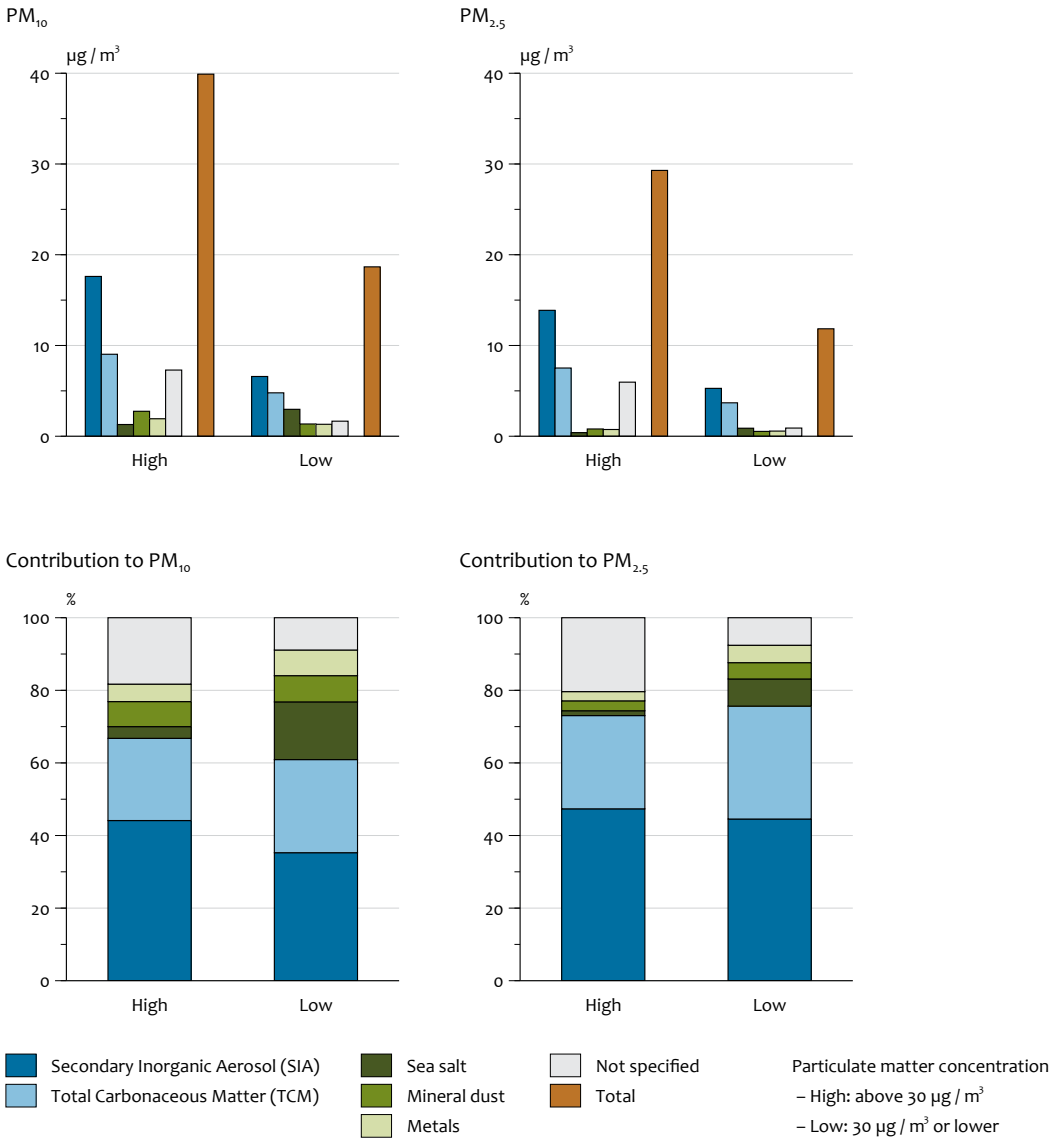
Model results for Rotterdam have shown large differences between local concentrations of elementary carbon (EC) near busy roads and the urban background concentration of this constituent. These differences were much larger than those for $PM_{2.5}$ and $PM_{10}$ (Figure 6). EC appears to be a good indicator for the dispersal of exhaust emissions of particulate matter from road traffic. From that health perspective, measures that aim to reduce metals and carbonaceous particulate matter, especially soot, have priority. Moreover, in the urban area and near roads, policy measures focusing on these fractions are even more effective; this is because their contributions at these locations have been shown to be even larger than those at measurement locations in the rural area.

### The $PM_{2.5}$ situation has now been mapped out

In 2007, the National Institute for Public Health and the Environment (RIVM) began taking $PM_{2.5}$ measurements according to the European reference method. These measurements showed that the yearly average concentrations of $PM_{2.5}$ in the Netherlands frequently fluctuated between 16 and 18 µg/m³. As expected, simultaneous measurements of the composition of $PM_{10}$ and $PM_{2.5}$ have shown that the anthropogenic contribution to $PM_{2.5}$ is significantly larger. However, the spatial gradients of the $PM_{2.5}$ concentrations were smaller than those for $PM_{10}$.

In 2008, European standards for $PM_{2.5}$ were established because this fraction was seen as having a larger health impact than $PM_{10}$. Member States must comply with the limit values for $PM_{2.5}$ beginning in 2015. Outlook studies based on measurements and calculations have shown that current and planned air-quality policy – which is being implemented for purposes such as complying with the limit values for $PM_{10}$ – will probably be sufficient for complying with the standards for $PM_{2.5}$ as well. This was a point of concern because the limit value for the 24-hour average concentrations of $PM_{10}$ is more stringent than the new standards for $PM_{2.5}$, at least in the Netherlands. Moreover, the 'best efforts obligation' to reduce the concentration of $PM_{2.5}$ in the urban living environment by at least 15 per cent during the next ten years appears to be attainable with the proposed emission policy. The target that will be imposed on the Netherlands to reduce the average $PM_{2.5}$ concentration in cities between 2010 and 2020 will

**Composition of high (elevated) and low particulate matter concentrations, 2007 – 2008** **Figure 5**




*On days with elevated concentrations (above 30 µg/m³), the proportion of secondary particulate matter from gaseous nitrogen oxides, ammonia and sulphur dioxide is relatively larger.*

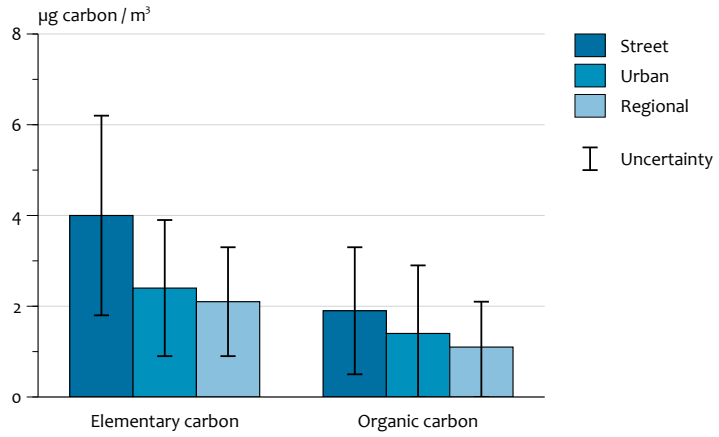
depend on the measurement results in 2009, 2010 and 2011. A reduction target of 15 per cent appears likely, but 20 per cent is also possible. In the latter case, the current and proposed emission policy will probably be inadequate.

### Wood combustion is sometimes an important source of particulate matter

According to the Netherlands Pollutant Release & Transfer Register, the particulate matter emissions caused by wood combustion in wood-burning stoves, fireplaces and furnaces for small-scale energy production amount to less than 5 per cent of the primary anthropogenic emissions of particulate matter in the Netherlands. However, it is uncertain how much air pollution actually results from the use of wood stoves; the pollution is strongly dependent on the type of fuel, the type of stove and how the stove is operated. In any case, the direct emission of particulate matter from wood-burning stoves is assumed to have a relevant health impact. Because this emission takes place in residential areas, increased exposure to particulate matter resulting from wood combustion by private parties can occur quickly and can lead to nuisance, especially for susceptible groups. The progression in the emissions from word-burning stoves during the next 15 years is very uncertain. This is because many factors can affect this process, including the energy price.

Initial measurements have shown that wood combustion, averaged year-round, contributes from 0.1 to 0.2 µg/m³ to the particulate matter concentrations in the urban area. This is equivalent to a contribution of approximately 1 per cent of $PM_{10}$. During the winter, this contribution is higher (0.2-0.4 µg/m³), and the 24-hour average can occasionally rise to several µg/m³. The contribution to particulate matter concentrations from wood combustion is seasonal. During the winter months, the contribution from wood combustion in Amsterdam was approximately 8 times higher than in the

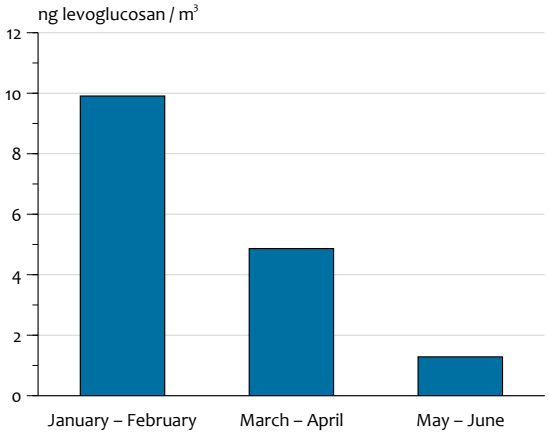
**$PM_{10}$ concentration of carbonaceous components, 2007 – 2008** **Figure .6**




*There are major differences in the contribution of elementary and organic carbon to the particulate matter concentrations, depending on the type of location.*

**Concentration due to wood combustion, 2006** **Figure 7**

Measurement location Amsterdam-Vondelpark




*Levoglucosan is a good indicator of the contribution of wood combustion to particulate matter concentrations.*

summer (Figure 7). In areas where ample supplies of firewood are available, incidentally higher concentrations of particulate matter caused by wood combustion have been measured, which were as high as 6 µg/m³ on average per month. In these cases, wood combustion can lead to exceedances of the limit value for the 24-hour average $PM_{10}$ concentration (50 µg/m³). In contrast to the local contribution to particulate matter concentrations from agriculture, storage/transshipment of goods and traffic, the local contribution from wood combustion has been mapped out much less thoroughly with models and measurements. Based on the measurements, it is advisable to improve our understanding of the contribution of wood combustion to particulate matter in the Netherlands, for example with models. However, for this purpose an improved temporal and spatial description of the emissions from wood combustion is required.

### Particulate matter policy in a broader context

The air-quality policy for reducing particulate matter concentrations in the Netherlands also affects other environmental themes. Measures that reduce emissions of sulphur dioxide, nitrogen oxides and ammonia are effective for attaining the limit values for both $PM_{10}$ and $PM_{2.5}$. Moreover, these measures reduce the deposition of nitrogen and sulphur on ecosystems and other vulnerable areas. Policy that aims to reduce emissions of nitrogen oxides and volatile organic compounds not only benefits the particulate matter concentrations at ground level, but also the ozone concentrations. As for climate change – and to a lesser extent for ozone at ground level – there are advantages and disadvantages. For example, the policy to reduce the emissions of sulphur dioxide has led to a decline in particulate matter concentrations. But at the same time this probably led to increased global warming due to a reduced direct or indirect cooling effect of particulate matter from sulphur

dioxide. In contrast, the reduction in black particles, such as those in the form of elementary carbon, is probably beneficial for both the climate and public health.

### Representativeness of the measurement data

The measurement campaigns of the BOP research programme provide a 'snapshot' of the particulate matter situation between 2007 and 2008. Particulate matter has many different and variable sources in the Netherlands and abroad. As a result, the representativeness of a measurement campaign to approximate the particulate matter situation in the Netherlands is limited by the number of measurement points and the measurement frequency. BOP researchers measured the total particulate matter concentration and its composition at six locations, consisting of three regional locations, one urban location and two street locations. The composition and concentration of particulate matter and the differences according to type of location are therefore only indicative for other parts of the Netherlands during the BOP research. The concentration differences for $PM_{10}$ and $PM_{2.5}$ between the urban location and the three regional locations turned out to be small on average: 0 to 3 µg/m³ ($PM_{10}$) and 0 up to 1 µg/m³ ($PM_{2.5}$). These differences were less than or about the same as the mutual concentration differences between the three regional locations. This measurement setup could therefore not be used to determine the significance of the contribution to particulate matter from urban sources. In subsequent research, the urban contribution will be studied with measurements taken specifically for this purpose.

### Consistency of the measurement data

To investigate the consistency of the BOP measurement data with other data on particulate matter, the various studies in BOP were expanded wherever possible with a European component. The BOP measurement results were compared with existing measurements from the Netherlands and the rest of Europe and calculations from the LOTOS-EUROS model. This chemical transport model describes the air quality for all of Europe and is used to describe the entire air quality chain, from emission to concentration and removal. In addition, the BOP measurement data was analysed with a statistical method in order to identify sources. This independent review of the measurement data provided additional support for the conclusions about the sources of particulate matter.

### Comparison with previous research

Between 1998 and 1999, a measurement campaign was conducted in the Netherlands: the *Bronstofonderzoek*. Since then, the knowledge about the various health aspects of particulate matter and the source contributions has changed and improved. The *Bronstofonderzoek* can be characterised as the first major study of the sources of particulate matter in the Netherlands. However, primarily for technical reasons, the results from that study are not directly comparable with those from BOP. For example, at that time there were no agreements about reference methods for sampling $PM_{10}$ and $PM_{2.5}$. Consequently, the contribution from natural sources in the BOP estimate (approximately 5 µg/m³) was at least 40 per cent lower than indicated by the earlier *Bronstofonderzoek*.

### Measurement campaigns in other countries from a Dutch perspective

Measurement campaigns have also been conducted in Belgium and Germany to determine the composition and sources of particulate matter. The most recent campaign in Germany coincided partly with the campaign in the Netherlands. The study in Belgium took place in 2006 and 2007. When making conclusions based on the Dutch data, the results of these other campaigns were taken into account as much as possible. Striking differences were found between the mineral dust contribution in Belgium, Germany and the Netherlands. In Belgium, the contribution of mineral dust to the $PM_{10}$ concentrations was approximately twice as high as that in the Netherlands. In Germany, the mineral dust concentrations were only slightly higher than those in the Netherlands. As yet, there is no definitive explanation for these differences. Differences were also found in contributions from elementary carbon and organic carbon to the increase in urban particulate matter concentrations relative to the regional background.

## Possible subsequent steps

To apply the acquired knowledge for implementing air-quality policy, additional analysis and research are required. At the same time, there are still major gaps in the knowledge about some aspects of particulate matter. Acquiring this knowledge is essential for the policy agenda on particulate matter during the period up to 2020 and possibly beyond. The policy agenda is being driven by European legislation on air quality and climate change.

The following aspects will still be important in the future:

- The recent insights into the magnitude of the anthropogenic contribution to particulate matter (caused by human activities) have consequences for particulate matter diagnoses and projections. To quantify these consequences, the following steps are required.
  First, follow-up research is required to determine the extent to which anthropogenic source contributions that may have been disregarded or underestimated in the calculations for air quality reporting can be included in this reporting.
  Second, measurements are required to determine why the particulate matter from sulphur dioxide, nitrogen oxides and ammonia was previously underestimated. This information would allow a trend in secondary inorganic aerosols to be determined more precisely.
  Third, models must be adapted to improve the description of anthropogenic contributions to particulate matter. This not only concerns particulate matter from sulphur dioxide, nitrogen oxides and ammonia, but also particulate matter from volatile carbonaceous compounds, mineral dust and water that is bound to anthropogenic particulate matter constituents.
  Finally, the contribution of various economic sectors in the Netherlands and abroad to the $PM_{10}$ and $PM_{2.5}$ concentrations must be recalculated. These contributions are especially relevant when particulate matter concentrations are elevated.

After this knowledge is acquired, it will become possible for the Netherlands and other countries in the region to determine an optimal strategy to reduce the large contribution to particulate matter from sulphur dioxide, nitrogen oxides and ammonia on days with elevated $PM_{10}$ concentrations.

- It would be desirable to revise the sea salt deduction that is applied during exceedances of $PM_{10}$ limit values. The contribution of sea salt to $PM_{10}$ concentrations has turned out to be lower on average than previously assumed.
- A supplementary indicator is required to monitor the effect of particulate matter policy measures that are relevant to human health, but which are not measurably expressed in the $PM_{10}$ or $PM_{2.5}$ concentrations. Elementary carbon and black smoke, both candidates for a supplementary indicator, are still topics for discussion and additional research.
- Measurements and models of emissions relating to the contribution of carbon compounds and carbonaceous particulate matter must be improved, especially for urban areas. These improvements are also essential for an adequate description and verification of the link between climate change and air quality.

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# Deepening

# 1 Introduction

The Policy Research Programme on Particulate Matter (BOP) is a national research programme on $PM_{10}$ and $PM_{2.5}$ that was financed by the Ministry of Housing, Spatial Planning and the Environment (VROM). The programme was implemented by four cooperating research institutes in the Netherlands: the Energy Research Centre of the Netherlands (ECN), the PBL Netherlands Environmental Assessment Agency, the National Institute for Public Health and the Environment (RIVM) and TNO Built Environment and Geosciences. In addition, the DCMR Environmental Protection Agency, the municipality of Breda, the Municipal Health Service of Amsterdam and Wageningen University and Research Centre contributed to the research programme by providing measurements, measurement facilities and/or research capacity.

BOP aims to reduce the number of dilemmas that occur when enforcing European standards for particulate matter. The uncertainties about particulate matter make it difficult to develop and implement suitable policy measures that the government can use to effectively alleviate the health impact of particulate matter. However, BOP did not conduct explicit research into the uncertainties about the health impact of particulate matter itself. Nevertheless, the health impact is important, and the BOP results will therefore be placed in that perspective.

The research programme, which ran from 2007 through 2009, has been completed and has provided a great deal of new information. The results have been published in a separate series of 15 reports. As the research proceeded, the results were used whenever possible to support air-quality policy at the national and European levels and to give shape to this policy in practice. For example, the explorative study on $PM_{2.5}$ (Matthijsen & ten Brink 2007) played a role in establishing the European standards for $PM_{2.5}$. In addition, the partial results were used where possible for preparing the National Air Quality Cooperation Programme (NSL) and for the future outlook studies on particulate matter (Matthijsen et al. 2009; Velders et al. 2009; PBL 2009a).

The present report reviews the most important results, summarises their relevance for policy and discusses the remaining uncertainties and possible subsequent steps. This chapter briefly discusses why and how the research aims were chosen against the background of the developments surrounding particulate matter during approximately the past 15 years.

The objective of this report is to inform policymakers and researchers, but without going exhaustively into the scientific details that emerged from the underlying research. In this regard, we limited ourselves to a number of publications that are relevant to particulate matter policy in the Netherlands. BOP was built upon this previous research. This concerned the following studies:

- The research programme on particulate matter (*Bronstof*), that took place at the end of the 1990s (Buringh & Opperhuizen 2002; Visser et al. 2001).
- *Particulate Matter: a closer look*, which established the facts on particulate matter (Buijsman et al. 2005). The PBL Netherlands Environmental Assessment Agency and the RIVM published this report on the newly imposed $PM_{10}$ limit values in response to the hectic activity in the Netherlands surrounding particulate matter.
- *New insights into the scale of the particulate matter problem in the Netherlands* (MNP 2006).

## 1.1 Developments concerning particulate matter

During the past 15 years, European legislation on air pollution and its transposition into Dutch legislation has resulted in many developments related to particulate matter. The increased attention for particulate matter in the Netherlands was primarily because the limit values for $PM_{10}$ were being exceeded on a large scale, while at the same time it appeared that Dutch policy could have only a limited effect on those exceedances. Moreover, failure to meet the deadlines for the standards had – and still has – major economic consequences. This is because spatial development projects are reviewed according to compliance with air-quality standards. Due to new insights, however, the picture of the feasibility of compliance with $PM_{10}$ and $PM_{2.5}$ limit values in the Netherlands has changed greatly, especially during the past four years. The following section summarises the developments surrounding particulate matter in the Netherlands in the areas of standards, measurements, model calculations, research and health impact.

### 1.1.1 Standards for $PM_{10}$ and $PM_{2.5}$

During the 1990s, European legislation was established to improve the air quality in Europe (Framework Directive on Air Quality: EU 1996). In 1999, to limit the potentially adverse health effects of particulate matter, limit values and target values for constituents such as $PM_{10}$ were established (1st Air Quality Daughter Directive: EU 1999a). At that time, $PM_{10}$

had been routinely measured in the Netherlands since 1992 by the RIVM in the National Air Quality Monitoring Network (LML). In 2008, standards for $PM_{2.5}$ were established with the revised Air Quality Directive in 2008 (EU 2008b). The following textbox (Summary of Standards for $PM_{10}$ and $PM_{2.5}$) lists the current standards for $PM_{10}$ and $PM_{2.5}$. It also cites the most stringent standard and the possibility of postponing compliance with air-quality standards (derogation).

### 1.1.2 Measurements of $PM_{10}$ and $PM_{2.5}$

Measurements, combined with model calculations, are used to determine whether the limit values for $PM_{10}$ and $PM_{2.5}$ are exceeded in the Netherlands. The reference methods for measuring $PM_{10}$ and $PM_{2.5}$ were established in the European air-quality directive (EU 2008b). Routine measurements of $PM_{2.5}$ in the National Air Quality Monitoring Network are conducted according to the reference method. Routine measurements of $PM_{10}$ in the National Air Quality Monitoring Network are conducted with automatic monitors, instead of using the reference method. This was permitted because it has been shown that these measurements are equivalent with the reference method (within the allowable uncertainty margin of 25 per cent). The automatic $PM_{10}$ measurements are also used to comply with the information requirements set in EU Air Quality directive (EU 2008b): the public has a right to be informed promptly about air pollution. With automatic monitors, particulate matter concentrations are measured hourly, which are then presented to the public via the Internet and teletext

Routine $PM_{10}$ measurements began in the Netherlands in 1992 at 10 measurement locations. In 2005, at the time when the limit values for $PM_{10}$ went into force, there were 39 measurement locations for $PM_{10}$. Approximately half of these

## *Summary of Standards for $PM_{10}$ and $PM_{2.5}$*

*Summary of the prevailing limit values and target values for $PM_{10}$ and $PM_{2.5}$ in the Netherlands. Limit values are results obligations, and target values are best efforts obligations. The legal implications for European Member States of failure to comply with target values are much less severe than failure to comply with limit values. In addition, failure to comply with particulate matter limit values has economic consequences in the Netherlands; this is due to the suspension of development plans that results from the link between land-use planning and air quality.*

### Which currently applicable particulate matter standard is the most stringent?

*Based on current information, the standard for 24-hour average $PM_{10}$ concentrations is still the most stringent. If the indicative limit value for yearly average $PM_{2.5}$ concentrations of 20 µg/m³ goes into force in 2020, this will possibly be more stringent.*

### Due to the National Air Quality Cooperation Programme (NSL), the Netherlands can postpone compliance with the $PM_{10}$ standards

*In July 2008, the Netherlands submitted a request for derogation to the European Commission. In April 2009, the European Commission approved this request. As a result, the deadline for compliance with limit value for $PM_{10}$ was postponed until 11 June 2011. The postponement was based on the National Air Quality Cooperation Programme (NSL).*

| Substance | Standard | Level | Status | Scope of application |
|---|---|---|---|---|
| $PM_{10}$ | Annual average | 40 µg/m³ | Limit value in force since 2005 (2011 with derogation) | all locations [a] |
| | 24-hour average, exceedance is permitted on no more than 35 days per year | 50 µg/m³ | Limit value in force since 2005 (2011 with derogation) | all locations [a] |
| $PM_{2.5}$ | Annual average | 25 µg/m³ | Limit value in force from 2015 | all locations [a] |
| | Average exposure index [b] | 20 µg/m³ | Limit value in force from 2015 | b) |
| | Average exposure index [b] | 15-20% reduction [c] | Target value in 2020 relative to 2010 | b) |
| | Annual average | 25 µg/m³ | Target value in force from 2010 | all locations [a] |
| | Annual average | 20 µg/m³ | Indicative limit value from 2020 [d] | All locations [a] |

a) The air quality directive from 2008 stipulates limit values for both $PM_{10}$ and $PM_{2.5}$. The directive also defines where the standards apply: at all locations in the public living environment, except at locations 'that are customarily inaccessible to the public'.
b) The average exposure index (AEI) is based on measurements taken at urban background locations, and is calculated as a three-year average. The AEI for 2010 in the Netherlands is determined for the years 2009-2011. The AEI for other years is determined as the average of that year and the two previous years. Therefore, the AEI for 2015 is the average for 2013-2015, and the AEI for 2020 is the average for 2018-2020.
c) The exposure reduction target (ERT) concerns possible targets for the Netherlands. In 2013, the air-quality directive will be revised. Among other things, this revision aims to convert the ERT value into a limit value.
d) At present, non-compliance with the indicative limit value has no legal consequences. As part of the revision of the air-quality directive in 2013, the indicative limit value will possibly be transposed into a target value or limit value that is obligatory for the Member States.

## *Measurements of $PM_{10}$ and $PM_{2.5}$ in the National Air Quality Monitoring Network of the Netherlands*

*In the National Air Quality Monitoring Network, $PM_{10}$ and $PM_{2.5}$ are measured automatically by determining the attenuation of beta radiation caused by particles on a filter band. Research has shown that this method systematically underestimates the measured particulate concentrations relative to the European reference measurement method. Measurement data from automatic monitors are therefore calibrated. This calibration factor can depend on the location and the monitor type.*

*Chronological overview (beginning in 1996) of European and national legislation and activities concerning the measurement of $PM_{10}$ and $PM_{2.5}$ and the reduction of measurement uncertainty.*

***1996** In 1996 the European Framework Directive on Air Quality (EU 1996) specified regulations for conducting measurements of constituents such as $PM_{10}$. In the Netherlands, $PM_{10}$ had been measured systematically since 1992. At the beginning of the 1990s, the automatic $PM_{10}$ measurements were compared with a predecessor of the most recent reference method for $PM_{10}$. This comparison showed that the automatic measurements systematically underestimated the $PM_{10}$ concentrations. To correct this underestimate, a calibration factor was implemented (see for example Putten et al. 2002).*

***1999** In 1999 the 1st Air Quality Daughter Directive (EU 1999a) was enacted, which included limit values for $PM_{10}$. This directive contained technical specifications for measuring $PM_{10}$. In 1999, the most recent reference method for measuring $PM_{10}$ (EN: 12341 1999) was also established.*

***2001** In 2001 the European Commission (EC 2001) decreed that particulate matter concentrations must be reported as measured with ambient temperature and humidity, in contrast to other air quality indicators, to which reference conditions applied. Previously, there were no European stipulations for measurement conditions for particulate matter. In the Netherlands, the new method produced measurements of yearly average $PM_{10}$ concentrations that were approximately 4 per cent higher.*

***2003** Between 2003 in 2004, the particulate matter measurement network in the Netherlands was structurally improved. Automatic $PM_{10}$ monitors were replaced by a newer type, and the measurement unit housings in the National Air Quality Monitoring Network were modified. Due to these changes in the measurement network, the equivalence with the reference method equivalent was once again determined.*

***2005** The standard reference method for measuring $PM_{2.5}$ was established (EN 14907 2005).*

***2007** RIVM, in consultation with administrators of local measurement networks in the Netherlands, made technical agreements in 2007 (NTA 8029 2008) to limit the number of degrees of freedom in the European reference method. In March 2008, reference measurements of $PM_{10}$ and $PM_{2.5}$ in the National Air Quality Monitoring Network were conducted based on these agreements. The concentrations turned out to be 1 to 2 µg/m³ lower than previously measured. This difference consisted of water vapour, which could become attached to the dust filters and was erroneously measured as particulate matter.*

***2008** In 2008, the revised European air quality directive (EU 2008b) went into force, including limit values for $PM_{2.5}$. This directive introduced technical specifications for measuring $PM_{2.5}$ and its constituents.*

*In the Netherlands, the automatic $PM_{10}$ measurements in the National Air Quality Monitoring Network were found to be equivalent to the reference method as stipulated in the European directive. This was reported to the European Commission (Beijk et al. 2007, 2008). The resulting calibration factor for $PM_{10}$ measurements was implemented in 2007 with retroactive effect for the entire $PM_{10}$ measurement series. In 2007, the measurement network consisted of 20 'old' monitors (primarily in urban areas) and 20 monitors of a newer type (primarily at regional locations). In 2007 and 2008, the 20 'old' monitors were replaced by the newer type.*

### More stringent monitoring of particulate matter and the consequences for policy

*In 2006, the adjustment of the calibration factor for regional locations led to new insights into the magnitude of the particulate matter problem (MNP 2006). At that time, as a result of the above-mentioned changes, the yearly average $PM_{10}$ concentrations were shifted downwards by 3-5 µg/m³ for all types of measurement locations. In 2006, this took place for regional locations and in 2007 for urban and street locations. The changes were implemented with retroactive effect for the entire measurement series. As a result, there was a drastic change in the picture surrounding $PM_{10}$ and exceedances of the European limit values in the Netherlands. Until about 2005, based on measurements and calculations, limit values were exceeded on a large scale in the Netherlands. After implementation of the technical improvements in the measurement network and the recalibration, it turned out that $PM_{10}$ limit values were exceeded to a much smaller extent than was previously assumed. There were still exceedances in urban areas, along motorways, in the vicinity of storage and transshipment facilities for dry bulk goods and at large agricultural operations. However, these particulate matter 'hotspots' could be remediated with general and local policy in the Netherlands, in supplementation to existing European air quality policy. Before the changes, it appeared that even draconian measures would often be inadequate to comply everywhere in the Netherlands with the $PM_{10}$ limit values. Although the changes were within the uncertainty margins of $PM_{10}$, there were major consequences for policy, because at many locations the $PM_{10}$ concentration fell from just above to just below the limit value. This is because the statistical significance of the exceedance is not taken into account when an exceedance of a limit value is ascertained.*

were at regional locations in the rural area and were used for measuring large-scale $PM_{10}$ background concentrations. The remaining measurement locations were urban and street locations.

Measurements of particulate matter concentrations in the Netherlands have an uncertainty of approximately 20 per cent (Beijk et al. 2007). This uncertainty has often turned out to be larger than the uncertainty of measurements of other air pollutants, such as sulphur dioxide. This larger uncertainty is because the measurement method is difficult to standardise, and is also due to the specific constituents of particulate matter. A large proportion of particulate matter consists of semi-volatile compounds that can evaporate or even condense during measurement. This is why the measurement uncertainty allowed by the EU directive is relatively large

### *Model calculations of $PM_{10}$ and $PM_{2.5}$*

#### Particulate matter concentration maps and uncertainty

*The $PM_{10}$ and $PM_{2.5}$ concentration maps are made by calibrating dispersion calculations of known anthropogenic emissions (from human activity) with measurements of $PM_{10}$ or $PM_{2.5}$. The methodology for making these maps is summarised in Velders et al. (2009) and is described in detail in Matthijsen & Visser (2006). A value of 40 per cent is used as the best estimate of uncertainty in the modelled large-scale concentrations of $PM_{10}$. If uncertainties caused by annual differences in weather conditions and emission projections are not taken into account, the uncertainty for projections of $PM_{10}$ is of the same magnitude. The uncertainty in the calculated contributions of the various economic sectors to the $PM_{10}$ concentration is extremely high (more than 40 per cent). For an historical year, the model uncertainty is approximately twice as high as the uncertainty in the measurements. However, due to calibration with the $PM_{10}$ measurements, the final uncertainty in the $PM_{10}$ concentration maps falls to about 20 per cent.*

#### Which anthropogenic constituents of particulate matter are modelled?

*The anthropogenic contribution to particulate matter takes place through primary emissions of particulates and through secondary particulate formation in the atmosphere from gaseous sulphur dioxide, nitrogen oxides, ammonia and from volatile organic compounds.*

*The contributions to particulate matter from all registered European sources are modelled, except the formation of particulates in the atmosphere from volatile organic compounds. The modelled contribution to $PM_{10}$ from the registered anthropogenic sources is calculated at approximately one-half (12-14 µg/m³) of the measured $PM_{10}$ concentration at regional locations. This does not include the contribution from volatile organic compound emissions to the $PM_{10}$ concentration because the model-based description of this constituent is still very uncertain, and not enough measurements are available for verification. The current contribution of anthropogenic volatile organic compound emissions to the $PM_{10}$ concentration is estimated at roughly 1 µg/m³.*

#### The 'non-modelled constituent'

*The relatively large difference between the measured and calculated particulate matter concentration, known as the 'non-modelled constituent', has been previously studied (Visser et al. 2001). Buijsman et al. (2005) and Matthijsen & Visser (2006) described the probable composition of the 'non-modelled constituent'. Besides natural constituents of particulate matter such as sea salt, the non-modelled constituent consists of systematic deviations in the measurements and model results. The total particulate matter concentration shown on the concentration maps is relatively insensitive to systematic deviations in the model results, such as disregarded or incorrectly modelled contributions from anthropogenic sources. This is because deviations are largely compensated by the calibration to the measurements. However, the systematic deviations do affect the magnitude of the calculated contributions per economic sector, and they also affect the $PM_{10}$ projections. Moreover, systematic deviations in the measurements of particulate matter have a direct effect on the concentration maps. To make particulate matter projections, the non-modelled constituent in future years must be estimated. For this purpose, the average of the non-modelled constituents from past years is used. The methodology for determining the non-modelled constituent is described in detail in Matthijsen & Visser (2006) and Velders et al. (2009).*

#### Changes and adaptations to the modelling of $PM_{10}$ and $PM_{2.5}$

*Every year, changes and adaptations are made to the models that are used to produce the large-scale $PM_{10}$ and $PM_{2.5}$ concentration maps. The changes relative to maps from the previous reports are specified in the GCN publications. These changes concern the following:*

- *For the majority of substances and sectors, the spatial distribution of the emissions is derived from the Netherlands Pollutant Release & Transfer Register (ER 2010). Updates to the Register are therefore immediately available for use on the concentration maps.*
- *Improvements are also implemented that are not derived from the Register. For example, in 2007 and 2008 the spatial distribution of $PM_{10}$ emissions from agriculture, emissions from aviation and the emissions of $PM_{10}$ from the storage and transshipment of dry bulk goods were updated.*
- *For the projections, the emission data were modified relative to the insights into the effectiveness of current policy and the accuracy of previous projections.*
- *Modifications were made to input data such as the emission characteristics of sources and model processes (e.g. the description of ammonia deposition).*
- *The model resolution was refined. The current calculation resolution is 1x1 km²; until 2007, this was 5x5 km².*

*The effects of these annual adaptations and changes are generally limited to less than 1 µg/m³. Larger changes in concentration were the result of modifications of the calculation resolution and changes in the source strength of large sources and/or sources close to the ground. The higher resolution has led to many changes at the local level – both higher and lower calculated concentrations.*

(25 per cent). This measurement uncertainty was taken into account in the trend analysis of $PM_{10}$ (see Chapter 5). The following textbox (Measurements of $PM_{10}$ and $PM_{2.5}$ in the National Air Quality Monitoring Network) lists when the most important European legislation was enacted that affected the measurement of particulate matter, and when new methods were implemented in the National Air Quality Monitoring Network to measure $PM_{10}$ concentrations more accurately. In addition, the policy consequences of these technical refinements in the monitoring network are discussed.

### 1.1.3 Model calculations for $PM_{10}$ and $PM_{2.5}$ and uncertainty

Model calculations of particulate matter have played an important role in assessing air quality regarding compliance with the limit values for $PM_{10}$ and $PM_{2.5}$. Since 2002, the PBL Netherlands Environmental Assessment Agency, in cooperation with RIVM, has published annual maps of large-scale concentrations of various air pollutants in the Netherlands (GCN maps). These maps are used for reporting exceedances of the EU air-quality directive, for defining general national and local policy and for reviewing land-use plans (see for example Velders et al. 2009).

The calculated $PM_{10}$ concentrations are calibrated to the measurements of particulate matter. The uncertainty in the concentration maps for $PM_{10}$ is therefore related to the measurement uncertainty. The following text box, Model calculations of $PM_{10}$ and $PM_{2.5}$, summarises the most important uncertainties in the concentration maps for $PM_{10}$ and $PM_{2.5}$, and lists the modifications and changes made to the modelling instruments.

### 1.1.4 Particulate matter and health impacts

Elevated atmospheric concentrations of particulate matter are associated with adverse health effects and are the most important environment-related disease burden in the Netherlands and Europe (Knol & Staatsen 2005; Holland et al. 2005). Particulate matter is a mixture of many chemical constituents originating from a wide range of anthropogenic and natural sources[1)]. Epidemiological studies have shown that both $PM_{10}$ and $PM_{2.5}$ concentration levels are related to adverse health effects (Pope & Dockery 2006). However, much less data is available for $PM_{2.5}$ than for $PM_{10}$. Studies have also shown that the correlations with specific constituents of particulate matter are stronger than for particulate matter as a whole (Laden et al. 2000; Grahame & Schlesinger 2010). It is still unknown exactly which substances or sources are responsible for the observed health impacts. However, there is more and more proof for the hypothesis that combustion aerosol – particulate matter that is released during combustion processes – has a greater health impact than particulate matter consisting of non-organic salts such as sea salt and the secondary particulates that are formed from gaseous sulphur dioxide, nitrogen oxides and ammonia (see for example Fischer et al. 2000). Other constituents, such as mineral dust, are also associated with adverse health affects (Brunekreef & Forsberg 2005; Perez et al. 2008). Water-soluble metallic compounds are generally viewed as being a potential health hazard. However, the adverse health effects appear to differ significantly for each element. The chemical form of the metal also plays a role. Transition metals such as copper are thought to be especially hazardous in ionic form (Costa & Dreher 1997).

In addition, a wide range of adverse health effects is associated with particulate matter and its constituents, where each constituent appears to have a specific relationship with a specific health effect (Gerlof-Nijland et al. 2009; Steerenberg et al. 2006).

Adverse health effects that can occur during short-term exposure to particulate matter are better understood than those that occur with chronic exposure. The adverse health effects of chronic exposure to particulate matter – on a time scale of many years – are difficult to determine and have a high degree of uncertainty. If American studies are declared valid for Europe, then the estimation is that long-term exposure to particulate matter leads to premature mortality of one year, relative to lifelong absence of exposure to particulate matter.

#### *Combustion aerosol*

*Combustion aerosol is a suspension of particulates that are released during combustion processes. Traffic, wood combustion and energy generation from fossil fuels are typical sources of combustion aerosol. The sources of combustion aerosol in the Netherlands are almost entirely anthropogenic. The composition of combustion aerosol is related to the source and can differ between sources. Generally speaking, combustion aerosol consists largely of carbonaceous particulate matter, with a small component of heavy metals such as nickel and vanadium. Nickel and vanadium are primarily released during combustion of oil (in the petrochemical industry and shipping). Polycyclic aromatic hydrocarbons (PAHs) are another constituent of combustion aerosol. The adverse health effects of this group of substances are relatively well known. For purposes of source recognition, measurement techniques are used to split the concentration of carbonaceous particulate matter into elementary carbon (EC) and organic carbon (OC) constituents. The proportion of EC and OC varies according to the source. Soot is an important constituent of combustion aerosol; it consists of a mixture of elementary and organic carbon. To monitor combustion aerosol, besides EC and OC, constituents such as black smoke and black carbon are also measured. The concentration of black smoke and black carbon is measured using simple optical methods. However, these measured parameters are not defined with sufficient precision to allow them be compared directly with the measured or calculated concentration contribution from traffic. The measurements of EC and OC themselves are sufficiently well defined, but the measurement methods are still unwieldy and the measured concentrations have a high level of uncertainty. European standards are currently being prepared for the measurement methods for EC and OC. Keuken & ten Brink (2010) summarised the various indicators for constituents of combustion aerosol that are assumed to be relevant to health.*

The acute effects caused by exposure to particulate matter take place during a timespan of one to several days. Public health studies that analyse these types of effects show that several thousand people in the Netherlands have a shorter estimated lifespan due to acute exposure. This lifespan reduction is probably limited to several days to several months. Particulate matter adversely affects cardiac and lung function, among other functions. For example, between one and two per cent of emergency hospital admissions for cardiovascular disorders in the Netherlands can be attributed to particulate matter exposure. Particulate matter can also aggravate allergies. Such results have not only been found in the Netherlands, but everywhere in the world, and are relatively robust. In addition, increased exposure to particulate matter also appears to be linked to sub-optimal lung development in children.

No safe limit value can be derived from population screening. Consequently, even though the air quality improves with every reduction in particulate matter concentration, no major public health benefits are expected from a small decline in the particulate matter concentration from just above to just below a limit value. Therefore, health policy should aim for even cleaner air. This approach is supported by the World Health Organization (WHO), which recommends a long-term target value for yearly average $PM_{2.5}$ concentrations of 10 µg/m³ (WHO 2005).

The knowledge about the possible causes of the observed adverse health effects of particulate matter continues to improve. According to current insights, the concentrations of $PM_{10}$ or $PM_{2.5}$ are not the best indicators for the observed health effects. Other indicators could be more relevant for public health, such as ultra-fine particulate matter (smaller than one-tenth of a micron), the total number of particles and specific components of particulate matter, such as soot. In addition, other air quality factors are important, such as the history of exposure and the convergence of effects that occur when someone is exposed simultaneously to ozone and particulate matter. The picture of the health-relevant components of particulate matter is becoming increasingly clear. However, this continuous process requires flexibility from policymakers and public administrators, which is often incompatible or difficult to reconcile with existing regulations on particulate matter. This is because the health effects are also affected by the specific approach that is used to comply with existing standards. The standards for $PM_{10}$ and $PM_{2.5}$ rely implicitly on the principle that all components of particulate matter are equally hazardous. Current and proposed EU policy is still based on $PM_{10}$ and $PM_{2.5}$ as indicators for the associated adverse health effects. To improve the monitoring of the health-relevant fraction of particulate matter, additional indicators are in development.

## 1.2 Research aims and mode of operation

The Policy Research Programme on Particulate Matter (BOP) aimed to improve the knowledge about particulate matter to support policymaking in the future more adequately. The most important research aims of the programme were the following:

- Improving the knowledge about $PM_{10}$ and $PM_{2.5}$ concentrations and about the composition and sources of particulate matter.
- Improving the understanding of the behaviour of particulate matter in the urban area.
- Determining the trends in particulate matter concentrations and in the individual components of particulate matter.
- Clarifying the effects of past and future policy measures on $PM_{10}$ and $PM_{2.5}$ concentrations.

The research primarily involved situations where limit values were exceeded. Exceedances of the European limit values for $PM_{10}$ still occur in the urban area or near motorways and locations with high local emissions. The composition of particulate matter and the contributions of various anthropogenic and natural sources are still insufficiently known, especially in situations where limit values are exceeded. The knowledge about the spatial differences in particulate matter concentrations is also limited. This concerns not only the spatial variability of particulate matter in the urban area, but also the differences between urban and rural areas.

The BOP results have been published in a special series (ISSN 1875-2314) of 15 reports. Some of these reports concerned the composition and sources of particulate matter, with summaries and specific attention to the following constituents: sea salt, secondary inorganic aerosol, elementary carbon (EC) and organic carbon (OC). Other BOP reports addressed more specific topics, such as particulate matter concentrations in the urban area, trends in particulate matter concentrations, particulate matter emissions from shipping, EC and OC emissions from traffic, particulate matter emissions from wood combustion and the feasibility of new norms for $PM_{2.5}$. Technical details about the research programme were addressed in two background documents: one about measurements and one about model developments. Included at the end of this report is a list of the reports and other publications and workshops that resulted from the BOP research.

The following chapters correlate the most important results from these reports and indicate their policy relevance. As a result, the present report provides a revision and/or confirmation of the knowledge on particulate matter with respect to previous reports such as *Particulate Matter: a closer look*.

Chapter 2 addresses the knowledge about $PM_{10}$ and $PM_{2.5}$ concentrations and about the composition and sources of particulate matter. Chapter 3 discusses the behaviour of particulate matter in the urban area. Chapter 4 considers the policy implications. Chapter 5 integrates the results of the BOP studies about the trends in particulate matter and its components. It illustrates the effects of past and future policy on the $PM_{10}$ and $PM_{2.5}$ concentrations. Chapter 6 discusses the unanswered questions and the new questions that have emerged. It also formulates the subsequent steps, including an indication of their priority for the implementation of particulate matter policy.

**Note**

1) The distinction between natural and anthropogenic origin certainly does not mean that natural particulate matter is free from adverse health effects.

# 2 Composition and sources of particulate matter

- In the Netherlands, roughly three-fourths of $PM_{10}$ and $PM_{2.5}$ in 2007-2008 consisted of sulphur, nitrogen and carbon compounds. The remainder consisted of sea salt, metals, mineral dust and a 'non-specified' constituent, which was probably water.
- Measurements showed that the contribution to the particulate matter concentration of atmospheric transformation products of sulphur dioxide, nitrogen oxides and ammonia was 50% greater than had been previously measured and calculated.
- Elevated $PM_{10}$ concentrations (above 30 µg/m³) turned out to be largely the result of an increase in the particulate matter contribution from nitrogen oxides and ammonia. During these elevated $PM_{10}$ concentrations, there was also a striking increase in the relative share of components that could not be specified chemically with the measurements. In these situations, the proportion of sea salt was three times smaller on average than was the case with low $PM_{10}$ concentrations.
- On average, the absolute contribution of the various particulate matter constituents did not increase significantly from the regional background locations to the urban locations. However, there was a clear increase in individual constituents from the urban locations to the street locations. As a natural constituent, sea salt was unrelated to the type of measurement location.

## 2.1 Data base

The composition of $PM_{10}$ and $PM_{2.5}$ was mapped out in the Netherlands by conducting measurements at six locations for a period of approximately one year, from August 2007 through August 2008. These locations were: Schiedam, Rotterdam and Breda ($PM_{10}$ only), Cabauw, Vredepeel and Hellendoorn (see Figure 2.1). In addition, specific sources were studied in detail with focused, shorter measurement campaigns and model studies in order to increase the knowledge about particulate matter sources, processes and trends. These measurement campaigns and studies concerned the emission or contribution of a number of anthropogenic, combustion-related sources: wood combustion, traffic and shipping. Technical details concerning the measurement data have been worked out in a separate report (van Arkel et al., in preparation). The uncertainties in the concentrations of the various particulate matter constituents, which are addressed in the appendix, are taken into account in the conclusions.

The PM composition was also studied with models. For this purpose, models were further developed and tested in order to study the relationship between concentration and the various sources. Technical details about the model development were discussed in Schaap et al. (2009).

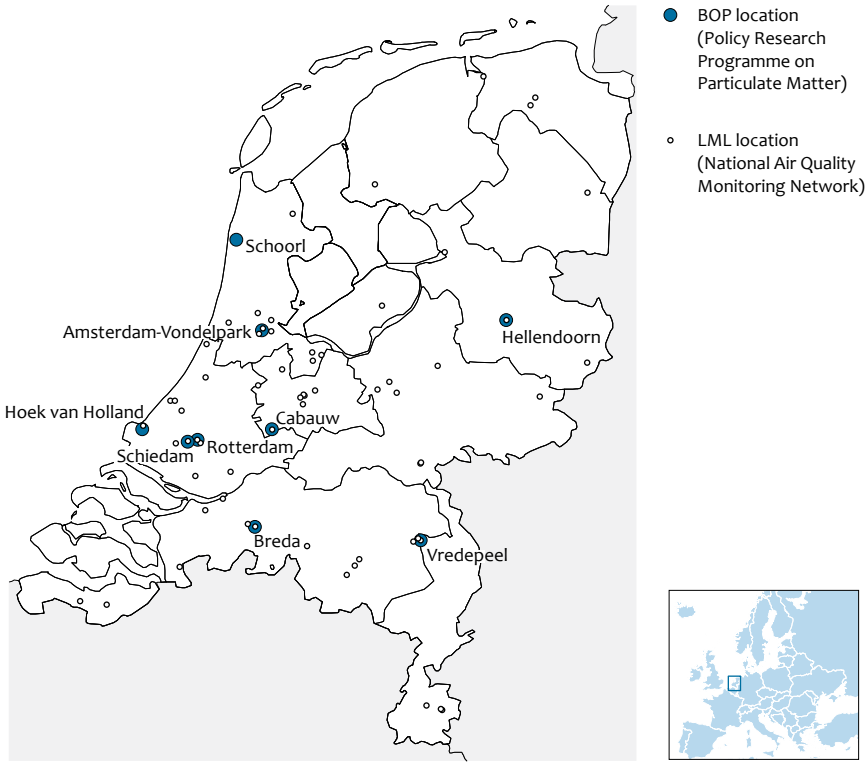
### 2.1.1 Measurements

#### What was measured?

The contribution from most sources of particulate matter could be determined based on 24-hour average measurements of the following constituents of $PM_{10}$ and $PM_{2.5}$: sulphate, nitrate, ammonium, elementary (EC) and organic carbon compounds (OC), silicon and 30 other metals.

Besides 24-hour average measurements, hourly average measurements were also conducted for sulphate, nitrate and ammonium, as well as sodium and chloride, using measurement equipment with a high temporal resolution that is specially suited for such composition measurements (MARGA, monitoring aerosol and gases; ECN 2006). This equipment was also used for researching the contribution of elementary and organic carbon from road traffic; for this purpose it was specially adapted to measure carbon. The measurement setup used in this research was discussed in Keuken & ten Brink (2009).

**Measurement locations, BOP and relevant LML 2007 – 2008** **Figure 2.1**




*BOP measurement locations and overview of standard measurement locations from the National Air Quality Monitoring Network (RIVM) and the local measurement networks of Rotterdam (DCMR) and Amsterdam (GGD Amsterdam).*

The temporal and spatial differences in the urban particulate matter concentrations were mapped out with measurements at 11 fixed locations and supplementary mobile measurements. The measurement locations in the Rijnmond area that were established for this research have been specified in Voogt et al. (2009). The main conclusions of the latter study are addressed in Chapter 3.

To estimate the contribution of wood combustion to particulate matter, the substance levoglucosan was measured. This substance is used as an indicator for the combustion of plant fibres and wood (see Chapter 3).

### When and where were measurements conducted?

The 24-hour average concentrations of $PM_{10}$ and $PM_{2.5}$ were measured during the period August 2007 through August 2008 at locations that were chosen for their differing particulate matter loads (see Figure 2.1). Three regional locations – Hellendoorn, Cabauw and Vredepeel – provided a picture of the regional background concentrations. The three regional locations were dispersed in such a way that spatial differences in the regional background concentrations in the Netherlands could be measured. Measurements at an urban location in Schiedam were used to map out urban background concentrations for the urban area near Rotterdam. The street locations in Rotterdam and Breda were chosen to measure particulate matter concentrations with a relatively high contribution from traffic. For approximately one-fourth of the measurements (once every four days on average), the composition of the $PM_{10}$ and $PM_{2.5}$ samples was determined for the six locations. To study specific episodes, additional samples were analysed.

The results in Section 2.2 are based on constituent measurements from five of the six measurement locations. The measurements taken at the street location were too limited for that analysis, because only $PM_{10}$ and its constituents were measured, and not $PM_{2.5}$. In Schaap et al (2010), the results for $PM_{10}$ from Breda were compared with the results from the street location in Rotterdam.

For hourly average measurements, three MARGA instruments were used during the period 2007-2008 at the locations in Cabauw (August-July), Schiedam (August-February) and Hoek van Holland (September-October); the latter is located directly on the coast. These rapid series measurements provided additional input about the level and spatial-temporal variability of the contributions to particulate matter from sulphate, nitrate, ammonium and sea salt.

The indicator for wood combustion, levoglucosan, was measured at two locations: Vondelpark in Amsterdam and Schoorl in North Holland. The concentrations measured in Amsterdam are indicative of an average contribution from wood combustion in an urban area. The concentrations in Schoorl are indicative of a situation with relatively high levels

of particulate matter from wood combustion. Schoorl is located in an area where firewood is plentiful and a relatively high proportion of residents burn wood. The measurements in Amsterdam ran from January through June 2008, so that samples were taken during both a cold season and a warm season. In Schoorl, measurements were conducted during the month of February 2009.

### Measurements from the National Air Quality Monitoring Network (LML)

The BOP research used the standard measurement data from the National Air Quality Monitoring Network (RIVM 2010). The most relevant data originated from the routine measurements of the fractions $PM_{10}$ and $PM_{2.5}$ and the constituents black smoke, heavy metals, sulphate, nitrate, ammonium, sodium and chloride. The locations where these measurements were conducted are shown in Figure 2.1. In addition, routine measurements from the LML of particulate matter precursors, such as sulphur dioxide, nitrogen oxides and ammonia, were relevant.

## 2.1.2 Models

For particulate matter policy, it is essential to quantify the relationship between emissions, atmospheric conditions and the concentration of air pollutants. For this purpose, a number of models are used in the Netherlands. The BOP

### *Models in BOP*

*The following models played a role in the Policy Research Programme on Particulate Matter (BOP). The levels of the particulate matter concentrations and constituents that were calculated with models were often very uncertain (see textbox Model calculations $PM_{10}$ and $PM_{2.5}$ in Section 1.1). The uncertainties for the BOP model studies are described in Schaap et al. (2009).*

#### LOTOS-EUROS model.

*The LOTOS-EUROS model (Schaap et al. 2008) is a chemical transport model for the entire European domain. In the Netherlands, the LOTOS-EUROS model is used for research and policy support (for example, see Denier van der Gon & Schaap 2009). In BOP, the LOTOS-EUROS model is the central research model. It has been validated with measurements and has been further developed by the addition of improved and new routines for calculating the contributions to particulate matter from sea salt, mineral dust and biogenic secondary organic aerosol. In addition, a link was created with the global chemical transport model TM5. This link allows the effects of global emission changes on European air quality to be calculated. The results from the LOTOS-EUROS model were compared with those from the OPS model and the EMEP model for primary $PM_{2.5}$ and the secondary particulate matter from sulphur dioxide, nitrogen oxides and ammonia (Schaap et al. 2009). The model-based description of the contribution of sea salt to particulate matter in LOTOS-EUROS was compared with the same description from the OPS model (Manders et al. 2009).*

#### OPS model.

*The PBL Netherlands Environmental Assessment Agency, in cooperation with RIVM, uses this model (Van Jaarsveld 2004) to make annual, large-scale concentration maps for various air pollutants that are subject to European air-quality norms. These maps are used for reporting exceedances of the EU air-quality directive, for defining general national and local policy and for reviewing land-use plans (see for example Velders et al. 2009). The results of the OPS model were therefore used as a point of departure for evaluating the feasibility of norms for $PM_{10}$ and $PM_{2.5}$ (Matthijsen et al. 2009) and for interpreting particulate matter trends between 1993 and 2007 (Hoogerbrugge et al. 2010). The OPS model provides a higher resolution (1x1 km²) than the EMEP and LOTOS-EUROS models. The results of the OPS model are limited to the air quality in the Netherlands.*

#### TM5 model.

*The TM5 model (Krol et al. 2005) is chemical transport model and covers the entire world and the entire troposphere (the lowermost 16 km of the atmosphere). The model has a resolution of 6x4 degrees of longitude and latitude and can locally zoom into a resolution of 1x1 degrees. The TM5 model is used for atmospheric-chemical research in relation to aspects such as climate change. Recent applications of the TM5 model include the multi-model studies: a global study on nitrogen oxides (Sanderson et al. 2008), and a global study on ozone at ground level (Fiore et al. 2009). In BOP, global particulate matter concentration fields calculated with the TM5 model have been made suitable for use as boundary conditions for the LOTOS-EUROS model.*

#### EMEP model.

*The 'unified EMEP model' (EMEP 2003) is a chemical transport model for the European domain. The results of the EMEP model are used as input for the 'GAINS integrated assessment model' for European policy development on air quality and climate. For each EU Member State, annual air-quality summaries are made based on EMEP model calculations for all of Europe (EMEP 2009a, 2009b, 2009c). As a result, the EMEP model plays an important role in policy preparation for the European Union. $PM_{10}$ and $PM_{2.5}$ are part of the model results at a resolution of 50x50 km². The comparison with the LOTOS-EUROS and OPS models showed large differences between the Netherlands and Germany regarding contributions to particulate matter from nitrogen oxides and ammonia.*

#### RAINS/GAINS models.

*The RAINS/GAINS models (RAINS/GAINS 2007) are integrated assessment models that are used to calculate air quality and climate forcing (GAINS) for all of Europe, including the costs of policy measures (Wagner et al. 2006, 2007). The models generate integrated evaluations of emissions across the entire chain, from source to affect and the reverse, and can be used for scenarios. A RAINS/GAINS version was also developed specifically for the Netherlands (Aben et al. 2005). The results of this model, GAINS-NL, were used to investigate the feasibility of the $PM_{2.5}$ standards (Matthijsen et al. 2009).*

**Relative and absolute contributions of the particulate matter constituents of $PM_{10}$ and $PM_{2.5}$** Table 2.1

| | $PM_{10}$ | | $PM_{2.5}$ | |
|---|---|---|---|---|
| | µg/m³ | % | µg/m³ | % |
| *Secondary Inorganic Aerosol (SIA)* | 8.4 | 38% | 6.7 | 45% |
| *Total Carbonaceous Matter (TCM)* | 5.5 | 25% | 4.3 | 29% |
| *Sea salt* | 2.7 | 12% | 0.8 | 5% |
| *Mineral dust* | 1.6 | 7% | 0.6 | 4% |
| *Metals* | 1.4 | 6% | 0.6 | 4% |
| *Not specified* | 2.6 | 12% | 1.7 | 12% |
| *Total* | 22.1 | 100% | 14.7 | 100% |

programme used the LOTOS-EUROS model, as well as the OPS model. LOTOS-EUROS calculates air quality at the European scale, while OPS focuses on the Netherlands. The model development concentrated on the description of the contribution of natural sources to particulate matter, because the knowledge on this topic was still limited. Schaap et al. (2009) provided an overview of the model development and described the model results in the perspective of measurements and other model results. BOP also used three other models: EMEP, GAINS-NL and TM5 (see textbox Models in BOP). Model results have played an important role in the interpretation of the measurement data and the analyses of the $PM_{10}$ trend and projections of $PM_{2.5}$ (see Chapter 5). For various reasons, model calculations of particulate matter concentrations are difficult to verify with measurement data. Schaap et al. (2009) also described the uncertainty in the model calculations of the modules developed in BOP.

## 2.2 Composition and sources

In BOP, measurements of the chemical constituents of $PM_{10}$ and $PM_{2.5}$ in the Netherlands have led in part to revision of the particulate matter composition, but also to confirmation of previous findings, as documented in Buijsman et al. (2005) and Visser et al. (2001).

On average, approximately 90 per cent of the mass of particulate matter can be explained based on the measured constituents. These constituents are: secondary inorganic aerosol (SIA), total carbonaceous matter (TCM), mineral dust, metals and sea salt. The remainder, approximately 10 per cent, cannot be allocated according to the measurements to the above constituents, and is therefore referred to here as 'not specified'. In this chapter, the following questions will be answered:

- What is the composition of particulate matter?
- What is the composition at high (elevated) and low particulate matter concentrations?
- How does the composition of particulate matter differ according to the measurement location (regional, urban and street)?

### 2.2.1 What are the constituents of $PM_{10}$ and $PM_{2.5}$?

The constituents of $PM_{10}$ and $PM_{2.5}$ appear to be very similar. The differences are shown in Table 2.1. The proportions of secondary inorganic aerosol and total carbonaceous particulate matter are dominant. Together, they contribute to approximately two-thirds of $PM_{10}$ and approximately 80 per cent of $PM_{2.5}$. Of these two constituents, secondary inorganic aerosol (SIA) is the largest contributor to both $PM_{10}$ (38 per cent) and $PM_{2.5}$ (44 per cent). In the BOP measurements, total carbonaceous matter (TCM) contributed 25 per cent to $PM_{10}$ and 29 per cent to $PM_{2.5}$. Compared to $PM_{2.5}$, $PM_{10}$ contained relatively more mineral dust (7 per cent vs. 4 per cent), metals (6 per cent vs. 4 per cent) and sea salt (12 per cent vs. 5 per cent).

As expected, the coarse fraction of particulate matter – particles with a diameter of 2.5 to 10 µm – turned out to be rich in particles that enter the atmosphere due to mechanical processes. Nevertheless, approximately one-third of the concentration of mineral dust, metals and sea salt turned out to be present in the fine fraction ($PM_{2.5}$). The contributions varied between the measurement locations and between days with high and low particulate matter concentrations. The differences were due to the proximity to the sources of particulate matter and the effects of the weather, i.e. the wind direction in relation to particulate matter source areas and the weather conditions that played a role in the dilution, formation and removal of particulate matter. The results are explained in more detail below, particularly to indicate the contribution of various sources of particulate matter.

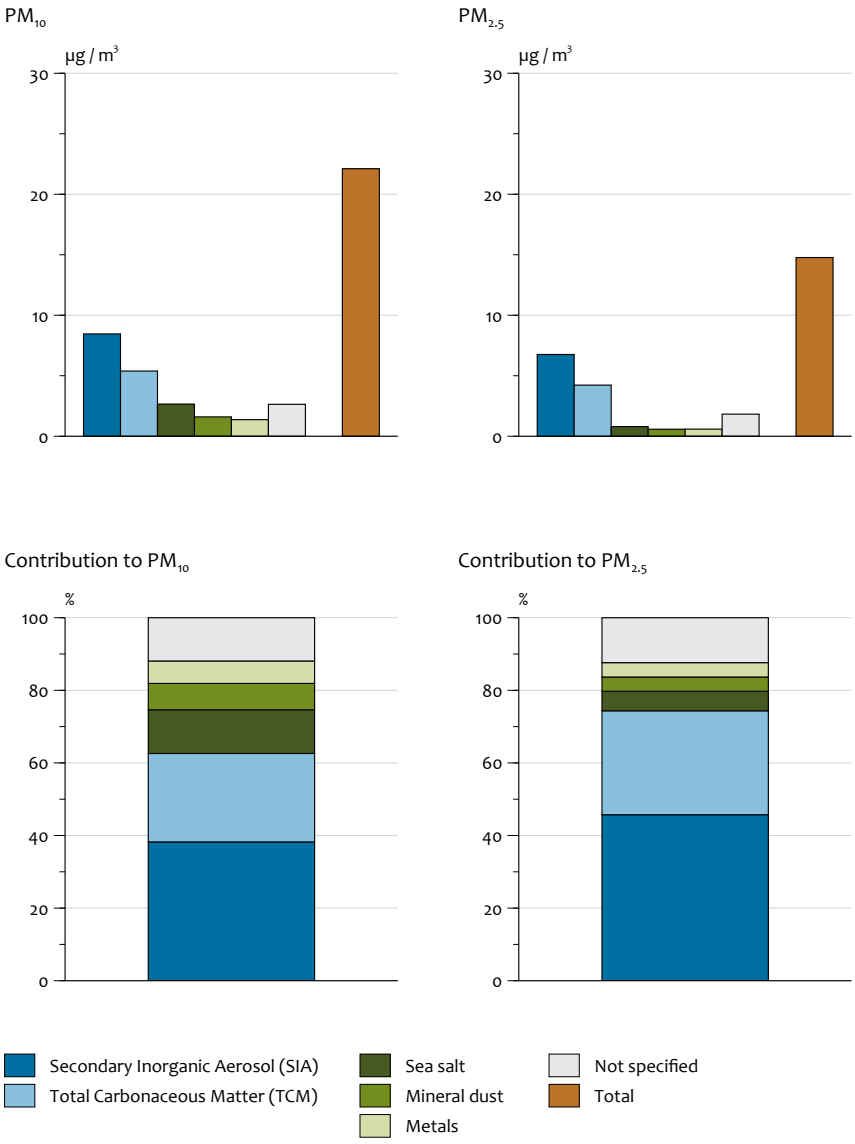
The concentration contributions and relative proportions are averages for 5 BOP measurement locations: the regional locations Hellendoorn, Cabauw and Vredepeel, the urban location Schiedam and the street location Rotterdam.

### 2.2.2 Secondary inorganic aerosol (SIA)

#### Sources

Secondary inorganic aerosol consists primarily of ammonium sulphate and ammonium nitrate. These secondary components are almost entirely of anthropogenic origin (caused by human activities). According to estimates, between 1 and 6 per cent of these components are of natural origin, and on average approximately 10 per cent originate outside Europe. Ammonium sulphate and ammonium nitrate are formed in the atmosphere from gaseous sulphur dioxide, nitrogen oxides and ammonia. The most important sources of sulphur dioxide are industry, energy generation, refineries and ocean shipping. The most important sources of nitrogen oxides are combustion processes in traffic, industry, the energy sector, refineries and ocean shipping. A small percentage is of natural origin and is released from bacterial processes, lightning and natural fires, such as forest

**Composition of particulate matter concentration, 2007 – 2008** **Figure 2.2**




*Average composition of $PM_{10}$ and $PM_{2.5}$ (in µg/m³ and per cent) 2007-2008.*

fires. Agriculture is by far the largest source of ammonia (approximately 90 per cent of the emissions).

### Contributions

The contribution to particulate matter from the precursor gases sulphur dioxide, nitrogen oxides and ammonia was approximately 8.4 µg/m³ on average for $PM_{10}$ and 6.7 µg/m³ for $PM_{2.5}$. For both particulate matter fractions, the SIA concentration consisted of 50 per cent nitrate, 20 per cent ammonia and 30 per cent sulphate. The $PM_{2.5}$ fraction contained almost all of the ammonia (>95 per cent), 90 per cent of the sulphate and about 80 per cent of the nitrate. As a result, the coarse component of SIA was about 1.7 µg/m³ on average.

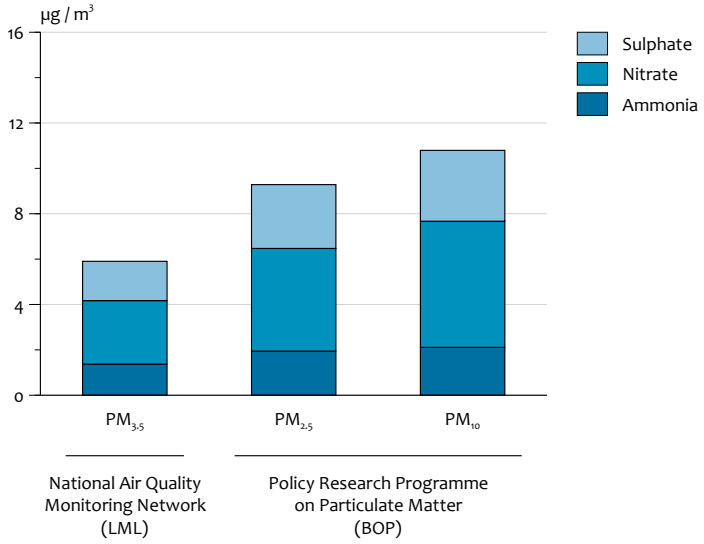
On days with elevated $PM_{10}$ concentrations (above 30 µg/m³), the relative contribution of SIA appeared to be especially high (see Section 2.3).

Between 1993 and 2007, the particulate matter concentrations fell by at least 6 µg/m³ as a result of the declining SIA contribution to $PM_{10}$ (Hoogerbrugge et al. 2010). This was shown by historical SIA measurements from the National Air Quality Monitoring Network. SIA concentrations declined sharply in the 1990s due to emission reductions, especially reductions of sulphur dioxide. However, beginning in 2001 the decline in concentrations flattened out (see Section 5.1). This pattern was largely consistent with changes in the European emissions of precursor gases (Weijers et al. 2010).

The SIA trend, as it has been measured and modelled until now, is possibly different than assumed. This is because the BOP measurements have shown that the SIA concentrations in the Netherlands are higher than those according to the existing SIA measurement series from the National Air Quality Monitoring Network. This observation also has consequences for the OPS model calculations of SIA concentrations for the

**Concentration of Secondary Inorganic Aerosol (SIA), 2007 – 2008** **Figure 2.3**

Measurement location Vredepeel




*SIA concentrations synchronously measured at the LML and BOP measurement location in Vredepeel in 2007-2008 according to the old (LML) and new (BOP) method for determining the contribution of SIA on particles (Weijers et al 2010).*

large-scale concentration maps in the Netherlands (GCN). This is because the calculations have been verified until now based on the existing SIA measurement series, with measurements taken since 1993. The models that are used to calculate the effects of measures to control SIA may not describe all complex formation processes with sufficient precision.

### SIA concentrations in the Netherlands are 50 per cent higher than previously thought

A comparison with the historical SIA measurements from the National Air Quality Monitoring Network showed that the SIA measurements from BOP were approximately 50 per cent higher than the periodic LML measurements that began in 1993. It is very probable that the higher SIA concentrations from BOP provide a better picture of the SIA contribution to $PM_{10}$ than the SIA concentrations from the LML. This conclusion is also supported by SIA levels in Belgium and Germany that are comparable with those from BOP (Weijers et al. 2010). The measurement precision of the SIA measurements in BOP is greater than the corresponding LML measurements. Moreover, the size of the particles for which the SIA concentration was determined in BOP ($PM_{10}$ and $PM_{2.5}$) was determined more precisely than those in the SIA measurements from the LML. The latter measured only particles of approximately 3.5 µm in diameter, while BOP measured two fractions: 2.5 and 10 µm.

The LML has measured SIA in $PM_{10}$ since 2008, sampled according to the reference method, an approach which is consistent with the SIA measurements in the $PM_{10}$ fraction in BOP. The LML has used the new approach to measure SIA since 2009. A comparison between the old and new SIA measurements from the LML confirmed that the SIA concentration from the BOP research was approximately 50 per cent higher (Hafkenscheid et al., report in preparation). Previously, Putten & Mennen (1998) showed that the SIA concentrations were rather uncertain due to the older measurement method and possibly underestimated the atmospheric SIA concentrations.

The finding that the SIA contribution was 50 per cent higher than previously assumed means that the implemented policy for the emissions of sulphur dioxide, nitrogen oxides and ammonia possibly had a greater share in the decline of the $PM_{10}$ concentrations than was previously calculated (see the $PM_{10}$ trend analysis in Chapter 5).

Moreover, planned measures focusing on the SIA precursor gases could have a greater effect than was initially calculated with current projections. The maximum effect between 2010 and 2020 is an additional concentration decline in the Netherlands of approximately 0.1 µg/m³ per year.

## 2.2.3 Total carbonaceous matter (TCM)

### Sources

The concentration contribution of carbonaceous particulate matter is measured as elementary carbon (EC) and organic carbon (OC), both expressed in µg carbon/m³. For total carbonaceous matter (TCM), an additional 30% is added; besides the carbon in EC and OC, the other elements in carbonaceous particulate matter, such as oxygen and hydrogen, are included (ten Brink et al. 2009). The measurement distinction between EC and OC is done for reasons of source recognition. EC appears to be a good indicator for the contribution from combustion engines, especially diesel engines. All EC and a portion of OC are emitted directly to the atmosphere. The other part of OC, the secondary organic aerosol, is formed in the atmosphere from volatile organic compounds (VOCs).

The particulate carbon compounds can be of natural or anthropogenic origin. In the Netherlands, however, elementary carbon is almost entirely of anthropogenic origin and is released in particulate form during combustion processes. Two-thirds of the anthropogenic VOC emissions are from solvents or are released during production processes and during the use of solid and liquid fuel. Volatile organic compounds are also emitted naturally by trees and plants. In addition, organic dust particles (usually larger in diameter) are emitted from agricultural activities in barns and outdoors. Such particles can also be of natural origin.

The current knowledge about the concentration and origin of TCM is limited. Recent measurements from two locations showed that approximately one-fourth to one-third of the organic carbon in the Netherlands originates from fossil fuels (ten Brink et al. 2010). According to these measurements, at least two-thirds of the organic carbon originated from carbonaceous particulate matter, which was produced much more recently (non-fossil origin). This constituent can be both natural and anthropogenic in origin. According to estimates, 25 to 50 per cent of the organic carbon in the Netherlands is of natural origin. Based on current knowledge, the magnitude of the anthropogenic contribution to TCM in the Netherlands therefore remains uncertain.

#### Contributions

The average contribution of carbonaceous particulate matter to $PM_{10}$ was about 5.5 µg/m³ and to $PM_{2.5}$ about 4.3 µg/m³. For $PM_{10}$ the TCM varied from 4.8 µg/m³ at the regional locations to 7.7 µg/m³ at the street location in Rotterdam. The $PM_{2.5}$ fraction contained approximately 80 per cent of the TCM and about 90 per cent of the elementary carbon. The TCM contribution is partly natural and partly anthropogenic in origin.

With elevated particulate matter concentrations (above 30 µg/m³), the contribution of TCM increased, especially in urban areas and near streets. In relative terms, the contribution to the increased particulate matter concentration fell slightly. The spatial variation in TCM was greater than that in SIA, due to the contribution of directly emitted particles with carbon. At elevated particulate matter concentrations, the differences in TCM concentration between measurement locations increased sharply.

There were no measurement data from which a trend in the TCM contribution to particulate matter could be derived directly. However, there were indications that concentrations of elementary carbon have declined sharply during the past two decades. The exhaust emissions from traffic and the concentrations of black smoke, an indicator for soot and elementary carbon, have declined by about 50 per cent since 1990. In the Netherlands and surrounding countries, the anthropogenic emissions of VOCs have declined by nearly 60 per cent since 1990. The anthropogenic component of TCM has therefore probably declined since 1990 as well.

### 2.2.4 Sea salt

#### Sources

Sea salt aerosol has a natural origin and is created in the atmosphere when wind blows across the ocean surface. Fresh sea salt consists primarily – about 85 per cent – of sodium chloride (table salt), with smaller amounts of sulphate, magnesium, calcium and potassium compounds. When sea salt particles remain in the air, they undergo chemical reactions with other components. As a result, the ratio between sodium and chloride changes.

#### Contributions

The average sea salt concentration in $PM_{10}$ during the measurement period ranged from 4 µg/m³ in Rotterdam, located near the coast, to 2 µg/m³ in Vredepeel, located inland. With a relative proportion of approximately 12 per cent, sea salt is the third most prevalent component in $PM_{10}$ after SIA and TCM. Based on the 24-hour average, the contribution was sometimes much higher or even lower. On days with elevated particulate matter concentrations (above 30 µg/m³) the relative contribution from sea salt was three times lower on average compared to the yearly average. Approximately one-third of the sea salt was contained in the $PM_{2.5}$ fraction.

Model calculations confirmed the previous spatial picture for the Netherlands and Europe, where sea salt concentrations decline in proportion to the distance from the coast. However, elevated sea salt concentrations were overestimated, sometimes by more than 100 per cent. This means that models by themselves are still insufficiently reliable for determining the 'sea salt deduction' (see Section 4.3 for more information about the sea salt deduction).

### 2.2.5 Mineral dust

#### Sources

Mineral dust is both anthropogenic and natural in origin. It is therefore difficult to make a clear distinction between the sources. Agricultural activities (harrowing, ploughing, harvesting) and road dust that is resuspended by traffic are the most important sources of mineral dust in $PM_{10}$ in the Netherlands and Europe (Schaap et al. 2009; Denier van der Gon et al. 2010). In Europe, the mineral dust contribution from wind erosion appears to have little or no importance. For example, Sahara dust – which is an important source of mineral dust globally and in southern Europe – has an insignificant effect on average on the particulate matter concentration in the Netherlands, with the exception of occasional episodes (Schaap et al. 2010). Mineral dust consists primarily of oxides of silicon, aluminium, calcium, iron and potassium.

#### Contributions

The contribution from mineral dust to $PM_{10}$ averaged 1.6 µg/m³ and varied between 1.2 and 2.1 µg/m³. The average contribution to $PM_{2.5}$ was 0.6 µg/m³, varying between 0.5 and 0.7 µg/m³. The mineral dust consisted primarily (60-70 per cent) of particles larger than $PM_{2.5}$. The contribution of mineral dust to $PM_{10}$ and $PM_{2.5}$ was approximately 50 per cent lower relative to previous estimates (Visser et al. 2001). Compared

with adjoining countries, the mineral dust concentrations in the Netherlands also appeared to be lower. One complication of comparisons with other studies is that the measured periods are not equivalent, because the predominating weather conditions (dry or wet years) appear to affect the contribution of mineral dust to $PM_{10}$ and $PM_{2.5}$. In addition, mineral dust contributions differ according to how they are derived from the measured concentrations of metals. On days with elevated $PM_{10}$ concentrations (> 30 µg/m³), the absolute concentration contribution of mineral dust also increases. The relative share of mineral dust was 7 per cent in $PM_{10}$ and 4 per cent in $PM_{2.5}$, and appeared to be constant with the $PM_{10}$ concentration.

### 2.2.6 Metals

#### Sources

Metals that are not part of mineral dust are released during various types of wear processes and in the metal industry. The metals component can be considered as entirely anthropogenic. The presence of metals in particulate matter can therefore be linked to specific anthropogenic sources. For example, zinc is linked to tyre wear, copper to brake wear, and cadmium to waste incineration and cement production. During the analysis of measurement data, the metals are therefore used as indicators for the contribution of specific source groups to particulate matter.

#### Contributions

The contribution from metals was 0.8 to 2.4 µg/m³ on average for $PM_{10}$ and 0.4 to 0.8 µg/m³ for $PM_{2.5}$. As a result, the metals component consisted largely (50-70 per cent) of particles larger than $PM_{2.5}$. The contributions of metals and mineral dust to particulate matter are similar in magnitude and behaviour. However, the contribution from metals could be measured with much greater precision than the contribution from mineral dust.

On days with elevated particulate matter concentrations (>30 µg/m³), the concentration contribution of metals also increased, but the relative proportion decreased slightly. Of all the particulate matter components referred to here, the contribution from metals showed the highest relative differences between regional, urban and street measurements; the lowest contribution was found in the regional measurements and the highest contribution in the street measurements. This indicated that the sources of metals have a more anthropogenic and local character than the sources of mineral dust. Road traffic is probably an important source of the metals.

### 2.2.7 Not specified

#### Sources

This constituent is the difference between the total $PM_{10}$ or $PM_{2.5}$ concentration and the sum of the other constituents. On average, the not-specified constituent comprised 12 per cent of the particulate matter, and differed between locations and according to the particulate matter concentration. The magnitude of the not-specified constituent was related to systematic deviations in the measurements of the other particulate matter constituents. The net result of the systematic deviations appeared in the not-specified constituent. It is especially the determination of TCM that appeared to play a role in this process (Schaap et al. 2010). Comparable studies suggested that the non-specified constituent consists largely of water. Until now, measurements of the relationship between water and hygroscopic particulate matter constituents, such as the secondary inorganic aerosol, have provided the only support for this hypothesis.

#### Contributions

The average contribution from the not-specified constituent in $PM_{10}$ was 2.6 µg/m³ (12 per cent); in $PM_{2.5}$ it was 1.8 µg/m³ (also 12 per cent). Consequently, approximately one-third (35 per cent) of the not-specified constituent was located on particles larger than $PM_{2.5}$.These contributions are in the same order of magnitude as the estimates of the concentration contribution for water that is bound to SIA in $PM_{10}$.

The contribution of the water bound to the organic secondary constituents was estimated at approximately 1.5 to 2.0 µg/m³ (about 5-10 per cent) of the total $PM_{10}$ concentration (Hoogerbrugge et al. 2010). Sea salt and the secondary organic aerosol also contain water, but no estimate was made of this percentage. However, these contributions are substantially smaller.

On days with elevated $PM_{10}$ concentrations (>30 µg/m³), the concentration contribution of the not-specified constituent increased sharply, to about 7 µg/m³ in $PM_{10}$ and about 6 µg/m³ in $PM_{2.5}$. The relative proportion is therefore about 20 per cent at the various measurement locations. At the regional locations, the proportion of the non-specified constituent in both $PM_{10}$ and $PM_{2.5}$ on these days was roughly twice as large relative to the street and urban locations.

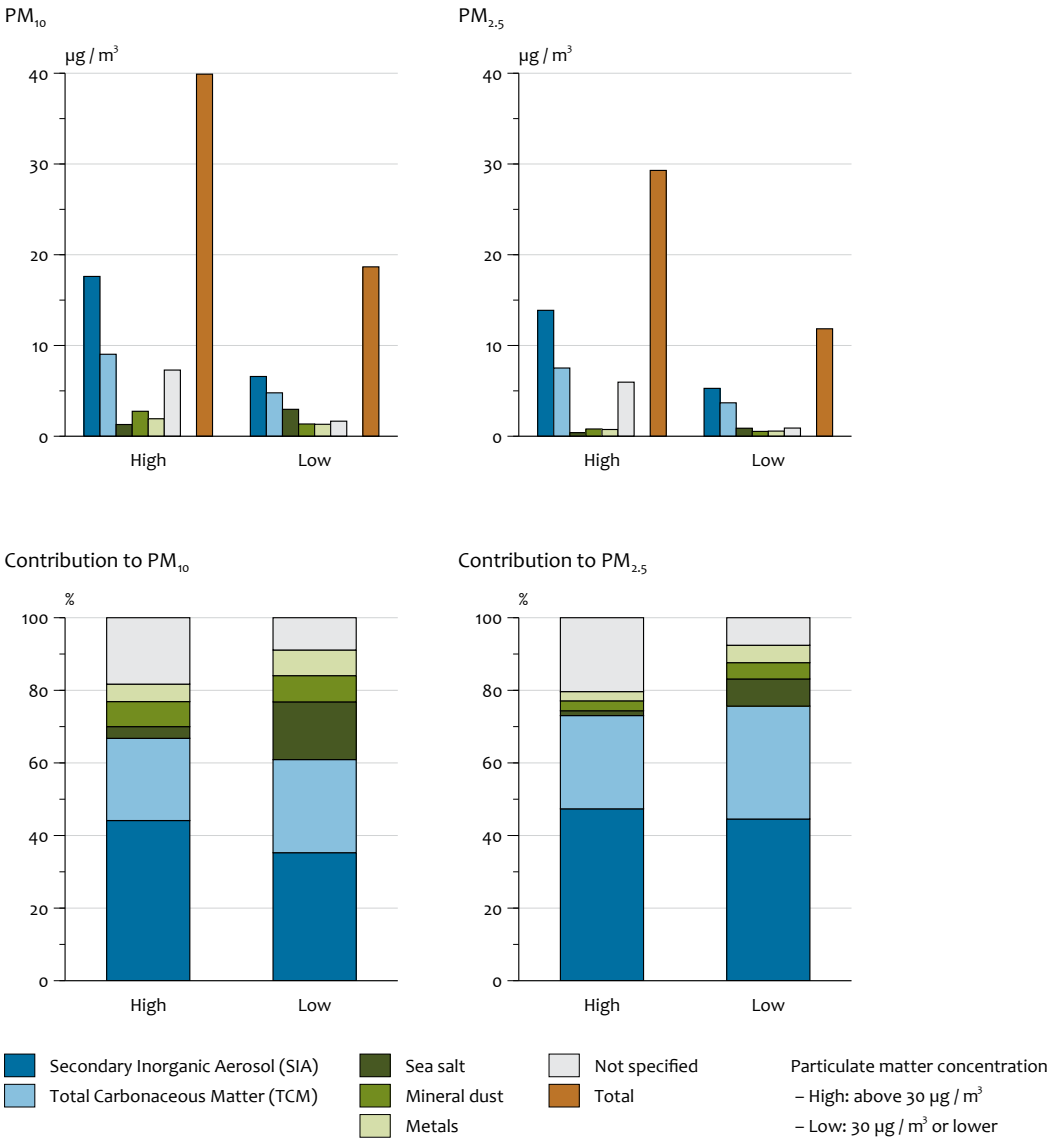
When allocating the particulate matter constituents to anthropogenic or natural sources, the not-specified constituent is proportionally distributed between the other constituents.

## 2.3 Are the constituents of particulate matter different at elevated PM concentrations?

According to the data, the composition of $PM_{10}$ and $PM_{2.5}$ changed when the concentration changed. Elevated particulate matter concentrations turned out to be largely the result of an increase in the SIA constituent. At elevated $PM_{10}$ concentrations (>30 µg/m³), the relative proportion of SIA increased up to 50 per cent. The relative proportion of the 'not-specified' constituent also increased to approximately 20 per cent in both $PM_{10}$ and $PM_{2.5}$. This is an additional indication for the presence of water in the 'non-specified' constituent, water which is bound to SIA. The anthropogenic contribution increased, and the contribution from sea salt declined.

Elevated $PM_{10}$ concentrations occur especially when the air is transported from eastern and southern directions and during extended periods without rain. Other meteorological factors also play a role in the level of particulate matter concentrations (see also Hoogerbrugge et al. 2010). In

**Composition of high (elevated) and low particulate matter concentrations, 2007 – 2008** **Figure 2.4**




*Composition of $PM_{10}$ and $PM_{2.5}$ in µg/m³ at elevated (above 30 µg/m³) and low (30 µg/m³ or below) $PM_{10}$ concentrations.*

addition, incidental source contributions, such as fireworks during New Year celebrations, result in elevated particulate matter concentrations. Particulate matter can also increase relative to average levels due to the contribution of natural sources such as sea salt or mineral dust. However, this occurs only once or twice per year on average (Manders et al. 2009; Denier van der Gon et al. 2010). Elevated particulate matter concentrations in the Netherlands are certainly anthropogenic in origin.

The $PM_{10}$ concentration was above 30 µg/m³ in approximately one-sixth of the measurements. In those cases, the contribution of all constituents was higher, except that of sea salt. On days with $PM_{10}$ concentrations above 30 µg/m³, the contribution of sea salt was 1 µg/m³, compared to 3 µg/m³ at $PM_{10}$ concentrations of 30 µg/m³ or below. Low sea salt concentrations indicate air movement over land.

On average for the measurement locations, the relative proportion of SIA increased from 35 to 44 per cent, and the relative proportion of the not-specified constituent doubled from 9 to 18 per cent. In contrast, the collective contribution from mineral dust, metals and sea salt fell by half, from 30 per cent to 15 per cent of the total. The relative proportion of TCM also declined by several per cent. At elevated $PM_{10}$ concentrations, the relative proportion of the particulate matter constituents in the $PM_{2.5}$ fraction changed in a similar way as the constituents in $PM_{10}$.

The increase in SIA was primarily the result of more particles being formed from nitrogen oxides and ammonia (ammonium nitrate). Calculations of yearly the average concentrations (Velders et al. 2009) showed that 20 per cent of the SIA in the Netherlands originated from sources inside the country. This is a relatively small proportion; the remaining 80 per cent

came from sources in other countries. However, on days with elevated particulate matter concentrations, the proportion originating in the Netherlands is probably significantly higher than 20 per cent. This is because the residence time of the air within the country is longer than average on such days; air is transported over land and the wind speed is low. Moreover, under these conditions the emissions of nitrogen oxides and ammonia, which lead to an increased contribution from ammonium nitrate, are relatively high in the Netherlands.

## 2.4 How does the composition of particulate matter differ according to measurement location (regional, urban and street)?

Sea salt in the atmosphere declined in proportion to the distance from the coast. It is entirely of natural origin and also appeared to be unrelated to the type of measurement location (regional, urban, street). But the other constituents varied according to the type of location as a result of differences in anthropogenic contributions. On average, the absolute contribution of the various particulate matter constituents did not increase, or increased only slightly, from the regional background locations to the urban locations. From the urban locations to the street locations, however, there was a clear increase in individual constituents (see Section 3.1). Metals, and to a lesser extent TCM, showed the strongest increases between these locations. At the street locations, the absolute contribution from metals was three times higher than that at the regional locations. TCM increased especially as a result of higher concentrations of elementary carbon. There was a small increase in the contribution of mineral dust between the regional, urban and street locations, but this was not significant. An increase could be attributed to the road dust that is resuspended by traffic. At other street locations in the Netherlands and abroad, the absolute contribution from dust was twice as high on average as the contribution at the urban locations. The fact that this effect did not appear in the BOP measurements could not be explained.

The contributions of SIA to $PM_{10}$ and $PM_{2.5}$ differed significantly at the regional background locations (7-10 µg/m³). These differences were difficult to explain due to the large-scale character of SIA.

The concentration differences of $PM_{10}$ and $PM_{2.5}$ between the urban location and the various regional locations turned out to be small on average; 0-3 µg/m³ ($PM_{10}$) and 0-1 µg/m³ ($PM_{2.5}$). These differences were smaller or of the same magnitude as the mutual concentration differences between the three regional locations. Therefore, the measurement setup was inadequate to determine the significance of the contribution from urban sources to particulate matter. In subsequent research, the urban contribution will be studied with measurements taken specifically for this purpose.

# 3

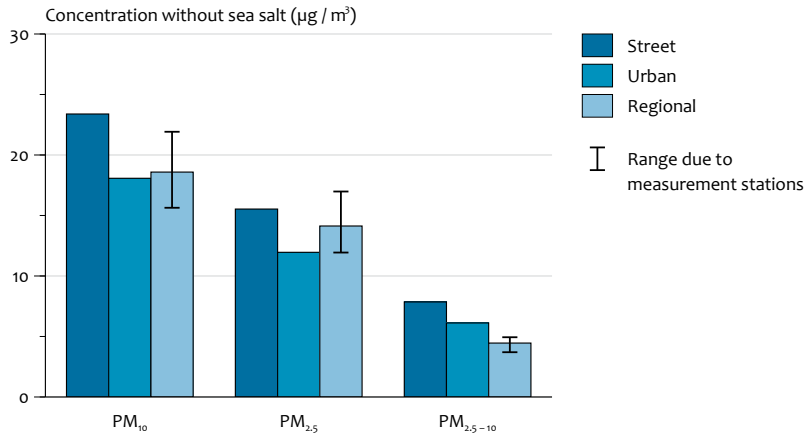
# Particulate matter in the urban area

- The urban background concentrations[1] of $PM_{10}$ and $PM_{2.5}$ from anthropogenic sources were not significantly higher than the corresponding regional background concentrations. Therefore, measures taken in a city to reduce the urban background concentration of $PM_{2.5}$ have a limited effect.
- Compared to the urban background, an additional anthropogenic contribution of 5 µg/m³ to $PM_{10}$ was measured at the street location, along with a contribution of 3 µg/m³ to $PM_{2.5}$. These contributions at the street location consisted of carbonaceous particulate matter, especially elementary carbon, and metals. The contributions to particulate matter from dust and from sulphur dioxide, nitrogen oxides and ammonia were not significantly higher at the street location relative to the urban location.
- The urban background concentration of particulate matter within the urban area of Rotterdam turned out to be virtually constant, i.e. within a margin of 10 per cent for $PM_{10}$ and 5 per cent for $PM_{2.5}$. These measurements suggest that there is a type of particulate matter plateau, where there is a small difference between the concentrations at locations outside the urban area and a more constant level for the urban background concentration.
- The negligible differences in concentration at various urban locations within a city mean that a single measurement point is representative for determining the exposure in that city to $PM_{10}$ and $PM_{2.5}$.
- The contribution from wood combustion (for example in wood stoves and fireplaces) to the urban background in Amsterdam was relatively small. On average during a six-month period (summer and winter) there was a contribution to the $PM_{10}$ concentration of 0.1 to 0.2 µg/m³.
- The spatial and temporal differences in the contribution from wood combustion were large: in the winter the contribution was 10 times higher than in the summer. Locally, the contribution of wood combustion to particulate matter can be as high as 6 µg/m³. In these cases, the relative contribution from wood combustion was up to 30 per cent for $PM_{10}$ and 40 per cent for $PM_{2.5}$.

The importance of the urban area regarding the exposure of the population to particulate matter was acknowledged during the preparation of the European standards for $PM_{2.5}$ (EU 2008b). A target has been formulated for exposure reduction: a relative decrease between 2010 and 2020 of the $PM_{2.5}$ concentration in the urban area, where the exact target depends on the $PM_{2.5}$ levels in 2009, 2010 and 2011. Based on the current knowledge about the $PM_{2.5}$ levels in the urban areas, a reduction target of 15 per cent in the Netherlands is foreseen. However, depending on the de $PM_{2.5}$ measurements in 2010 and 2011 this target could possibly be 20 per cent.

This chapter first summarises the results of the BOP research on the elevated levels of particulate matter and its constituents in the urban area. It then discusses the conclusions of independent measurement campaigns in the urban area near Rotterdam that focused on the spatial and temporal variability of particulate matter. Finally, it presents the most important results of the studies on sources that contribute to combustion aerosol. Combustion aerosol, particulate matter that is released during combustion processes, is assumed to be the component of particulate matter with the greatest health impact. Road traffic, seagoing and inland shipping and wood combustion all contribute to combustion aerosol. Because there were many unknowns concerning the emissions of these sources and their contribution to particulate matter, the BOP research paid special attention to them.

**Particulate matter concentration without sea salt, 2007 – 2008** **Figure 3.1**




*Average concentrations (µg/m³) without sea salt: $PM_{10}$ minus sea salt, $PM_{2.5}$ minus sea salt and the difference between $PM_{10}$ and $PM_{2.5}$ ($PM_{2.5\_10}$ minus sea salt) for the street, urban and regional locations, 2007-2008. The range for regional locations provides the minimum-maximum of the concentration contributions at the three regional locations.*

## 3.1 $PM_{10}$ and $PM_{2.5}$ and their constituents at urban and street measurement locations

In and near urban areas, the emissions of particulate matter and a number of precursor gases are high. When BOP began, the knowledge about the composition of and contributions to urban PM was still very limited. The measurements from the BOP measurement campaigns specified the differences. $PM_{10}$ and $PM_{2.5}$ concentrations differed according to the type of location (regional, urban, street).

Figure 3.1 shows the average concentrations (without sea salt) of $PM_{10}$ and $PM_{2.5}$ and the difference between $PM_{10}$ and $PM_{2.5}$ ($PM_{2.5}$ is the fraction of finer particles in $PM_{10}$). The contribution of sea salt has been subtracted from the total to clarify the concentration differences between the regional, urban and street locations due to anthropogenic sources. The contribution of sea salt to particulate matter is natural in origin and depends on the type of location; as a result it distorts the analysis of anthropogenic contributions.

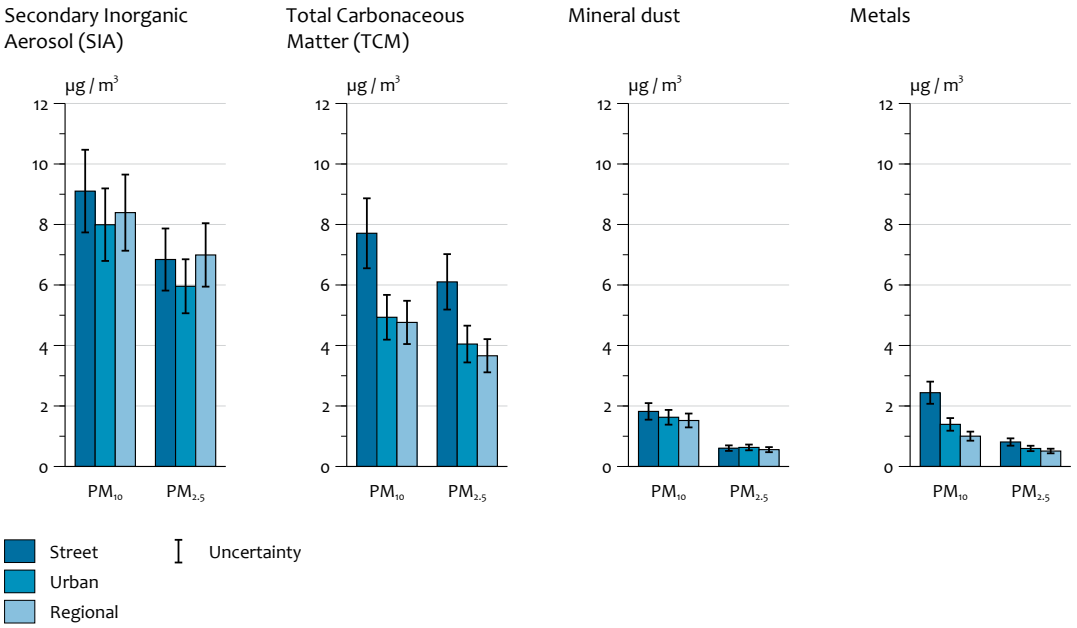
Relative to the regional contribution, the contribution of the urban area it is not very large in the Netherlands – no more than several µg/m³ (Voogt et al. 2009). The regional measurements were of limited suitability for measuring the increase in the various particulate matter constituents from the regional to the urban locations. For the coarser fraction, which is part of $PM_{10}$ but not of $PM_{2.5}$, the increase from regional to urban measurement locations was approximately 1 - 1.5 µg/m³. However, this difference is probably not significant. The concentration difference in $PM_{10}$ between urban and street locations was caused by particulate matter components in both the fine ($PM_{2.5}$) and the coarse ($PM_{10}$-$PM_{2.5}$) fractions.

The difference between the urban and street locations was determined based on a single set of two linked measurement locations, Schiedam and Rotterdam. This set will probably not be representative for all increases from urban to street locations, but is probably sufficiently representative to make general conclusions about the Randstad (the urban agglomeration in the western part of the country). The increase between urban and street locations was as high as several µg/m³; approximately two-thirds of this difference was due to finer particles and one-third to the coarser particles.

For each constituent, Figure 3.2 shows the concentration differences between the regional, urban and street measurement locations. This figure confirms the picture of a small difference between regional and urban locations. The spatial variance of the SIA contribution to $PM_{10}$ and $PM_{2.5}$ was relatively limited. The gradients in the SIA concentrations between the regional locations were larger than those between the regional locations and the urban locations. The concentration increase between the urban and street locations was largely attributed to TCM and metals. TCM increased as a result of elementary carbon (see Figures 3.4 and 3.5). There was a small increase in the mineral dust contribution between regional and urban locations, and between urban and street locations. Such an increase could be attributed to the road dust that is resuspended by traffic. However, the concentration differences were not significant. Other measurement data from the Netherlands and abroad have shown that the dust contribution in streets was twice as high on average as that in the urban background, probably as a result of road dust resuspended by traffic (Denier van der Gon et al. 2010). It is unclear why this general behaviour of dust did not emerge from the BOP measurement data.

As part of the BOP measurement campaign, the concentration and composition of $PM_{10}$ were measured at a street location in Breda. These measurements supported the conclusions about the composition of particulate matter at street locations. Except for the sea salt contribution, the relative proportions of the $PM_{10}$ constituents at the street location in Breda were about the same as those at the

**Particulate matter concentration per component, 2007 – 2008** **Figure 3.2**




*Constituents of $PM_{10}$ and $PM_{2.5}$ (in µg/m³) at street, urban and regional locations.*

street location in Rotterdam. However, the concentration contributions of the various constituents were lower in Breda than in Rotterdam, except for SIA. Breda had the highest SIA contribution of all BOP locations.

## 3.2 How does the particulate matter concentration vary in the urban area?

The European directive (EU 2008b) established a target value for the urban background concentration, which is defined as the concentration at 'Urban background locations': these are urban locations where the levels are representative for the exposure of the general urban population. This concerns urban locations, and not street and road locations in a city; at street locations, the particulate matter concentration is often higher due to the contribution of road traffic, and is therefore not representative of the average concentration in the urban area.

To map out spatial concentration differences, two measurement campaigns were conducted in the urban area of Rijnmond. These campaigns were conducted in de period 2007-2008 using mobile apparatus and particulate matter measurements taken at a network of 11 fixed locations (Voogt et al. 2009).

The results indicated that the urban background concentration of $PM_{10}$ or $PM_{2.5}$ could be determined by using a single measurement point. However, the uncertainty is relatively high (20 per cent). Logically, more measurement points would reduce the uncertainty in the measurement. The current measurement strategy to ascertain $PM_{2.5}$ concentrations in the urban area was developed by using the knowledge about the variability in the concentration at the urban locations.

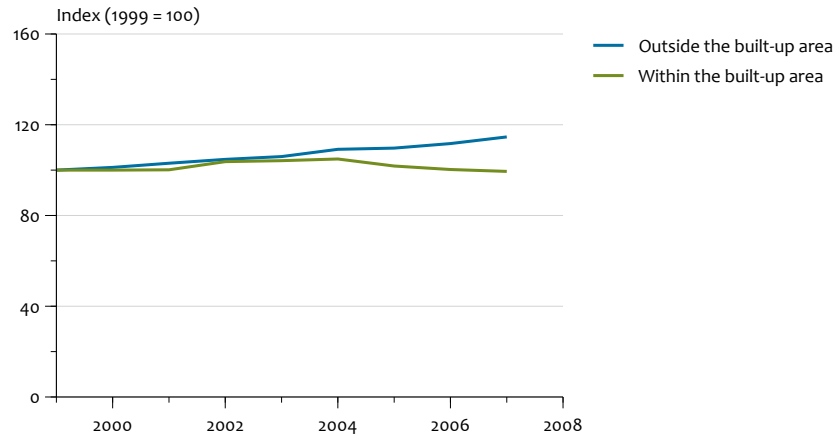
Moreover, the research of Voogt et al. (2009) showed that on average there was only a small difference between particulate matter concentrations in the urban area of Rotterdam and those outside this area. As a monthly average, this difference was between 1 and 3 µg/m³ for $PM_{10}$ and less than 1 µg/m³ for $PM_{2.5}$. These relatively small differences are consistent with previous estimates and are characteristic of the situation in the densely populated Netherlands. Measurements from other parts of Europe have shown much larger differences between regional and urban background concentrations. For Europe as a whole, 7 µg/m³ ($PM_{10}$) and 4 µg/m³ ($PM_{2.5}$) are typical values for the increase in concentration between urban locations and street locations (Mol et al. 2009).

Traffic is an important source of particulate matter in the urban area. A spatial and temporal trend in the traffic emissions affects the distribution of particulate matter concentrations in the urban area. Exhaust emissions of particulate matter from road traffic have declined at the national and European levels since 1990 (see Chapter 5). Due to traffic circulation management, traffic emissions have changed spatially at the urban level.

An analysis has been made about the spatial changes of traffic volume in various Dutch cities based on figures from the *Nationale Mobiliteitsmonitor* (see Hoogerbrugge et al. 2010). The results are relevant to a better understanding of the particulate matter concentration at the urban scale. Within the built-up area, the developments in traffic volume were more or less stable, but outside this area there was a constant growth of approximately 2% per year in traffic volume (Figure 3.3). However, the developments in traffic volume were not the same for all urban areas.

Spatial differences within an urban area are caused by changes in traffic circulation. Between 1999 and 2007 in

**Passenger car kilometres** **Figure 3.3**




*The trend in average vehicle distances travelled inside and outside the built-up area in the Netherlands. (Source: Mobiliteitsonderzoek Nederland 1999-2007, recalculation by Goudappel Coffeng; Hoogerbrugge et al. 2010).*

Utrecht and Amsterdam – which are urban areas with old city centres – the vehicle distance travelled within the built-up area remained approximately level, but outside the built-up area it increased. In Rotterdam – which is a city with a more open traffic infrastructure – there was not such a clear difference. However, these types of differences are difficult or impossible to measure as changes in the $PM_{10}$ and $PM_{2.5}$ concentrations.

## 3.3 Combustion aerosol

The particles that are released during combustion processes are defined as combustion aerosol. These particles are strongly associated with the adverse health effects of particulate matter. Black smoke is an indicator for the contribution of combustion aerosol to particulate matter (see Particulate matter and health impact in Section 1.1). According to the Netherlands *Environmental Balance* (Milieubalans; PBL 2009a), the concentration of combustion aerosol appears to be a better indicator for the impact of the local mixture of air pollutants on public health than $PM_{10}$.

The Netherlands Pollutant Release & Transfer Register (ER 2010) contains annual estimates of the anthropogenic emissions, including those from combustion processes. However, there are still large uncertainties in some areas. The BOP research contributed explicitly to improving the knowledge about combustion aerosol. This concerned the following research activities:

- Researching the contribution and emission factors of elementary and organic carbon from road traffic in the urban area.
- Systematically mapping out the methods used in the Netherlands for determining emissions from seagoing shipping and shipping on inland waterways.
- Estimating the contribution from wood combustion to $PM_{10}$ and $PM_{2.5}$ at an urban location and at a regional location in the rural area.

### 3.3.1 Road traffic

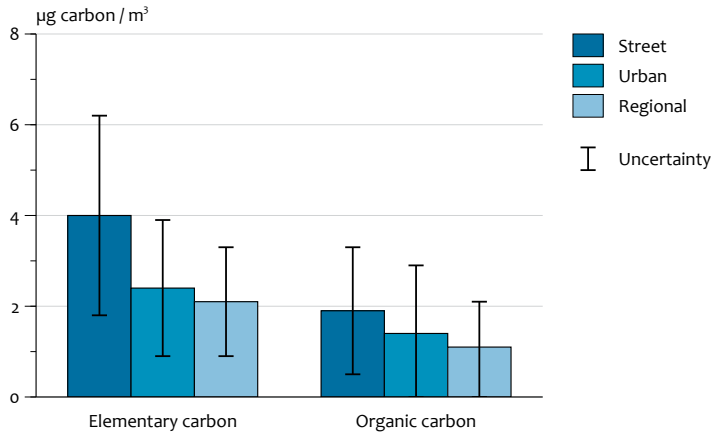
The contribution of traffic emissions to EC and OC was studied at street and urban locations with measurements and model calculations (Keuken & ten Brink 2009; ten Brink et al. 2009). These studies aimed to improve the knowledge about EC and OC and about the increase in EC and OC concentrations at urban and street locations relative to concentrations outside the urban area. In addition, an EC emission factor of 10 mg carbon per travelled km was empirically determined for urban traffic on location. This factor is near the bottom of the range reported in international literature of 8 to 20 mg carbon per travelled km. Because no significant increase for OC was measured at street locations, an OC emission factor could not be determined for urban traffic (see Figure 3.5).

#### Contribution of carbonaceous particulate matter in the urban area

Knowledge about the contribution of EC and OC to particulate matter in the urban area helps to determine whether the European standards for $PM_{2.5}$ in the urban area will be attained. In the Rijnmond area, measurements and models were used to study the contribution of elementary carbon and organic carbon to the particulate matter concentration in the urban area. The measurements showed a significant increase in the contribution of TCM to $PM_{10}$ and $PM_{2.5}$ only between urban and street locations. The small observed increase between regional and urban locations corresponded approximately with an increase in the EC and OC concentrations. The observed increase between urban and street locations appeared to be primarily the result of an increase in EC (Figure 3.4).

The contribution of EC and OC emissions from road traffic in the urban area to the large-scale $PM_{2.5}$ and $PM_{10}$ concentrations was approximately 0.5 µg/m³. As a result, the potential to reduce particulate matter in the urban area on a large scale by limiting exhaust emissions of EC and OC is very limited. If all road traffic were to be stopped in a large area, the $PM_{2.5}$ concentration would decline by a maximum of only 5% annually due to the reduction of exhaust emissions.

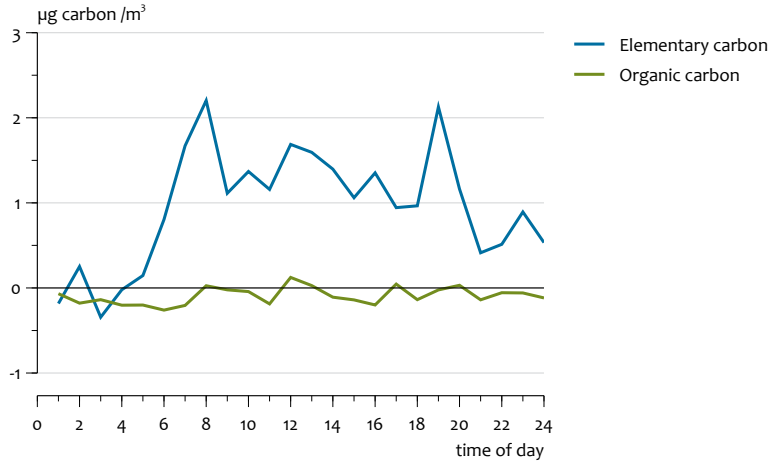
**$PM_{10}$ concentration of carbonaceous components, 2007 – 2008** **Figure 3.4**




*Elementary and organic carbon concentrations (µg carbon/m³) in $PM_{10}$ at street, urban and regional locations, 2007-2008 (Keuken & ten Brink 2009).*

**Street contribution to $PM_{10}$ concentration of carbonaceous components, 2007 – 2008** **Figure 3.5**

Concentration difference between street station Bentickplein Rotterdam and urban station, weekdays




*Hourly variations in the street contribution of elementary and organic carbon (µg carbon/m³) to $PM_{10}$: the increase measured at the street location Rotterdam Bentinckplein, relative to a measurement point in the vicinity without contribution from traffic (Keuken & ten Brink 2009).*

However, the effect of such a hypothetical measure on the particulate matter concentrations would probably be significantly greater, because the other traffic contributions would also be eliminated. This concerns the contribution from wear of tyres, brakes and the road surface and the contribution in streets from dust resuspended by traffic. The traffic contribution to the particulate matter concentration that does not originate from vehicle exhaust is uncertain, and is possibly larger than the contribution from exhaust emissions (Hoogerbrugge et al. 2010). According to the measurements, the contributions of other anthropogenic sources in the Rijnmond area (such as industry, refineries, air traffic and shipping) to the large-scale EC and OC concentrations in the urban area were not significant (Keuken & ten Brink 2009).

### Contribution of carbonaceous particulate matter at the street level

At the Bentinckplein street location in Rotterdam, the EC concentration was 4 µg/m³ on average. The OC concentration was about half this level. Traffic appeared to be the source of EC at the street level, because clear traffic peaks were observed (Figure 3.5). OC concentrations in the street were not significantly higher than those measured away from the street. Moreover, the increase in the OC concentration in the urban area relative to the region was not significant. This means that the background contribution to OC dominated the concentrations at the urban and street locations.

The model results for Rotterdam also showed a large difference between EC concentrations near busy roads and large-scale EC concentrations in the urban area; these were

much larger than the corresponding differences for $PM_{2.5}$ and $PM_{10}$. This confirms that EC is a good indicator for the dispersion of particulate matter emissions from traffic.

### 3.3.2 Shipping

A good estimate of shipping emissions is essential for specifying the impact of shipping on air quality and health in harbour cities and coastal areas. In the Netherlands, shipping is an important – but poorly documented – emission source of particulate matter and precursor gases. Since 2000, estimation methods have been developed specifically for shipping emissions on the North Sea and in harbours, as well as for traffic on the inland waterways. Denier van der Gon & Hulskotte (2010) summarised and described the methods used to estimate particulate matter emissions from shipping, as they are currently used in the Netherlands Pollutant Release & Transfer Register. Additional attention was given to the current emission factors and activity data that are needed to estimate emissions from moored ships and traffic on the inland waterways. A methodology was formulated to derive the particulate matter emissions from sulphur dioxide emissions, which are changing due to regulation. At present, the estimated emission from traffic on the inland waterways in the Netherlands is approximately 1 kilotonne. This is approximately 15 per cent of the particulate matter emissions from traffic on the inland waterways in all of Europe.

### 3.3.3 Wood combustion

During the combustion of wood, gases and particulates are released. The released particles consist primarily of organic carbon, with a smaller component of elementary carbon (soot). It had been previously assumed that the contributions to particulate matter in the Netherlands due to wood combustion were very small, because natural gas is the dominant fuel in the Netherlands for domestic heating and energy generation. Virtually no particulate matter is released during the combustion of natural gas. According to the Netherlands Pollutant Release & Transfer Register, the particulate matter emissions caused by wood combustion in stoves, fireplaces and combustion in small-scale energy production amounts to less than 5 per cent of the primary anthropogenic emissions of particulate matter in the Netherlands. The actual amount of air pollution caused by the use of wood stoves is uncertain, because this is strongly dependent on the type of fuel, the type of stove and how the stove is operated.

In many other European countries, such as Germany and France, wood combustion is one of the most important sources of primary $PM_{2.5}$. Because estimates of particulate matter emissions from wood stoves are notoriously uncertain, larger contributions are possible than those that have been assumed until now. To estimate the contribution from wood combustion to $PM_{10}$ and $PM_{2.5}$, initial measurement campaigns have been conducted in the urban area near Amsterdam and in the rural area near Schoorl, where large amounts of wood are customarily burned in stoves and fireplaces (Weijers et al. 2010, article in preparation).

#### The average contribution is small

The measurements resulted in a differentiated picture of the contribution from wood combustion to particulate matter. In the urban area, the average contribution was indeed limited to approximately 1 per cent (0.1 to 0.2 µg/m³). This level was consistent with the yearly average contribution from wood combustion as calculated annually for the large-scale $PM_{10}$ concentration map (GCN). During the winter months and in rural surroundings where more wood is burned, the contribution to $PM_{10}$ was substantially larger: between 10 and 30 per cent. During the winter months, the local contribution from wood combustion to the $PM_{10}$ concentration can be quite large. Wood combustion, which can result in local annoyance for susceptible groups, could result in exceedances of the limit value for 24-hour average $PM_{10}$ concentrations (50 µg/m³). This is probably only the case in forested areas where wood is a customary fuel.

#### Large spatial and temporal differences

During the winter months, the contribution from wood combustion in Amsterdam was approximately 8 times higher than during the summer (Figure 3.6). At a shorter time scale, the differences between summer and winter were even greater: on a weekly basis there were maximums in the winter of several µg/m³ compared to less than 0.1 µg/m³ in the summer. The measurement campaign in Schoorl aimed to map out a situation with a high contribution from wood combustion. In February 2009, a contribution of 1 to 4 µg/m³ from wood combustion was derived from the measurements. Relative to total $PM_{10}$, this is a maximum of 30 per cent. This contribution was of local origin.

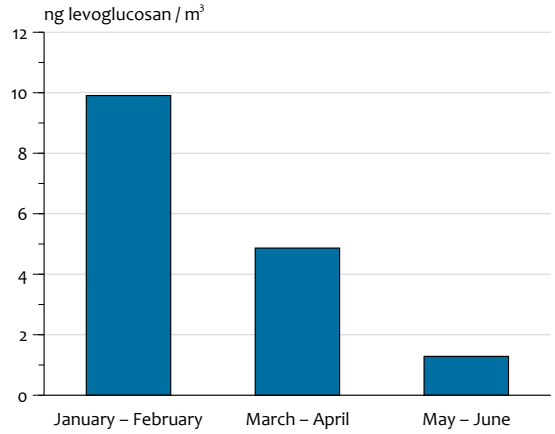
#### Knowledge about local contributions from wood combustion is limited

As a source of primary particulates, the emission contribution from wood combustion is included in the Netherlands Pollutant Release & Transfer Register under the item *vuurhaarden* (fireplaces). This item includes open and closed wood stoves and open hearths that are used as a heat source by private individuals. The type certification for wood stoves was abolished in the Netherlands in 2004, leaving the existing, less stringent European certification system in place. The EU is working on a more stringent certification system for wood stoves. In addition, there are modern wood stoves that are fuelled with wood pellets, which are subject to measures to limit the emissions of sulphur dioxide, nitrogen oxides and $PM_{10}$.

The measurements of the emissions and spatial/temporal dispersion of particulate matter from wood combustion appear to be sufficient to establish the yearly average contribution in cities by using models. However, the concentration contribution to particulate matter from wood combustion is seasonal, and on some days or some months – and in wooded areas – is sometimes much higher. Compared to the knowledge about the local contribution at the street level from sources such as road traffic, the knowledge about the local contribution from wood combustion has scarcely developed. The local contribution from agriculture, storage and transshipment locations and traffic has been mapped out with models and measurements. Regarding the contribution from wood combustion, only initial measurements have been taken, even though the local contribution can be high. Moreover, the contribution is relevant to health. It is unclear to what extent wood combustion plays a role

**Concentration due to wood combustion, 2006** **Figure 3.6**

Measurement location Amsterdam-Vondelpark




*Concentrations of an indicator (levoglucosan) for the contribution of wood combustion to particulate matter, in ng/m³. Concentrations were measured at the urban location Amsterdam Vondelpark; they are averages for the periods January through February, March through April and May through June 2006.*

during exceedances of the standard for 24-hour average $PM_{10}$ concentrations; the human exposure to combustion aerosol from this source is also unclear. To clarify the local contribution of wood combustion to particulate matter, the estimates of particulate matter emissions from wood stoves and the spatial and temporal distribution must be improved. These improvements will become increasingly urgent if wood becomes more popular as a fuel for heating. Boersma et al. (2009) recently published emission projections for $PM_{10}$, $PM_{2.5}$ and other components as a result of wood combustion.

**Note**

[1] The European directive (EU 2008b) has established a target value for the urban background concentration, which is defined as the concentration at 'Urban background locations': locations in urban areas where the levels are representative for the exposure of the urban population in general.

# 4 Policy implications

- On average, anthropogenic constituents make up 75-80 per cent of $PM_{10}$ and 85-90 per cent of $PM_{2.5}$. These percentages are larger – by approximately 25 per cent for $PM_{10}$ and 20 per cent for $PM_{2.5}$ – than the figures currently used in air-quality reports. Relative to previous knowledge, the natural component is approximately one-third smaller.
- The contribution of certain anthropogenic constituents to particulate matter has not yet been determined or only partly determined; these constituents are: mineral dust, carbonaceous particulate matter, secondary particulate matter from sulphur dioxide, nitrogen oxides and ammonia and the water that is associated with anthropogenic particulate matter.
- The emission reductions that were implemented in the Netherlands and other European Member States as a result of the broad European approach have had a positive net effect on the large-scale particulate matter concentrations; this applies not only to compliance with limit values but also to the various anthropogenic constituents that are assumed to have adverse health effects according to current knowledge.
- From this health perspective, measures focusing on reducing metals and TCM, especially soot, have priority. Moreover, in the urban area and near streets, measures focusing on these constituents are even more effective because their contributions are larger at these locations than in the rural area.
- Revision of the Air Quality Assessment Scheme (Regeling beoordeling luchtkwaliteit) for the sea salt deduction is desirable, because the contribution of sea salt to the particulate matter concentrations is smaller than was previously assumed.

Based on the composition data for $PM_{10}$ and $PM_{2.5}$, a number of questions have been answered with an eye to the possibilities for reducing concentrations in order to comply with the European limit and target values for $PM_{10}$ and $PM_{2.5}$ (see textbox Summary of standards for $PM_{10}$ and $PM_{2.5}$ in Chapter 1). In addition, compliance with limit values alone does not necessarily result in health benefits, because not all constituents of particulate matter are equally hazardous to health. In that perspective, the results can be seen as supporting policy that aims for both objectives.

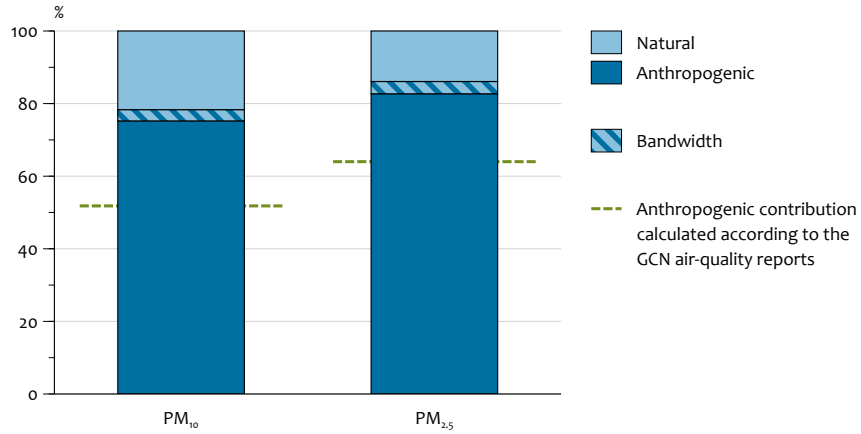
- Which constituents of particulate matter are anthropogenic or natural in origin?
- What are the policy strategies for reducing particulate matter concentrations?
- Is the current sea salt deduction from the Air Quality Assessment Scheme compatible with the new insights?

## 4.1 What proportion of particulate matter is anthropogenic or natural?

The anthropogenic contribution to particulate matter is that portion which is related to human activities and can therefore be influenced by policy measures – at least theoretically. Particulate matter is both natural and anthropogenic in origin. It was previously uncertain which components were natural and which were anthropogenic. The composition measurements in BOP made it possible to clarify the distinction between anthropogenic and natural sources. In the figure below we place these new insights in the perspective of the anthropogenic contribution as it is currently calculated for the large-scale concentrations for the Netherlands (GCN maps; Velders et al. 2009).

The GCN calculations showed that at least half of the measured yearly average $PM_{10}$ concentration originated from anthropogenic sources in the Netherlands and Europe, as shown in Figure 4.1. It was known that the other half, the 'non-modelled' part, also contained anthropogenic components, but the amount was unclear (Matthijsen & Visser 2006). Schaap et al. (2010), based on the BOP measurements, made a conservative estimate of the anthropogenic contribution to yearly average $PM_{10}$ concentrations of 75 to 80 per cent. Consequently, the BOP measurements have earmarked an additional 25 per cent (approximately) of $PM_{10}$ as anthropogenic in origin.

**Anthropogenic and natural contributions to particulate matter, 2007 – 2008** **Figure 4.1**




*Anthropogenic and natural contributions to $PM_{10}$ and $PM_{2.5}$. The striped lines show which contributions were defined as anthropogenic in the reports on air quality – the GCN maps – (Velders et al. 2009). The figures are the averages for the urban and regional locations Schiedam, Hellendoorn, Cabauw and Vredepeel for 2007-2008. The bandwidth indicates the uncertainty about the anthropogenic component in total carbonaceous matter (TCM); it is estimated that half to three-fourths of carbonaceous particulate matter is anthropogenic in origin.*

The anthropogenic contribution ascertained by the GCN calculations was two-thirds of the measured yearly average $PM_{2.5}$ concentrations. The additional component allocated to anthropogenic sources by the BOP measurements therefore turned out to be approximately 20 per cent. The anthropogenic contribution to $PM_{2.5}$ is now thought to be about 85 to 90 per cent. For example, the approximate proportion of particulate matter of natural origin is now estimated at 5 µg/m³ for $PM_{10}$ and 2 µg/m³ for $PM_{2.5}$, which is around 40 per cent lower than estimated by Visser et al. (2001) and Buijsman et al. (2005).

This additional anthropogenic component of $PM_{10}$ (about 6 µg/m³) and $PM_{2.5}$ (about 3 µg/m³) consists of contributions from anthropogenic sources that are disregarded or only partly included in the GCN calculations:

1. ***SIA***; this concerns an additional SIA contribution. The current GCN calculations underestimate SIA concentrations by about 35 per cent. This has been shown from BOP measurements and has been confirmed by other independent measurements in the Netherlands and other countries (see Section 2.2.2).
2. ***TCM***; this concerns the primary and secondary TCM contributions.
   a. The contribution from primary emissions of carbonaceous particles. The emission data are highly uncertain because TCM consists partly of semi-volatile components. The actual contribution could therefore deviate from the contribution calculated in GCN.
   b. TCM also contains a contribution from secondary organic aerosol that is formed from volatile organic compounds (VOCs). The formation of secondary organic aerosol from VOCs was not included in the GCN calculations. The secondary organic aerosol component is both natural and anthropogenic in origin. Based on current measurements and models, its magnitude is still very uncertain.
3. ***Mineral dust***; this concerns the contribution from dust that enters the atmosphere when it is resuspended by vehicles on roads and by agricultural activities. This fraction is not included in the GCN calculations.
4. ***'Not specified'***; this component has not been specified and probably also contains anthropogenic components. Possibly this concerns water that is associated with other components, especially SIA, which is almost entirely of anthropogenic origin. The contribution of water is not currently included in the GCN calculations.

These anthropogenic constituents should be included in the GCN calculations to improve the specification of particulate matter concentrations and the effects of policy measures.

Modifications to include the additional contribution of SIA in the GCN calculations have priority. For this purpose, follow-up research is foreseen. In addition, the 'not specified' anthropogenic constituent can be included in the GCN calculations, if it is confirmed that it consists largely of water. This constituent would then 'hitchhike' on the changes in the fractions with which water is associated.

However, modifications to the contribution of TCM and mineral dust are less self-evident. The modelling of the anthropogenic components of mineral dust and TCM is still very uncertain. The first solutions to this problem, which were introduced in BOP (Schaap et al. 2009; Denier van der Gon et al. 2010), must be worked out in greater detail. Nonetheless, a rapid improvement of the modelling instruments for mineral dust and TCM cannot be expected; this is especially because the modelling of carbonaceous matter remains poorly developed. Moreover, the effectiveness of policy measures to reduce the anthropogenic contribution of mineral dust in the Netherlands is probably limited (Keuken et al. 2009).

## 4.2 Effectiveness of policy strategies

Not all locations in the Netherlands comply with European legislation on particulate matter concentrations. Dutch policy therefore aims to reduce these concentrations, so that these locations can comply with the legislation in the future.

### Particulate matter constituents have different health impacts

The current understanding is that the adverse health effects of particulate matter differ per constituent and in relation to the exposure to specific constituents. Some constituents of particulate matter are much more harmful than others (see Particulate matter and health impacts, Section 1.1). However, there is still insufficient knowledge to classify specific fractions or constituents as harmless, with the exception of sea salt. This means that the public health benefits of compliance with the limit values for $PM_{10}$ and $PM_{2.5}$ depend on the specific emission policy.

### European legislation takes a broad approach to air quality

The international – especially European – legislation on air pollutants is so diverse that virtually all anthropogenic sources of particulate matter are being dealt with. The sources are being addressed for different reasons (see Section 6.2). Compliance with $PM_{10}$ and $PM_{2.5}$ limit values was actually one of the secondary aims. European legislation is leading to large-scale reductions of nitrogen and sulphur emissions to the atmosphere, and in this way to reducing SIA concentrations. In addition, the anthropogenic emissions that contribute to the TCM and metals are being reduced.

### Effects of measures focusing on nitrogen and sulphur emissions

Measures to reduce emissions of nitrogen oxides and ammonia have had the greatest impact on large-scale particulate matter concentrations. The large-scale reduction of particulate matter concentrations also has a local impact. This approach is supported for the Netherlands by previous studies (Erisman & Schaap 2004; Denier van der Gon et al. 2009). Moreover, if these measures are implemented in the Netherlands itself or in the adjacent region, their impact could be even greater if elevated particulate matter concentrations are reduced.

In recent decades, the contributions to particulate matter from sulphur dioxide have been sharply reduced by European emissions legislation. The remaining large sources that are important for the Netherlands are shipping (seagoing and inland), industry, energy production and refineries. Emission reductions in shipping are expected to lead to substantial reductions in particulate matter.

Reducing atmospheric emissions of sulphur dioxide, nitrogen oxides and ammonia also has a major effect on the quality of ecosystems (less acidification and eutrophication). Northwest Europe has a very high emission density of nitrogen components, and a relatively high proportion of natural habitats that are susceptible to acidification and eutrophication. Critical values for deposition are still being exceeded, especially those for nitrogen. Nevertheless, national and European policy has reduced the exceedances (Hettelingh et al. 2010). According to current insights, however, the health benefits of nitrogen and sulphur policy in relation to particulate matter appear to be limited for the Netherlands.

### Effects of measures focusing on carbon and metals

From that health perspective, measures focusing on reducing metals and TCM, especially soot, have priority. Moreover, in the urban area and near roads, policy measures focusing on these constituents are even more effective; this is because their contributions at these locations have been shown to be even larger than those at measurement locations in the rural area. Such measures are more effective in urban areas, where the contribution of these constituents is high and where many people live.

The reduction of combustion aerosols, which contain metals and carbonaceous particulate matter, is partly the result of European standards for vehicle exhaust emissions. In the past, the Netherlands has taken additional measures that aimed to reduce combustion aerosol (see Hammingh et al. 2005). Considering the health impact of this component, this was a wise choice.

### Mineral dust measures

There is no European policy to reduce the contribution from mineral dust. These measures would be essentially different than 'end-of-pipe' technologies that can be used for purposes such as reducing sulphur dioxide emissions. Measures to control mineral dust include counteracting continuous wind erosion. Measures such as wet cleaning or sweeping roads to reduce the contribution from road dust resuspended by vehicles have proven to be ineffective in the Netherlands and Germany (see Keuken et al. 2009). Ploughing techniques are available to reduce the mineral dust contribution from agricultural activities (Baker et al. 2005). These are used primarily to counteract loss of soil fertility; the component of soil that is lost to wind erosion is also the most fertile component. For reducing particulate matter concentrations, the effectiveness of measures to limit dust generated by agricultural activities is unknown.

### European approach to large-scale particulate matter concentrations has a positive net effect

The emission reductions that were implemented in the Netherlands and other Member States as a result of the broad European approach have had a positive net effect on the large-scale particulate matter concentrations; this applies not only to compliance with the limit values, but also to the various anthropogenic constituents that are assumed to have adverse health effects according to current knowledge (see Chapters 5 and 6). In the broad European perspective on air quality, which includes other air-quality themes besides particulate matter, the Member States have limited scope to independently reduce the emissions of specific substances on a large scale. For example, there is no risk that a Member State will focus exclusively on reducing the less health-relevant sulphur and nitrogen emissions with aim of complying with more quickly the particulate matter limit values.

### Specific local policy may be required

The European air-quality policy primarily affects large-scale concentrations. At the local scale, it appears that $PM_{10}$ and $PM_{2.5}$ are not good indicators, especially for locations with heavy traffic or other locations where the concentration of combustion aerosol is high but the contribution to total particulate matter is limited. At the local scale, there is a risk that measures will be taken which reduce the particulate matter concentration, but which are not relevant to health. One example is the installation of barriers along roads without nearby housing in order to comply locally with the $PM_{10}$ or nitrogen dioxide limit value.

To further reduce concentrations at the local scale, measures that focus specifically on the direct emission of particulates are effective. In streets, this primarily concerns measures to reduce the emissions of metals and soot (characterised as elementary carbon in TCM). Reducing the emission of organic carbon (OC) particles appears to make only a limited contribution to reducing the local particulate matter concentration.

Smeets et al. (2007) showed that the cost optimisation strategy for reducing the exposure of the general population to $PM_{10}$ will generally also provide cost-effective solutions for reducing the number of $PM_{10}$ hotspots. More stringent European $PM_{10}$ emission norms for trucks (soot filters) are an exception. This measure has a much larger effect on the $PM_{10}$ concentration along motorways than on the $PM_{10}$ exposure of the general population. From a health perspective, this measure is especially relevant.

## 4.3 Is the current sea salt deduction from the Air Quality Assessment Scheme compatible with the new insights?

The European air quality directive stipulates that the contribution of natural sources to $PM_{10}$, including sea salt, can be subtracted from exceedances of limit values. There are arguments both for and against the application of this 'sea salt deduction' (see textbox Sea salt deduction).

The most important insights from the BOP research about sea salt are the following:

- The yearly average contribution of sea salt during the BOP period 2007-2008 turned out to be smaller than previously assumed. The measurements indicated a contribution of 2 to 4 µg/m³. A higher local deduction (which varies per municipality) of 3 to 6 µg/m³ is currently used for the yearly average $PM_{10}$ concentration.
- The current sea salt deduction, based on multi-annual average sea salt concentrations, is too high in case of elevated yearly average sea salt concentrations and too low in case of lowered yearly average sea salt concentrations. The year-to-year variability in the sea salt concentration is large (±30 per cent).
- The contribution of sea salt to the 24-hour average $PM_{10}$ concentrations is extremely variable in time and space. Days on which the limit value is exceeded for the 24-hour average $PM_{10}$ concentrations are attributed to incidental source contributions (fireworks, bonfires) or to higher background levels caused by transport of air over land instead of over the North Sea or Atlantic Ocean. There are few exceedances of the limit value for 24-hour average $PM_{10}$ concentrations on days with a sea wind.
- When determining the number of exceedance days, the current sea salt deduction is based on a constant contribution from sea salt for the entire country. However, the most recent measurement data from the National Air Quality Monitoring Network indicate that the contribution of sea salt on exceedance days differs between the eastern and western regions of the Netherlands, where the western region is approximately 1 µg/m³ higher than the eastern region.
- The number of exceedance days that remain with a fixed sea salt deduction depends on the yearly average $PM_{10}$ concentration. With a further decline in particulate matter

### *Sea salt deduction*

*The European air-quality directive stipulates that the contribution of natural sources to $PM_{10}$, including sea salt, can be subtracted from exceedances of limit values. The reason for this provision is to create a level playing field for Member States for tackling their air quality problems, so that no additional effort is required because there happens to be a high natural contribution in a particular Member State. The disadvantage of this policy is that the limit values for particulate matter, as a result of deducting the natural contribution, provide a somewhat lower level of health protection. This is because the deduction leads de facto to a relaxation of the standard. This expansion of the limit value also offers the possibility for an additional anthropogenic contribution, which – in contrast to sea salt – probably does have adverse health effects. Essentially, the sea salt contribution is replaced with additional anthropogenic contributions up to the limit value. Member States which claim an exceptionally high contribution from natural sources must be able to prove this. In the Netherlands, the sea salt deduction is used to comply with both the limit value for 24-hour average and the yearly average $PM_{10}$ concentrations. The amount of sea salt that can be deducted is defined in the Air Quality Assessment Scheme (Regeling beoordeling luchtkwaliteit - Staatscourant 2007). For the yearly average $PM_{10}$ concentration, a deduction of 3 to 6 µg/m³ applies, which differs according to the municipality. For the 24-hour average $PM_{10}$ concentration, the number of exceedance days can be reduced by six days. This means that 41 days with 24-hour average $PM_{10}$ concentrations of 50 µg/m³ or higher are permitted every year instead of 35 days.*

*The basis for the current sea salt deduction scheme has become uncertain due to the limitations of the measurements and model calculations (Hoogerbrugge et al. 2005). To reduce these uncertainties, a separate study in BOP was devoted to sea salt (Manders et al. 2009).*

concentrations, in accordance with the projections, the number of exceedance days will also decline. However, this reduction of the effect of the sea salt deduction on the exceedance days has not been included in the current projections.

- Due to national and international policy, it appears that the $PM_{10}$ concentrations will continue to decline (see Section 5.1). According to LML measurements, the number of days on which the standard for 24-hour average $PM_{10}$ concentrations is exceeded – without the sea salt deduction – has remained below the permitted annual total of 35 days since 2008 (see www.lml.rivm.nl). According to calculations, at some locations with high local emissions due to traffic, agriculture or storage and transshipment activities, there are still more exceedance days than permitted (see Velders et al. 2009).

Based on the above insights into the contribution of sea salt to the particulate matter concentrations in the Netherlands, revision of the sea salt deduction in the Air Quality Assessment Scheme is desirable. Sodium measurements (an indicator for sea salt) from the LML, combined with model calculations, could establish the basis for an improved sea salt deduction, one which takes account of the large spatial and temporal variability in the sea salt concentration.

# 5 How effective is the policy?

- Between 1993 and 2007, the $PM_{10}$ concentrations fell by 0.7 to 1.0 µg/m³ per year on average. During this period, there was a relative concentration decline of 24 to 32 per cent. Within the large uncertainty margins, the observed concentration decline corresponded with the anthropogenic emission developments (emissions due to human activities).
- Approximately two-thirds of the decline was caused by reduced emissions of sulphur dioxide, nitrogen oxides and ammonia. The remainder of the decline (one-third) was the result of fewer primary particulates, secondary carbonaceous particulates and particulates to which water is bound. For the Netherlands, the $PM_{10}$ trend is now interpreted based on the registered anthropogenic emissions, domestic and international.
- The progression of the decline from year to year is difficult to interpret due to the large measurement uncertainty, in combination with weather-related annual variations in particulate matter, which are as high as 2.5 µg/m³. Since 2000, the rate of reduction in emissions and concentrations has declined. The trend between 2000 and 2007 was not significant.
- There are differences between the overall $PM_{10}$ trend and the trend in the anthropogenic constituents of this fraction. As a result, the benefits to health depend on which component is the best indicator for adverse health effects.
- If black smoke is used as the most important indicator for the adverse health effects of particulate matter, then the implemented policy has provided greater public health benefits than implied by the $PM_{10}$ trend alone.
- All $PM_{2.5}$ standards are probably attainable with current and planned national and European emission policy. Approximately one-third of the expected decline in the urban $PM_{2.5}$ concentration in the Netherlands between 2010 and 2020 will probably result from the reduction in primary $PM_{2.5}$ emissions.
- The target that will be imposed on the Netherlands – to reduce the average $PM_{2.5}$ concentration in the urban area between 2010 and 2020 – will depend on the measurement results in 2009, 2010 and 2011. A reduction target of 15 per cent appears likely, but 20 per cent is also possible. In the latter case, current and planned national and European emission policy will probably be insufficient.

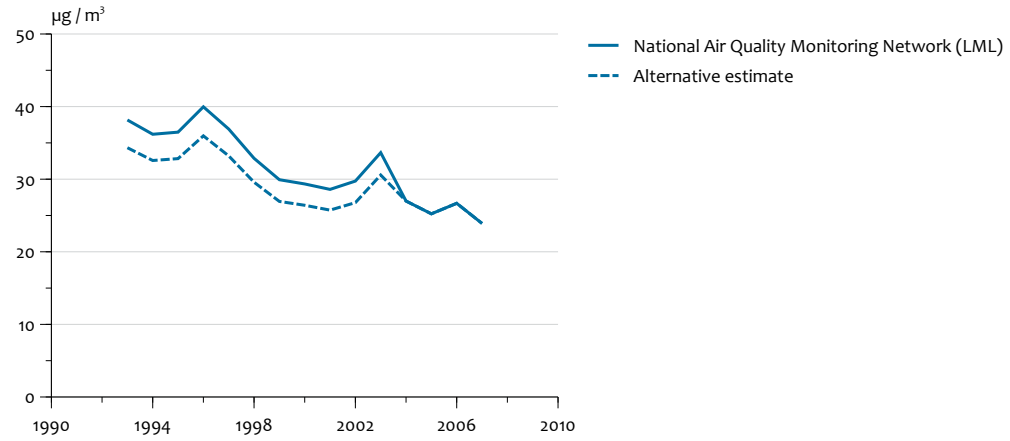
In BOP, the historical progression of the particulate matter concentrations was analysed, and future outlook studies were conducted based on implemented or planned policy for particulate matter (Matthijsen & ten Brink 2007; Matthijsen et al. 2009; Hoogerbrugge et al. 2010). In this chapter, the most important results from these studies will be reviewed based on the following questions:

- What are the trends in $PM_{10}$ and its constituents, and which anthropogenic emission reductions are responsible for these trends?
- What are the current levels of $PM_{2.5}$, and are the European norms for $PM_{2.5}$ attainable in the Netherlands?
- Based on the current trends, what can be said about the health impact?

## 5.1 Trends in $PM_{10}$ and its constituents

There are many questions about the particulate matter trend, both in the Netherlands and elsewhere in Europe. Recently

**$PM_{10}$ concentration** **Figure 5.1**




*Yearly average $PM_{10}$ concentrations for regional locations (1993-2007). The solid line shows the RIVM-reported concentrations with the changes that were implemented retroactively in 2007 (see Section 1.1); the dotted line shows an alternative (lower) estimate of the average $PM_{10}$ concentration (Hoogerbrugge et al 2010).*

published research has indicated that $PM_{10}$ concentrations in Europe are no longer falling, even though the relevant emissions are still declining. Understanding the particulate matter trend is a precondition to evaluating the effectiveness of the policy.

During the past two decades, much air-quality policy has been implemented at the European and national levels. This policy has various themes, but has certainly affected the $PM_{10}$ and $PM_{2.5}$ concentrations in the Netherlands. In addition, specific constituents of particulate matter have been sharply reduced. $PM_{10}$ has many constituents with different source characteristics. The trend in the $PM_{10}$ concentrations is determined by factors such as the emission, dispersion, chemistry and removal of these constituents. A trend in $PM_{10}$ can be attributed to policy measures at the local and national scales, and especially at the European scale.

The measurements of $PM_{10}$ in the Netherlands at regional locations (Figure 5.1) show that particulate matter concentrations have been declining since 1993. However, there are large variations from year to year. High concentrations occur during dry (and warm) summers and dry (and cold) winters. The measurement data for 2004 are very uncertain. This is shown with the alternative – lower – estimate. The progression of the alternative estimate between 1993 and 2007 appears to be more plausible relative to the progression at the urban and street locations, and falls within the large uncertainty margin of the measurements for 2004 (see Hoogerbrugge et al. 2010). Based on the bandwidth that is formed by the two series, the $PM_{10}$ concentrations fell by 0.7 to 1.0 µg/m³ per year between 1993 and 2007.

Particulate matter components that are largely of anthropogenic origin have also shown a declining trend in the Netherlands (Figure 5.2).

The trends in the various anthropogenic constituents (Figure 5.2) correspond in terms of their behaviour with the trend in $PM_{10}$: the largest decline took place between 1990 and 2000, after which the decline levelled out or even stabilised. However, in order to distinguish a statistically significant stabilisation in the concentration from a decline, at least 10 years of measurements are required. After 2000, only black smoke measurements continued to show a continuous decline.

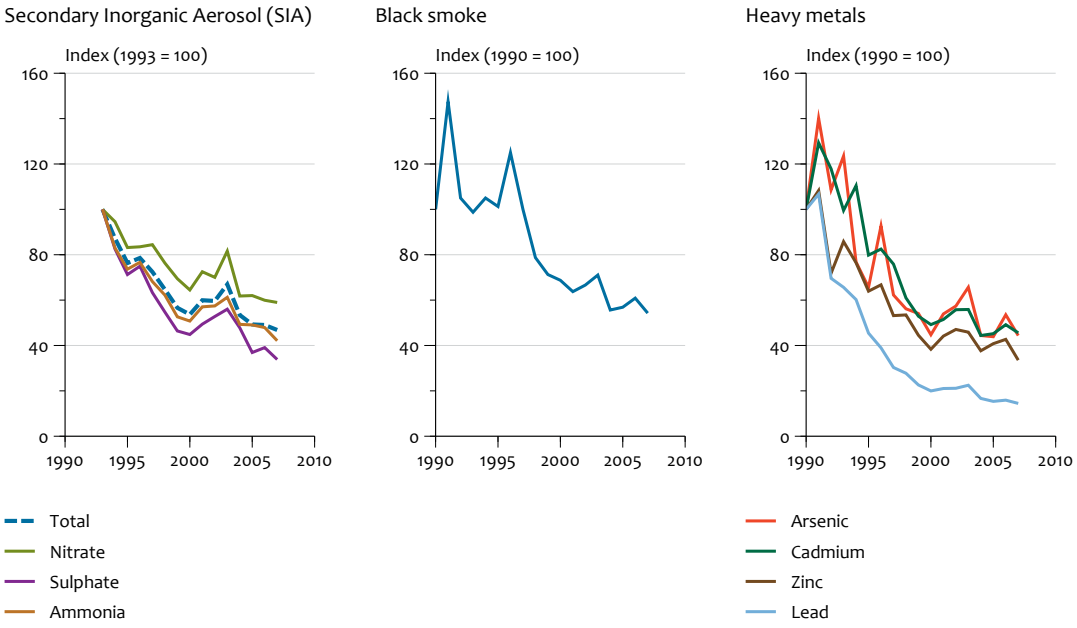
If black smoke is used as an indicator for combustion aerosol, then the contribution of combustion aerosol to particulate matter has also declined proportionally. This decline is the result of the application of a many emission-limiting measures, such as the introduction of emission standards for vehicles and the use of soot filters on diesel-powered vehicles. In urban areas, the trend in combustion aerosol based on black smoke measurements is difficult to ascertain due to the effect of local contributions. At the LML measurement location in Vlaardingen, the black smoke concentration appears to be stable or perhaps even increasing slightly (Beijk et al. 2009), while the black smoke concentration at other locations in the Rijnmond area declined sharply relative to the region (Keuken & ten Brink 2009).

Anthropogenic emission changes in Europe, and even elsewhere, probably led to a slight decline in the $PM_{10}$ concentration in the period 1993–2007. Calculations with the OPS model, combined with measurements, were used to estimate the effect of the anthropogenic emission changes on the $PM_{10}$ concentrations between 1993 and 2007 (Figure 5.3). Relevant emissions for $PM_{10}$ are sulphur dioxide, nitrogen oxides, ammonia, volatile organic compounds and primary particulates.

The analysis took account of all anthropogenic contributions that could be determined based on registered anthropogenic emissions. These are:

- The contribution from particulate matter originating from sulphur dioxide, nitrogen oxides and ammonia (SIA). The

**Regional background concentration of particulate matter components in the Netherlands** **Figure 5.2**



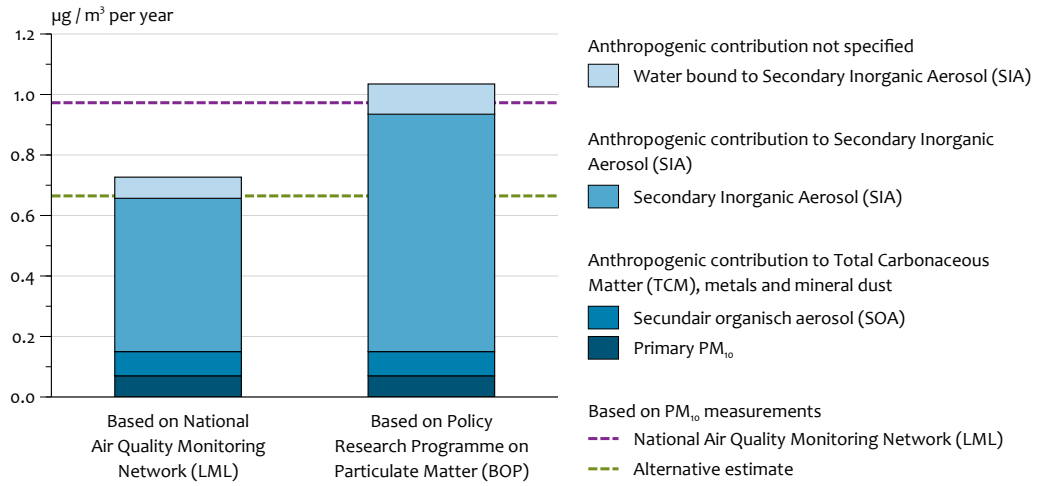

*Average trend of a number of particulate matter constituents at regional locations in the Netherlands, based on measurements from the National Air Quality Monitoring Network: secondary inorganic aerosol on particulate matter[1]: nitrate, sulphate and ammonia, and the total; black smoke; heavy metals on particulate matter[1]: arsenic, cadmium, zinc and lead (Hoogerbrugge et al. 2010).*

BOP measurement data showed that the SIA contribution to $PM_{10}$ was about 50 per cent higher than the estimate based on historical measurements and calculations (see Section 2.2.2). As a result, the implemented policy on the emissions of sulphur dioxide, nitrogen oxides and ammonia probably contributed more to the decline in $PM_{10}$ concentrations that was calculated previously. The composition of the $PM_{10}$ trend based on the components has therefore been split into two possibilities:

- The left bar in Figure 5.3 shows the contribution of the change in the SIA concentration to the decline in the $PM_{10}$ concentration between 1993 and 2007, according to the historical SIA measurements from the LML.
- The right bar in Figure 5.3 shows the maximum contribution of the change in the SIA concentration to the decline in the $PM_{10}$ concentration between 1993 and 2007. This larger contribution is possibly the result of a 50 per cent larger proportion of SIA in $PM_{10}$, as indicated by the BOP measurements. The possible additional decline in the SIA concentrations between 1993 and 2007 is estimated to be a maximum of 0.3 µg/m³ per year.
- The contribution of water to the $PM_{10}$ concentration decline. Water is a component of particulate matter. As hygroscopic particulate matter components decline, the associated quantity of water also declines. The contribution from SIA dominates the total quantity of hygroscopic components in particulate matter. According to estimates, the quantity of water that is bound to SIA has declined in proportion to the SIA contribution, to 0.1 µg/m³ per year.
- The contribution from anthropogenic secondary organic aerosol to the $PM_{10}$ concentration decline. Between 1990 and 2007, anthropogenic emissions of volatile organic compounds declined by approximately 60 per cent. As a result, the contribution of anthropogenic secondary organic aerosol to $PM_{10}$ probably declined as well – according to estimates by slightly less than 0.1 µg/m³ per year.
- The contribution of primary $PM_{10}$ to the $PM_{10}$ concentration decline. Primary $PM_{10}$ contains all directly emitted particulates and has many constituents. The most important constituents are elementary and organic carbon, metals and mineral dust. The registered primary $PM_{10}$ emissions are uncertain for the carbonaceous constituents and contain only a small part of the anthropogenic dust: the primary $PM_{10}$ emissions from road surface wear. The remaining anthropogenic dust emissions, including emissions from dust resuspended by vehicles on roads, are extremely uncertain and have not been included in the contribution of primary $PM_{10}$ in Figure 5.3.

The observed trend in the $PM_{10}$ concentration and its most important constituents corresponds with the registered developments in the emissions. But there seems to be a contradiction: 'concentration stable, emissions declining slightly'. However, there are too many uncertainties to support the apparent contradiction. The uncertainty in the measured $PM_{10}$ trends is large, not only due to the strong effect of weather conditions (and other aspects) on the concentrations, but also because the measurement uncertainty for particulate matter is high (20 per cent). This measurement uncertainty is inherent to the European reference method for determining particulate matter concentrations; it is unlikely that it can be reduced any further with technical modifications. However, by increasing the

**Change in background concentration $PM_{10}$, 1993 – 2007** **Figure 5.3**




*$PM_{10}$ concentration change (µg/m³ per year) between 1993 and 2007, dotted lines based on the $PM_{10}$ measurements (see Figure 5.1) and anthropogenic components: secondary inorganic aerosol, primary $PM_{10}$, secondary organic aerosol and water bound to secondary inorganic aerosol (Hoogerbrugge et al. 2010).*

number of measurements, the average concentration can be ascertained with more certainty.

## 5.2 $PM_{2.5}$ and the new standards

In 2008, the European air-quality directive established standards for the finer fraction of particulate matter ($PM_{2.5}$). To comply with the European standards for $PM_{2.5}$, the Netherlands is modifying the policy, the monitoring methods and the models for particulate matter. BOP supported this process with two outlook studies. The first study supported the European policy process for establishing the standards (Matthijsen & ten Brink 2007), and the second study supported Dutch policy by determining the feasibility of the various agreed standards for $PM_{2.5}$ in the Netherlands and the EU (Matthijsen et al. 2009).

### The progression of $PM_{2.5}$ concentrations in the Netherlands

In the Netherlands, $PM_{2.5}$ has been measured for several years using the reference method that dates from 2005 (EN 14907: 2005). Figure 5.4 shows the progression of the yearly average concentrations at the regional, urban and street locations since 2006. The average and the range are shown for each type of location.

Concentrations are stable or appear to decline slightly. However, the database is still too small to determine a statistically significant trend. $PM_{2.5}$ concentrations in 2009 had a relatively broad range of 11 to 20 µg/m³ (average 15 µg/m³). Generally, we see that the levels at the various types of locations differ only slightly on average. These small differences also appeared in the BOP measurements (see Chapter 2).

The $PM_{2.5}$ concentrations in the urban area (at urban background locations) are important because they establish the basis for the average exposure index (AEI) – the 3-year average concentration at a number of urban locations. The Netherlands has measured this concentration at 12 locations (see Matthijsen et al. 2009 for the location of the 12 urban locations). In 2008, two European standards were established for the AEI (see textbox Summary of standards for $PM_{10}$ and $PM_{2.5}$ in Chapter 1). The average concentration in 2009 was 17.4 µg/m³. The average for 2007, 2008 and 2009 was 17.8 µg/m³, and is consequently just below 18 µg/m³. If the AEI in 2010 – the average of 2009, 2010 en 2011 – were higher than or equal to 18 µg/m³, then a 20 per cent reduction target for the AEI in 2020 (relative to 2010) would apply; this would be the case if the average $PM_{2.5}$ concentration in the urban area in 2010 and 2011 was 18.3 µg/m³ or higher. In case of lower $PM_{2.5}$ concentrations in 2010 and 2011, a reduction target of 15 per cent would apply. It will therefore be a close call about which target reduction will be imposed on the Netherlands. For that matter, a difference of only 0.3 µg $PM_{2.5}$/m³ (18.0 vs. 18.3 µg/m³) cannot be measured with significance. The Association of European Air Quality Reference Laboratories (AQUILA 2009) has questioned the significance of an AEI reduction and how this must be dealt with when complying with the $PM_{2.5}$ standards.

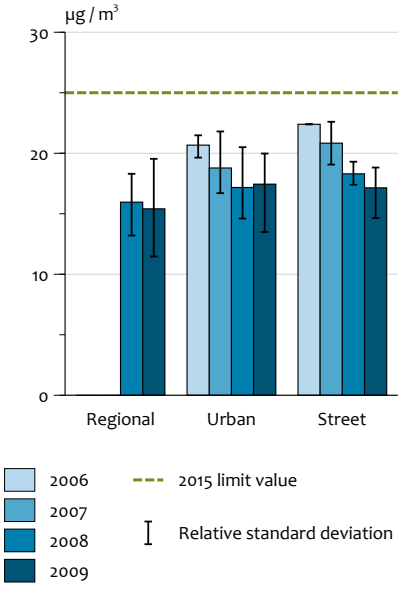
### Is compliance with the $PM_{2.5}$ standards feasible in the Netherlands?

Assuming average weather conditions, with current and planned national and European policy, it is likely that all target values and limit values for $PM_{2.5}$ can be attained on time. Consequently, the $PM_{2.5}$ standards do not appear to be more stringent than the existing $PM_{10}$ limit values. But the uncertainties are great. Consequently, the possibility of a very limited number of exceedances of the 25 µg/m³ target value near busy streets in 2010 cannot be excluded.

Adverse weather conditions could also lead to more exceedances in 2015 – possibly of the 25 µg/m³ limit value as well. The exposure reduction target (ERT) is a target value for the national average $PM_{2.5}$ concentration at urban background

**$PM_{2.5}$ measurements** **Figure 5.4**

Concentration




Number of measurement stations

| | Regional | Urban | Street |
|---|---|---|---|
| 2006 | - | 3 | 1 |
| 2007 | - | 4 | 2 |
| 2008 | 9 | 10 | 6 |
| 2009 | 8 | 11 | 9 |

*Yearly average $PM_{2.5}$ concentration (µg/m³) for regional, urban and street locations based on measurements taken according to the reference method from 2006 through 2009. Data sources: National Air Quality Monitoring Network (LML), DCMR Environmental Protection Agency, Municipal Health Service of Amsterdam.*

locations between 2010 and 2020. Theoretically, an ERT of 15 per cent can be measured with just enough significance, given $PM_{2.5}$ monitoring setup in the study. The ERT appears to be the most difficult $PM_{2.5}$ standard to comply with.

At present, the ERT is still a target value (a best efforts obligation), but during the evaluation of the directive in 2013, the European Commission could propose to make the exposure reduction legally binding for urban areas (a results obligation).

Figure 5.5 shows that an ERT of 15 per cent will probably be attained with current and proposed national and European policy. To attain a 20 per cent decrease would probably require additional national and European policy. The uncertainty in the estimates takes account of a potentially greater affect on the particulate matter concentration of measures to reduce the emissions of sulphur dioxide, nitrogen oxides and ammonia.

The concentration declines in Figure 5.5 are the result of calculations based on a number of emission scenarios used in the study to prepare for the revision of the National Emissions Ceilings Directive, according to Amann et al. (2008).

## 5.3 Effects of implemented policy and health impacts

The implemented policy on particulate air pollution covers most anthropogenic sources of particulate matter (particulates and precursor gases). Moreover, policy on other environmental themes, such as counteracting global warming, also affects particulate matter (see Section 6.2; Viewing particulate matter policy in a broader context).

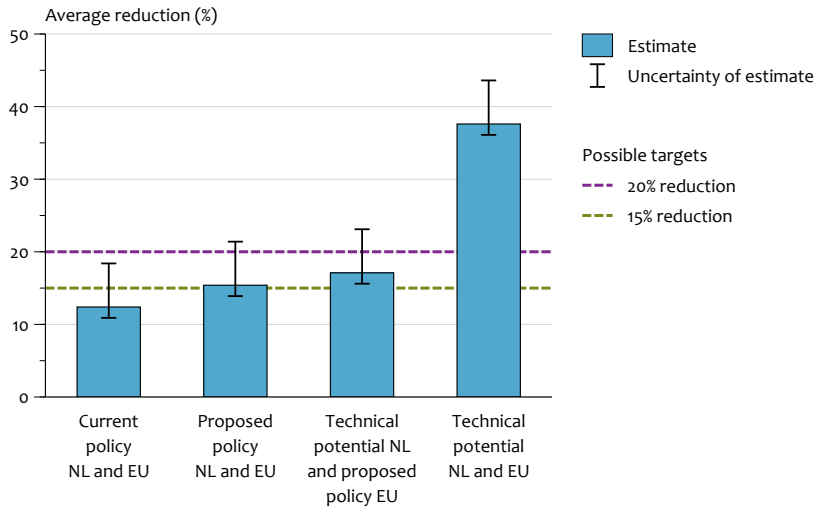
Between 1993 and 2007, $PM_{10}$ concentrations at regional locations decreased by 24 to 32 per cent. Large differences have been measured between the trends of $PM_{10}$ as a whole and the trends of its anthropogenic constituents. As a result, the expected health benefits depend strongly on which constituent is the best indicator for adverse health effects.

Approximately two-thirds of the decline in the $PM_{10}$ concentration is the result of reductions in the emissions of sulphur dioxide, nitrogen oxides and ammonia. According to current insights, the health benefits from these reductions are limited.

Emission reductions of primary particulates would be expected to provide more health benefits, because an important part of this fraction consists of carbonaceous components and metals. The decline in the concentration of primary $PM_{10}$ emissions from registered sources contributed approximately 10 per cent to the decline in $PM_{10}$ between 1993 and 2007. During this period, according to calculations, the primary $PM_{10}$ fraction at regional locations declined by 16 per cent on average. No measurements are available to support these calculated declines. If the concentration of primary $PM_{10}$ emissions is assumed to have the greatest health impact, then the calculated improvement since 1993 is probably smaller than when the entire $PM_{10}$ concentration is used as a health impact indicator.

Exhaust emissions from road traffic have declined by approximately 50 per cent in the Netherlands, corresponding with the average concentration of black smoke at regional locations since 1990. If black smoke is used as the health impact indicator for particulate matter, then the implemented

**Reduction in urban $PM_{10}$ concentration, 2010 – 2020** **Figure 5.5**




*Decline of the $PM_{2.5}$ concentration in urban areas – the average exposure index – between 2010 and 2020, relative to 2010, with various policies (Matthijsen et al. 2009; PBL 2009a)*

policy possibly provided greater health benefits than indicated by the $PM_{10}$ concentration as a whole.

At a more local scale, the composition of $PM_{10}$ differs between locations, and there are trend differences as well. Depending on the location, large declines and small increases have both been measured. One conclusion (PBL 2009a) was that the exposure to combustion aerosol is much more unevenly distributed across the population than exposure to $PM_{10}$ as whole. Residents in the large cities and those who live near motorways and busy streets are exposed to the highest concentrations of combustion aerosol.

If such local differences have a significant health impact – and this appears to be the case – then $PM_{10}$ and $PM_{2.5}$ by themselves are inadequate for monitoring the health effects of policy measures. In that case, more knowledge about the fractions with the greatest health impact is required. Moreover, a supplementary indicator is required in practice to monitor measures that are relevant to health, but which are not sufficiently expressed by the $PM_{10}$ or $PM_{2.5}$ concentrations alone. Elementary carbon and black smoke, both candidates for a supplementary indicator, are still the topic of discussion and additional research.

**Note**

[1] The particle size in the sampled particulate matter is uncertain: for the secondary inorganic aerosol concentrations it concerns particles with a diameter of approximately 3-4 µm; for the heavy metal concentrations, this is approximately 7-8 µm.

# 6 What is next?

- The recent insights into the magnitude of the anthropogenic contributions to particulate matter have consequences for particulate matter diagnoses and projections. To quantify these consequences, steps must be taken to revise the models.
- Revision of the Air Quality Assessment Scheme for the sea salt deduction during exceedances of the limit values for $PM_{10}$ is desirable. The contribution of sea salt to $PM_{10}$ concentrations has turned out to be lower on average than was previously assumed.
- A supplementary indicator is required to monitor the effects of particulate matter policy measures that are relevant to human health, but which are not measurably expressed in the $PM_{10}$ or $PM_{2.5}$ concentrations. Elementary carbon and black smoke, both candidates for a supplementary indicator, are still the topic of discussion and additional research.
- Measurements, models and data on emissions relating to the contribution of carbon compounds and carbonaceous particulate matter must be improved, especially for urban areas.

BOP has provided a clearer focus on particulate matter. As intended, this has reduced a number of uncertainties. Besides providing new understanding of the composition of particulate matter, existing knowledge has been refined. To use the acquired knowledge for implementing air-quality policy, additional analysis and research are required. At the same time, after the completion of BOP, major questions about particulate matter remain unanswered. The policy agenda will be driven by European legislation on air quality and climate change.

## 6.1 Most important actions and open questions

The contributions of various sources of particulate matter have been clarified by BOP, but there are still uncertainties in specific areas that hamper adequate policy choices about air quality. Given the new insights from BOP, the most important actions related to the open questions about particulate matter are the following:

- Revision of models.
  The measurements and models with which diagnoses and projections for particulate matter concentrations are made – for the GCN – must be revised to take account of the underestimated constituents of particulate matter. For this purpose, a number of steps must be taken. These steps are being partly addressed in the subsequent research programme. No obstacles are foreseen a priori to the implementation of this programme.

First of all, follow-up research is required to determine the extent to which anthropogenic source contributions, which have apparently been disregarded or underestimated in the calculations for air quality reporting, can be included in these reports.

Secondly, measurements are required to determine why the secondary particulate matter from sulphur dioxide, nitrogen oxides and ammonia has been underestimated until now. This information would allow any trend in secondary inorganic aerosols to be determined more precisely.

Thirdly, models must be adapted to improve the description of anthropogenic contributions to particulate matter. This not only concerns secondary particulate matter from sulphur dioxide, nitrogen oxides and ammonia, but also from volatile carbon compounds and the dust and water that are bound to the anthropogenic constituents.

Finally, the anthropogenic contribution to the $PM_{10}$ and $PM_{2.5}$ concentrations of various economic sectors in the Netherlands and abroad must be recalculated.

After this knowledge is acquired, it will become possible for the Netherlands and other countries in the region to determine an optimal strategy to reduce the major contribution to particulate matter from ammonium nitrate and ammonium sulphate on days with elevated $PM_{10}$ concentrations. The contribution from Dutch sources to

elevated $PM_{10}$ concentrations, for instance exceedances of the limit value for 24-hour average concentrations, is higher than the contribution to average $PM_{10}$ concentrations. The contribution from other countries dominates the yearly average. Quantification of the contribution from Dutch sources, especially during periods with elevated PM concentrations, would therefore be useful information, which is also amenable to research.

Specification of the constituents of primary emissions of anthropogenic particulates is required in order to compare models with composition measurements. The contributions of sea salt and mineral dust have been mapped out with models and can be implemented more completely in the policy instruments. Improved understanding of the semi-volatile constituents of particulate matter will reduce the uncertainties in particulate matter measurements and modelling.

- Revision of the sea salt deduction.
  The contribution of sea salt to $PM_{10}$ concentrations has turned out to be lower on average than was previously assumed. It is therefore desirable to revise the Air Quality Assessment Scheme (*Regeling Beoordeling Luchtkwaliteit*) for the sea salt deduction during exceedances of the limit values of $PM_{10}$.

  Sodium measurements from the LML (an indicator for sea salt), combined with model calculations, could establish the basis for an improved sea salt deduction, one which also takes account of the large spatial and temporal variability in the sea salt concentration. When $PM_{10}$ concentrations decline, the effect of the sea salt deduction on the number of days with exceedances of the $PM_{10}$ limit value will become smaller; $PM_{10}$ projections must take account of this situation. The current measurement scheme stipulates that six days can be deducted (due to the contribution of sea salt) when determining the number of days on which the limit value for 24-hour average $PM_{10}$ concentrations is exceeded. However, the number of days on which sea salt actually causes the exceedance varies annually, and six days is probably too high for most years.
- *Mapping out the local contribution from wood combustion.*
  For health protection, it would be useful to revise projections for the direct emissions from wood combustion, linked to an outlook study of simple measures to limit the emissions. As part of the improvement in the sustainability of energy sources, it is conceivable that the contribution to particulate matter from wood combustion – including that from efficient, small-scale furnaces for energy production – will increase. An obvious strategy is to link up with countries such as Italy, Germany and Austria, which have much more experience with wood combustion and measures to limit the emissions from this source.

  Another obvious step is to make a broader scenario study of local particulate matter situations related to wood combustion with models, emission estimates and measures. The local contribution from wood combustion to $PM_{10}$ has been studied with initial measurements. The contribution turned out to be seasonal and differed per location, sometimes by a factor of 10. Moreover the contribution from wood combustion has a relevant health impact. Other important local contributions have already been mapped out more clearly with models and measurements, such as the contributions from storage and transshipment facilities and from traffic.

- Carbonaceous particulate matter.
  The knowledge about volatile organic compounds and carbonaceous particulate matter in the atmosphere is still very limited. Carbon is an important component of combustion aerosol, which is assumed to be relevant to health. The revision of the national emission ceilings at the European and UN level stipulates a relative emissions ceiling for primary $PM_{2.5}$. Carbonaceous particulate matter is a major component of this fraction.

  The knowledge about the contribution of carbon compounds to particulate matter is still very limited. Measurements and model calculations are still affected by fundamental problems. For example, it is unclear which constituents of carbonaceous particulate matter originate from human activities in the Netherlands and which originate from abroad. To more clearly determine the reduction potential for carbonaceous particulate matter and volatile organic compounds, a further adaptation of the emission and modelling instruments is required. At present, the models are unable to verify the European agreements with sufficient precision. Measurements can help in this process, especially by providing more understanding about determining source contributions at various spatial scales.

- Particulate matter and health impact
  There is no doubt that particulate matter has adverse health effects. However, there are still many unanswered questions about specific aspects of particulate matter and human health. According to current insights, the adverse health effects are more strongly associated with combustion aerosol and metals on particles than with other constituents, such as sea salt and secondary inorganic aerosol. $PM_{10}$ and $PM_{2.5}$ concentrations by themselves have limited suitability as indicators for monitoring measures that focus on the more health-relevant constituents. A supplementary indicator that is suitable for this purpose is needed to support *smart policy* on both particulate matter and health. This is especially important for local policy, where it is possible to reduce the exposure to combustion aerosol by means of focused measures and spatial planning.

- Concentration contribution from the urban area
  Based on BOP research, the contribution of various particulate matter constituents in the urban area relative to the rural area appears to be rather uncertain, especially regarding dust, organic carbon and secondary inorganic aerosol. This hampers adequate policy choices regarding particulate matter in the urban area. Consequently, research is needed to clarify the elevated $PM_{10}$ and $PM_{2.5}$ in the urban area relative to the region in terms of source contributions and particle size. Improved understanding of the particulate matter in the urban area has a high priority,

due to the combination of the large urban population and the frequently elevated concentrations.

- Obligation to measure constituents of $PM_{2.5}$
  The European air-quality directive (EU 2008b) obligates Member States to measure the total $PM_{2.5}$ mass concentration and the concentrations of individual chemical constituents of this fraction at a regional background location, and to report these measurements as yearly averages. The Netherlands has not yet complied with this obligation. The Cabauw measurement location, with its long history as an atmospheric research location, appears to be suitable for this purpose. It is advisable to measure the constituents of both $PM_{10}$ and $PM_{2.5}$, because the norms for $PM_{10}$ are more stringent than those for $PM_{2.5}$. The provisions in the European directive make it possible to comply with this obligation in cooperation with other countries.

Particulate matter is one of the many indicators for air quality. It also plays a role in climate change. An integrated approach to these themes is still a relatively new field. More coherence and improved knowledge about the connections between the systems is required, also to make the correct policy decisions in a timely fashion.

## 6.2 Particulate matter policy in a broader context

BOP has provided information about a number of topics, including the constituents and sources of $PM_{10}$ and $PM_{2.5}$, with the intention of preventing exceedances of limit values. However, particulate matter is only one of the environmental themes in the air-quality dossier.

The substances that contribute directly or indirectly to particulate matter concentrations also play a role in other themes in the air-quality dossier. The atmosphere is dominated by processes with varying temporal and spatial scales. This brings various policy fields together, which in practice are sometimes isolated from each other by the scale on which an air-quality problem manifests itself and by the administrative scale on which measures are taken.

This section provides a qualitative sketch of the effectiveness of the emission-reducing measures. The underlying question is: how effective are the various measures for reducing particulate matter when the following environmental policy objectives are viewed in an integral fashion?

- Complying with the limit values for $PM_{10}$ and $PM_{2.5}$
- Reducing adverse health effects associated with particulate matter
- Reducing the deposition of acidifying and eutrophying substances on wildlife habitats
- Reducing the impact of ozone on health and vegetation
- Counteracting climate change

The objectives 'complying with the limit values for $PM_{10}$ and $PM_{2.5}$' and 'reducing adverse health effects associated with particulate matter' are purposefully separated here. This is because compliance with the standards for $PM_{10}$ and $PM_{2.5}$ does not necessarily serve the underlying objective of the standards, which concerns health benefits (see Section 5.3 and PBL 2009a).

### Legislation that impacts particulate matter

The emissions of substances that affect the particulate matter concentrations in the Netherlands are being approached with a wide range of policy instruments that are applied at various scales. This concerns the emissions of primary particulate matter (elementary carbon, organic carbon and other particles, including metals), and emissions of precursor gases that can lead to the formation of secondary particulate matter (sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds).

The National Emissions Ceilings Directive and the Gothenburg Protocol are international agreements with national emission targets for sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds. Reducing primary particulate matter emissions and complying with these emission targets is achieved by means of emission-reducing measures applied to vehicles, products and production processes (such as Euro standards for vehicles, IPPC, LCP, HM protocol – see textbox Legislation relevant to particulate matter).

Besides implementing the international legislation, the Netherlands, by means of the National Air Quality Cooperation Programme (NSL), has taken additional measures for reducing the particulate matter concentration, including subsidies and financial incentives for cleaner vehicles and the particulate matter action plan for industry.

At the local scale, many different measures are being taken that can lead to a reduction of particulate matter concentrations. These measures focus primarily on the direct emissions of particulate matter, and are also included in the NSL. In addition, measures such as barriers or changes in traffic circulation have been taken that can lead to reduced particulate matter concentrations. The introduction of environmental zones aims to reduce traffic emissions that are assumed to be relevant to health; only vehicles that comply with a specific Euro standard are permitted in these zones.

At the European scale, an integrated approach using national emission ceilings is already being taken towards air pollution and its effects on ecosystems (EU 2001b; UN-ECE 1999). In this approach, the most cost-effective national emission ceilings are determined, while taking account of various endpoints of air-quality policy (health and nature): this is known as the multi-pollutant/multi-effect approach (GAINS/RAINS; Wagner et al. 2006, 2007).

However, the individual substances are not weighted according to their health impacts. This is because the formal standpoint of the World Health Organization is observed: all constituents of $PM_{10}$ and $PM_{2.5}$ are considered to be equally harmful (WHO 2000). At the same time, the WHO acknowledges that there are differences (WHO 2006). But the health impact of reducing individual constituents is unknown, which also plays a role. The effects on climate change of the changes in the emissions are not yet considered when determining the priorities in air-quality policy.

### The effect of air-quality policy on $PM_{10}$ between 1993 and 2007

Between 1993 and 2007, $PM_{10}$ concentrations at regional locations fell by between 8 µg/m³ and 12 µg/m³ (an approximate decline of 24 to 32 per cent). Approximately two-thirds of this decline could be attributed to emission reductions of the precursor gasses sulphur dioxide, nitrogen oxides and ammonia. The emission reductions in Europe played a role in this process, but those in the USA probably made a small contribution as well. The reduction in the emissions of primary particulate matter probably contributed no more than 10 per cent to the decline in the $PM_{10}$ concentrations. The remainder of the decline is probably the result of the concentration decline of secondary particulate matter that is formed from volatile organic compounds, combined with a reduction in the average quantity of water bound to particulate matter. Water is a component of particulate matter, and is bound to organic and inorganic salts. These contributions to particulate matter declined between 1993 and 2007, along with the quantity of water.

The concentrations at regional locations of some particulate matter constituents have fallen more sharply than the $PM_{10}$

## *Legislation relevant to particulate matter*

*With the aim of limiting the emissions of air pollutants, various binding international agreements have been made by the European Union or as part of the United Nations 1979 Convention on Long Range Transboundary Air Pollution and as part of the United Nations 1973 MARPOL Convention and amendments to prevent pollution from ships. The following legislation is most relevant to particulate matter:*

### National Emissions Ceilings Directive

*The National Emission Ceilings Directive (EU 2001b) assigns ceilings to each EU Member State for the total emissions of sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds, which take effect in 2010. If the emissions in the Netherlands in 2010 exceed the stipulated levels, a non-compliance procedure will be initiated by the European Commission and a severe penalty can be imposed.*

### Gothenburg Protocol

*The Gothenburg Protocol (UN-ECE 1999) is part of a step-by-step process from the United Nations 1979 Convention on Long Range Transboundary Air Pollution. This Protocol aims to protect health and ecosystems by bringing levels of deposition and concentrations of pollutants to below critical levels. For this long-term objective, the Protocol establishes national emission ceilings and emission limit values that must be attained in 2010 as interim targets. Criteria for the national emission targets in the protocol are cost-effectiveness, equal rights to clean air (equity), and progress towards the long-term environmental targets.*

### National emissions ceilings for 2020

*The Gothenburg Protocol and the National Emissions Ceilings Directive have emission ceilings for 2020. Relative emission ceilings are foreseen for primary $PM_{2.5}$. In 2011 a decision is expected about the revised version of the Protocol. This decision will give shape in part to the revision process for the National Emissions Ceilings Directive.*

### IPPC Directive

*The Integrated Pollution Prevention and Control (IPPC) directive (EU 2008a) aims at integrated prevention and control of pollution. The IPPC directive obligates the EU Member States to regulate the emissions of environmental pollutants to water, atmosphere and soil (including measures for wastes) from large polluting firms and from the intensive animal husbandry sector. This is done by means of an integrated permit, which must be based on the best available technologies. A revised version of the IPPC directive is currently in the final phase of the decision process. The revised directive, which integrates seven different directives, is expected to be approved in 2010.*

### LCP directive

*The revised Large Combustion Plant directive (EU 2001a) aims to reduce acidification, ozone at ground level and particulate matter by means of a Europe-wide limitation of the emissions of specific air pollutants by large combustion plants.*

### Euro standards for vehicles

*Since the beginning of the 1990s, European emission standards for vehicles have been in force. Emission standards have become increasingly stringent. As a result, the emissions of carbon monoxide, nitrogen oxides, volatile organic compounds and particulate matter from vehicles have steadily decreased. There are separate standards for passenger vehicles, delivery vans and trucks, and a distinction is also made according to fuel: diesel or petrol. In 2009, the Euro 5 standard (EU 1999b) for passenger vehicles went into effect.*

### Heavy Metals Protocol

*The Heavy Metals Protocol (UN-ECE 1998) aims to control the anthropogenic emissions of heavy metals that are subject to long-distance transboundary transport and which can have significant adverse effects on public health or the environment. The Protocol provides for the reduction of the total annual atmospheric emissions of cadmium, lead and mercury to below the levels in 1990, as well as the application of the best available technologies for product management. The Protocol requires lead in petrol to be phased out. A side effect of the Heavy Metals Protocol is that other primary particulate emissions are automatically reduced along with the heavy metals (UN-ECE 2007).*

### MARPOL

*In 2008 the Marine Pollution convention was revised concerning emission reductions of sulphur dioxide and nitrogen oxides from marine shipping (IMO 2008). For the Netherlands, which is a coastal country with busy shipping routes, the intended emission reductions are important. These emission reductions will be taken into account when the national emission ceilings for 2020 are determined.*

concentration as a whole. For example, between 1990 and 2007, the black smoke concentration declined by more than 50 per cent due to the implemented policy, and the lead concentration on particles declined by more than 80 per cent due to the phasing out of leaded petrol.

### Limit values for particulate matter and health impacts

The legal obligation to comply with the air quality standards before the deadlines and the policy pressure that has resulted from this obligation in the Netherlands – partly due to the economic consequences of suspension of development plans – are the driving forces for national and local particulate matter policy.

The most effective policy for complying with the particulate matter limits has been the measures to reduce sulphur dioxide, nitrogen oxides and ammonia. These precursor gasses contribute approximately 40 per cent to the mass of particulate matter in the air, and on days with exceedances of the $PM_{10}$ 24-hour standard, that contribution is even larger. Unfortunately, few health benefits are expected from the reduction of sulphur dioxide, nitrogen oxides and ammonia. Measures that limit the emissions of combustion aerosol are thought to be more relevant for health. But these measures contribute little to the reduction of the total particulate matter concentration. Measures to reduce combustion aerosol continue to have the greatest impact near busy streets and motorways.

In the past, the emission reduction of sulphur dioxide and nitrogen oxides was often linked automatically to a reduction of the emissions of elementary and organic carbon. In this way, two things were accomplished at once: lower particulate matter concentrations and benefits to health. However, due to the rapidly increasing energy efficiency of combustion motors and the transition from oil to gas in many sectors in the Netherlands, this link has almost disappeared. As a result, $PM_{10}$ and $PM_{2.5}$ are no longer optimal indicators at the local scale for the adverse health effects of particulates.

A supplementary indicator is therefore being urged internationally. Besides $PM_{10}$ and $PM_{2.5}$ as leading indicators

## *Particulate matter in relation to other environmental themes*

### Particulate matter and deposition on the living environment

*Although measures to reduce the emissions of sulphur dioxide, nitrogen oxides and ammonia probably result in few health benefits, they do significantly improve the environmental conditions for natural habitats due to a reduction of acidifying and eutrophying deposition.*

*In the Netherlands, wildlife habitats are still under severe pressure from acidification and eutrophication due to high emissions of nitrogen oxides and ammonia (PBL 2009a, 2009b). In the atmosphere, nitrogen oxides and ammonia can be converted into nitrogen-bearing particles. For other countries near the Netherlands, the tentative emission ceilings for 2020 that are proposed for the revision of the National Emissions Ceilings Directive and the Gothenburg Protocol are at most only slightly lower than the emission ceilings for 2010 (Amann et al. 2008). Consequently, the national emission ceilings for 2020 are not expected to result in a major decline in the particulate matter concentration from nitrogen-bearing particles. The nitrogen deposition in the Netherlands will also not decline very much after 2010.*

*The direct particulate emission of heavy metals such as arsenic and cadmium also affects the quality of nature reserves (Hettelingh et al. 2010). Heavy metals are deposited on these areas via atmospheric deposition. In addition to the deposition route, heavy metals can also be inhaled by people, leading to adverse health effects. Generally speaking, the current atmospheric concentrations of heavy metals in the Netherlands are below the established limit values.*

### Particulate matter and ozone at ground level

*Particulate matter and ozone at ground level are often strongly correlated on days with summer smog. In recent decades, peak concentrations of ozone have declined, but the background concentrations now appear to be climbing due to the emissions of nitrogen oxides and volatile organic compounds from outside Europe. The policy to reduce ozone concentrations focuses on reducing emissions of nitrogen oxides and volatile organic compounds, which also leads to lower particulate matter concentrations. Moreover, the secondary particulate matter that is formed in the air from volatile organic compounds is assumed to have an adverse health impact (Biswas et al. 2009).*

### Particulate matter and climate

*The relationship between particulate matter policy and climate change is characterised by benefits and detriments. Depending on its composition, particulate matter can have either a cooling or warming effect (IPCC 2007; Shindell et al. 2009; Jacob & Winner 2009; SEPA 2009).*

*Particles that are formed from sulphur dioxide have a significant cooling effect, which is also true to lesser extent for nitrogen oxides and ammonia. The strong reduction in the emissions of these gases in Europe and the USA – 20 per cent or more in recent decades – has possibly played an important role in the warming in Europe during the past 30 years, which has been stronger than the global average (Philipona et al. 2009). In the future, more far-reaching emission reductions of sulphur dioxide, nitrogen oxides and ammonia could lead to world-wide to an even stronger warming effect (Raes & Seinfeld 2009). However, the emission reduction of elementary carbon (soot) benefits both sides: it leads to health benefits and reduced warming. Indirect climate effects, both positive and negative, can also be expected from emission changes that lead to reduced particulate matter levels. The magnitude of the indirect effects is very uncertain. On balance, however, climate policy leads to less air pollution (Daniels et al. 2008; Hammingh et al. 2008).*

**Policy agenda on particulate matter for 2010 - 2020** **Table 6.1**

| | |
|---|---|
| *2010* | – Limit values for $PM_{10}$ in force since 2005, with derogation until June 2011<br>– Target value for $PM_{2.5}$ in force since 2008 (25 µg/m³)<br>– European Commission makes proposals for sustainability criteria for the production of biofuels (has consequences for air-quality policy).<br>– Enactment of revised IPPC directive.<br>– Ongoing process on the revision of national emission targets (Gothenburg Protocol, UN ECE; National Emissions Ceilings Directive, EU): a new aspect is a relative emission ceiling for the emission of primary $PM_{2.5}$.<br>– Measurement obligation for $PM_{2.5}$ concentration beginning no later than 2009. According to the directive, the Netherlands has an adequate number of $PM_{2.5}$ measurement points.<br>– Measurement obligation, composition of $PM_{2.5}$.<br>– Euro-V emission standard for delivery vans goes into force. |
| *2011* | – June 2011, the Netherlands must comply with $PM_{10}$ limit values everywhere in the country.<br>– Possible decision about revised Gothenburg-Protocol. |
| *2012* | – Possible decision about revised National Emissions Ceilings Directive. |
| *2013* | – Revision of European air-quality directive; provisions in the directive on $PM_{2.5}$ and $PM_{10}$ will be reviewed. Will the exposure reduction target become a limit value? Will the Phase-2 indicative value of 20 µg/m³ for 2020 become a limit value? |
| *2014* | – Euro-VI emission standards for passenger cars go into force. |
| *2015* | – 1 January 2015, $PM_{2.5}$ limit values from EU (2008b) go into force.<br>– Euro-VI emission standards for diesel vans go into force. |
| *2020* | – EU Member States must comply with the national emission ceilings for sulphur dioxide, nitrogen oxides, ammonia, volatile organic compounds and $PM_{2.5}$ as stipulated in the revised NEC directive and the Gothenburg Protocol.<br>– EU Member States must comply with international exposure reduction targets for the average $PM_{2.5}$ concentration in urban areas between 2010 en 2020. In the Netherlands, the exposure reduction target will be 15% or 20%. |

for the health impact of particulate air pollution, an indicator is required that is more suitable for determining the effectiveness of health-relevant measures. The World Health Organization has investigated black smoke as an indicator for soot emitted by traffic (WHO 2003), but the question of the most suitable indicator is still a topic of discussion and further research.

The air-quality policy that influences particulate matter concentrations in the Netherlands also affects other environmental themes (see textbox Particulate matter in relation to other environmental themes). Measures that reduce emissions of sulphur dioxide, nitrogen oxides and ammonia are not only effective for attaining $PM_{10}$ and $PM_{2.5}$ limit values, but they also reduce the environmental deposition of nitrogen and sulphur. Ozone concentrations at ground level benefit from particulate matter policy that focuses on nitrogen oxides and volatile organic compounds. For climate change – and to a lesser extent for ozone in the living environment – there are advantages and disadvantages. In particular, the policy to reduce sulphur dioxide has not only led to a reduction of particulate matter concentrations, but at the same time to increased global warming. Reducing black particulates (soot) is probably good for both the climate and health.

During the next 10 years, various topics in the air-quality dossier will certainly require support. To be prepared for this, the national policy instruments will have to be modified so that the effects of measures on air-quality, climate change and deposition can be explored and assessed in an integrated fashion.

## 6.3 Policy agenda for particulate matter 2010 - 2020

Policy that is relevant to the concentrations of particulate matter in the Netherlands is implemented at various scales. European directives are often the driving force behind this policy. At the global level, the amendments to the MARPOL convention on marine emissions are important for the Netherlands. Table 6.1 lists the most important items on the policy agenda for particulate matter.

# Appendix Uncertainties

The conclusions of the BOP research should be seen in the context of a number of uncertainties. This concerns the uncertainty in the various types of measurements and calculations and uncertainty about the representativeness of the measurements. In short, the uncertainty concerns the scope of the conclusions in the various BOP reports. The uncertainty in the calculations was discussed in Schaap et al. (2009) and Velders et al. (2009).

## Measurement uncertainties

The particulate matter measurements in the BOP measurement campaign were made according to the European reference methods for $PM_{10}$ and $PM_{2.5}$ (EN 12341; 1999 and EN 14907; 2005). These standards define particulate matter in relation to the limit values. The approach in the BOP measurement campaign was therefore optimal for relating the composition of particulate matter to compliance or non-compliance with the European limit values for $PM_{10}$ and $PM_{2.5}$.

The section below addresses the uncertainties for each particulate matter constituent that are the result of the uncertainty in the composition measurements. The uncertainties described here are discussed in greater detail in Schaap et al. (2010). The uncertainty margins here are a lower limit. For each particulate matter constituent, the margins become significantly larger if other technologies and methods for measuring the concentration of particulate matter constituents that are permitted by the European directive are taken into account. For example, the contribution of the sea salt constituent to particulate matter can be determined in various ways from the measurement data. For sea salt, the margins that result from the various calculation methods can be as high as 30 to 40 per cent. Similar margins apply to other constituents.

The average uncertainty in the total $PM_{10}$ and $PM_{2.5}$ concentration is approximately 20 per cent. Measurements performed according to the reference methods allow room for systematic deviations relative to atmospheric concentrations. This is related to the fact that a significant proportion of particulate matter consists of semi-volatile compounds: ammonium nitrate, semi-volatile carbonaceous particulate matter and water. The evaporation and condensation in these fractions leads to deviations relative to atmospheric concentrations. For the analysis of the measurements, however, this did not have any major consequences because similar systematic deviations occur when determining the particulate matter constituents in the total $PM_{10}$ and $PM_{2.5}$ concentrations.

During the BOP 2007-2008 measurement campaign, the reference method became more stringent (see textbox Measuring $PM_{10}$ and $PM_{2.5}$ in the National Air Quality Monitoring Network in Section 1.1). Consequently, after February 2008 the $PM_{10}$ and $PM_{2.5}$ concentrations systematically became 1 to 2 µg/m³ lower. The uncertainty in the BOP measurement data therefore increased slightly: the uncertainty about the average contributions of the particulate matter constituents increased by several per cent.

### SIA

The uncertainty in measuring the contribution of secondary inorganic aerosol to particulate matter is approximately µg/m³ (10-15 per cent); this measurement is therefore relatively precise. However, there were probably systematic differences between the measured SIA concentrations and those in the atmosphere. Measurements conducted in the Netherlands according to the European reference method, as used in BOP, would have underestimated the contribution of SIA – and therefore the particulate matter concentration in the air as well – by several µg/m³ as an annual average. This was shown by a comparison between SIA measurements according to a method where volatilisation does not play a role (MARGA) and according to the European reference method, where volatilisation does play a role (Weijers et al. 2010).

### TCM

There are major uncertainties surrounding concentrations of carbon compounds and carbonaceous particulate matter. The total of elementary carbon (EC) and organic carbon (OC), with an uncertainty of approximately 1 µg/m³ (10-15 per cent), is the most robust measure for carbon. The uncertainty in the TCM concentrations is greater, and is estimated at about 25 per cent. The relative proportion of EC and OC is much more uncertain. This is an undesirable situation, because carbonaceous particulate matter, especially in the form of EC but also OC, is thought to have a greater health impact

than other 'major' particulate matter components (SIA and sea salt). Volatile organic compounds, which can lead to the formation of TCM, are also a source of ozone at ground level and play various roles in climate change, especially indirect roles as particles.

The uncertainty is due to the volatile character of organic carbon. As a result, the emissions and concentration measurements are both uncertain. In addition, the EC/OC ratio is uncertain. The relative contribution of EC to EC+OC turned out to vary between 20 and 60 per cent, depending on the measurement method. Moreover, most methods only measure elementary carbon, so that the contribution from the other elements in the carbon compounds must be estimated. For this purpose, the EC+OC mass is multiplied by 1.3 in accordance with the American EPA approach. This factor is relatively uncertain; according to the literature, it can vary between 1.2 and 2.0. Agreements are currently being made at the European level by the CEN Working Group 35 on EC and OC in particulate matter about a standard method for measuring EC and OC (Kuhlbusch et al. 2009).

The measurements showed that the $PM_{10}$ and $PM_{2.5}$ concentrations, which were sampled according to the reference method, contained about 0.7 µg/m³ gaseous carbon compounds on average (ten Brink et al. 2009). According to the European reference method, this component was defined as particulate matter and was therefore included in the concentration contribution of TCM. This component is not expected to decline if anthropogenic VOC emissions decline.

The modelling of the contribution of TCM to particulate matter is not yet sufficiently developed, partly due to the uncertainties in the measurements and emissions. As a result, the quantitative effect on the particulate matter concentrations of measures to reduce VOC emissions and emissions of primary EC and OC particulates is uncertain.

### Mineral dust

The uncertainty in measurements of mineral dust is at least 30 per cent, according to estimates. The measured silicon and aluminium concentrations are the best way to determine the contribution of mineral dust to particulate matter. This is because silicon and aluminium are the dominant components in mineral dust (clay with sand) from Northwest Europe. However, the measurement uncertainty in the concentration measurements, especially of silicon, are large (30 per cent). Not all of the silicon (quartz) can be measured with the standard method. A correction is therefore made for the missing silicon. This correction introduced the greatest uncertainty.

### Metals

The uncertainty in the metals component is relatively small (10 per cent). A correction has been applied to include the other elements chemically bound to the metals, such as oxygen. This correction introduces uncertainty, because it is uncertain how much of these other elements must be included.

### Sea salt

The uncertainty in the contribution of sea salt to particulate matter is estimated at approximately 15 per cent. The contribution of sea salt to particulate matter is determined by using the ratios of the various elements that are found in fresh sea salt, such as sodium and chloride. However, when sea salt particles remain in the air, they undergo chemical reactions with other components, and the sodium/chloride ratio changes. Therefore, various methods are used to calculate the contribution of sea salt to particulate matter from the measured concentration of sea salt elements. Schaap et al. (2010) discussed the consequences of this calculation method for the sea salt contribution.

## Representativeness and consistency of the measurement data

The BOP measurement campaigns provided a snapshot of the particulate matter situation between 2007 and 2008. Particulate matter has many different and variable sources in the Netherlands and abroad. As a result, the representativeness of a measurement campaign to approximate the particulate matter situation in the Netherlands is limited by the number of measurement points and the measurement frequency. BOP measured the total particulate matter concentration and its composition at six locations, including three regional locations, one urban location and two street locations. The composition and concentration of particulate matter and the differences according to type of location can therefore only provide an indication for the rest of the Netherlands during the BOP research. The concentration differences of $PM_{10}$ and $PM_{2.5}$ between the urban location and the various regional locations turned out to be small on average: 0-3 µg/m³ ($PM_{10}$) and 0-1 µg/m³ ($PM_{2.5}$). These differences were smaller or of the same magnitude as the mutual concentration differences between the three regional locations. The measurement design was therefore inadequate to determine the significance of the contribution to particulate matter from urban sources. However, the urban contribution could be determined for particulate matter constituents that are largely contained in $PM_{10}$ and are less prevalent in $PM_{2.5}$, such as sea salt, mineral dust and metals.

To investigate the consistency of the BOP measurement data with other data about particulate matter, the various BOP studies were expanded as much as possible with a European component. The measurement results were compared with existing measurements in the Netherlands and the rest of Europe and with calculations from the LOTOS-EUROS model. This chemical transport model describes the air quality for all of Europe and is used to describe the entire air quality chain, from emission to concentration and removal. In addition, the BOP measurement data was analysed with a statistical method in order to identify sources (Schaap et al. 2010). This independent review of the measurement data provided additional support for the conclusions about the sources of particulate matter.

In the Netherlands, a previous particulate matter measurement campaign was conducted in 1998/1999: the *Bronstof* study (Visser et al. 2001; Buringh & Opperhuizen 2002). Since then, the knowledge about the various health impacts of particulate matter and about the source

contributions has changed and improved. The *Bronstof* study can be characterised as the first major study of the sources of particulate matter in the Netherlands. However, primarily for technical reasons, the results of that research are not directly comparable with the BOP research. For example, at that time there were no agreements about reference methods for sampling $PM_{10}$ and $PM_{2.5}$. Relative to the *Bronstof* results, the contribution from natural sources according to the BOP estimate (4 – 5 µg/m³) is at least 40 per cent smaller (estimates according to Visser et al. (2001)).

In Belgium (VVM 2009) and Germany (IUTA 2004; Quass et al. 2004, 2009) measurement campaigns also took place to clarify the constituents and sources of particulate matter. The recent campaign in Germany took place partly at the same time as the campaign in the Netherlands. The study in Belgium took place in 2006 and 2007. When drawing conclusions based on Dutch data, the results of the studies in Belgium and Germany were considered where possible (see Weijers et al. 2010; Denier van der Gon et al. 2010; Schaap et al. 2010). Striking differences were found between the mineral dust contribution in Belgium, Germany and the Netherlands. In Belgium, the mineral dust contribution to $PM_{10}$ turned out to be about twice as high on average as that in the Netherlands. In Germany, the mineral dust concentrations, although slightly higher, were reasonably comparable with those in the Netherlands. Until now, there has been no conclusive explanation for this situation. The difference is probably related to the large margins in the mineral dust concentrations due to the various methods that are used to derive the mineral dust contribution from the measured concentrations of elements and due to differences between measurement locations. Differences were also found in contributions from elementary carbon and organic carbon to the increase in urban particulate matter concentrations relative to the regional background. These differences are probably the result of differences between the measurement methods that are used to determine the concentrations of elementary and organic carbon.

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### PBL press releases

Agreement on the 'new' European Air Quality Directive
http://www.pbl.nl/nl/dossiers/fijn-stof/content/Akkoord_over_nieuwe_Europese_Richtlijn_Luchtkwaliteit.html
The Netherlands will probably attain the EU standards for $PM_{2.5}$
http://www.pbl.nl/nl/nieuws/nieuwsberichten/2009/20091119-Nederland-gaat-EU-normen-voor-kleine-deeltjes-fijn-stof-waarschijnlijk-halen.html

### Workshops

$PM_{2,5}$ in the Netherlands, Informative workshop for stakeholders in relation to the introduction of the European standards for $PM_{2,5}$ in 2008, Utrecht, 18 January 2008, 13.30 - 16.30.
Measurements and Modelling of $PM_{2,5}$ in Europe, International workshop on $PM_{2,5}$, Bilthoven, 23 - 24 April 2009.
Programme: http://www.pbl.nl/en/dossiers/Transboundaryairpollution/content/Programme-Workshop-Measurements-and-Modelling-of-PM2-5-in-Europe.html
Presentations: http://www.pbl.nl/en/dossiers/Transboundaryairpollution/content/Presentations-Workshop-Measurements-and-Modelling-of-PM2-5-in-Europe.html
Report: http://www.mnp.nl/bibliotheek/rapporten/500099017.pdf
BOP, the Netherlands research programme on particulate matter, VVM Workshop, Bilthoven, 11 December 2009. Programme and presentations: http://www.vvm.info/main.php?id=43.

# Colophon


### Final responsibility
PBL Netherlands Environmental Assessment Agency

### With contributions from
Ernie Weijers (ECN), Harry ten Brink (ECN), René Otjes (ECN), Aline Kraai, (ECN), Emel Sahan (ECN), Marita Voogt (TNO), Astrid Manders (TNO), Elise Hendriks (TNO), Martijn Schaap (TNO), Menno Keuken (TNO), Hugo Denier van der Gon (TNO), Arjo Segers (TNO), Magdalena Jozwicka (TNO), Mhtenkeni Gondwe (TNO), Frits van Arkel (RIVM), Ronald Hoogerbrugge (RIVM), Dennis Mooibroek (RIVM), Lan Nguyen (RIVM), Margreet van Zanten (RIVM), Ferd Sauter (RIVM), Frank de Leeuw (PBL), Guus Velders (PBL), Hans van Jaarsveld (PBL), Winand Smeets (PBL), Benno Jimmink (PBL), Jelmer Cnossen (PBL), Dave de Jonge (GGD Amsterdam), Sef van den Elshout (DCMR).

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### Project assistant
Karin van Doremalen

### Text editing
Ester Goddijn

### Figure editing
Marian Abels, Raymond de Niet, Jan de Ruiter, Joost van Wollingen, Wilco de Vries

### Design
Uitgeverij RIVM

**Latest research shows that particulate matter is largely anthropogenic in origin**

Recent research has clarified a number of facts about particulate matter in the atmosphere and has overturned a number of assumptions. For example, it is now clear that a much greater proportion of particulate matter is caused by human (anthropogenic) activities than was previously assumed. Measures that focus on reducing anthropogenic emissions are therefore potentially more effective for reducing particulate matter concentrations. However, measurements of $PM_{10}$ and $PM_{2.5}$ concentrations – the standard size fractions of particulate matter – have been shown to be less effective than assumed as indicators of local health impact. This applies especially to assessments of measures to control emissions from health-relevant combustion processes, such as traffic exhaust emissions. Further research must be conducted to determine whether specific components of particulate matter, such as black smoke or elementary carbon, would be more suitable indicators.

These are several conclusions from the 'Policy Research Programme on Particulate Matter' (BOP) of the Netherlands Environmental Assessment Agency, the Energy Research Centre of the Netherlands, the National Institute for Public Health and the Environment and TNO. This programme took place from 2007 to 2009 and was funded by the Ministry of Housing, Spatial Planning and the Environment (VROM). BOP aimed to reduce the number of policy dilemmas that occur when enforcing European standards for particulate matter. The research has led to various new insights into the composition and sources of particulate matter and into the progress in particulate matter policy. These insights have led to policy recommendations, and the results of the various components of the research have been published in 15 reports. The present report summarises the results of the entire research programme and its policy implications.

This study is a BOP publication produced under the auspices of PBL.

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

PO Box 303, 3720 AH Bilthoven, The Netherlands