Trends in Particulate Matter

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

PBL Netherlands Environmental Assessment Agency

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TNO

BOP – report

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ECN Energy research Centre of the Netherlands
PBL Netherlands Environmental Assessment Agency
TNO Built Environment and Geosciences
RIVM National Institute for Public Health and the Environment

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Rapport in het kort

De trend in fijn stof (PM_{10}) en zijn belangrijkste samenstellende componenten zijn niet in tegenspraak met de geregistreerde ontwikkelingen in de emissies. Hoewel er op het oog een contradictie is; "concentratie stabiel, emissie (licht) dalend" blijkt bij nadere studie dat het onderscheidend vermogen niet hoog genoeg is om dit daadwerkelijk te concluderen.

De onzekerheid in de gemeten trends is relatief groot vooral door de sterke invloed van de weersomstandigheden op de concentraties maar ook doordat het meten van fijn stof moeilijk is.

In deze studie is op verschillende manieren gecorrigeerd voor dit effect van meteorologie waardoor de onzekerheid gereduceerd is. Zowel voor de gemeten concentraties als voor de emissies is geconstateerd dat de afname in de periode 1993-2000 duidelijk groter is (typisch 2-4 % per jaar) dan de afname in de periode 2000-2008 (0-2 % per jaar). Een gedetailleerd onderzoek naar verkeersemissies, belangrijk in verband met gezondheidseffecten, laat zien dat de daling als gevolg van de invoering van schonere motoren grotendeels verdwijnt door stijging van het aantal gereden kilometers en het zwaarder worden van de voertuigen.

Het blijkt dat de intrinsieke onzekerheid waarmee trends over korte periodes, minder dan 10 jaar, kunnen worden vastgesteld te groot is om de huidige ontwikkelingen eenduidig te interpreteren. Na correcties voor weersinvloeden kunnen systematische dalingen sneller worden aangetoond.

Summary

The trend in particulate matter (PM_{10}) and its most important constituents does not contradict the developments in registered emissions. At first glance there appears a contradiction; 'concentrations stable and emissions (slightly) decreasing', however, after a detailed study the statistical power appears to be insufficient to prove the contradiction.

The uncertainty about the trend in the measured results is relatively large due to meteorological influences and also due to measurement uncertainties. In this study, several methods are presented for correcting meteorological effects that reduce uncertainties.

For both emissions and measurements, the decreasing trends over the 1993-2000 period were obviously larger (typically 2 to 4%, annually) than over the period between 2000 and 2008 (0 to 2%, annually). A detailed study on traffic emissions, important with respect to health effects, has shown that any reductions due to cleaner car engines are cancelled out by the increase in total distance driven and the increasing weight of vehicles.

The study has also shown that the uncertainty in the estimation of PM trends over short periods of time (<10 years) is too large for an unambiguous interpretation of the developments. After correction for meteorological influences, decreasing trends can be proven more early.

Contents

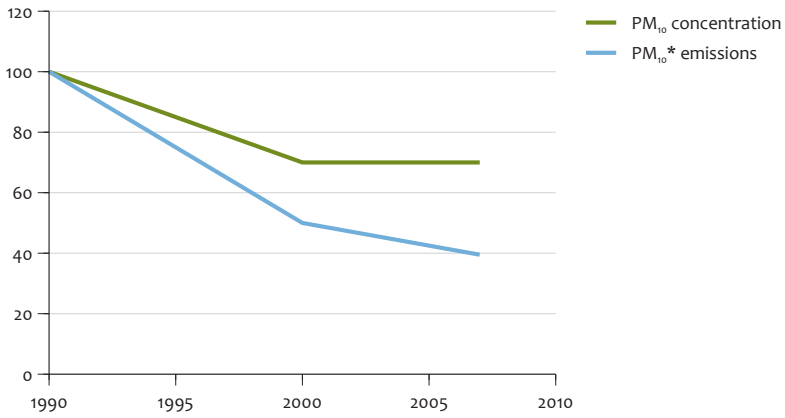
Executive summary

This report gives an analysis of the trend in particulate matter in the Netherlands over the 1990-2007 period. The purpose of this study is to improve our understanding of the observed trends in PM_{10} and other particulate substances. Our understanding of the PM_{10} trend was challenged because, since 2000, PM_{10} concentrations have appeared to be stabilising. This could not be easily understood in terms of changes in the registered emissions relevant to PM_{10} concentrations in the Netherlands. These emissions have in fact continued to decrease since 2000, although at a slower rate than in the nineties (Figure S.1).

Main conclusions

- All trends in anthropogenic PM components and their emissions between 1990 and 2007 show a general similarity: decreases took mainly place in the nineties; after 2000, trends have levelled off. Between 1993 and 2007, the trend in average annual concentrations of PM_{10} at rural locations in the Netherlands was 0.7 to 1.0 µg/m³ per year. This trend was found to be in keeping with the known changes in the emissions of registered anthropogenic sources relevant to PM_{10}, in spite of the apparent discrepancy between concentrations and emissions.
- Two thirds of the decrease in PM_{10} concentrations between 1993 and 2007 was attributed to reductions in anthropogenic emissions of sulphur dioxide, nitrogen oxides and ammonia. The remaining one third, was attributed in equal amounts to primary particles, particle-bound water and secondary organic aerosol.
- Reported decreases in anthropogenic emissions relevant to PM_{10}, since about 2000, have not led to a significant downward trend in measured rural PM_{10} concentrations. The absence of a significant trend was found to be largely due to fluctuating weather conditions and uncertainties in the measurements.
- Fluctuating weather conditions cause large variations in year-to-year PM_{10} concentrations, with a standard deviation of about 2.5 µg/m³. Correction of average annual PM_{10} concentrations for these fluctuations provides a better picture of the trend due to anthropogenic emission changes. Nevertheless, to detect significant, small trends of about 0.3 µg/m³ per year requires at least 10 years of measurements. Therefore, whether PM_{10} concentrations have continued to go down significantly after 2000, could probably not be detected at all in the measured PM_{10} time series that are currently available.
- The PM_{10} trend since 1990 may differ between traffic locations due to several, sometimes counteracting,

Apparent trends in the averaged annual PM_{10} concentration and in the registered emissions **Figure S.1**



*Schematic representation of the apparent trend in the average annual PM_{10} concentration and in the registered emissions relevant to PM_{10} in the Netherlands. * anthropogenic gaseous sulphur, nitrogen, carbon and primary particle emissions weighted for their contribution to PM_{10}, according to De Leeuw (2002).*

factors. Consequently, since 2000, for this type of location, both downward and upward trends in average annual PM_{10} concentrations have been found. PM_{10} at urban background locations may also be affected by local changes in traffic emissions.

PM_{10} consists of many different components with different source characteristics. Therefore, the trend in PM_{10} is determined by many factors concerning the emission, dispersion, chemistry and removal of all these components. This executive summary integrates the main results from separate chapters of this report. These separate chapters on different aspects of the trend in particulate matter addressed the following questions:

- What are the trends in concentrations of other components related to PM_{10}?
- What is the effect of fluctuating weather conditions on PM_{10} and sea salt concentrations?
- How many years of data are necessary before a trend could be detected with any significance?
- Which anthropogenic emission changes are relevant to PM_{10}, and on which scale?
- Can we explain the observed PM_{10} trends at rural locations, bottom-up, in terms of PM_{10} components and their emissions, using models and measurements?
- What are the spatial and temporal trends in traffic emissions, including from resuspension of particles?

Introduction

Particulate matter is hazardous to human health. Health studies have shown that there is a significant relationship between premature death and both short-term and long-term exposure to, especially, fine particles. Other important effects include aggravation of respiratory and lung diseases, asthma attacks, heart attacks and irregular heartbeat (WHO, 2000; 2003; 2006a; 2006b).

To reduce effects of air pollution on human health and the environment, European and national legislation has been developed to lower emissions and concentration levels of air pollutants. PM_{10} consists of many different components. Anthropogenic emissions of substances that are relevant to PM_{10} concentration levels are the directly emitted particles (primary PM_{10}), sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and non-methane volatile organic compounds (NMVOC). The gases are partly converted, chemically and/or physically, to PM_{10} in the air.

Over the last two decades, policy measures have led to decreases in the emission of these substances. In addition, emission projections for 2020 show a further decrease, except for ammonia. However, in the Netherlands and also in other European countries, ambient PM_{10} concentrations seem to have stabilised in the period between 2000 and 2007. This appears to contradict the continuing decrease in anthropogenic emissions. The question of whether or not PM_{10} concentrations are in keeping with relevant emission changes, implies a serious uncertainty of our knowledge on particulate matter. This uncertainty influences our ability to evaluate reduction policies and to assess trends in future scenarios. The latter, in turn, is essential to determine plausibility of compliance with targets as those set by the EU thematic strategy.

Trend in measurement data

- All trends in the emission of anthropogenic PM components and related substances between 1990 and 2007 shared a similarity: concentration decreases mainly took place in the nineties, and after 2000 trends levelled off.
- The trend in average annual concentrations of PM_{10} between 1993 and 2007, at rural locations in the Netherlands, was 0.7 to 1.0 µg/m³ per year. This range signifies a rather large uncertainty in the measurements, due to several reasons. This uncertainty can only partly be reduced.

Figure S.2 shows measurement data for PM_{10} in the Netherlands, for rural, urban background and traffic locations. Uncertainty in these measurements is about 20%. The figure shows that:

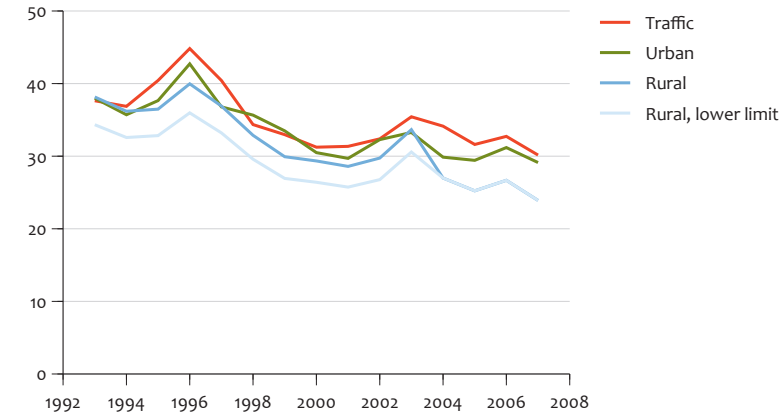
- Concentrations were decreasing during the period from 1992 until approximately 2000;
- In the 2000-2007 period, there appeared to be no significant trend;
- From year to year, large variations occurred; for instance, in 1996 and 2003 concentrations were particularly elevated;
- The concentration range for PM_{10} at rural locations, before 2004, indicates a large systematic uncertainty in the measurements due to instrumental changes.
- On average, concentration differences between the three types of locations was relatively small.

To place these observations into the context of other air pollutants, the trends for a number of anthropogenic PM constituents are shown in Figure S.3. For all components, year-to-year variations occurred, although decreases during the 1992-2000 period exceeded those in the subsequent period.

The influence of weather conditions

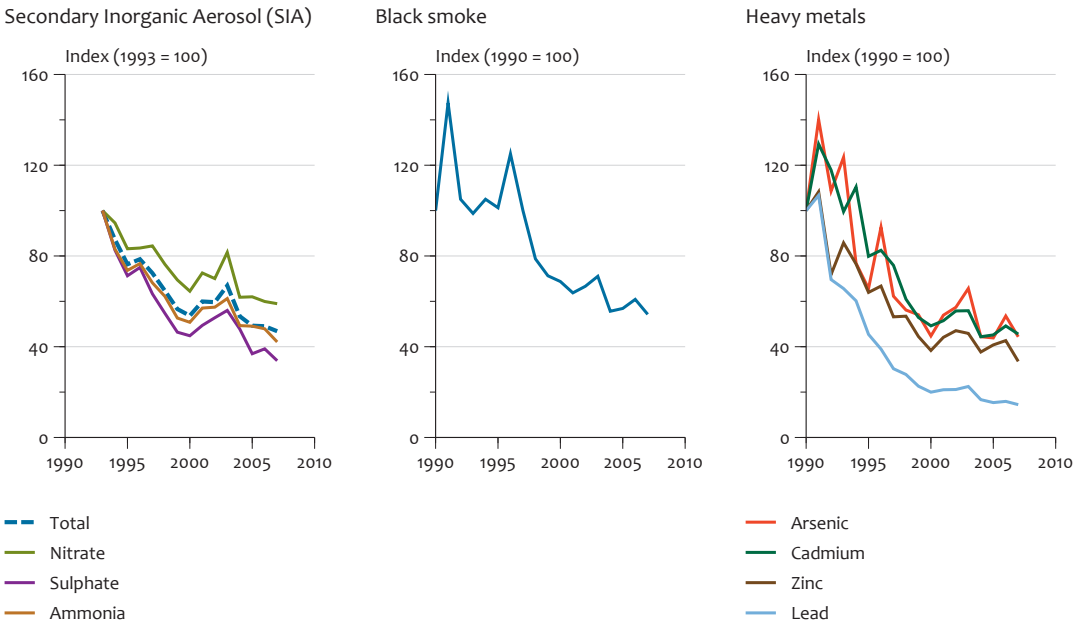
- Fluctuating weather conditions cause large year-to-year variations in PM_{10} concentrations, with a standard deviation of about 2.5 µg/m³.
- The impact of fluctuating weather conditions on PM_{10} concentrations can partly be eliminated by means of a meteorological correction derived from model results or measurements. The resulting meteorological correction differs per method used. The methods agree especially for the years with similar weather conditions throughout large parts of Europe.
- Correction of average annual PM_{10} concentrations for these fluctuations provides a better focus on the trend due to anthropogenic emission changes. Still, at least 10 years of measurements would be necessary to detect significant small trends of about 0.3 µg/m³ per year. Therefore, whether PM_{10} concentrations continued to go down after 2000 can probably not be detected with any significance from the measured PM_{10} time series that are currently available.

Measured trend in averaged annual PM_{10} concentrations in the Netherlands **Figure S.2**



Measured trend in average annual PM_{10} concentrations at rural, urban and traffic locations in the Netherlands.

Regional background concentration of particulate matter components in the Netherlands **Figure S.3**



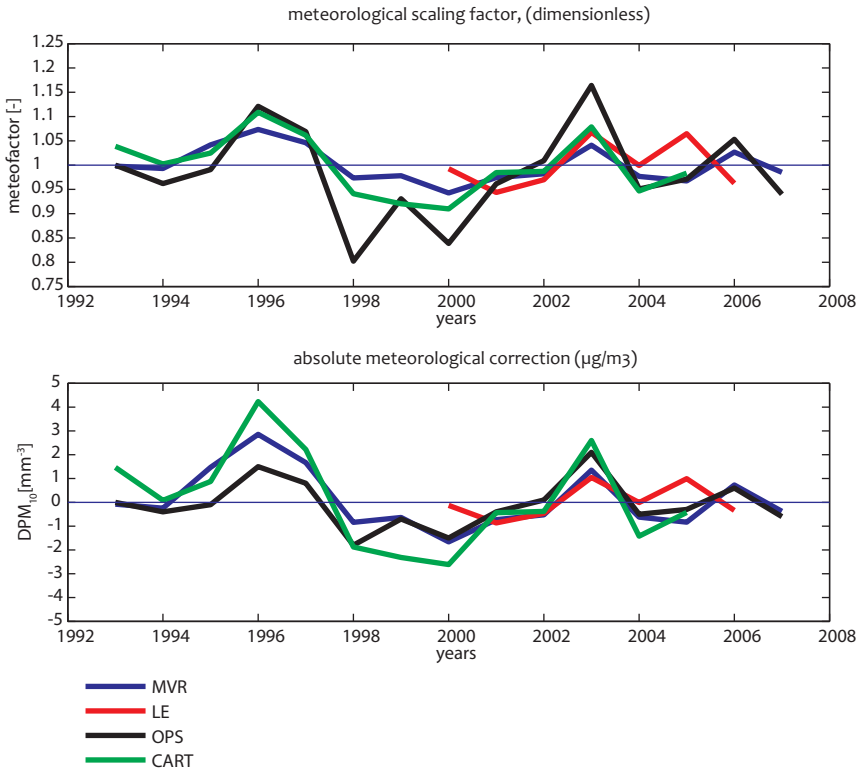
Measurement results for different anthropogenic PM constituents at rural background locations in the Netherlands: a) Secondary Inorganic Aerosol concentrations; indexed concentrations of b) black smoke and c) heavy metals in PM.

The trend in the concentration of particulate matter components show large year-to-year variations. For PM_{10}, these variations are dominated by the influence of fluctuating weather conditions, with a standard deviation of 2.5 µg/m³. Such variations hamper a trend analysis, because they can obscure relatively small anthropogenic emission changes.

Weather conditions influence PM_{10} concentrations through several different processes:

- *Origins of air masses, and mixing, formation and removal of PM*; different wind directions cause correlations with different origins of air masses, and, therefore, with different amounts and compositions of particulate pollutants and precursors. Dispersion of emitted substances affects PM_{10} concentrations, because of dilution and mixing with other air masses. Chemical processes which lead to the formation of PM_{10} depend on weather conditions; temperature plays an important role, as does the presence of precursors. Percipitation is also important because rain efficiently removes particulate matter and certain precursors from the air,
- *Human adaptation to weather*; different weather conditions can indirectly cause different anthropogenic emissions. For instance, extreme cold weather conditions lead to an increase in combustion emissions.

Meteorological corrections for average annual PM_{10} concentrations at rural background locations **Figure S.4**



Meteorological corrections for average annual PM_{10} concentrations at rural background locations, resulting from the different methods; a) relative meteorological scaling factor, (dimensionless), and b) absolute meteorological correction (µg/m³). Note that meteorological corrections derived with the LOTOS EUROS model are limited to the years between 2000 and 2006

- *Wind dependency of PM sources*; weather directly influences source strengths. For example, production of sea spray depends on wind velocity. Furthermore, strong winds over dry surfaces can induce emissions of mineral dust.

Correction of average annual PM_{10} concentrations may provide a better focus on the trend in anthropogenic emission changes. However, it is not possible to eliminate all impacts of fluctuating weather conditions. This report presents different methods for deriving a correction for the influence of the year-to-year meteorological variations on PM_{10} concentrations. The approaches were based on measurements or on model results.

Meteorological correction based on measurements

MVR.

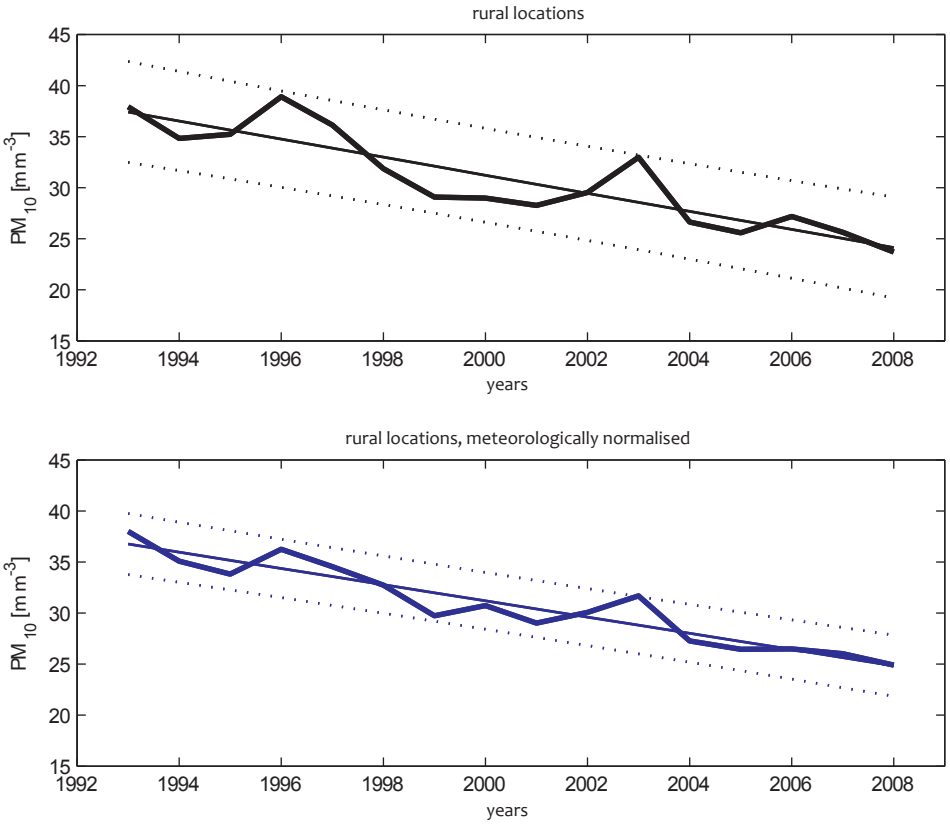
Measured average daily PM_{10} concentrations were correlated with meteorological parameters from one meteorological station in the Netherlands. This particular station provided the most representative information on large-scale (> 100 km) dispersion processes. To focus on large-scale phenomena, a statistical model was developed, using only a limited number of parameters: temperature, wind direction and precipitation. In this way, about 36% of the year-to-year variation in PM_{10} concentrations could be attributed to large-scale variations in weather conditions.

CART.

Higher percentages of weather induced variations were attributed, using a similar method. In this case, a statistical model was developed that used the relationship between average monthly variations in PM_{10} concentrations and meteorological parameters collocated with the PM_{10} measurement site. The higher percentage could be attributed to fluctuating weather conditions, because, in addition to the first approach, coincidental correlations between PM_{10} concentrations and local weather conditions might be used.

Meteorological correction based on model results

The two model-based methods both use the same input: a) a set of registered anthropogenic emissions of a given year, and b) meteorological input parameters which could vary between different years. It should be noted that the model-based approaches could only derive a meteorological correction for modelled part of PM. Model calculations, generally, only explain half to two thirds of measured PM_{10} concentrations. The non-modelled part of PM_{10} can behave differently, possibly counteracting.



Average annual PM_{10} measurement data and trend (straight solid line) for rural locations: a) without, and b) with MVR meteorological correction (measurement-based approach). The dashed lines indicate the 95% confidence limits

OPS

The OPS model is used with the meteorological data of 1990-2007 and fixed emissions for the year 2005 used for the whole period. The OPS model is a quasi-Lagrangian trajectory model with simplified chemistry and a spatial resolution of 1x1 km. The ransport of pollutants is based on statistics of meteorological parameters in the Netherlands. The OPS model approach, therefore, focuses on Dutch weather patterns.

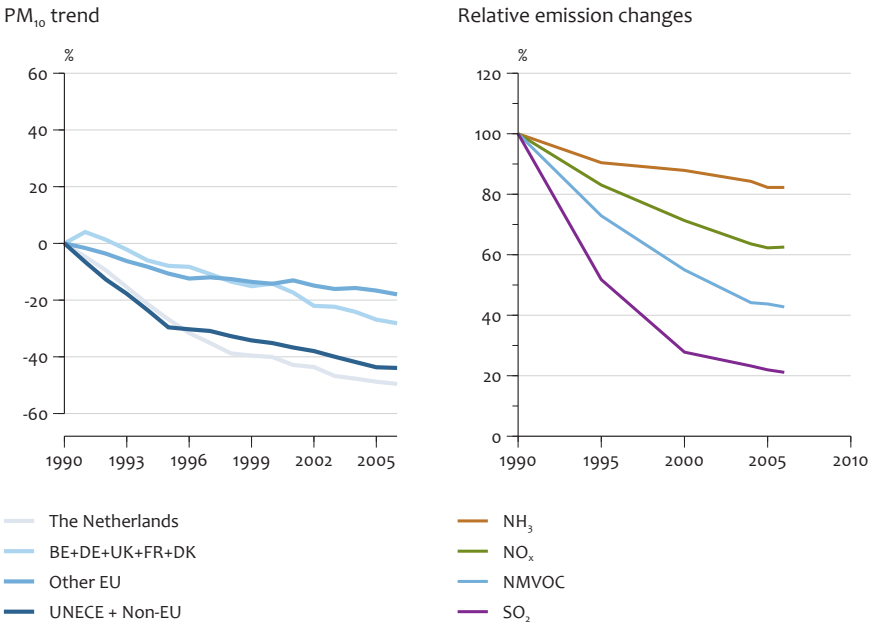
LE

The LOTOS-EUROS model, is used with the 2000-2006 meteorological data and fixed emissions for the year 2000. The LOTOS-EUROS model is a Eulerian chemistry transport model that operates on a European scale with a spatial resolution of 30x30km. The LOTOS-EUROS model approach, therefore, incorporates data on meteorological variation across Europe. The LOTOS-EUROS model describes the sea salt contribution to PM_{10}. Formation of sea salt particles depends strongly on wind velocity over sea.

Figure S.4 shows empirical meteorological corrections. Meteorologically correlated increments that were especially relevant, were found for several years, two of which (1996 and 2003) had pronounced winter and/or summer episodes in large parts of Europe.

Differences between the various meteorological corrections (Figure S.4) could not be explained straightforwardly, due to methodical differences. The discrepancies between the approaches (e.g. for 2006) appeared to be a matter of scale. The MVR, CART and OPS methods are all based on meteorological parameters of the Netherlands only, whereas the LE approach incorporates meteorological variation across Europe. Prevailing weather conditions in the Netherlands do not always show a strong correlation with weather conditions in the rest of Europe. In addition, PM_{10} concentrations, especially their secondary components, are largely determined by long-range transport of air pollutants. Consequently, meteorological variations occurring at greater distances also can have an impact on Dutch PM_{10} concentrations. The LE approach, theoretically, is the best approach for studying long-range contributions to PM_{10}. However, for studying the impact of large-scale weather fluctuations on contributions to PM_{10} from nearby emission sources, the other approaches appeared to be more valid.

PM_{10} Emission trend **Figure S.6**



(on the left) Relative emission changes in primary particles (PPM_{10}) on different scales, and (on the right) relative emission changes from source areas relevant to PM_{10} in the Netherlands, for the PM_{10} precursor gases of ammonia (NH_3), nitrogen oxides (NO_x), (non-methane) volatile organic compounds (NMVOC), and sulphur dioxide (SO_2).

The meteorological corrections shown in Figure S.4 can be applied to average annual PM_{10} concentrations. For this study, we considered the application of the MVR method; year-to-year variation in PM_{10} concentrations have reduced from 5 to about 3 µg/m³. The average influence of weather conditions on the measured average annual concentrations is shown in Figure S.5. Although the trend in PM_{10} from 2000 to 2007, after meteorological correction, inclines slightly more downward, it still could not be called statistically significant.

In theory, around 15 years of data on PM_{10} would be needed before a relatively small trend of 0.3 µg/m³ per year (around 1% per year) could be detected with statistical significance - given a standard deviation of 2.5 µg/m³ because of year-to-year variations in PM_{10}. Meteorological correction, using the MVR approach, reduces the year-to-year variation in PM_{10} concentrations, thus also reducing the number of years needed to detect a trend with any significance, by about 30%. Therefore, meteorological correction of average annual PM_{10} concentrations gives a better view of the trend that is caused by anthropogenic emission changes. Nevertheless, there are still atleast 10 years of measurements required for small trends of about 0.3 µg/m³ per year to be detected with statistical significance.

Trend in emission data

Between 1990 and 2000, large emission reductions relevant to PM_{10} in the Netherlands took place. After 2000, relevant emissions continued to go down, but at a significantly smaller rate. The reduction after 2000 seems to contradict with the apparent lack of statistical significant trend in the measured concentrations.

We also studied the apparent mismatch between trends in concentrations and in emissions from the emission perspective. Emissions are the drivers of concentrations, if emissions decrease, concentrations should follow suit. For this report, we investigated the trends in primary PM_{10} emissions and in emissions of precursor gases in secondary PM_{10}.

Trends in data on primary PM_{10} emissions were reviewed on a variety of scales. For analysis on the European scale, we defined four groups of countries, encompassing the Netherlands, its neighbouring countries, other EU countries, and non-EU European countries. Both reported European and Dutch PM_{10} emissions showed reductions in the 1990-2006 period. After 2000, annual reductions were still occurring, but were weaker (Figure S.6). A similar general pattern was found for the emission reduction in secondary precursor gases. We focussed on countries or areas that contribute three quarters or more to the Dutch secondary PM_{10} concentration levels, by emitting SO_2, NO_x and NH_3. These emissions come from the Netherlands, France, Belgium, Germany, the North Sea and the United Kingdom.

Emission reductions in primary PM_{10}, between 1990 and 2006, were of the order of 1 to 2%, per year, for the Netherlands, which is in keeping with emission trends of neighbouring countries. A similar conclusion was drawn from looking at reported data on ambient concentrations (figure not shown). Hence, the apparent contradiction between trends in PM_{10} measurements and emissions occurs not just in the Netherlands, but is a European-wide phenomenon.

Its understanding or solution should certainly include the European scale.

Primary PM_{10} emissions decreased between 1990 and 2006, by 1.5 to 3%, per year, on different scales. For the 2000-2006 period, a smaller but still clear annual trend of 0.7 to 2% remained. Precursor gases relevant to secondary PM_{10} in the Netherlands decreased between 1990 and 2006, by 1 to 5%, per year: 5% (SO_2), 4% (NMVOC), 2% (NO_x) and 1% (NH_3). In the 2000-2006 period, emission rates continued to decrease, relative to 1990, by 1 to 2%, per year: 1% (SO_2), 2% (NMVOC), 1% (NO_x) and 1% (NH_3).

Between 1990 and 2000, large emission reductions relevant to PM_{10} took place in the Netherlands. After 2000, relevant emissions continued to go down, but at a significantly smaller rate. However, from emission trends relevant to PM_{10} in the Netherlands, no evidence could be found for an absence of a trend in PM_{10} concentrations since 2000, although PM_{10} concentrations might have levelled off.

Trends in PM_{10} concentrations and their components, from model results and measurements

- The trend in average annual concentrations of PM_{10} between 1993 and 2007, at rural locations in the Netherlands, was 0.7 to 1.0 µg/m³ per year. This trend was found to be in keeping with known changes in registered emissions from anthropogenic sources relevant to PM_{10}, in spite of the apparent discrepancy between concentrations and emissions.
- Around two thirds of the decrease in PM_{10} concentrations between 1993 and 2007 was attributed to reductions in anthropogenic emissions of sulphur dioxide, nitrogen oxides and ammonia. The remaining third was attributed, in about even amounts, to primary particles, particle-bound water and secondary organic aerosol.

Is the observed trend in keeping with relevant anthropogenic emission changes?

Average annual PM_{10} concentrations, between 1993 and 2007, decreased by 0.7 and 1.0 µg/m³ per year, between 1993 and 2007, at rural locations in the Netherlands. The range is due to uncertainties introduced by instrument changes. This report also presents a bottom-up estimate of the PM_{10} concentration changes, between 1993 and 2007, starting from the registered anthropogenic emissions relevant to PM_{10}: directly emitted particles (primary PM_{10}), sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and non-methane volatile organic compounds (NMVOC). The gases are, in part, chemically and/or physically converted to secondary particles in the air.

Average annual contributions of primary and secondary anthropogenic components to PM_{10} were estimated for all years from 1993 to 2007, using model results and measurements. Figure 1.7 shows concentration decreases per component, between 1994 and 2006, with respect to the observed decrease in PM_{10} concentrations. The concentrations for 1994 (2006) are averaged over the years 1993, 1994 and 1995 (2005, 2006 and 2007), to eliminate interferences due to fluctuating weather conditions.

SIA;

The contribution of Secondary Inorganic Aerosol was estimated by using the OPS model and registered national and EMEP emissions of SO_2, NO_x, and NH_3. The model results were combined with a long time series of measured SIA concentrations. SIA concentration changes were 0.66 or 0.97 µg/m³ per year (see also below in *Sensitivity to SIA trend*).

Particle bound water;

We estimated the mass of particle bound water that is associated with SIA to be about 15% of the SIA concentration. Water, a natural component of PM_{10}, is attracted by hygroscopic salts on particles, such as secondary inorganic and organic aerosols. A decrease in SIA, which is abundant in PM_{10}, therefore, could lead to an additional significant decrease in PM_{10}. Changes in particle bound water concentrations, corresponding with the SIA changes, were 0.07 to 0.1 µg/m³, per year.

Primary PM_{10};

The contribution of primary PM_{10} emissions from registered anthropogenic sources in Europe was estimated by using the OPS model with national and EMEP expert emissions. Changes in primary PM_{10} concentrations were 0.07 µg/m³, per year.

SOA;

The contribution of Secondary Organic Aerosol to PM_{10} is very uncertain, and the fraction from anthropogenic NMVOC emissions is even more uncertain. We made a rough approximation of the changes in anthropogenic SOA concentrations, based on: a) the NMVOC emission decrease of 60%, between 1990 and 2007, and b) an estimate on anthropogenic SOA for 1990 of 1.5 µg/m³, and c) assuming a linear decreased in SOA along with the NMVOC emission reductions. SOA concentration changes were 0.08 µg/m³, per year.

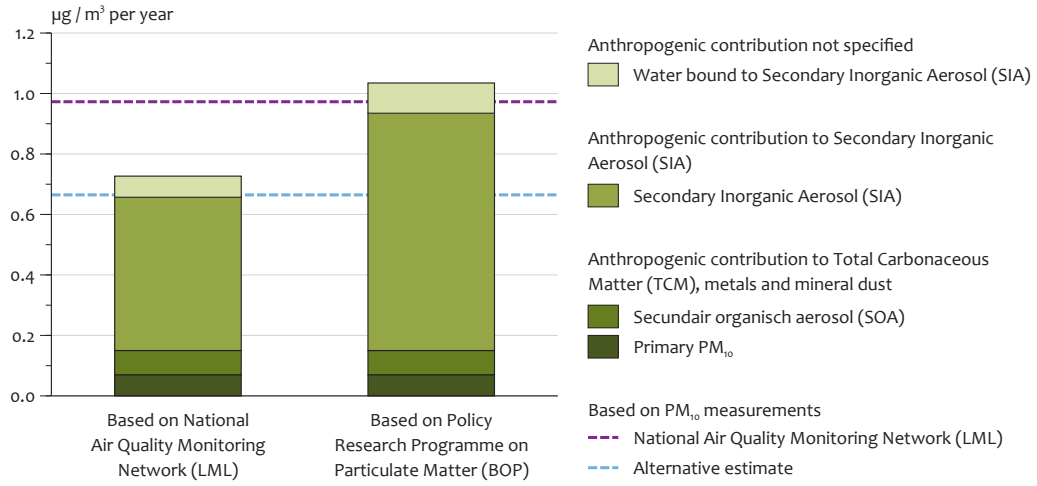
Sensitivity to SIA trend

The national BOP research programme found that routine SIA concentration measurements in were lower , by about 40%,than measured using the PM_{10} reference method . Consequently, the SIA trend was more uncertain than previously thought. The 'High SIA trend' concentration changes of Figure S.7 correspond with an upper limit for SIA concentration changes between 1994 and 2006, given the SIA uncertainties. It shows that the bottom-up approach to understand the trend in PM_{10}, based on changes in registered anthropogenic emission, is also rather uncertain.

The observed PM_{10} concentrations do not necessarily show a linear response to anthropogenic emission changes, for several reasons:

- *Measurement uncertainties*. Most of the relatively large PM_{10} measurement uncertainty is related to the semivolatile part of PM_{10}. A major part of this semivolatile fraction is of anthropogenic origin. The impact of anthropogenic emission changes on PM_{10} needs to be considerable for it to be detected with statistical significance.
- *Weather conditions*. Fluctuating weather conditions modify the impact of emission changes on PM_{10} concentrations.

Change in background concentration PM_{10}, 1993 – 2007 **Figure S.7**



Estimated concentration decreases (µg/m³, per year) per anthropogenic PM_{10} component, between 1994 and 2006, compared with the observed PM10 concentration decreases in the Netherlands.

- *Chemical saturation*. Ambient conditions in north-western Europe are such that, since 2000, ammonium concentrations in the Netherlands are hardly dependent on the ammonia emissions. Also sulphate and nitrate concentrations in the Netherlands no longer react linearly to precursor changes.
- *Natural PM_{10} fraction*. The contribution from natural sources to PM_{10} in the Netherlands is several tens of percentage points. A fixed annual contribution, therefore, would subdue any anthropogenically induced concentration change. In addition, model results and long-term chloride measurements indicated that the contribution of sea salt particles, coincidently, counteracted the anthropogenic downward trend between 2000 and 2007 with an upward trend of 0.1 to 0.3 µg/m³ per year.

The anthropogenically induced concentration changes which resulted from the "High SIA trend" case indeed appeared to be an upper limit, with about 1 µg/m³ per year. To improve the source attribution in our model estimates for assessments of PM_{10} in historical and future years, it is imperative to reduce the current uncertainty in the SIA trend.

PM_{10} concentration changes that were derived, bottom-up, from the anthropogenic emission changes per PM_{10} component, between 1993 and 2007, were about equally uncertain as the observed changes. Given the uncertainties, both observed and estimated PM_{10} concentration changes for the Netherlands were consistent and in line with each other.

Traffic emissions

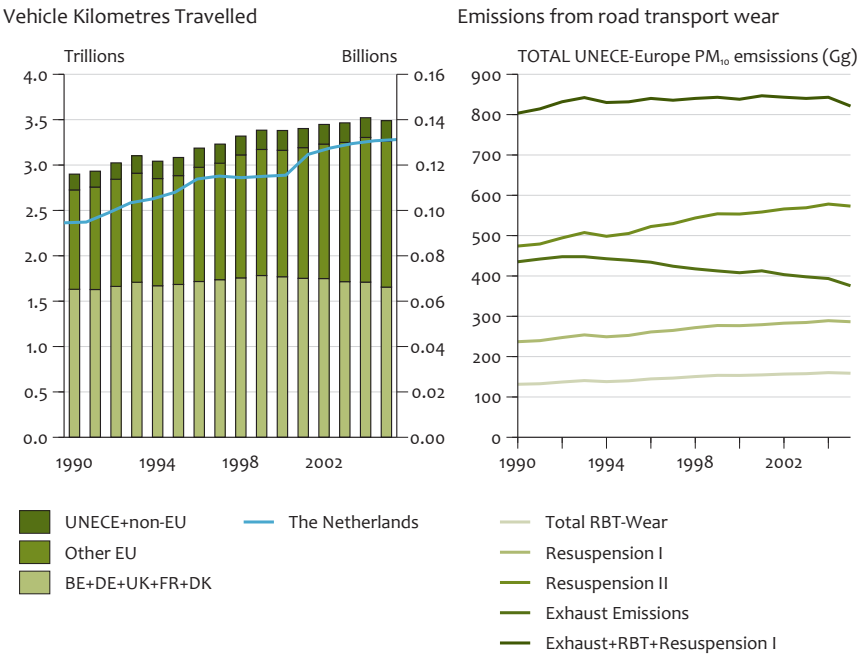
- On a national scale, particle from vehicle exhausts emissionshave decreased, probably by 50% since 1990. Over the same period, particle emissions from brake wear, road wear, tyre wear and dust brought into resuspension by traffic have increased, as these are related to traffic volumes which have been steadily growing at a rate of about 3% per year.
- The net trend in particulate emissions related to road traffic appears to be very small. However, PM from resuspension has a rather broad range of uncertainty; resuspension, as a source of particles, is not included in the Dutch Pollutant Release & Transfer Register.
- On a local scale, spatial distribution of emissions from road traffic in urban areas has altered, due to policies that changed road traffic circulation within municipalities. In the Netherlands, road transport within most major municipalities shows a decreasing trend. In contrast, road traffic to and from these urban areas has increased.

In general, emissions from road traffic contribute significantly to PM_{10}. Along city roads, traffic can even be the dominant emission source, and this is especially important since traffic emissions are suspected to have a relatively high impact on human health. The contribution of traffic emissions to PM was investigated using a trend analysis of traffic and traffic induced emissions at different spatial scales.

Road traffic emissions in Europe

Figure S.8 shows the development of road traffic (expressed in kilometres travelled) and of PM emissions from road traffic for Europe. Although road traffic volumes in Europe continue to increase, European vehicle exhaust emissions are slightly decreasing, because of cleaner technology. However, other emissions related to road traffic, such as from brake wear, tyre wear, road wear (BTR wear) and resuspension, are increasing. Levels of resuspension emissions are highly uncertain, but their existence is undisputed. We have taken a moderate estimate ('resuspension I') and a doubling of this estimate ('resuspension II') just to illustrate that, theoretically, a growth in traffic volume may cause increased emissions despite cleaner technology. Resuspension emissions are not officially registered, which makes them an important unknown factor.

Trend in kilometres travelled and in calculated PM_{10} emissions from road transport **Figure S.8**



Trend in average vehicle distances travelled (left panel), and in calculated PM_{10} emissions from road transport: exhaust emissions, road wear, tyre wear, brake wear, and approximated resuspension of dust due to traffic (right panel).

Road traffic emissions in the Netherlands

Although the reduction in exhaust emissions in the Netherlands and its neighbouring countries is larger than is suggested by the overall European picture presented in Figure 1.8, the limited decrease contradicts the general assumption that emission regulation and general technological improvements should lead to (large) emission decreases. Explanations for this contradiction are:

- The largest reductions were achieved in the first EURO engine classes. In later engine classes, reduction percentages were still substantial, but absolute emissions from BTR wear and resuspension now approach the level of exhaust emissions.
- Increases in road traffic volumes affect both types of emissions, and especially non-exhaust emissions will grow substantially, because they are not influenced by implemented emission reduction technology.
- It was expected that, over time, engine technology would further improve and road transport would become more fuel efficient, thereby using less fuel per vehicle kilometre (vkm), and hence producing less emissions per vkm. However, improvements in engine performance have been countered by increases in luxury (e.g., air conditioning) and safety (weight). This has resulted in a virtually unchanged energy use per vehicle kilometre, over the past 10 years.

Road traffic emission changes, on sub-national and urban scale

Apart from national and European trends in road traffic, relevant changes may also occur on an urban scale. Observational stations in cities are placed in fixed locations. Changes in traffic circulation within a city, therefore, could cause different trends in observational data.

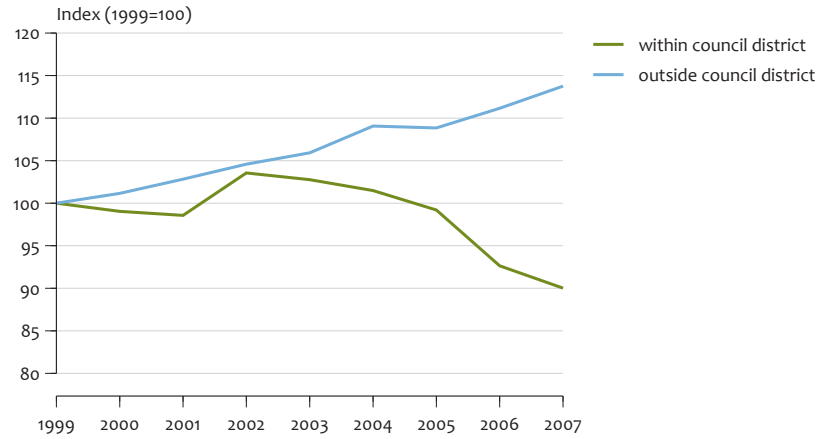
For example, if inner-city traffic were to be reduced because of policy measures, while non-urban traffic would increase, we could see an increasing trend at rural locations and one that stabilises or decreases at traffic locations. Unfortunately, accurate traffic monitoring data (including fuel use and vehicle ages) needed to answer such questions, were lacking.

A spatial analysis was made of trends in traffic volumes in various cities within the Netherlands, based on available data from the 'Dutch mobility monitor'. The results from this analysis are relevant to understand the patterns on sub-national and urban scales. They showed that spatial differences in traffic emission trends were not the same for all cities:

- In Rotterdam, a major port and industrial city, the trend in inner-city traffic volume was similar to that on its ring roads.
- In Amsterdam and Utrecht, both representative of major Dutch cities with old historical city centres, inner-city traffic volumes were declining, while on transit roads they showed an increase (Figure S.9).

The contribution from traffic emissions to PM has been studied by means of a trend analysis of traffic and traffic induced emissions between 1990 and 2005, at different spatial scales. The net trend in particulate emissions related to road traffic appears to be very small since 2000. However, PM brought into resuspension by traffic has a rather broad range of uncertainty. The decrease in PM from exhaust emissions

Number of kilometres (absolute volume) passenger vehicles **Figure S.9**



Difference in development of traffic volumes (average distances travelled by passenger vehicles) within and outside council district.

per vehicle kilometre during the 2000-2007 period has been counteracted by increased traffic volumes and their related non-exhaust emissions (BTR).

1 Introduction

Particulate matter is hazardous to human health. Health studies have shown that there is a significant connection between both short-term and long-term exposure to, especially, fine particles and premature death. Other important effects include aggravation of respiratory and lung disease, asthma attacks, heart attacks and irregular heartbeat (WHO, 2000; 2003; 2006a; 2006b).

The main purpose of this report was to achieve a better understanding of the trends in measured PM_{10} concentrations and other components of particulate matter. Therefore, this report gives an analysis of the trend in particulate matter and other component in the Netherlands, over the 1990-2007 period. Understanding of the trend is necessary for effective PM reduction policies. We studied trends in PM_{10} concentration levels at both rural and traffic locations, in terms of dispersion conditions and emission changes.

First, we focused on the factor of 'dispersion conditions' by following both a measurement-based and a model-based approach to estimate the effect of meteorological fluctuations on average annual PM_{10} concentrations.

Second, we looked at the factor of 'emission changes' by analysing registered anthropogenic emission changes relevant to PM_{10}, on different scales, since 1990. A more detailed trend analysis was performed of traffic and traffic induced emissions from the European scale, down to urban scales.

The overall effect of dispersion conditions and emissions changes on PM_{10} were investigated for large-scale rural concentrations, by using models in combination with measurements.

In the past, European and national legislation has been introduced to curb the effects of air pollution on human health and the environment (e.g. EU Air Quality Directive (EU 2008); National Emission Ceilings Directive (EU 2001), Gothenburg protocol (UN-ECE 1999)). These policy instruments have taken a three-track approach to reducing the negative effects of air pollution. They did this by establishing national emission ceilings and air quality standards, and by achieving reductions in emissions from road traffic and production processes. For particulate matter, air quality standards have been set for the fraction PM_{10} and, recently, also $PM_{2.5}$, to protect public health in general.

As a consequence, important emission reductions have taken place, over the last two decades, which have altered the concentration levels of particulate matter over Europe and the Netherlands, in spite of increased economic activity during that period. Particulate matter consists of many components originating from many different sources. Particulate mass as a whole has been regulated instead of its individual components, because all particulate matter was considered to be harmful – although some components were believed to be more harmful than others – and more importantly, because the health benefits of reducing the components individually are still unknown. An approach of regulating individual PM components would also carry the risk of some harmful components not being addressed in reduction measures.

Currently, PM_{10} and other fractions are being monitored throughout Europe at an ever increasing number of locations (Mol *et al.*, 2007; 2008). Studies on the trend in PM_{10} generally have reported a strong decline throughout the nineties. However, since about the year 2000, PM_{10} concentrations have levelled off and have even seemed to stabilise. This trend of levelling off concerns both secondary and primary components. Such a levelling trend in particulate matter has been observed in many European countries, such as France, Germany, the United Kingdom, Italy, Poland (Mol *et al.*, 2008; Harrison *et al.*, 2008), and also in the Netherlands (Van der Zee and Woudenberg, 2006; Beijk *et al.*, 2007b; Wesseling and Beijk, 2008; Woudenberg *et al.*, 2008).

The question is whether observations could be explained based on the current understanding of PM_{10}, including the estimates of current anthropogenic emissions, or if we are missing something? Answers to this question are important regarding the validity of current projections of future emissions and concentrations, which form the basis for policy measures to improve air quality.

Assessment of the trend in particulate matter in relation to anthropogenic emissions, however, is being seriously hampered by large uncertainties about measurements, emissions and model results that have their origin in the complex physio-chemical behaviour of PM_{10} and its sub-fractions. This is particularly true for semivolatile components such as water, ammonium nitrate (NH_4NO_3) and volatile particulate organic material. Furthermore, it seems that current concentration levels of secondary inorganic aerosol in the Netherlands are underestimated (by around 40%) from

current routine measurements in the national monitoring network and by model simulations used for national assessments of current and future air quality (Weijers e*t al*., 2010).

The long-term trend in PM_{10} caused by anthropogenic emission changes is also obscured for other reasons. In the Netherlands, several tens of percentage points of PM_{10} consist of natural and semi-natural fractions, such as sea salt and wind-blown dust (Schaap *et al.,* 2010). Therefore, the impact of anthropogenic emission changes on PM_{10} is a priori reduced in the long term by the contribution from natural sources. Furthermore, average annual PM_{10} concentrations in the Netherlands, show a response of around 9% to large-scale meteorological variabilities (Velders and Matthijsen, 2009).

More insight into the effect of emission reductions on PM_{10} in the past, and a better understanding of how PM_{10} concentrations are brought about by different sources, may help us to improve assessment of future levels of PM_{10} and $PM_{2.5}$, using models based on emission scenarios. With present knowledge and data, we aimed to determine whether relevant emission changes would be consistent with observations of PM_{10} concentration levels and other particulate components in the Netherlands.

The following gives a general outline of this report:

Chapter 2, *Influence of weather conditions on measured PM_{10} concentrations* In this chapter we address the trend of PM_{10} observations and the influence of weather conditions on the PM_{10} concentrations for the Netherlands. Therefore, we first analysed observed trend between 1993 and 2007 in PM_{10} concentrations in the Netherlands. To investigate the effect of weather conditions on the concentrations we use the correlation between PM_{10} observations and more or less co-located meteorological measurements. From these data meteo corrected PM_{10} trends are built for PM_{10} measurements at rural, urban and traffic stations.

Chapter 3, *Influence of weather conditions on modelled PM_{10} concentrations.* This chapter describes another approach for investigating the effect of weather conditions on PM_{10} concentration levels. With the use of the same PM emission data, model calculations were performed for seven consecutive years (2001 to 2007). This approach highlights the effect of fluctuating weather conditions on PM_{10} and excludes possible effects from changes in anthropogenic emissions. Such an exercise can only be done with the use of models.

Chapter 4, *Detection of trends and meteorological summary.* In this chapter, the previous chapters on several approaches of meteorological influences on PM are summarised. In addition, some statistical properties of trend detection are elaborated. Combining these statistical properties with hypothetical trends clarifies why small trends, and small changes in trends, are difficult to detect.

Chapter 5, *Trends in PM_{10} and other components of particulate matter at rural locations, in emissions and model results.* This chapter describes our interpretation of the observed trend in PM_{10}. We compared the trend in PM_{10} with relevant trends in emissions and concentrations of other components of particulate matter; these are sulphate, nitrate, ammonium, black smoke and heavy metals. In addition, we estimated the contribution from changes in anthropogenic emissions to observed changes in PM_{10} concentrations between 1993 and 2007. In this study, we accounted for the large uncertainties in both measurements and model results.

Chapter 6, Trends in *emissions and air quality on national to European scales.* This chapter zooms out from the national perspective, and places air quality and emissions trends in a bigger frame with trends taking place in other European Member States and in Europe as a whole. A question here is whether the trends in the Netherlands are in keeping with European developments.

Chapter 7, *Trends in emissions from traffic.* The trend in PM_{10} concentrations at traffic locations can be distinctly different from that at rural locations, which represent the large scale. Local traffic conditions may determine local trends. This chapter describes our exploration of local traffic conditions for several cities in the Netherlands. In addition, we looked at the trend in primary PM and NO_x emissions at different traffic locations to investigate the possible effect of these emissions on PM concentrations at these locations.

2

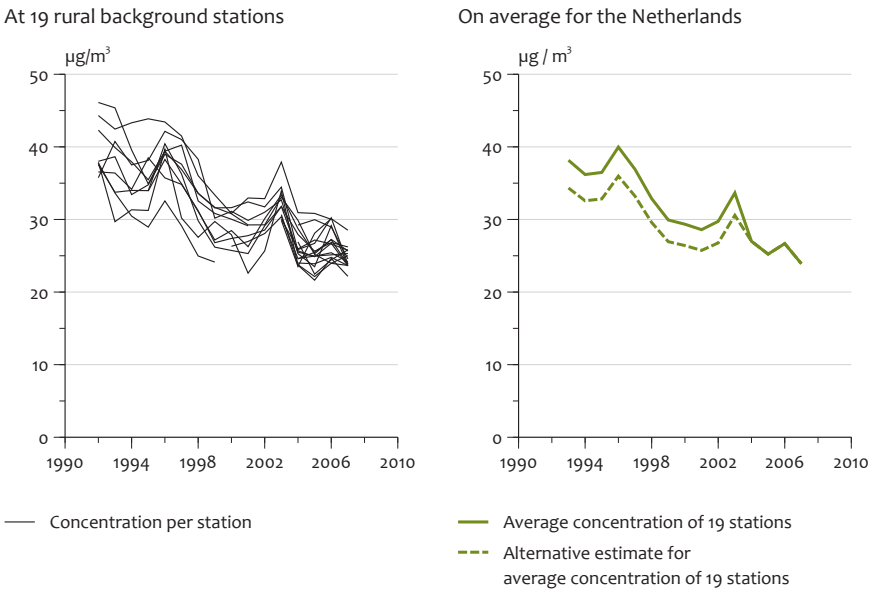
Influence of weather conditions on measured PM_{10} concentrations

This chapter addresses the trend in measured PM_{10} concentrations and the influence of weather conditions on these concentrations for the Netherlands. Measurements of PM_{10} concentrations are part of the Dutch National Air Quality Monitoring Network (LML) (Beijk, 2009). PM_{10} was also measured during the measurement campaign of the BOP research programme, in 2007 and 2008.

2.1 PM_{10} observations

PM_{10} is measured with automated instruments using the beta attenuation method. In 1992, the Dutch LML network started with 13 measuring stations; 10 were placed in rural locations, 1 in an urban background location and 2 in traffic locations. Since then, the number of stations has increased gradually to a total of 40 stations in 2007: 17 rural, 7 urban background and 16 traffic locations. Figure 2.1 (left panel) shows average annual PM_{10} concentrations for all rural locations, and the right-hand panel shows the average for measurements with a time series of over 10 years and a data availability of 80% per year. In 2003, an update was started of the PM_{10} monitoring network, whereby instruments, heating configuration and housing were renewed. As a consequence, absolute PM_{10} concentrations that were measured before 2003 are believed to be much less certain than measurements that have been performed since then. The thick and thin lines indicate the

Average annual PM_{10} concentration **Figure 2.1**



Average annual PM_{10} concentrations (µg/m³) in the Netherlands (1992-2007) for stations at rural locations, left panel. Average annual PM_{10} concentrations (green line), right panel. The green dashed line signifies the lower estimate for the average concentration measured in rural locations for the years 1992 to 2003 (see Text box PM_{10} measurement limitations).

range considered in this report (for more details, see Text box *PM_{10} measurement limitations*).

2.2 PM_{10} concentration trend

In this section, the consequences of two possible cases for the PM_{10} trend are presented. Instrumental changes in 2003 have hampered a trend analysis of the PM_{10} series of measurements between 1993 and 2008. From 1993 to 2003, PM_{10} levels at rural locations decreased on average by 0.8 to 0.9 µg/m³ per year, while for the 2004-2008 period, the average decrease was about 0.7 to 0.8 µg/m³ per year (Figure 2.1). The uncertainties about the absolute PM_{10} level between 1993 and 2003 result in two possible cases for the PM_{10} trend for the whole 1993-2008 period:

1. a trend of about 0.9±0.3 µg/m³ per year (thick green line in Figure 2.1). The decrease is more or less continuous, apart from meteorology-related variabilities.
2. a lower estimate: an average trend of about 0.6±0.3 µg/m³ per year (dashed green line Figure 2.1). In this case, a levelling off could be observed around the year 2000; until 2000, a decrease of over 1±0.6 µg /m³ per year was observed, followed by a much more level decrease of no more than 0.2±0.5 µg/m³ per year between 2000 and 2007. Please note that the confidence intervals of the 1993-2000 and 2000-2008 trends overlap.

Therefore, the uncertainty in the measurements would allow for a decrease in PM_{10} over 14 years of either about 14 µg/m³ (case 1) or about 10 µg/m³ (case 2). The behaviour of the PM_{10} concentrations in case 2 – with a trend break around the year 2000 – appeared more in keeping with reported PM_{10} trends in countries nearby (Mol *et al.*, 2008) and with trends in other particulate substances (see Chapter 4).

2.3 Spatial and temporal variability

PM_{10} concentrations at rural locations should be representative of those on a relatively large scale (> 30 km). As a consequence, the spatial variability between average annual concentrations should be small. However, the spatial variability in PM_{10} annual averages (see Figure 2.1, left panel) is relatively large with an average standard deviation of about 3.5 µg/m³, up to 2003. After the instrumental changes of 2003, the standard deviation came down to a value of about 2.5 µg/m³. Therefore, the PM_{10} trend at individual locations is more uncertain than the trend in annual averages. Several sources have contributed to the observed spatial variability:

- Random errors of the monitoring instruments.

PM_{10} measurement limitations

The Dutch LML network uses automated instruments for measuring PM_{10}, as do most other networks in the EU. A major complication in this process is the fact that the air has to be dried first before it can be measured. In most automated monitors, this drying is done by heating the air. This leads to evaporation of semivolatile compounds that may contribute substantially to the total PM_{10} mass. The estimated average loss of PM_{10} in automated monitors in the EU is 30%.

In 2003, most of the automated monitors at rural locations in the monitoring network were replaced with new monitors. Like the old monitors, this new type of monitor was also calibrated to the European reference method for PM_{10} (NEN 12341), and equivalence was demonstrated in 2007 (Beijk et al., 2007a). For the old and the new types an uncertainty of 22% and 16% was found in the average daily PM_{10} concentration at rural locations, based on the comparison with reference measurements. Following the calibration of PM_{10} data in 2007 measurement results in AIRBASE were updated.

Since calibration of the PM_{10} measurement results, the Dutch monitoring data fully comply with the European standard. However, a trend analysis is sensitive to uncertainty in the systematic difference between the old instrument (pre-2003 data) and the new instrument (post-2003 data). This uncertainty is strongly related to the number of monitors involved in the equivalence study. For the old type of monitor, necessarily in two different configurations, the number of comparisons were quite limited, therefore, the uncertainty in the systematic difference might be 10 to 15%.

Another method of estimating the systematic difference between both types of monitors was that of recording the change in concentration levels at locations with new monitors, compared to those still operating with old monitors. Although there are considerable uncertainties attached to each of these changes, the average values were more accurate due to the large number of new monitors. Evaluation of this data showed, for the new types of monitors, a decrease in concentration levels of between 0 and 10%, compared to results from the old monitors. This systematic difference, is shown in Figure 2.1 (right panel, dashed line), by the alternative trend line for the data on measurements taken with the old instruments. This alternative was calculated by multiplying the PM_{10} data for the 1992-2003 period by 0.9.

This lower estimate gives a more consistent increment in urban concentrations between 1992 and 2003, which was the same as in the 2004-2007 period (5 µg/m³). This increment in urban concentrations has been defined as the difference in the measured average annual concentrations at urban and traffic locations, on the one hand, and rural locations, on the other.

The correction described above is most likely to reduce the uncertainty in 2003. There was no high-quality information available on the earlier data, therefore, the correction was applied to the data on the entire 1993-2003 period. Uncertainties related to data that go back further in time are presumed to be higher.

- The use of an unchanging equivalence factor to correct for evaporation losses of the continuous PM_{10} measurements at all rural locations, whereas the amount of semivolatile particulate matter varied per location. The set of available reference measurements was too small to derive differentiated equivalence factors for all 17 rural locations.
- Influences from local-scale meteorology in combination with location-specific source strengths, such as sea salt along the coast, also played a role.
- Location-specific dispersion characteristics, which could have caused the influence of long-range transport of pollutants to vary per locations.

Much of the year–to-year variability in the annual averages could have been attributed to meteorological circumstances (Matthijsen and Visser, 2006; Velders and Matthijsen, 2009; see Section 2.2). Meteorological circumstances have led to higher (1996 and 2003) and lower PM_{10} concentrations (1998, 1999 and 2000) than those of the average trend. This behaviour was due to large-scale meteorological variabilities and applied to large parts of the continent – at least to north-western Europe (Andersson *et al.,* 2007).

2.4 Meteorological normalisation of PM_{10} data

Meteorological conditions have a large influence on air quality levels, not only on daily levels, but also on annual averages. Thus, for the assessment of trends in air quality data series, meteorological induced variability had to be determined, in order to separate the effect of abatement policies and economic growth from natural variations due to the weather.

Therefore, we constructed a meteorological normalised time series of PM_{10} data, based on the most simple model framework that would still be physically plausible and statistically reliable.

We used in our study two data sets. One set contained hourly data on particular matter (PM_{10}), from between 1993 and 2008, obtained by the National Institute for public Health and the Environment in theirDutch National Air Quality Monitoring Network (LML). The other set contained hourly data on meteorological variables as measured by the Royal Netherlands Meteorological Institute (KNMI), covering the same 15-year period.

The PM_{10} data set contains data from all the locations where measurements have been conducted since the start of the PM_{10} network, and consists of 10 rural locations, 4 urban background locations, and 3 traffic locations (see Table 2.1). Please note that, for the rural location of De Zilk, the time series starts with January 1994, and that the rural locations of Witteveen and Wageningen have been coupled to the additional time series from Wekerom and Valthermond, respectively.

Both data sets have been downscaled into average daily values, as average daily PM_{10} values are more reliable than hourly values, and because our main interest concerned meteorological influences on PM_{10} values, on timescales of more than one hour. While calculating daily averages, the data was checked for gaps of more than six hours and data coverage of under 50%. This is conform EU legislation (97/101/EG) for air quality data and is only important for average daily PM_{10} values. The data coverage of the meteorological parameters is 100%. Of the PM_{10} daily averages from the rural locations, a total of 6.7% is missing, a significantly higher amount of which from the beginning of the time series than from the more recent years. The missing data has not been addressed in our analysis.

To illustrate the influence of weather on PM_{10} levels, three examples are presented of the correlation between meteorological variables and PM_{10} values. The first panel of Figure 2.3 indicates that low temperatures (and, to a lesser degree, high temperatures) correlate with higher PM_{10} levels. This is mainly because low temperatures usually correlate with shallow mixing layers. Mixing-layer height is an important meteorological variable, influencing PM_{10} concentrations (Rost *et al.*, 2009).

The correlation figure that presents wind direction versus PM_{10} shows that contaminated air was coming from the east (continental), while westerly (sea) winds transported cleaner air; a rather predictable result. The last panel of the figure shows that precipitation reduces particulate matter concentration in air. Even small amounts of precipitation have this effect, as is shown by the fact that only the average PM_{10} value of the first bin is above average.

Because of this effect, we introduced an extra meteorological parameter, named LPDD ('length of period of dry days'). The LPDD was derived from the daily amounts of precipitation and defined as the number of days without detectable rain (with zero denoting a day with precipitation). Statistically, the LPDD parameter is more significant than the mere indication of daily amounts of precipitation. The physical explanation of this is that the question of whether it has been raining or not is more important than that of how much actual rain has fallen. A further advantage of using the LPDD parameter is its less local character, compared to the parameter that indicates the spatially highly varied amount of rain.

We determined the optimal meteorological predictor set, after applying forward selection and backward elimination techniques in a multivariate regression analysis (Sokal and Rohlf, 1995). In the regression analysis, data from the ten rural locations were used, in combination with data from one meteorological location at the time. For six out of ten meteorological locations the optimal predictor sets were identical. This optimal predictor set consists of the following six meteorological parameters:

- the sinus of the wind direction;
- temperature in Celsius (minus a constant of 10 degrees Celcius) squared;
- relative humidity squared;
- the amount of daily precipitation;
- period of drought (LPDD);
- LPDD squared.

The data appeared very sensitive to the LPDD. Therefore, the LPDD was included in the analyses, both as a linear and as a squared term.

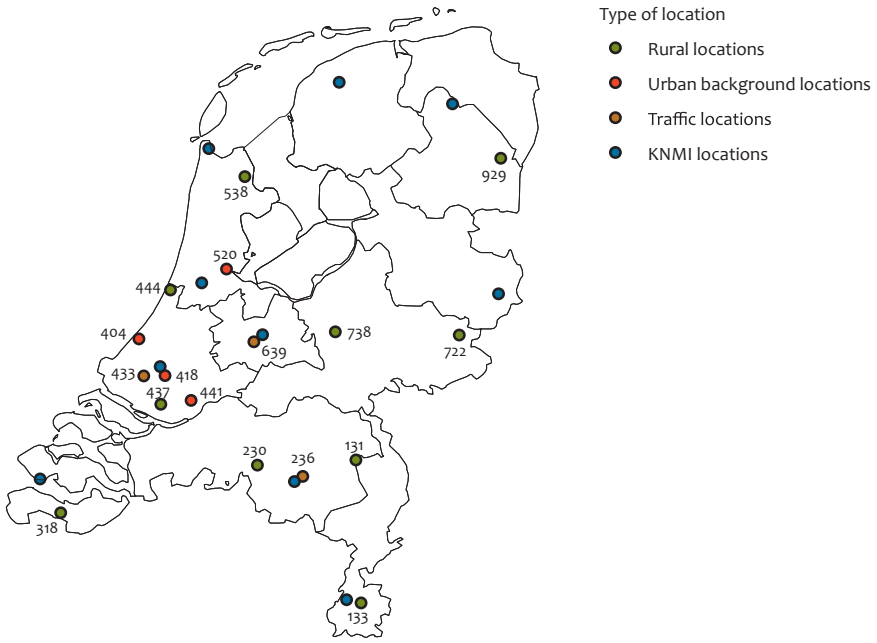
List of LML locations from which PM_{10} data has been included. Location numbers have been included for easy reference Table 2.1

Rural locations	Urban background locations	Traffic locations
131 Vredepeel	404 Den Haag - Rebecquestraat	639 Utrecht - Erzeijstraat
133 Wijnandsrade	418 Rotterdam - Schiedamsevest	236 Eindhoven - Genovevalaan
230 Biest	520 Amsterdam - Florapark	433 Vlaardingen - Floreslaan
318 Philippine	441 Dordrecht - Frisostraat	
437 Westmaas		
538 Wieringerwerf		
722 Eibergen		
444 De Zilk		
929 Witteveen/Valthermond		
738 Wageningen/Wekerom		

List of meteorological KNMI locations from which data has been included Table 2.2

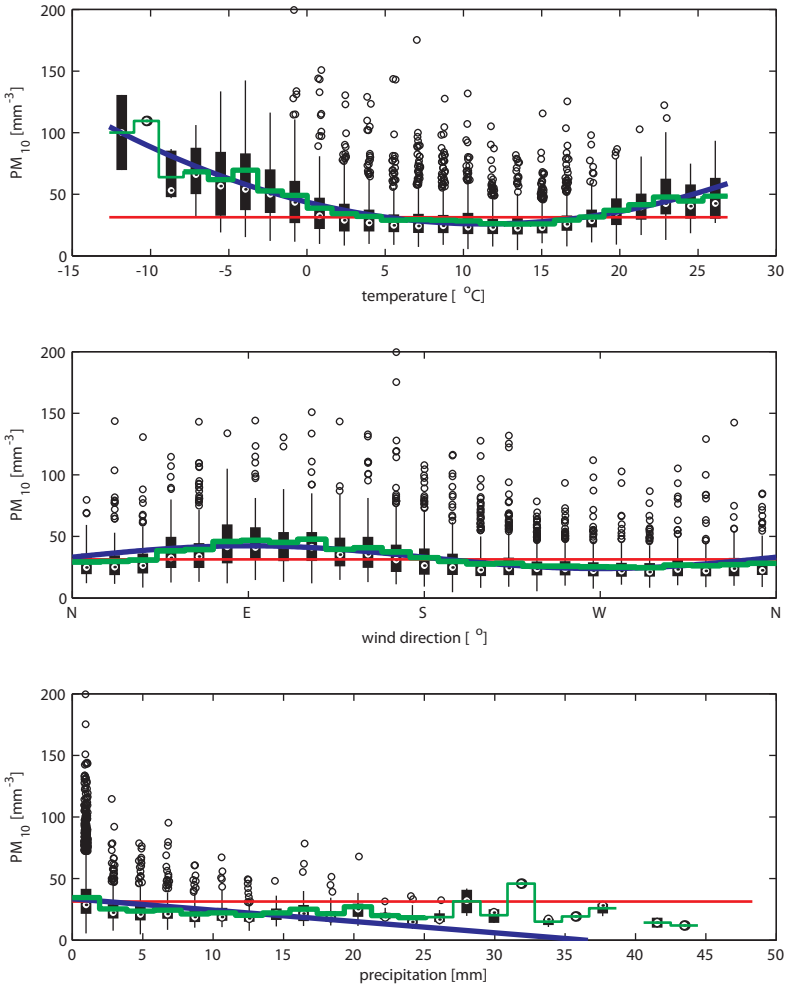
Meteorological locations
260 De Bilt
240 Schiphol
344 Rotterdam
290 Twenthe
310 Vlissingen
235 Den Helder
380 Maastricht
370 Eindhoven
270 Leeuwarden
280 Groningen

Location of LML sites and KNMI stations Figure 2.2



LML and KNMI locations used in this study (see Tables 2.1 and 2.2 for the locations and their names).

The multivariate analysis was performed of the PM_{10} data from all the rural locations, simultaneously. For the predictor set, we chose a minimalistic set of parameters by taking only variables into account with high absolute t-stat values (above 20). This extremely critical approach was selected, because the PM_{10} data set contained data from multiple locations,



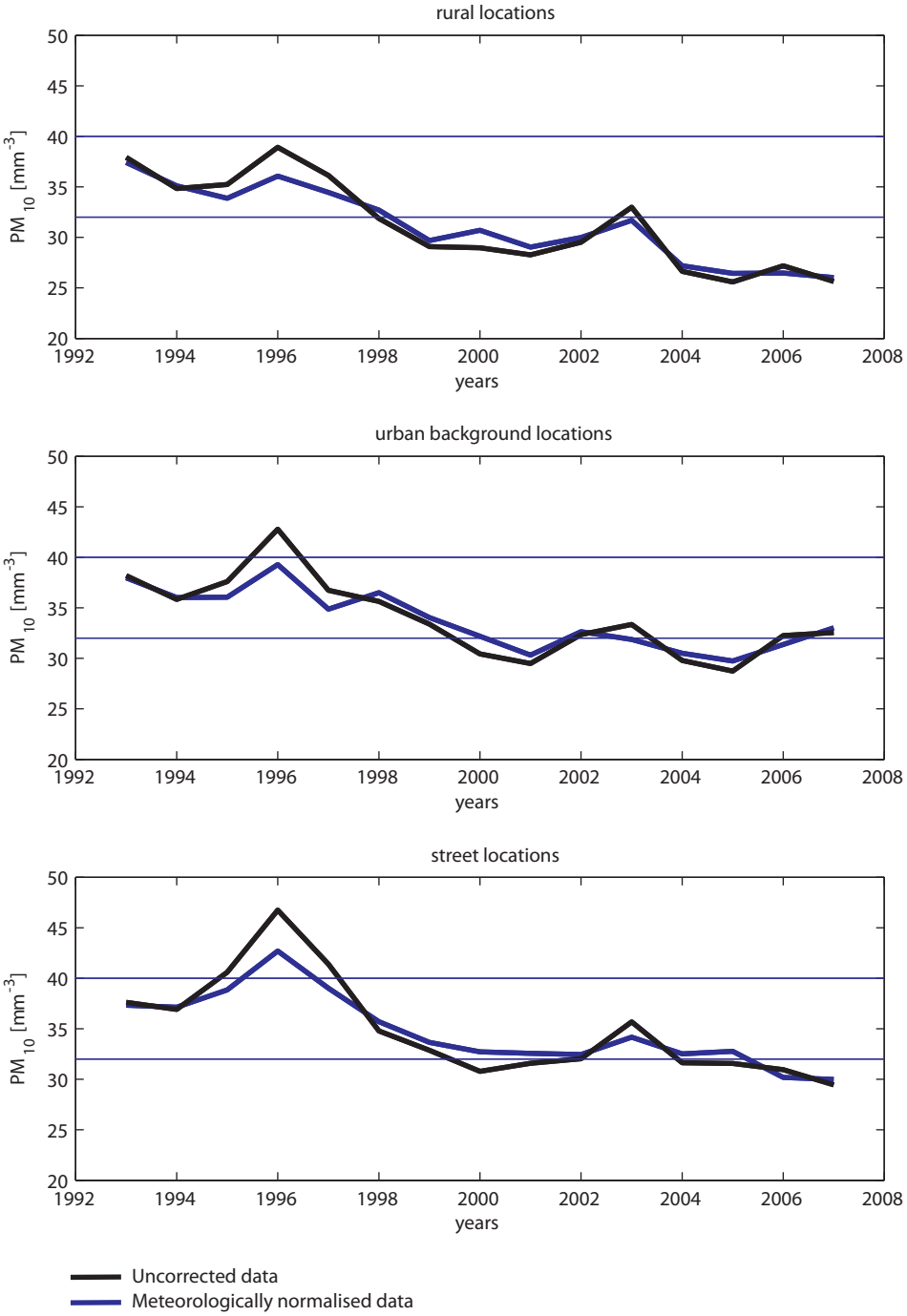
PM_{10} values as a function of three meteorological variables, namely, temperature, wind direction, and daily amount of precipitation. PM_{10} data represent the Westmaas-Groeneweg location, while the meteorological data refer to the location of De Bilt. The meteorological data was binned in 25 equally sized bins and is presented in the form of a box-and-whisker plot: the 25th, median and 75th quantiles are given and the upper and lower whiskers denote plus or minus 1.5(75th-25th) + median (or the most extreme data value within this range). Outliers are given with open circles. The thin red line represents the average PM_{10} value, the blue line denotes the best fit of the data (a quadratic form is assumed for temperature, and a sinus form for wind direction), and the green line stands for the average value of each bin. The green line is thick when more than 6 data values are available in the respective bin.*

which might introduce statistical dependencies between model residuals. The analysis was performed by using one meteorological data set (the sensitivity to the selection of meteorological locations is shown in Figure 2.6). When taking De Bilt as meteorological location, the set explained 26.5% of the variance in daily PM_{10} values in the ten data series on rural PM_{10}. Including all parameters meant a doubling of the number of parameters used, but raised the explained variance to only 27.9%.

In the multiple regression analysis the variable 'time' was included to take into account a decrease in PM_{10} over the years. In the calculation of the meteorologically normalised PM_{10} concentrations this variable was left out of the model. In this way, the decrease in PM_{10} over the years was accounted for, while the normalised PM_{10} were solely corrected for meteorological influences.

For a similar reason, PM_{10} values for every New Year's Day were disregarded in the multiple regression analysis. Firework emissions in combination with cold temperatures and light wind conditions may lead to exceptionally high PM_{10} values. This might have influenced the regression analysis and present unrealistic results. In actual practice, this influence

Yearly mean PM_{10} concentrations for the three types of locations **Figure 2.4**

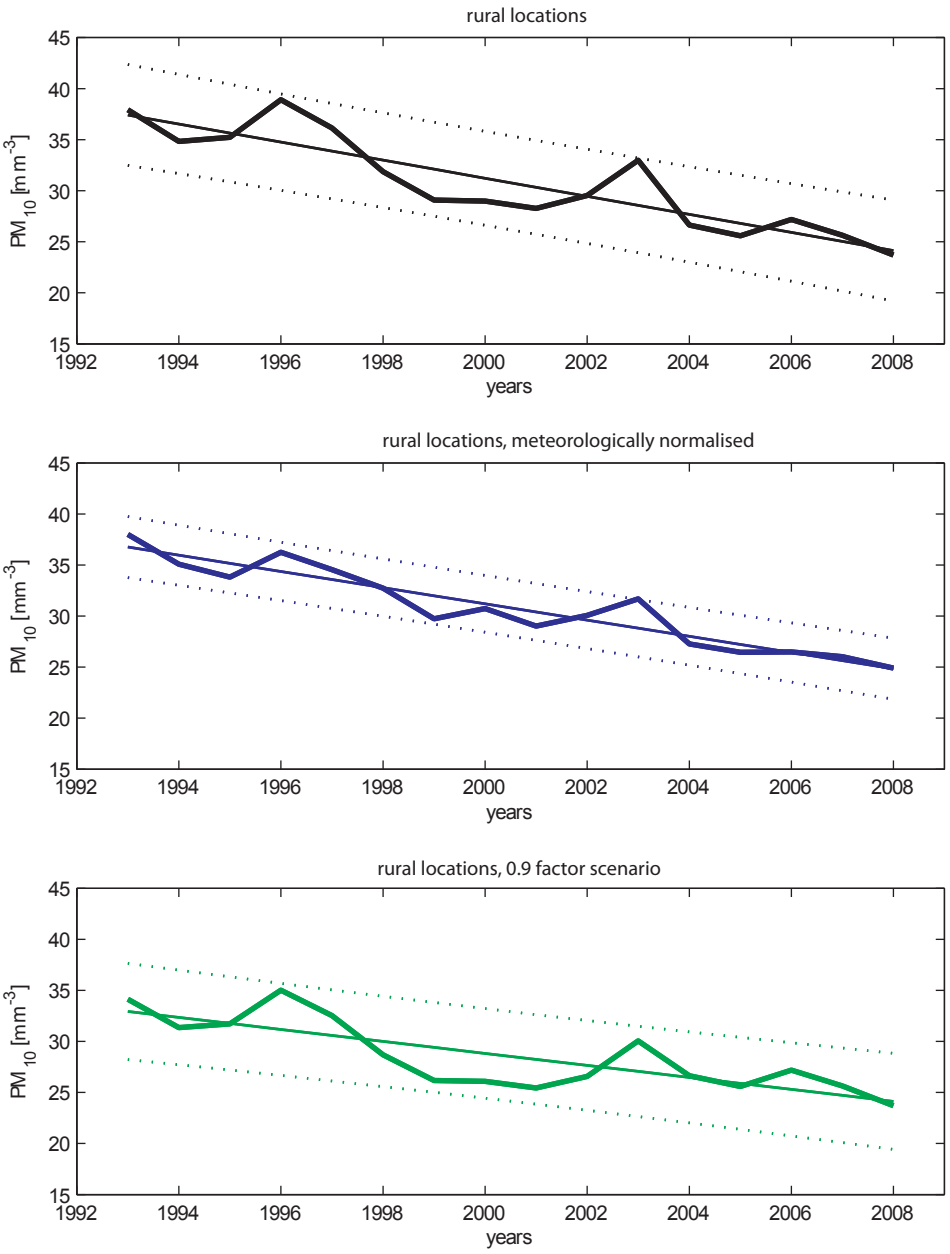


Average annual PM_{10} concentrations for the three types of locations. Uncorrected data are represented by a black line, while meteorologically normalised data are presented in blue.

was found to be small, most likely because such particular weather conditions only occurred on a small number of occasions between 1993 and 2007.

However, in the calculations of the normalised PM_{10} concentrations, all New Year's Days were included.

Figure 2.4 displays time series of the meteorologically normalised average annual PM_{10} concentrations for the three types of locations. For the urban background and traffic locations, the same predictor set was used as for the rural locations, but the multivariate regression analysis was repeated for each type of location, separately. Because of the normalisation, the peak PM_{10} values for 1996 and 2003 (related to cold and dry winter weather in both years, and to hot and dry summer weather in 2003 only) were reduced, while in the intervening years, PM_{10} values were slightly

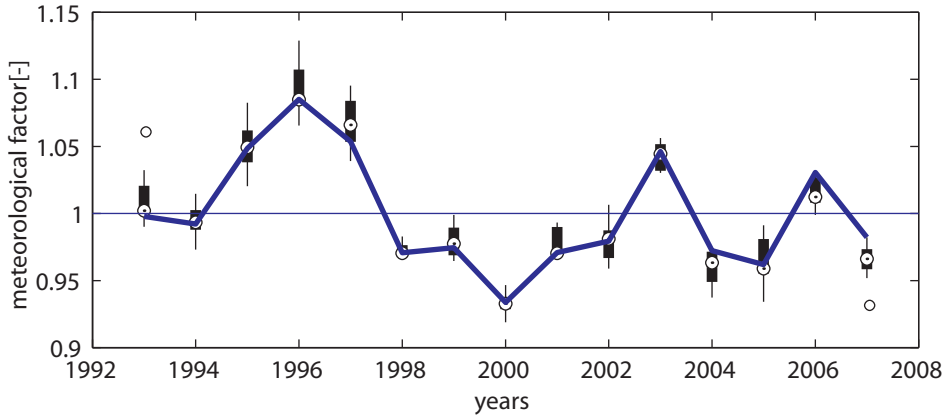


Trend line and 95% confidence limits for the trend line of average annual PM_{10} concentrations at the rural locations (cf. upper panel of 2.4, here the time series was extended with data of 2008). Uncorrected data are shown in black, while meteorologically normalised data are presented in blue, and in the bottom panel, a scale factor of 0.9 was applied to all uncorrected data of before mid November 2003.

elevated. As anticipated, meteorological normalisation led to a reduction in scatter in the annual averages.

Normalisation of the annual averages also had a small effect on the calculated long-term trend (see Figure 2.5). For the 1993-2008 period, the long-term trend with respect to the rural locations changed from -0.88 to -0.79 µg/m³/year; for the other two types of locations, the absolute difference in the trend was of the same order. The applied meteorological normalisation model reduced the width of the 95% confidence limits for the trend line by roughly a third, as shown. In Chapter 5, more statistical features on the detection of trends are elaborated. Figure 2.6 displays the same data, but in a different manner, namely, as factors with which the average

Scalingsfactor of the PM_{10} yearly mean values due to meteorologically normalisation **Figure 2.6**



Scaling factor of average annual PM_{10} values after meteorological normalisation. The blue line represents the meteorological factor for the KNMI location of De Bilt. The meteorological factors that were based on all ten available KNMI locations (see Table 2.2) are shown in the form of a box-and-whisker plot: the 25th, median and 75th quantiles are given, and the upper and lower whiskers denote plus or minus 1.5(75th-25th) + median (or the most extreme data value within this range). Open circles represent the outliers.*

annual values in specific years were elevated or reduced because of meteorological circumstances. The yearly factors have only been displayed for the rural locations; however, differences with the factors for urban background and traffic locations are small.

For simplicity and clarity, we carried out our multivariate regression analysis with data from only one meteorological location for all ten PM_{10} locations. The meteorological location chosen was De Bilt, which, because of its central location, we considered to be representative of the average weather patterns in the Netherlands. De Bilt was indeed the most optimal choice with respect to the multivariate regression analysis that we carried out. For certain specific years, other locations might be the better choice, but the weather pattern for De Bilt was the most representative (i.e. the lowest standard deviation from the average) of the full period. This can also be seen in Figure 2.6, which shows the information on the uncertainty of the meteorological scaling factor due to the choice of meteorological station. The spread was based on all ten meteorological stations.

A fifteen-year PM_{10} time series, normalised on the basis of a multivariate regression analysis of the 1993-1996 period only, led to similar results as those from a time series that had been normalised on the basis of a multivariate regression analysis of the 2004-2007 period. This indicates that the discontinuity in the time series as noted by Matthijsen and Visser (2006) is no longer present in the revalidated PM_{10} data series (Beijk *et al.*, 2007a). When comparing absolute PM_{10} values with those presented in Matthijsen and Visser (2006), please note that the time series had not yet been revalidated at the time.

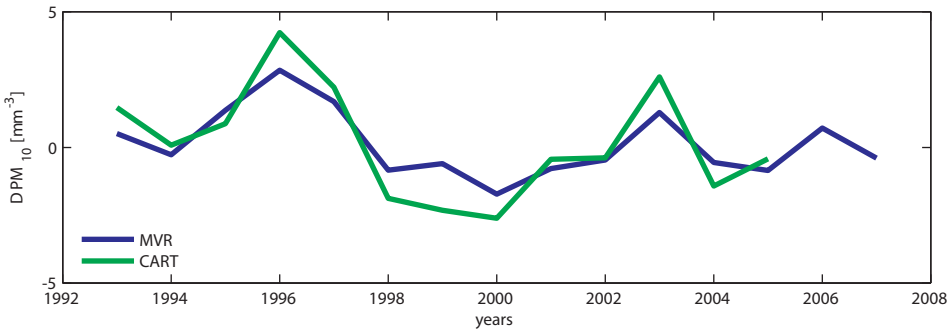
As stated above, starting point for our meteorological normalisation method was a good balance between simplicity and effectiveness. In this regard it would be interesting to compare the outcome of our straightforward approach with the concentration corrections derived from the Classification and Regression Tree (CART) analysis, as developed by Visser and Noordijk (2002). Their model is spatially more refined, both in model structure and in model parameters, but as such it is also more sensitive to local aspects and incidental measurements. The number of parameters taken into account in the CART analysis, consequently, is also at least one order of magnitude larger than in the multivariate regression analysis. Because of our choice of daily values, we avoided difficulties regarding the aggregation into monthly averages with respect to certain meteorological variables (e.g. wind direction).

Averaged nationwide, that is, averaged over the ten rural locations available, the concentration corrections estimated according to the two methods are comparable, as can be seen in Figure 2.7.

The PM_{10} corrections as estimated by the multivariate regression method had a smaller amplitude. This is as is expected, due to the choices made when designing the meteorological normalisation model.

2.5 Conclusion

PM_{10} concentrations are strongly influenced by weather conditions. This also affects the annual averages. By using a simple statistical approach, a meteorologically normalised data set can be generated. The variation in annual averages reduces by approximately one third.



Comparison between the multivariate regression meteorological normalisation method (as used in this study) and the CART method by Visser and Noordijk (2002).

3

Influence of weather conditions on modelled PM_{10} concentrations

This chapter presents our attempt to quantify the impact of meteorological fluctuations on PM concentrations in the Netherlands. We studied the influence of meteorological fluctuations in model-predicted PM concentrations. In order to do so, we performed calculations with the LOTOS-EUROS model, using a fixed set of anthropogenic emissions to predict PM concentrations over Europe for seven consecutive years (2000-2006), as well as fluctuating meteorological data as model input for each of these seven years.

Variation in meteorological conditions can influence source strengths. For example, changes in PM emission levels from residential combustion, due to warmer or colder years or increased resuspension of dust in dry years. Furthermore, meteorological variation influences the lifetime of PM in the atmosphere. More rainy days in a year will 'cleanse' more often the atmosphere, as the rain removes suspended particulates. Elucidating the influence of meteorological variation on PM concentrations is complicated further, because atmospheric transport of particles also plays an important role and impacts can vary across Europe. For instance, a hot or wet summer in one country does not necessarily mean hot or wet summers in all countries.

We have not included the effect of meteo variation on the source strengths except for the meteo dependent generation of sea salt particles. The effect of meteo variation on the anthropogenic source strengths is highly speculative and demands a thorough analysis of all individual European countries. The source strength for a particular year may change in one climate region and not in another. The generation of sea salt particles is driven by meteo data, this process included in the model (Manders et al., 2009). Therefore, calculations for each year have a unique sea salt contribution to PM and consequently the annual variation due to meteo-variation in this source is quantified.

Objectives

- Quantification of the inter-annual variability in model-predicted PM_{10} concentrations due to meteorological differences between years.
- Quantification of the inter-annual variation in sea salt particulates that depends on weather conditions.
- Identification of the extent to which the natural variability due to meteorological variation could obscure trends in emissions, and the resulting change in concentrations.

3.1 Methodology

The LOTOS-EUROS model is used for predicting PM concentrations over Europe, for different years. LOTOS-EUROS is a 3D-Eulerian model with complete atmospheric chemistry. The model has been described in the literature (Schaap *et al.*, 2008 + ref to technical BOP report), and has been used for studying the formation of secondary inorganic aerosol (Schaap *et al.*, 2004a) and the distribution of primary aerosols (Schaap *et al.*, 2004b). The model is run with fixed anthropogenic emissions set to predict PM concentrations over Europe, for seven consecutive years (2000-2006), using each year's own meteorological data. which were obtained from ECMWF (http://www.ecmwf.int/). The anthropogenic emission data that was used as input were emissions for the year 2000, consistent with the baseline projections from the EU CAFE model, gridded at a 0.25° x 0.125 ° longitude and latitude. For further details on the emission database we refer to Visschedijk and Denier van der Gon (2005).

The model-predicted PM concentrations were sampled from the model output for five rural background locations of the Dutch National Air Quality Monitoring Network (LML) (Table 3.1). These five locations were chosen as they are situated in different parts of the country, to investigate if any observed variation in the PM concentration would be the same, countrywide, or whether regional differences would also be important.

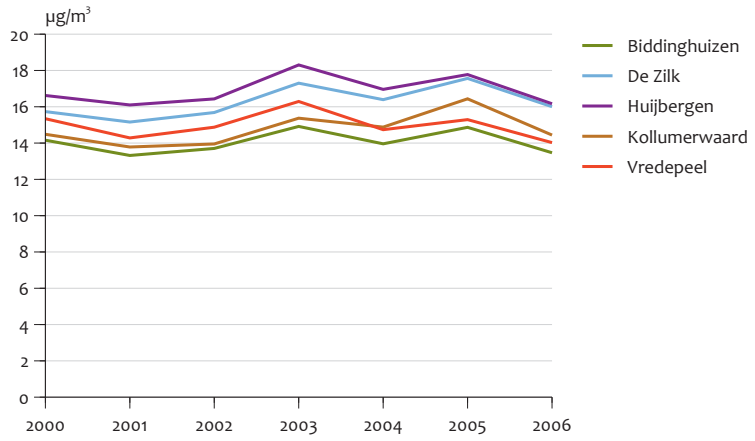
3.2 Results and Discussion

Model-predicted concentrations of PM_{10} in the 2000-2006 period, for five rural locations, are presented in Figure 3.1. The PM_{10} concentrations can be divided in contributions

Selected rural locations from the Dutch National Air Quality Monitoring Network (LML) Table 3.1

Location #	Name	Descriptive location in the Netherlands
131	Vredepeel-Vredeweg	Southeast
235	Huijbergen-Vennekenstraat	South
444	De Zilk-Vogelaarsdreef	Coastal
631	Biddinghuizen-Hoekwantweg	Central
934	Kollumerwaard-Hooge Zuidwal	North - coastal

Model-predicted PM_{10} concentrations for five regional background stations Figure 3.1



Model-predicted PM_{10} concentration levels at five rural background locations, using fixed emission input and year-specific meteorological data.

of sea salt particulates (Figure 3.2). and in PM_{10} due to anthropogenic emissions (Figure 3.3). The latter included emissions of primary PM_{10} and secondary PM_{10} due to precursor emissions (NO_x, SO_2, NH_3). Since not all sources of PM_{10} were represented in the model input (e.g., natural sources other than sea salt, resuspension of dust), the model would under predict actual PM_{10} concentrations. However, validation studies have shown that the variability and trends in concentrations had been captured accurately. This phenomenon is not further discussed here; for further information and discussion on model validation and performance, we refer to the BOP technical report on the LOTOS-EUROS model (Schaap *et al.*, 2009).

Variation in PM concentrations due to meteorological variations was spread equally across the country (Figure 3.1-Figure 3.3), although absolute concentrations varied from location to location. Sea salt contributions were highest for the two coastal locations (De Zilk and Kollumerwaard; Figure 3.2). Annual contributions of sea salt particulates ranged from as low as 1.5-2 µg/m³ at Vredepeel, to as high as 7 µg/m³ in 2005, at Kollumerwaard. The south and south-eastern locations (Huijbergen, Vredepeel) showed the highest concentrations due to anthropogenic emissions (Figure 3.3), which was not surprising as these locations are surrounded by highly populated areas and industrial sites (Rhine-Ruhr metropolitan region, Belgium, the Netherlands). Figure 3.1 to Figure 3.3 show that trends at the various locations are similar, there is thus no need for a separate discussion on each individual location.

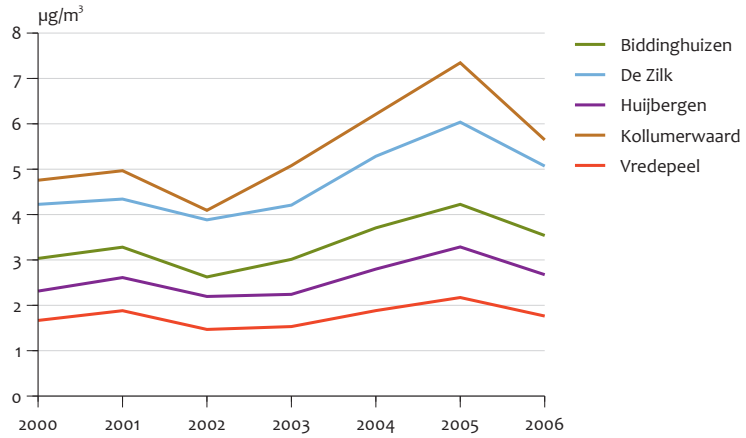
Since the model would under predict absolute concentrations, the most valuable information was the relative variation that could have been due to meteorological variation. This relative variation is a measure of how much observed values at monitoring locations, potentially, may have been influenced by meteorological conditions alone. Absolute concentrations of sea salt could also be of direct interest, as they were calculated only by the model, without needing additional correction.

Variation in total annual PM_{10} concentration levels was of the order of 10% (Table 3.2). Relative variation in sea salt PM_{10} only, was considerably higher (-25% - +35%), but since sea salt particulates are only a limited part of total PM_{10}, the influence of their variation is limited to a maximum of 2 µg/m³. Table 3.2 shows the extreme values modelled for five rural locations. For example, an absolute variation of 1.9 µg/m³ in sea salt PM was found at a coastal location, but this was not the average maximum value for all locations. However, relative variations did apply to all locations.

3.2.1 Variation in sea salt contribution

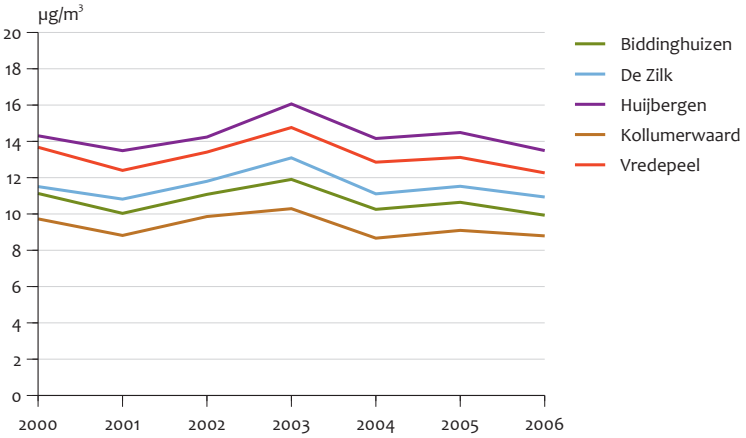
The absolute deviation in the sea salt contribution, compared to the annual average value per location, was approximately ±2 µg/m³ for coastal locations and less than ±1 µg/m³ for locations further inland (Figure 3.4). Relative variations in sea salt contributions were independent of location and could be as high as ± 25% (Figure 3.5). Please note that these variations concern the sea salt in PM_{10}, not total PM_{10}. The years 2002 (low) and 2005 (high), especially, saw extreme values for sea salt particulates.

Model-predicted PM_{10} concentrations due to sea salt particulates Figure 3.2



Model-predicted PM_{10} concentration levels due to sea salt particulates, for five rural background locations, using year-specific meteorological data.

Model-predicted PM_{10} concentrations excluding sea salt contribution Figure 3.3



Model-predicted PM_{10} concentration levels excluding sea salt contribution, for five rural background locations, using year-specific meteorological data.

Minimum and maximum annual variations in model-predicted PM_{10} due to meteorological variability, over the 2000-2006 period, for five rural locations Table 3.2

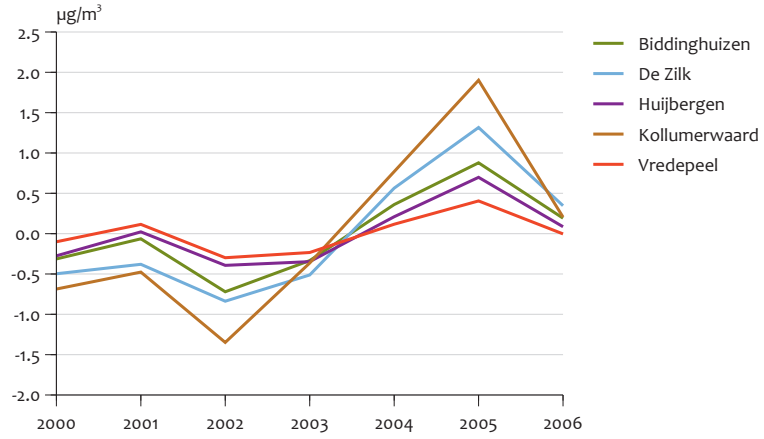
	Absolute variation		Relative variation	
	min	max	Min	max
	µg/m³		%	
Total PM_{10}	-1.1	1.7	-7%	11%
PM_{10}_sea salt	-1.3	1.9	-25%	35%
PM_{10} excl. sea salt	-1.0	1.7	-7%	13%

3.2.2 Variation in anthropogenic contributions

The variation in annual PM_{10} resulting from meteorological variability in anthropogenic PM_{10} (primary and secondary) was generally ± 5% (Figure 3.6), with extreme years causing larger variations, such that of 10 to 15% in 2003.

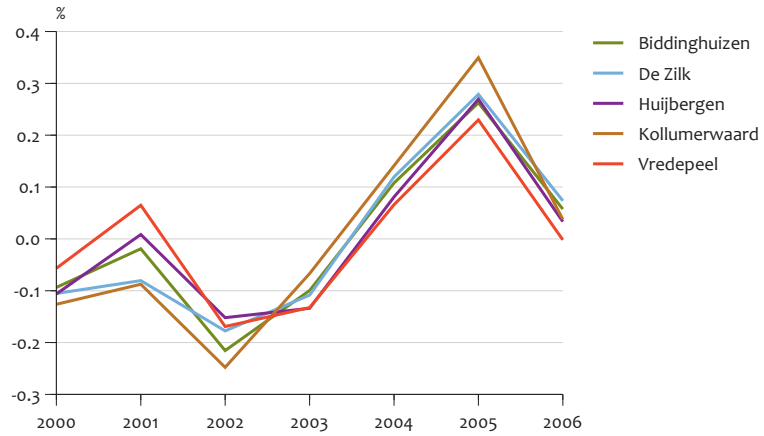
The variation in total model-predicted annual PM_{10} was slightly smaller, with maximum variations of ± 8% (Figure 3.7). This can be explained by the antagonistic behaviour in time of sea salt PM_{10} and non-sea salt PM_{10} (Figure 3.8). In certain areas that are favoured by strong westerly and north-westerly winds, large amounts of sea salt particulates may be generated, and carried to particular monitoring locations.

Absolute variation of sea salt contribution to PM_{10} compared to average sea salt contribution **Figure 3.4**



Absolute variation of sea salt contributions to PM_{10} compared to average SS contribution over 2000-2006 per location.

Relative variation of sea salt contribution to PM_{10} compared to average sea salt contribution **Figure 3.5**



Relative variation in sea salt contributions to PM_{10}, compared to average sea salt contributions in the 2000-2006 period, per location.

At these locations in the Netherlands, this implies that such air is coming from less-polluted regions, where strong winds also cause further dilution of PM_{10}. Such conditions reduce the build-up of anthropogenic PM_{10}. The existence of these opposite effects is well demonstrated in Figure 3.8.

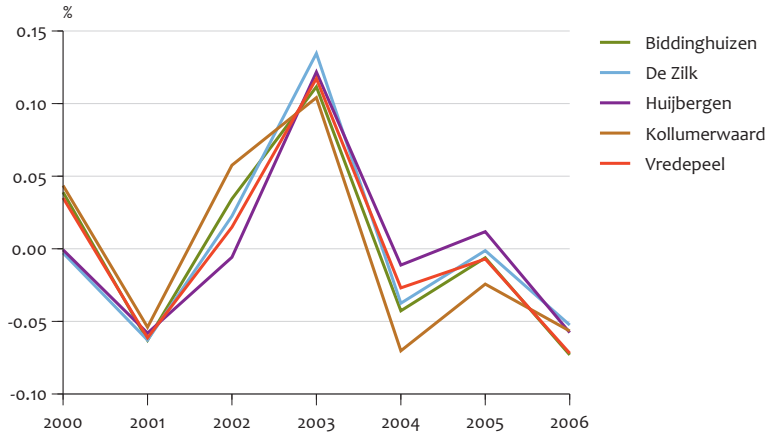
3.2.3 Variation in anthropogenic contributions, using the OPS model

The extent of exceedances of EU limit values for particulate matter (PM_{10}) concentrations are expected to decrease, significantly, within the Netherlands, in the coming years. Whether limit values will actually be exceeded in the next decade, depends not only on European, national and local policies, but also on the effects of inevitable interannual meteorological fluctuations. A study by Velders and Matthijsen (2009) provides an estimate of the effects of inevitable interannual meteorological fluctuations. Figure 3.9 shows the calculated fluctuations.

The analysis of results from calculations that were carried out with the OPS model (Van Jaarsveld, 2004) and of measurement results showed variations (1 sigma) in average annual concentrations of about 9% for PM_{10}, due to meteorological fluctuations that had their effect on the measurements. Andersson *et al.* (2007) found the same variability in PM_{10} concentration levels within Europe, based on simulations with a chemistry transport model for the 1958–2001 period.

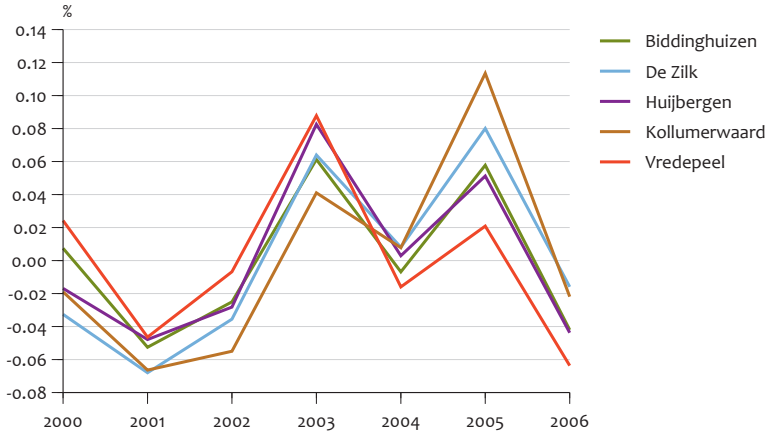
These deviations from long-term average concentration levels will affect assessments of future levels, when set against limit values. The limit value for average daily PM_{10} concentration levels is equivalent to an annual average of about 32 µg m³. At an estimated annual average of 29 µg m³, this threshold is unlikely to be exceeded for three years in a row (Velders and Matthijsen, 2009). Interannual variations in concentrations of PM_{10} are linked to large-scale meteorological fluctuations.

Relative varation in PM_{10} concentration excluding sea salt compared to average contribution **Figure 3.6**



Relative variation in PM_{10} concentration levels, excluding sea salt, compared to the average contribution in the 2000-2006 period, per location.

Relative variation in total PM_{10} concentration compared to average contribution **Figure 3.7**



Relative variation in total PM_{10} concentration levels, compared to the average contribution, in the 2000-2006 period, per location.

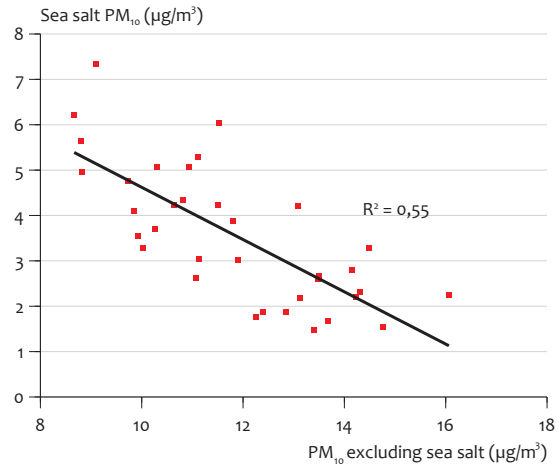
Therefore, similar results can be expected for other European countries.

3.2.4 Seasonal variation

This study has focused mostly on annual PM_{10} concentrations. However, the impact of meteorological variability on PM_{10} could also be studied on a seasonal scale. In order to do so, for each location, we grouped the monthly averages of the winter months (December, January, February), spring months (March, April, May), summer months (June, July, August) and autumn months (September, October, November). Subsequently, we determined the average concentration levels for winter, spring, summer and autumn, and for each season in the 2000-2006 period, we calculated the deviation from average seasonal concentration levels (Figure 3.10). An analysis of seasonal variations confirmed that all rural Dutch locations behaved quite similarly and followed the same trends. Coastal locations De Zilk and Kollummerwaard behaved slightly different, as they are more influenced by extreme sea salt events. Figure 3.10 shows that no trend over time was observed, which was as expected when looking at meteorological variation only.

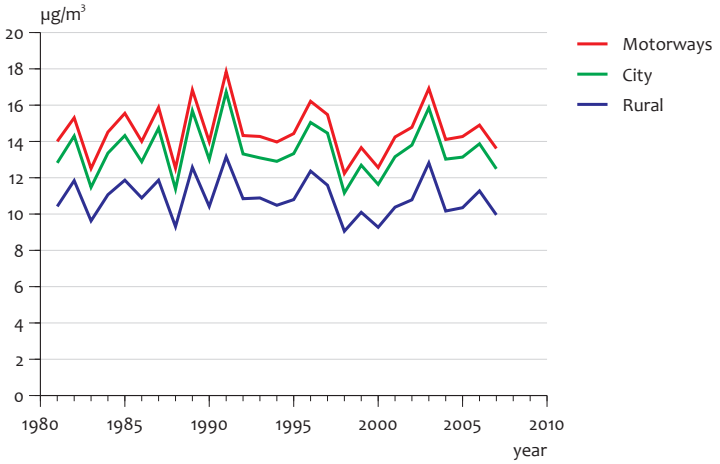
Figure 3.10 shows by how much the average concentration level for a particular season in a particular year, deviated from average seasonal concentration levels, in this example for winter PM_{10} concentration levels during the 2000-2006 period. We also studied whether model-predicted seasonal concentration levels would deviate, systematically. This was investigated by plotting the seasonal average for each season as a fraction of the overall average annual concentration level per location (see Figure 3.11). Strictly speaking, these variations were not only from meteorological effects, as, for instance in winter, additional PM_{10} would be emitted from residential heating. In general, winter and summer concentrations were higher than average annual

Model-predicted annual sea salt PM_{10} as a function of predicted annual anthropogenic PM_{10} **Figure 3.8**



Model-predicted annual sea salt PM_{10} as a function of predicted annual anthropogenic PM10, for five background locations, for the 2000-2006 period, using fixed emission data.

PM_{10} variability: Modelled **Figure 3.9**



The modelled calculations show the average background concentrations close to motorways, in cities and in rural areas. The same 2005 emission data were used in the model calculations for all years. Modelled concentrations were based on only the quantified direct 2005 anthropogenic emissions for all years. The absolute difference between measured and modelled PM_{10} concentrations arose because natural emissions or hemispheric contributions were not incorporated in the modelled concentrations. [figure adapted from Velders and Matthijsen, 2009].

concentration levels, in the 2000-2006 period (Figure 3.11). The most remarkable increases were found in the winter concentration levels at the coastal locations.

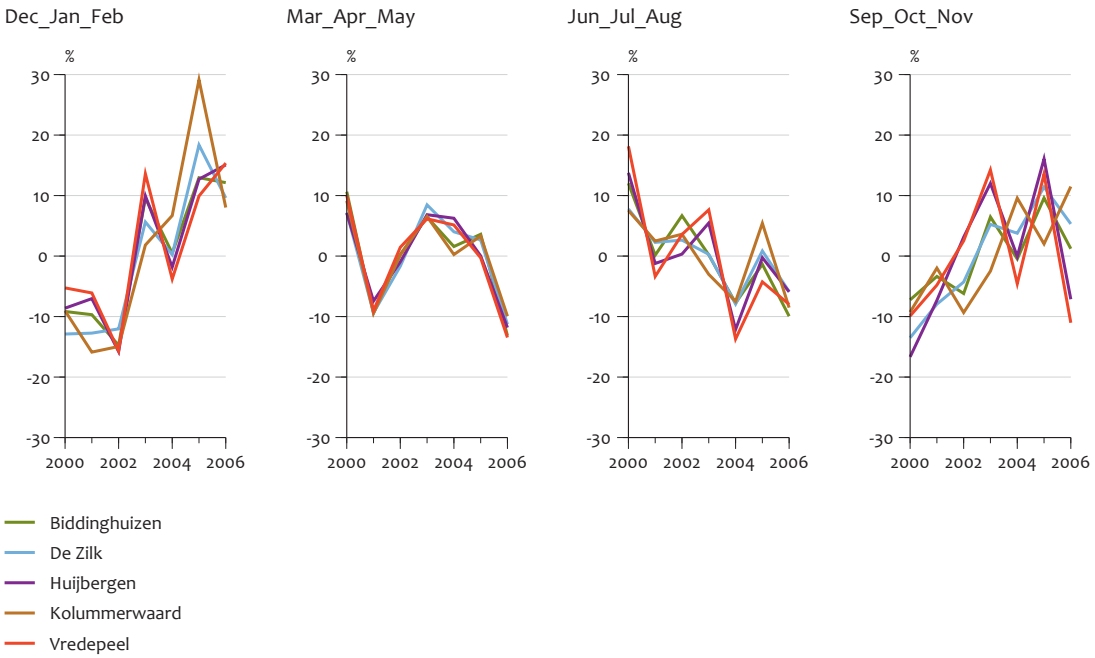
Considerable variability could also exist within seasons. This is illustrated by plotting minimum and maximum sea salt PM_{10} contributions according to season, for an inland and a coastal location Figure 3.12. Again, no trends in time could be observed. The monthly variability was not explored further.

3.3 Conclusions

The variation in model-predicted PM_{10} due to meteorological variability in the years 2000 to 2006 was calculated by using the LOTOS-EUROS model. Maximum variation in total annual PM_{10} concentrations was of the order of 10%, while the maximum variation in annual PM_{10} concentration excluding sea salt was slightly higher (around 13%). The maximum variation in sea salt PM_{10} contribution was around 25 to 35%.

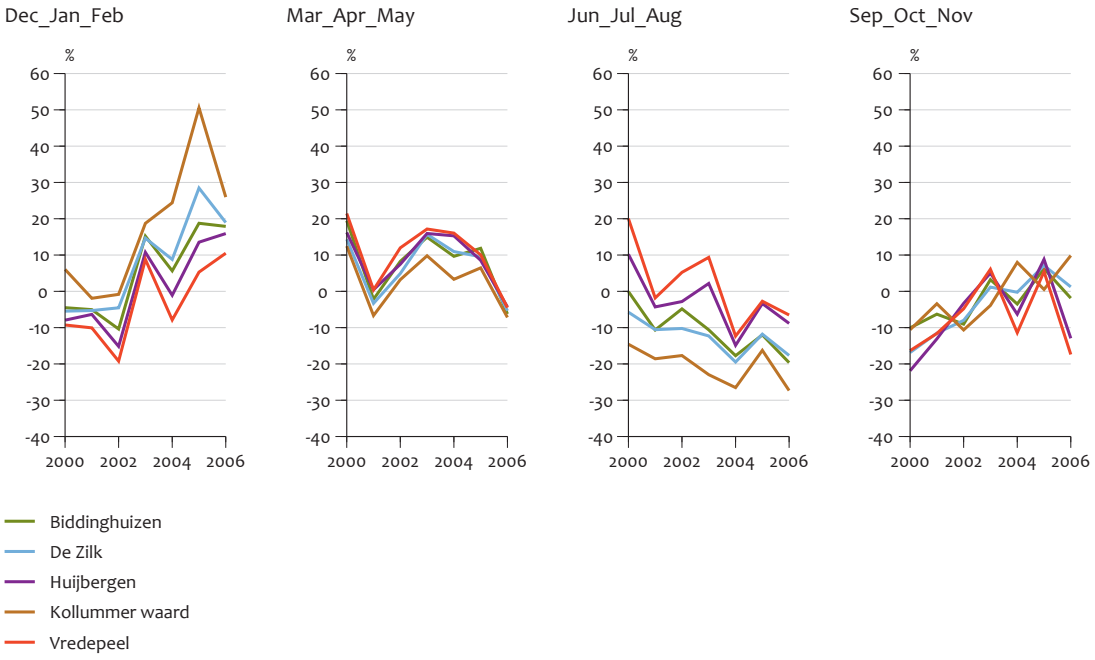
Absolute maximum deviations from the rural location average were 1.4 µg/m³ (total PM_{10}); 1.9 µg/m³ (PM_{10}_SS) and 1.7 µg/m³ (PM_{10}_excluding SS). The magnitude of this variation for sea salt was roughly in agreement with variations in observed chloride concentrations. For total PM_{10}, these variations appeared realistic, assuming that the model would underestimate absolute PM_{10} by approximately a factor of 2. Therefore, a maximum average variation in PM_{10} due to meteorological variation of around 3 µg/m³ is expected.

Relative variation in seasonal total PM_{10} for five background stations Figure 3.10



Relative variations in seasonal total PM_{10}, for five background locations (winter=DJF; spring = MAM; Summer = JJA; Autumn = SON).

Deviation of the seasonal average PM_{10} concentration from the annual average Figure 3.11



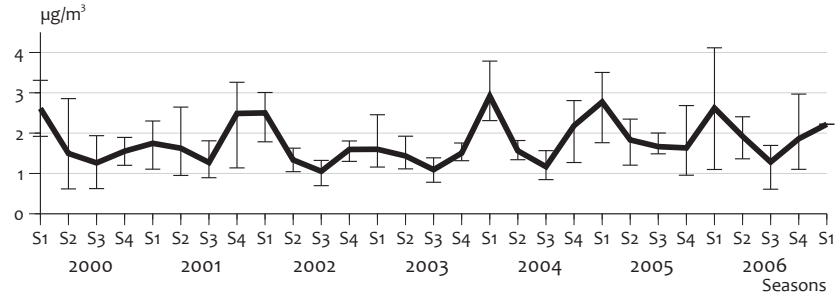
Deviation of seasonal average PM_{10} concentration levels from annual averages.

It should be noted that, for this study, we did not adjust (primary) PM_{10} source strengths for meteorological variability. Examples of such variability are increased residential heating in cold winters or elevated emissions from diffuse sources in dry years.

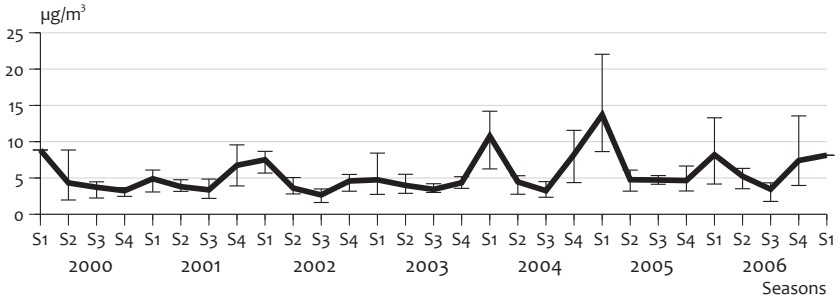
The results illustrate that meteorological variability in sea salt PM_{10} and non-sea salt PM_{10} is antagonistic. Years with strong north-westerly winds generate higher sea salt contributions to PM_{10}, while reducing PM_{10} from anthropogenic sources located in the south-eastern part of the Netherlands. During

Seasonal variation in sea salt in PM_{10} **Figure 3.12**

Location Vredepeel



Location Kollumerwaard



S1 = Dec_Jan_Feb
S2 = Mar_Apr_May
S3 = Jun_Jul_Aug
S4 = Sep_Oct_Nov

Seasonal variation in sea salt PM_{10} at an inland location (Vredepeel, top) and a coastal location (Kolummerwaard, bottom). Note the difference in scale along the Y axes.

years with strong opposite winds, the effect is vice versa. Therefore, total average annual PM_{10} behaves differently than sea salt PM_{10} or non-sea salt PM_{10}. For total PM_{10}, 2003 and 2005 were extreme years. In 2003, this was entirely due to non-sea salt contributions, while, in 2005, the deviation is mostly due to sea salt contributions.

A trend analysis of model-predicted data did not reveal any significant trends. This was expected, as we only investigated meteorological variation and assumed that the climate had not changed, dramatically, over the 2000-2006 period. The opposite is also true; the absence of a trend indicates that the 7-year period covered in the data set would be long enough to investigate meteorological variation. The exception is possibly the coastal location of De Zilk, which showed a small increasing trend for sea salt particulates caused by some relatively high seasonal data on 2005 and 2006.

Looking at the relative deviation of the average annual concentrations (Figure 3.7) and the seasonal average concentrations (Figure 3.10), we concluded that a trend in concentrations, for example, due to declining emissions, would need to be rather robust, and, in the 2000-2006 period, should exceed the 5 to 10%, otherwise it would not show up as significant.

A first step, to look more closely at possible trends due to changes in anthropogenic emissions, was to subtract model-predicted sea salt PM_{10} concentrations for a particular period (year/season/day) from measured concentrations at rural locations. However, an extreme meteorological year, such as 2003, in the middle of the 2000-2006 period, reduced the sensitivity of the measurement network for detecting trends in concentrations.

4 Detection of trends and meteorological summary

This chapter introduces some general statistical aspects of trend detection that apply to (PM_{10}) data sets, in general. The detection of trends is presented, starting from the hypothesis that the presence of a trend should be shown in contrast to the alternative hypothesis in which no trend is assumed. In the following section, the ability to detect a change in trend is introduced, for two different models. This ability critically depends on the variability in the measurement data from year to year. In previous chapters, several approaches have been described for reducing the effect of year to year variability. The third section of this chapter presents the results from these approaches, compared and summarised. The last section presents the statistical principles, applied to the original measurement data and to the meteorologically normalised data, thus providing answers into the key question of this study: do the measurement data support or conflict with emission estimates?

4.1 Statistical aspects of trend detection

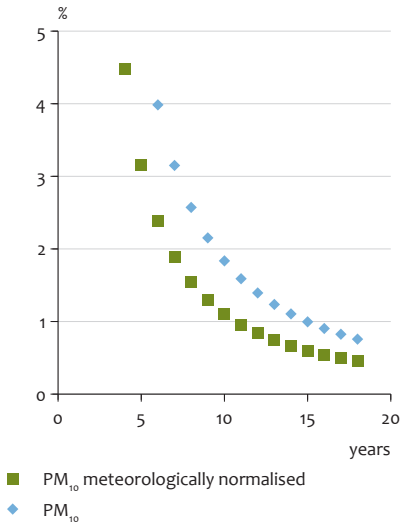
The time series of PM_{10} annual averages, for example in Figure 2.4, show fluctuations of up to 5 µg/m³ and a standard deviation of 2.5 µg/m³. With the use of this information, we estimated the length of the time series necessary for detecting a statistically significant trend. Using ordinary linear regression, the time series in Figure 2.4 is expressed as:

$PM_{10}(t_0+t) = PM_{10}(t_0)+bt.$

In this equation, the trend is described by regression coefficient b which is the average annual decrease in concentration. For a time series that includes n years, the series runs from t_0 to $t_0+(n-1)$. The standard deviation of uncertainty of b can be calculated using:

$$s_b^2 = \frac{s^2}{\sum_{t=0}^{n-1}(t-\bar{t})^2}$$

Uncertainty in a trend as function of the number of years **Figure 4.1**



Relation between the expected uncertainty about the trend in a PM_{10} time series as a function of the length of the time series.

In this equation, $\bar{t}$ is the average time from the start of the series, which is (n-1)/2. A significant trend, defined as probability < 5% for accidental occurrence, is detected approximately when $b > 2\ s_b$. Using these equations, the relation between the length of the time series and an expected significant trend, is shown in Figure 4.1. The uncertainty is expressed in the percentage of the average concentration by dividing the uncertainty in µg/m³ by 30 µg/m³. When applying meteorological normalisation, the standard deviation for annual averages typically reduces to 1.5 µg/m³. For comparison, Figure 4.1 also shows the detectable trends based on the meteorologically normalised data.

Figure 4.1 shows that, in order to detect a small trend of 1 %/year (0.3 µg/m³/year), a series of approximately 15 years would be necessary. This can be reduced to 11 years after the described meteorological normalisation. Large trends of, for example, 3 %/year (1 µg/m³/year) would still need 7 years to be detected without, and 5 years with meteorological normalisation.

4.2 When can changes in trends be observed?

Both measurements and emission data suggest that the downward trend in average PM_{10} concentrations was significantly larger in the 1993-2000 period than in the 2000-2008 period. To prove this hypothesis, we tried to detect a change in trend by using a second-order non-linear model ($PM_{10}(t_0+t) = PM_{10}(t_0)+bt+ct^2$). However, this yielded no statistically significant trend.

Using this model, the theoretical standard deviation of uncertainty s_c can be calculated using

$$s_c^2 = \frac{s^2}{\sum_{t=0}^{n-1}(t-\bar{t})^4}$$

An interesting change in trend might be that of a decrease of 2% annually, in the years at the beginning of a time series, which then changes to 1% annually, at the end of the time series. In such a situation, the uncertainty about c multiplied by the length of the time series should be lower than 2 * 1%.

Another approach, more close to the original observation, would be to explicitly determine the trends in two consecutive periods. For simplicity, we define t_0 as the year with the change of trend.

The model for the annual average would thus read:

$PM_{10}(t_0+t) = PM_{10}(t_0)+ b_1*t*\theta(-t)+ b_2*t*\ \theta(t)$

With $\theta(t)=0$ for $t<0$ and $\theta(t)=1$ for $t>0$

In this set-up, the change in trend $\Delta=b_1-b_2$.

$$s_\Delta^2 = s_{b1}^2 + s_{b2}^2 = \frac{s_1^2}{\sum_{t=0}^{n_1-1}(t-\bar{t})^2} + \frac{s_2^2}{\sum_{t=0}^{n_2-1}(t-\bar{t})^2}$$

When two time series are equal in length and the standard deviation of annual averages is assumed to be the same for both time series, the equation reduces to:

$$s_\Delta^2 = 2\frac{s^2}{\sum_{t=0}^{n_1-1}(t-\bar{t})^2}$$

When, for example, the first time series would be much longer than the second, the s_Δ can be approximated by the s_2 of the shortest time series, which would be a factor of $\sqrt{2}$ smaller than for time series of equal length.

Figure 4.2 shows the uncertainties for both theoretical approaches to detect a change in trends. Compared to Figure 4.1 obviously, the detection of a change in trend is much more uncertain than determining the trend itself. Figure 4.2 also shows that the difference in trend detection between the second-order approach and the piecewise, two-segments approach is quite small.

4.3 Overview of meteorological normalisation methods

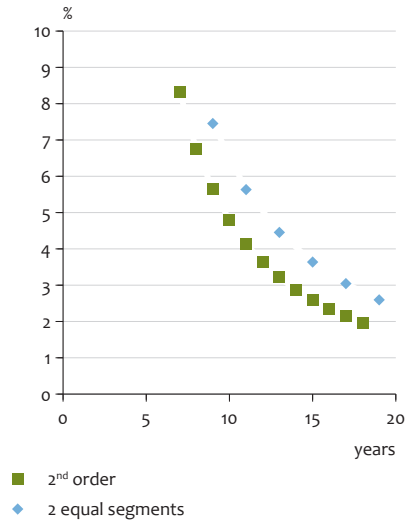
Chapter 2 presents the results from two empirical methods of meteorological normalisation (based on measurements). Chapter 3 presents the results from two meteorological normalisation methods that were based on runs of two different air quality models. Figure 4.3 shows the results from those four methods in one single figure, for comparison. Both a relative and an absolute measure of meteorological influence on PM_{10} concentrations are given. Both measures were calculated on the basis of data within one method, that is, the absolute difference was defined as $(PM_{10}$ concentrations$)_x$ minus $(PM_{10}$ concentrations meteorologically normalised with method X$)_x$, with X indicating either multivariate regression (MRV), classification and regression tree (CART), LOTOS-EUROS (LE) or OPS. For the relative meteorological factor, the absolute difference is scaled with $(PM_{10}$ concentrations$)_x$.

It is relevant that the meteorological normalisation results are provided both in relative and absolute terms, since the amount of modelled PM_{10} concentrations differed substantially from the amount of measured PM_{10} concentrations. As a result, result from a comparison of the model-based methods with the measurement-based methods can also differ, substantially, either in relative or absolute sense (see, e.g., the large relative meteorological factor for OPS model, for the years 1998 and 2000, which, in absolute terms, is comparable with that for the MVR and CART methods).

At first glance, the results from the four methods appeared to be similar. The two years (1996 and 2003) with the largest meteorological influence on PM_{10} concentrations were detected in all four methods, and also the general pattern for the other years is quite similar. However, a more detailed comparison revealed several anomalies between the four. For LOTOS-EUROS (LE), for example, the results, although comparable for most years, were rather different from those of the other three methods, for several of the years. For

Uncertainty in detecting a change in trend

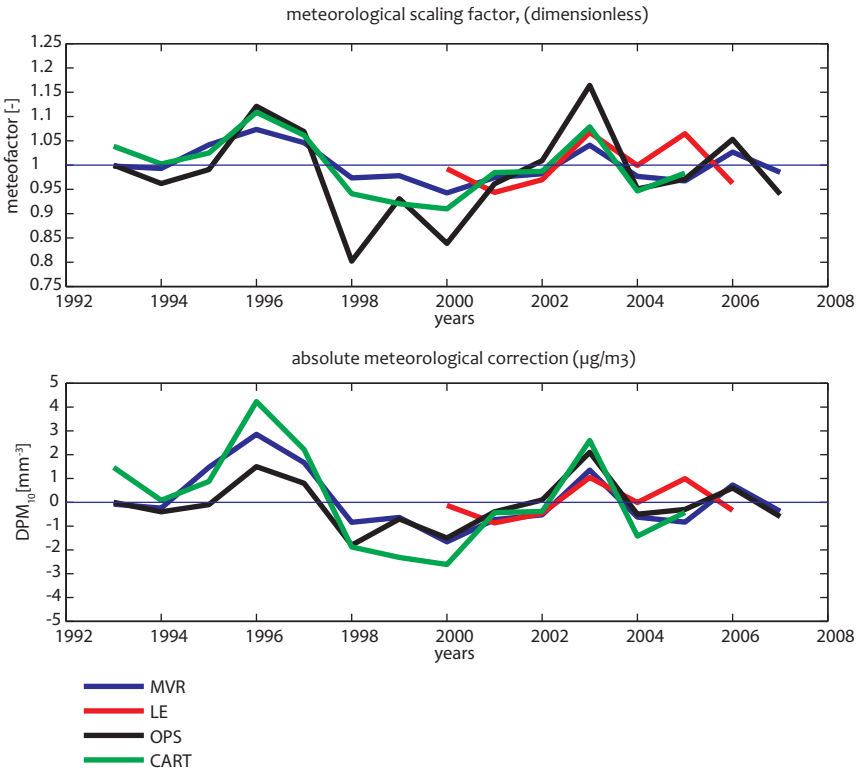
Figure 4.2



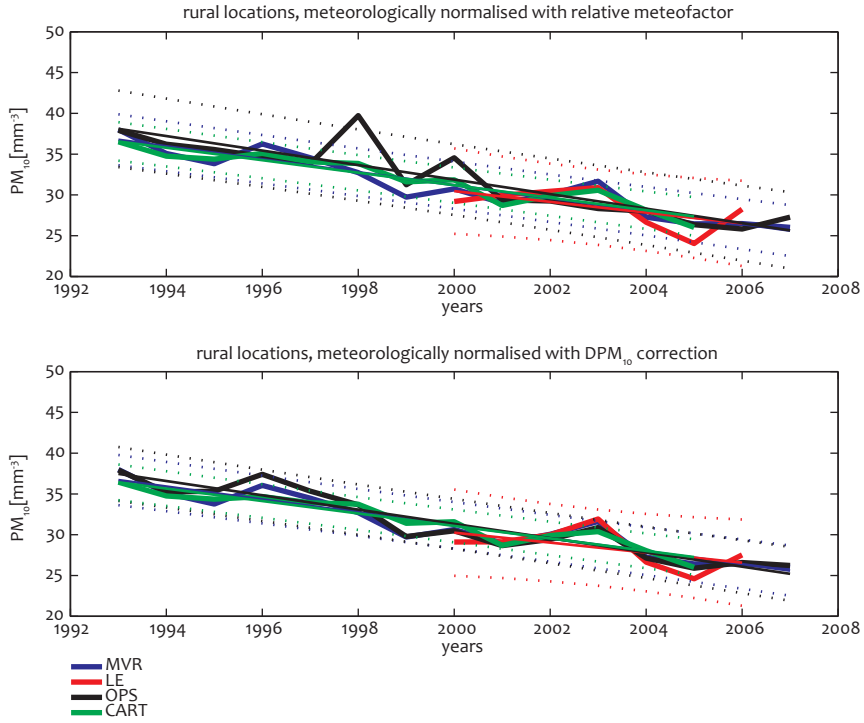
Uncertainty in the detection of a change in trend, for two approaches: the second-order approach and the two-segments approach (segments of equal length).

Meteorological corrections for average annual PM_{10} concentrations at rural background locations

Figure 4.3



Comparison of the multivariate regression (MVR) meteorological normalisation method to the classification and regression tree (CART) method of Visser and Noordijk (2002), and to the two meteorological normalisations based on LOTOS-EUROS (LE) and OPS model runs. The upper panel presents the relative meteorological normalisation per method; the lower panel presents the same results in absolute PM_{10} concentration differences.



Trend line and 95% confidence limits for the trend line of the average annual PM_{10} concentrations at the rural locations. The average annual PM_{10} concentrations were meteorologically normalised, either with the relative meteorological factor (upper panel) or the absolute PM10 concentration differences (lower panel).

the year 2000, the deviations were rather large but still of the same sign, while for 2005 and 2006, the meteorological influence was of opposite behaviour. It is difficult to judge which method would be more correct; the similarities between MVR (and CART) and OPS is encouraging, however, LE is the only method that takes the effect of long-range transport (which is important for PM_{10}) into account.

Applying meteorological normalisation to PM_{10} measurement time series from the rural locations would be another way of evaluating the issue. The meteorological normalisation method, which is the most realistic, was expected to present the smoothest time series with the smallest confidence limits. For this study, we took the PM_{10} time series as presented in Figure 2.4, and applied the meteorological normalisation, similarly as was described in Section 2.1. The results are presented in Figure 2.1. Both absolute and relative corrections were applied. For the MVR method, both corrections were identical, and for CART the differences were (and were expected to be) negligibly small. However, for OPS, the differences proved to be fairly large.

The MRV method had only slightly larger confidence limits than the CART method. However, the former method is easier to apply and currently available for a longer time series. Therefore, it was the preferred meteorological normalisation method in our study.

The LE and OPS methods proved less effective. On one hand, the superior effectiveness of the empirical methods could be understood, since they are explicitly optimised for this normalisation. On the other hand, especially the LE method has the potential for improvements, since it incorporates more relevant processes and larger scales. This potential seems not yet fully achieved, while the results from the OPS model appeared remarkably effective in the meteorological normalisation.

For the deterministic model-based methods, the absolute meteorological normalisation correction is thought to be slightly more realistic, since the relative scaling also influences components which might have completely different meteorological effects. For example, sea salt is not incorporated in the OPS model calculations.

The analysis of (OPS) model calculations of measurements yielded variations (1 s) in the average annual concentrations of about 9% for PM_{10}, due to meteorological fluctuations. Andersson *et al.* (2007) also found the same variability in PM_{10} concentrations within Europe, based on simulations with a chemistry transport model for the 1958–2001 period.

These deviations from long-term average concentrations will affect future assessments of concentration levels, set against limit values. The limit value for average daily PM_{10} concentrations is equivalent to an annual average of about

Standard deviations for original and normalised measurement data, with respect to a linear trend Table 4.1

Data set	s [µg/m³]	reduction
Measurements 1993-2007	2.2	
*Measurements*0.9*	2.1	
MVR	1.4	36%
LE (2000-2006)	2.2	0%
OPS	1.5	32%
CART (1993-2005)	1.0	55%

Results from a linear, second-order analysis, and piecewise first-order analyses, for different variations of the rural data set. Uncertainties reflect 95% confidence estimates. Table 4.2

	y=a+bx+cx^2			y=a+b1(x<2000)x+b2(x>2000)x			
Rural 1993-2008	**a**	**b**	**C**	**a**	**b1**	**b2**	**b1-b2**
Original	31.0	-0.9±0.3	0.01±0.06	30.4	-1.1±0.6	-0.7±0.5	-0.4±0.9
*original *0.9*	28.1	-0.6±0.3	0.03±0.05	27.2	-1.0±0.6	-0.2±0.5	-0.8±0.8
Meteo	31.0	-0.8±0.2	0.01±0.04	30.8	-0.9±0.4	-0.7±0.3	-0.2±0.6
*Meteo*0.9*	28.1	-0.5±0.2	0.03±0.03	27.6	-0.9±0.3	-0.2±0.3	-0.6±0.4
Typical emission trend %		-2±1%			-3±1%	-1.5±1%	
Typical emission trend µg/m³		-0.6±0.3			-0.9±0.3	-0.5±0.3	

Significant slopes are marked in green; significant changes in slopes are marked in yellow. For comparison, typical emission trends are shown.

32 µg/m³. This threshold is unlikely to be exceeded for three years in a row, when an annual average concentration of 29 µg/m³ is estimated. Interannual variations in concentrations of PM_{10} are linked to large-scale meteorological fluctuations. Therefore, similar results can be expected for other European countries.

4.4 Trend analysis based on the rural data set

This section presents the application of the statistical diagnosis as described in Sections 4.1 and 4.2, to the PM_{10} measurement data from the rural locations. The rural data set was selected, since the other data sets were much smaller. This analysis was done for both the original measurement data as the meteorologically corrected data, as calculated by using the multivariate empirical equations (MRV method). In addition, the sensitivity to uncertainty due to the change of instrument – as applied in 2003 – was studied. Therefore, we introduced an alternative data set with pre-November 2003 measurement results, scaled by an extreme value of to 0.9. These 2*2 options in total led to 4 data sets. From these data sets, the following statistics were obtained (all slopes and changes in slope with 95% confidence intervals):

- linear trend for the 1992-2008 period;
- second-order trend for the 1992-2008 period;
- linear trend for the 1992-2000 period;
- linear trend for the 2000-2008 period;
- difference between first and second period;

Table 4.2 presents a summary of the results from these analyses.

For the considered period of 1993 to 2008, the long-term trend for the rural locations changes only marginally, due to meteorological normalisation, from -0.9 to -0.8 µg/m³/year; (a similar difference was found for the other two types of locations). The applied meteorological normalisation model reduced the margins for the 95% confidence limits for the regression estimates by roughly a third, as is also shown in Figure 4.4. For the total data set, the influence of the uncertainty on the measurement calibration changes the trend from -0.9 to -0.6 µg/m³/year. For the data set on the whole period, the trends are highly significant, both to the length of period and the slope of the trend.

Two methods for a change in slope detection were applied. The first approach, for the second-order analyses, yielded statistically insignificant results for all four data sets (however, the scaled and normalised data set was quite close to being significant). The second approach, the piecewise regression, yielded significantly different results for both scaled datasets. Table 4.2 also summarises typical emission trends for primary PM_{10} and precursors, on various geographical scales (see Chapter 6 for more detailed data). Comparing the measured slopes with the estimated uncertainties showed the emission trends to be well within these ranges.

Each line in Table 4.2 contains uncertainty estimates based on statistical algorithms that assume variations from year to year to be mutually independent. Due to the change of monitor in this period we might introduce a systematic source of uncertainty that obviously violates the assumption of independence. A simple approach to test the influence of a systematic change on trend analyses was a comparison with the alternative '0.9' data set. In this approach, trends were significant only when this significance was shown for both alternatives. Applying these more complete uncertainty analyses, we arrived at the following three statements:

- For the 1993-2000 period, the trend is significant
- For the 2000-2008 period, the trend is not significant
- The change in trend is not significant

Please note that the statements seem to contradict.

These sensitivity analyses using two alternatives presumably have overestimated the uncertainties. For future research, a more sophisticated approach is in preparation.

Conclusions

1. Trends in PM_{10} measurements at rural locations, on three time scales – 1993 to 2008, 1993 to 2000, and 2000 to 2008 – are in line with estimated trends in emissions
2. For the measurement periods of 1993 to 2008 and 1993 to 2000, unambiguous significant trends could be proven
3. For the measurement period of 2000 to 2008, an unambiguous significant trend could *not* be proven
4. When using a second-order, curved trend, a significant *change* in trends could *not* be proven
5. When using a piecewise regression method, an unambiguous *change* in trend could *not* be proven.
6. Systematic measurement uncertainties influenced conclusions 3 and 5.
7. Meteorological variations had a large effect on annual PM_{10} averages (2sd = 4-5 µg/m³). Various methods for meteorological normalisation typically reduced the variation by 30% (2sd = 2-3 µg/m³).

5 Trends in PM_{10} and particulate components at rural locations; emissions, measurements and model results

Average annual PM_{10} concentrations decreased, on average, by 0.7 and 1.0 µg/m³ per year, between 1993 and 2007, at rural locations in the Netherlands (see Chapter 2). This broad range was caused by large uncertainties that were introduced by instrument changes. This chapter addresses the questions: Is the observed trend in line with relevant anthropogenic emission changes? Do we understand why the PM_{10} trend has levelled off after 2000?

Therefore, a bottom-up estimate on PM_{10} concentration changes between 1993 and 2007 is presented, starting from the registered anthropogenic emissions relevant to PM_{10}: directly emitted particles (primary PM_{10}), sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3), and non-methane volatile organic compounds (NMVOC). The gases are in part chemically and/or physically converted to secondary particles in the air.

In addition, concentrations and emission trends in anthropogenic PM_{10} components were investigated, to obtain a better general understanding of the trends in anthropogenic particulates. The Dutch National Air Quality Monitoring Network (LML) (RIVM, 2009) contains time series of PM_{10} observations and related species, starting back in the 1990s and sometimes even earlier. Table 5.1 shows the monitoring specifications on the measured particulate substances.

Subsection 5.1 presents trends in observed concentrations of secondary inorganic aerosol (SIA), collected at rural locations. Subsection 5.2, presents trends in emission data for the Netherlands and nearby countries, from registered anthropogenic sources that are relevant to PM_{10} concentrations in the Netherlands. Subsection 5.3 describes the analyses of Dutch trends in observed concentrations of the anthropogenic PM_{10} components black smoke and heavy metals in particulate matter. In addition, we investigated the relevant emission changes for these components, to substantiate our knowledge on the relation between anthropogenic particulate matter concentrations and emissions.

Subsection 5.4, presents an evaluation of the observed PM_{10} trend, via a bottom-up approach using measurements and model results, with European registered anthropogenic emissions for SO_2, NO_x, NH_3, NMVOC and primary PM_{10} as input. In Subsection 5.5, we present a discussion on the trend in PM_{10}, in the 1993-2007 period, by integrating the different pieces of information from the preceding subsections.

Monitoring specifications on particulate substances routinely measured in the Dutch National Air Quality Monitoring Network (LML) **Table 5.1**

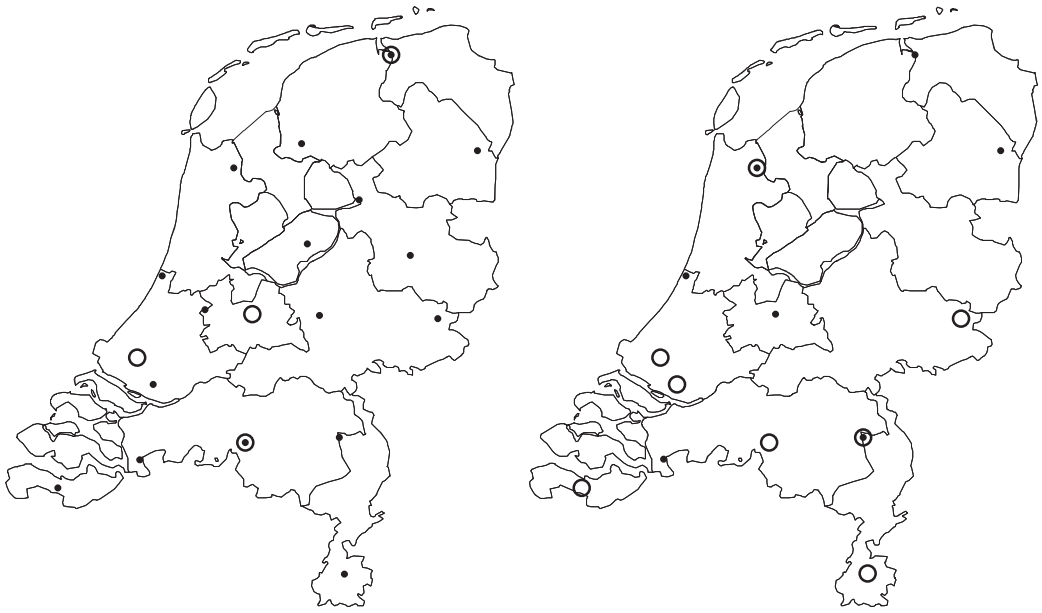
Substance	measurement method	measurement frequency	particle cut-off diameter (µm)
PM_{10}	Beta attenuation type: FH62 I-R/N [a)]	hour	10
Secondary inorganic aerosol NO_3^-, SO_4^{2-}, NH_4^+ and Cl^-	low volume sampler	day	3 – 4 [b)]
Heavy metals As, Cd, Pb, Zn	medium volume sampler	every other day	6 – 7 [b)]
Black smoke (BS)	EEL 43 [c)]	hour	n/a

a) in 2003, the PM_{10} network's old monitor types, FH62 I-N, were replaced by the newer type FH62 I-R.
b) up to 2008, in lvs and mvs measurements, an open tube inlet was applied. Cut-off estimate based on empirical data.
c) in 2002, the BS measurement method was adapted (Bloemen *et al.*, 2007, 863001004).

Rural measurement locations in the Netherlands. **Figure 5.1**

PM_{10} (dots) and heavy metals (circles)

secondary inorganic aerosol (dots) and black smoke (circles)



Rural measurement locations in the Netherlands. On the left: PM_{10} (dots) and heavy metals (circles); on the right: secondary inorganic aerosol (dots) and black smoke (circles).

5.1 Secondary inorganic aerosol

Secondary inorganic aerosol (SIA), consisting of nitrate, sulphate and ammonium, together with chloride ions, in particulate matter (abbreviated as: NO_3, SO_4, NH_4 and Cl) has been monitored since 1993, at seven rural locations throughout the Netherlands (see Figure 5.1). SIA components form an important fraction of the PM_{10} mass and are mainly the result of anthropogenic emissions of SO_2, NO_x and NH_3 in Europe. Sea salt sulphate and contributions to SIA from across the Atlantic Ocean, together, contribute a relatively small amount (about 10% or less). Therefore, understanding the trend in SIA from anthropogenic emissions in Europe, helps us to understand the trend in PM_{10}.

The SIA measurements obtained in the BOP research programme are believed to give a better estimate of current SIA concentrations in the Netherlands than those from the routine SIA measurements conducted in the Dutch National Air Quality Monitoring Network (LML). The latter, long time series of measurements of particulate sulphate, nitrate and ammonium were started in the early 1990s. The method used for capturing and sampling PM differs from the EU reference method for PM_{10} (EN 12341: 1998). Therefore, differences between 'BOP' measurements and those taken in the LML network are to be expected. The national BOP research programme found that the routine (LML) SIA measurements in 2007 and 2008 had underestimated (by about 40%) SIA concentrations measured in PM_{10} obtained with the reference method (Weijers *et al.*, 2010). These results have been confirmed by Hafkenscheid et al. (2010). The large differences between both measurement results constitute a major uncertainty for the trend analysis of PM_{10} and its secondary inorganic components (see also Text box *SIA measurement limitations*).

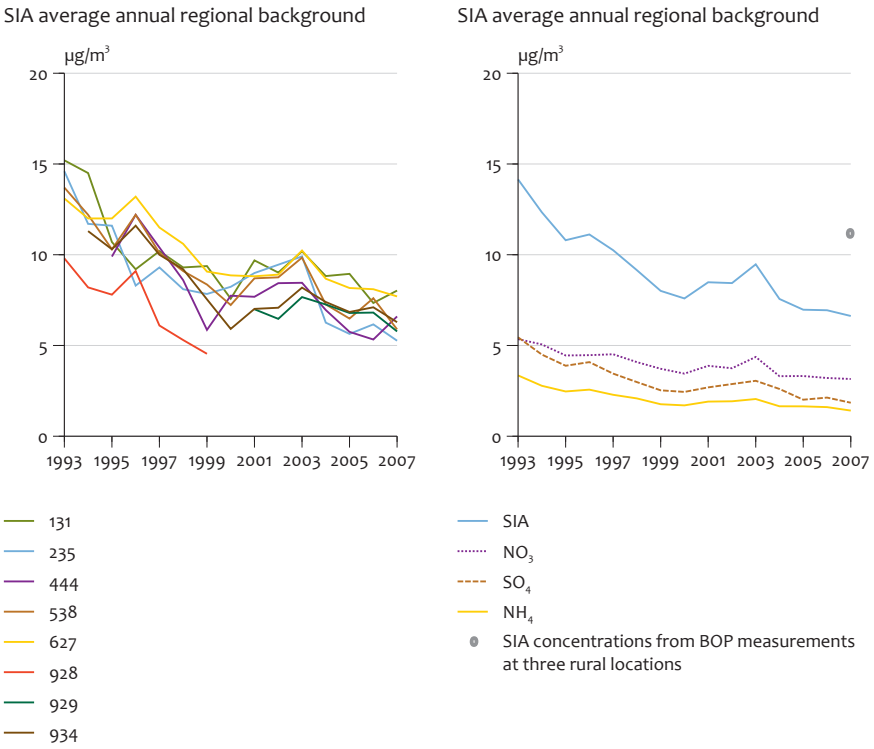
5.1.1 Trends in secondary inorganic aerosol

The subject of long-term trends in routine measurements of SIA and its components is treated in Weijers et al. (2010). Here, we have confined ourselves to providing an overview of their main findings.

The calculated linear trends at the various locations in the national network do not differ much per component. At 5 out of 6 locations the annual decrease is quite similar: 0.16-0.18 µg/m³ (SO_4), 0.12-0.14 µg/m³ (NO_3), and 0.08-0.09 µg/m³ (NH_4). The weakest trends over the entire, 14-year period were observed at one specific location (Vredepeel), which is situated within a region known for its high NH_3 emissions from agriculture. At this location, no trend was discovered for NO_3. Downward trends for the 1994-2000 period were (somewhat) stronger than between 2001 and 2007. However, during the second most recent period, negative regression coefficients were found for most locations, although these were of less significance. Therefore, it was concluded that the decreasing trends observed over the 1994-2000 period had slowed down after the year 2000, and in some cases became insignificant.

From year to year, considerable concentration variations took place, which could be related to specific meteorological circumstances. The observed spatial variabilities were similar for SO_4, NO_3 and NH_4 (on average about 15%), and from year to year the spatial variability varied within a range of 10 to 25%.

Average annual secondary inorganic aerosol concentrations **Figure 5.2**



Average annual secondary inorganic aerosol (NO_3+SO_4+NH_4) concentrations (µg/m³) in the Netherlands, between 1993 and 2007, measured at seven rural locations (left panel). Average annual SIA concentrations and components (right panel), based on measurements in a time series of over 10 years; NH_4 (thin solid line), SO_4 (dashed line), NO_3 (dotted line). The single grey dot indicates the average SIA concentration from observations at three rural background locations, obtained in the BOP research programme on PM_{10} and $PM_{2.5}$ (Weijers et al., 2010). The average SIA concentration (according to 'BOP') is representative of the 2007 annual average, for all rural PM_{10} measurement locations in the Netherlands.

5.2 Emissions of particulate matter and SIA precursor gases

Emissions of different substances lead to concentration levels of PM_{10} after transport in air, possible transformation and removal. PM_{10} is usually classified to be primary or secondary, depending to the formation mechanism. Primary particles are emitted directly into the atmosphere and are either anthropogenic or the result of natural emissions, such as of sea salt. Secondary particles are those formed in the air through chemical reactions of gases, such as sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and (non-methane) volatile organic compounds (NMVOC). Such secondary particles largely found in the fine fraction ($PM_{2.5}$) (Weijers *et al.*, 2010; Brink *et al.*, 2009).

5.2.1 Anthropogenic primary and secondary PM_{10}

The current knowledge on the chemical composition of PM_{10} in the Netherlands shows that SO_2, NO_x, and NH_3 are the main contributors to PM_{10} through the formation of secondary inorganic aerosol, about 7 to 10 µg/m³ at rural background locations. Secondary organic aerosol that results from NMVOC contributes far less to PM_{10}. Levels are very uncertain, but no larger than 1 to 2 µg/m³, on average, at rural background locations. Primary emitted particles constitute an important fraction of PM_{10}; they chemically consist of a range of components, including elemental carbon, organic carbon, metallic particles, and oxides of metals and silicium. At rural background locations, the anthropogenic primary fraction is believed to contribute about 4 to 5 µg/m³ to PM_{10}. Therefore, in the Netherlands, anthropogenic emissions contribute at least 50% to the PM_{10} concentration at rural background locations. Consequently, contributions from natural sources to PM_{10}, such as sea salt, is also important and even can be dominant under specific circumstances (Manders *et al.*, 2009; Denier van der Gon *et al.*, 2009).

Emission data are used as input for models to assess historical and future PM_{10} levels, origins and composition. In general, most models use anthropogenic primary emissions and SO_2, NO_x and NH_3 emissions, which are relatively well known. The other sources, including natural emissions, are still very uncertain, as is their effect on levels of particulate matter.

The PM_{10} model runs, presented in the next subsection, followed the general approach, that is, they were based on anthropogenic (European) emissions of primary PM_{10} and the PM precursor gases SO_2, NO_x and NH_3. Below, the emission data, relevant to the Netherlands, for SO_2, NO_x, NH_3 and NMVOC, are presented and discussed.

5.2.2 The origin of secondary inorganic aerosol

The countries and regions that, together, contributed 80% or more to the SIA concentration levels in the Netherlands, by emitting SO_2, NO_x and NH_3, are the Netherlands itself, France, Belgium, Germany, the North Sea and the United Kingdom (Ruiter *et al*., 2006; EMEP, 2007; Schaap *et al*., 2009). Hereafter, they are called the *relevant emission areas*. More than 70% of sulphate in the Netherlands originates from these areas; for nitrate this is more than 85%, and for ammonium more than 90% In addition, SO_2 emissions from Poland and the Czech Republic contribute significantly to concentrations in the Netherlands; in 2000, this was about 15%. Emission changes in other areas are of minor importance to the SIA concentrations in the Netherlands. The main contribution of primary PM_{10} to total PM_{10} also originates from the *relevant emission areas*.

The emissions of primary PM_{10} and precursor gases of secondary aerosols have been monitored for about two decades, and, during this period, the uncertainty about the emissions has been reduced (Figures 5.3 and 5.4, Tables 5.2 and 5.3).

For the *relevant emission areas,* the total emissions of SO_2, NO_x, NH_3 and NMVOC declined by about 80%, 35%, 20% and 55%, respectively, between 1990 and 2006. Relative emission reductions in SO_2 in the Netherlands are smaller compared to those in the *relevant emission areas* as a whole, whereas the relative reduction in NH_3 emissions was considerably larger in the Netherlands. The relative decline in primary PM_{10} emission reductions in the Netherlands was similar to the relative decline for the *relevant emission areas* as a whole. Primary PM_{10} emissions reduced by about 25%, between 1995 and 2006.

SIA measurement limitations

Weijers et al. (2010) and Hafkenscheid et al. (2010) quantified the difference between routine SIA measurement series (LML) of NO_3, SO_4 and NH_4 (Figure 5.2) and those SIA measurements based on filters obtained according to the reference methods for $PM_{2.5}$ and PM_{10}. The non-reference SIA measurements result in much lower concentration levels, between 30 and 40%, than those for the individual SIA components measured in $PM_{2.5}$ and PM_{10}. These findings were further corroborated by an earlier study and by SIA measurements abroad:

- Van Putten and Mennen, (1998) found an underestimation of 10 to 25%, based on an intercomparison of SIA measurements with different instruments in a laboratory setting.
- Preliminary results from comparisons with recent simultaneous SIA measurements in Belgium, the Netherlands and Germany confirm that routine measurements are likely to underestimate actual SIA concentration levels.

The instruments used in the LML network for the routine SIA measurements appear to introduce a bias, although it should be emphasised that the measurement results will need further corroboration before final conclusions can be drawn on these findings. The uncertainty about the magnitude of the bias clearly has influenced our trend analysis. It caused us to follow a two-track sensitivity approach, to view the trend in SIA and its contribution to the trend in PM_{10}:

1. We kept to the regular interpretation, *SIA 1993-2007*, of the routine measurements (and model results) that have been used, so far, although they would underestimate the PM_{10} SIA concentration levels.
2. We considered a second case, *high SIA trend*, simply by scaling measured and modelled time series to fit the PM_{10} 'BOP' concentrations for 2007 and 2008, using constant correction factors per SIA component. This case provided an upper-limit effect for the impact of SIA precursors on concentration changes between 1993 and 2007.

Although concentrations levels that resulted from routine LML observations of the SIA components NO_3, SO_4, and NH_4 might be biased low, the first approach was still considered worthwhile for a number of reasons:

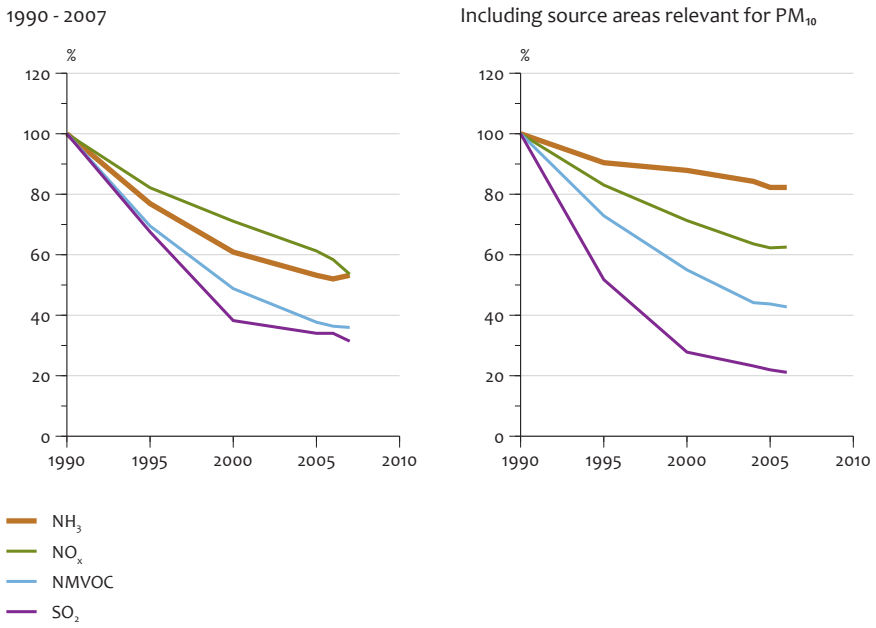
- The SIA trend according to the 1993-2007 measurements may still be correct. The SIA concentrations were measured with one and the same instrument between 1993 and 2007. Therefore, it is plausible that, although absolute concentrations were underestimated, the observed SIA trend is not far off from the actual SIA trend when measurements would have been obtained from PM_{10} filters used in the reference method. This hypothesis is subject to future research.
- The available routinely measured SIA concentrations constitute the only long time series of SIA measurements for the Netherlands.
- These measurements for the 1993-2007 period, together with model results for same period, are the basis for the current view on the effect of policies that affect SO_2, NO_x and NH_3 and, consequently, PM_{10} (e.g. Velders *et al*., 2009).
- The concentrations form the basis for our current understanding of SIA in the Netherlands.

The SIA 1997-2007 case also is affected by several other uncertainties; measurement uncertainty, detection limit, and the fact that SIA is measured from particles with a cut-off size that has 50% transmission of about 3 to 4 µm and, presumably, with a rather flat cut-off curve. Both the 50% transmission and the shape of the curve are different from those of the PM_{10} reference method.

The high SIA trend case, per definition, is considered bias free, compared to the reference method that was used for the for PM_{10} concentration measurements in 2007 and 2008, although the scaled concentrations for these years become more and more uncertain when going further back in time.

Both data sets have complications in comparison to modelled data, because particles on the filter may react with gases. The traditional method included a denuder to prevent these type of reactions. This comparability with models and between data sets is the subject of a consecutive study, to be conducted in the new research programme of BOP II.

Emissions trends of NO_x, SO_2, NH_3, and NMVOC for the Netherlands **Figure 5.3**



Emission trends in NO_x, SO_2, NH_3, and NMVOC, for the Netherlands between 1990 and 2007 (left panel), and for the Netherlands including the source areas relevant to PM_{10} in the Netherlands: Belgium, France, Germany, the North Sea, and the United Kingdom, for the same period (right panel). For absolute emission numbers see Table 5.2.

Annual emissions of NH_3, NO_x, SO_2, NMVOC and primary PM_{10} (kilotonnes), for the Netherlands (PBL, 2008; MB) **Table 5.2**

	1990	1995	2000	2005	2006	2007
NH_3	250	192	152	133	130	133
NO_x*	560	460	398	343	327	300
SO_2*	191	129	73	65	65	60
NMVOC*	459	319	224	173	167	165
PPM_{10}*	75	55	45	38	38	37

*Emission from sea shipping that reach the Dutch continental shelf have not been included

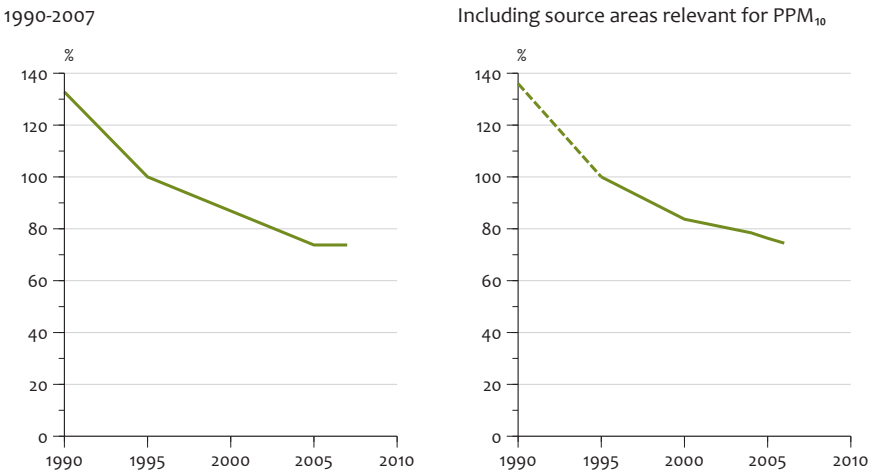
Between 1990 and 2006, the primary PM_{10} emissions probably halved.

All emissions from the *relevant emission areas* showed the largest decline between 1990 and 2000 (Figure 5.5). For these areas, the trends for the different substances varied between 1 and 7%/year, and, in the Netherlands between 3 and 6%/year. Following this period, emissions have continued to decrease, although trends have levelled off, more and more, especially for emissions of SO_2, NH_3 and primary PM_{10}. For the *relevant emission areas* as a whole, emission trends after 2000 varied between 1 and 2%/year, and in the Netherlands between 1 and 3%/year. In terms of mass, emission reductions were dominated by NO_x, SO_2 and NMVOC, before and after 2000.

5.2.3 Measured SIA concentrations as a function of precursor gas emissions

Figure 5.6 shows the relation between average annual concentrations of the different SIA components for the rural locations in the Netherlands, on the one hand, and of the precursor gas emissions for the *relevant emission areas*, on the other. Initially, all SIA components decreased in a linear way following emission reductions in precursor gases. Sulphate concentrations decreased by about 0.5 µg/m³ per megatonne SO_2. At a certain emission level, linear dependencies became weaker, and for ammonium and ammonia even seemed to disappear. The chemical formation process became saturated. Following the large SO_2 emission reductions of the 1990s, a strong decrease in SO_2 concentrations took place. At the same time, NO_x emissions and concentrations stayed relatively high. As a consequence, SIA formation from precursor gases showed saturation effects with NH_3. Under these circumstances, ammonium concentrations are less dependent on changes in NH_3 emission (see also Denier van der Gon, 2009). Moreover, the reduction in nitrate becomes less efficient per mass unit of NO_x emission reduced. This phenomenon is rather well known (e.g. Fagerli and Aas, 2008). To further efficiently reduce, in this case, ammonium and nitrate concentrations, NH_3 emission reductions should be combined with reductions in NO_x emissions (see also Schaap *et al.*, 2009). The design of an optimal reduction strategy for PM_{10} based on the SIA precursor gases NO_x, NH_3 and SO_2, is subject to future research.

Emissions trends of primary PM_{10} for the Netherlands **Figure 5.4**



Emission trends in primary PM_{10} for the Netherlands, between 1990 and 2007 (left panel), and for the Netherlands including the relevant emission areas: Belgium, France, Germany, the North Sea and the United Kingdom, over the same period (right panel). For absolute emission numbers see Table 5.3.

Annual emissions of NH_3, NO_x, SO_2 and primary PM_{10} (in kilotonnes) **Table 5.3**

	1990	1995	2000	2004	2005	2006
NH_3	2288	2069	2010	1927	1882	1882
NO_x	9077	7539	6473	5772	5653	5675
SO_2	11231	5816	3122	2607	2466	2372
NMVOC	9208	6710	5065	4065	4026	3936
PPM_{10}	-	1302	1090	1021	994	969

Total emissions summed for the Netherlands, Belgium, France, Germany, the North Sea and the United Kingdom (EMEP, 2009a; CEPMEIP, 2005). Emissions of primary PM_{10} for 1990 are not in the EMEP emissions database. Extrapolation to 1990 (Figure 5.4, Figure 5.5) was based on primary PM_{10} emission estimates for 1990 for all 27 EU Member States, as a whole, from gap-filled emission data published by the European Environment Agency (EEA, 2008a).

5.3 Trends in black smoke and heavy metals in PM at rural locations

Other indicators for particulate matter that are monitored in the national network are black smoke (BS) and heavy metals; the latter consisting of arsenic (As), cadmium (Cd), lead (Pb) and zinc (Zn). Relatively long time series of average annual background concentrations are available and their relative trends are shown in Figure 5.7, to illustrate the trend in particulate matter from specific anthropogenic sources. The differences and similarities between the trends in PM_{10} and SIA substantiated our knowledge on anthropogenic emission changes in relation to particulate matter of the last decades.

5.3.1 Black Smoke

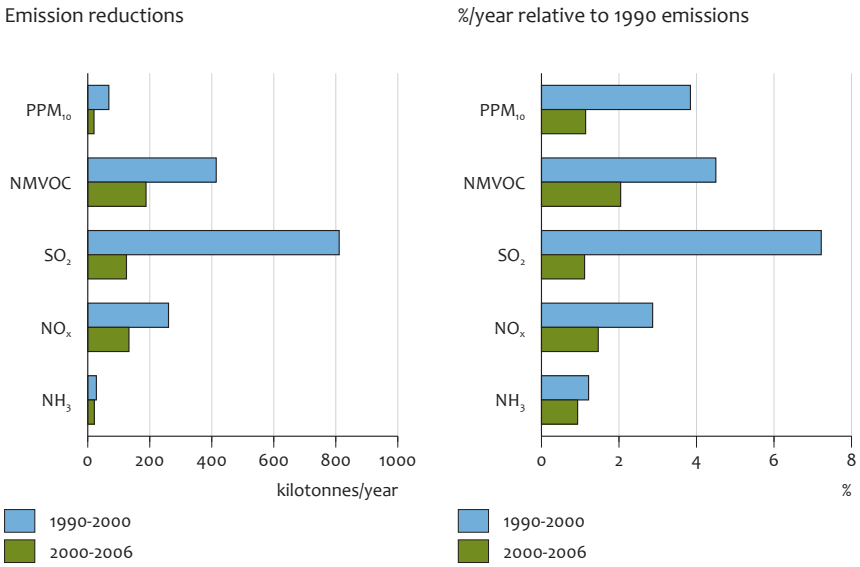
Black smoke (BS) is related to the combustion of organic fuel. Roughly half of BS concentrations in the Netherlands can be attributed to road transport, and about a third to industrial and domestic oil combustion (Bloemen *et al.*, 2007). Important BS emission changes have taken place due to the introduction of soot filters on diesel engines, and a switch from oil- to gas-fired petrochemical industry in the Netherlands. Soot has also been reduced because of a further introduction of filters in industry. Measured concentrations of Black Smoke (indicator for soot) at rural locations have shown a continuous decrease, from 2000 onwards. Although no significant decrease in PM_{10} concentrations could be observed between 2000 and 2007. Soot is considered as a more health-relevant fraction of PM_{10} than sulphate, nitrate, ammonium and sea salt. Therefore, air quality health conditions may have improved more than the trend in PM_{10} would suggest.

Long-term Black Smoke observations at urban and traffic locations have shown different trends: a levelling off after 2000 was observed at traffic locations, together with a decline at a single urban background location (Keuken and Ten Brink, 2009). Black Smoke trends at urban and traffic locations are subject to larger impacts from contributions of local emissions and, therefore, are more relevant to local situations and are probably less indicative of large-scale concentrations in the Netherlands.

5.3.2 Arsenic, cadmium, lead and zinc

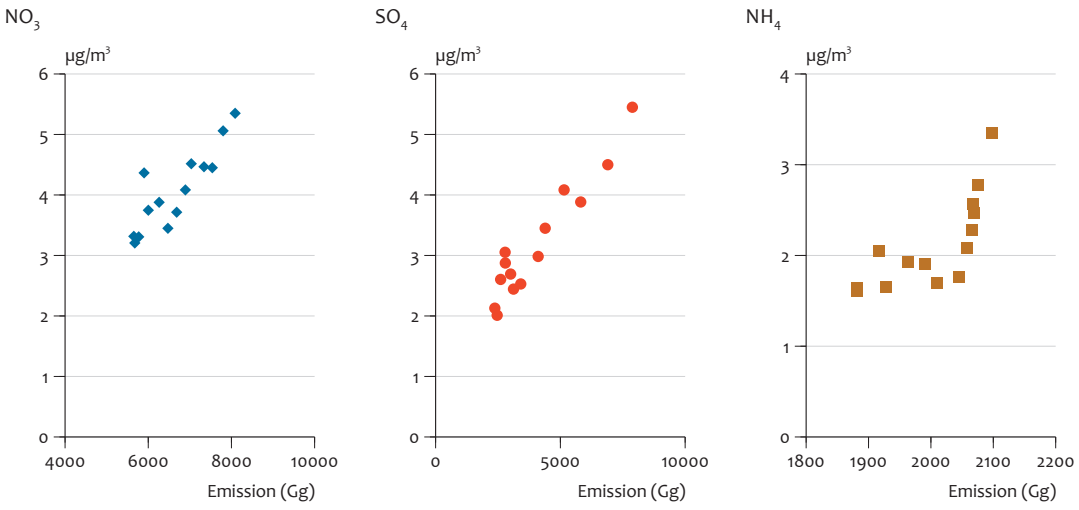
Heavy metals, arsenic (As), cadmium (Cd), lead (Pb) and zinc (Zn), are also related to anthropogenic emissions. For heavy metals, the main cause of anthropogenic emissions to air are industry, energy production, traffic, waste treatment and domestic sources. Moreover, heavy metals are released as particles in combustion processes, for instance, at refineries and waste incineration. Therefore they can be registered in PM_{10} measurements. While the contributions of As, Cd, Pb

Emission reductions in kilotonnes/year and in %/year relative to 1990 **Figure 5.5**



Emission reductions in kilotonnes/year (left panel) and in %/year, relative to 1990 emission levels (right panel), for NH_3, NO_x, SO_2, NMVOC, and primary PM_{10}, realised in the relevant emission areas: the Netherlands, Belgium, France, Germany, the North Sea, and the United Kingdom, for 1990 to 2000 and 2000 to 2006.

Average annual concentrations of NO_3, SO_4, NH_4 in the Netherlands **Figure 5.6**

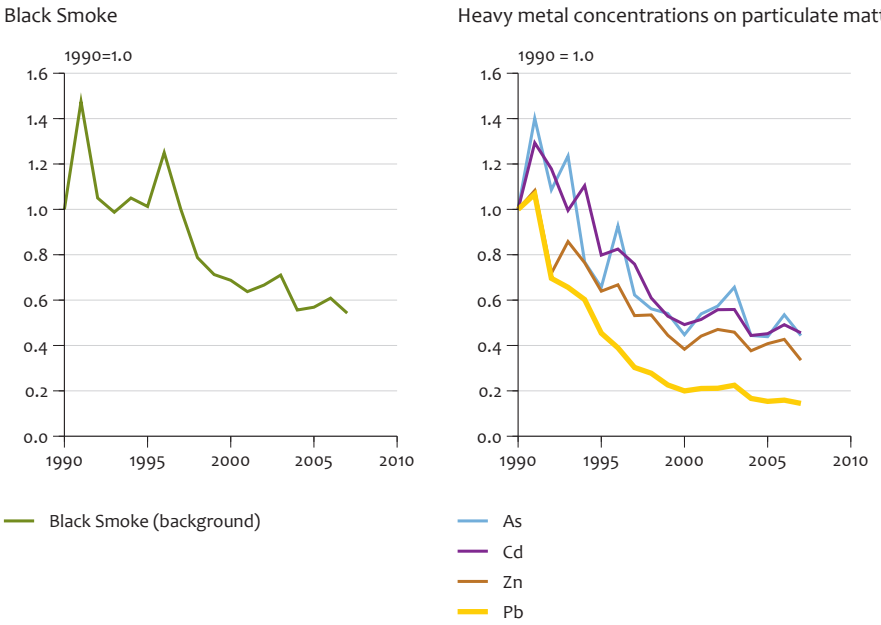


Average annual concentrations of NO_3, SO_4 and NH_4 in the Netherlands, as a function of SIA precursor gas emissions in the relevant emission areas: NO_x, SO_2, and NH_3, respectively. For SIA concentrations see Figure 5.2 (right panel), and for emission data see Table 5.3 (EMEP, 2009a).

and Zn to PM_{10} concentrations are negligible, the trend in the heavy metal concentrations can be seen as an indicator for the emissions trend of all components emitted by the heavy metal sources.

In 1998, in order to reduce the emission of heavy metals, the Netherlands, together with other European countries, signed The 1998 Aarhus Protocol on Heavy Metals, under the Convention on Long-range Transboundary Air Pollution by the United Nations Economic Commission for Europe (UNECE, 1998). Consequently, heavy metal emissions have declined over the last two decades (see Figure 5.8). Important emission reductions have taken place in the industrial sector, especially in waste incineration. Lead emissions, mainly form traffic, clearly plummeted after the use of lead in petrol was banned.

Measured trends of Black Smoke and heavy metal concentrations **Figure 5.7**



Measured trends in black smoke (left panel) and heavy metal concentrations in particulate matter (right panel), normalised to 1990 concentration levels; lead (yellow line), arsenic (bleu line), cadmium (purple line) and zinc (brown line). The concentrations were measured at rural locations in the Netherlands.

5.3.3 Similarities and differences between trends in particulate substances

For all components in Figure 5.7, a large decrease could be observed for the 1990s, followed by a levelling off of the trend after 2000. This behaviour is similar to the trend in PM_{10} (see Chapter 2), although the decline in PM_{10} concentrations in the 1990s appears to have been much smaller. The decline in black smoke concentrations continued, after 2000, at a larger rate than that in heavy metal and PM_{10} concentrations. Apparently, emissions of elemental carbon was efficiently abated by the European emission standards for vehicles between 1990 and 2007, in spite of increased traffic volumes over that period. The decline in heavy metal concentrations can be explained by emission trends as shown in Table 5.2. All components showed fluctuations due to meteorology, but their amplitude response differed. An explanation for these differences in response to meteorological variations could be related to the behaviour in time and space of the different sources and the lifetime of the components. In this study, however, this was not further explored.

5.4 Modelling trends in PM_{10} at rural locations

This subsection presents an analysis of the PM_{10} trend, using results from a dispersion model that calculates fractions of PM_{10} from emissions of primary PM_{10} and SIA precursor gases. The analysis offers more insight into the relation between the observed trend in the measurements and emission changes over the years 1993 to 2007. In the Netherlands, air quality model calculations play an important role in official national air quality assessments (e.g. Velders *et al.*, 2009).

5.4.1 Approach

First, we considered the contribution of secondary inorganic aerosol to PM_{10}, by comparing measurements and model results for the years 1993 to 2007. There are large uncertainties in both model results and measurements. We explored the magnitude and effect of the main biases involved.

Second, we added modelled concentrations of primary PM_{10} particles from European anthropogenic sources to the observed concentrations of secondary inorganic aerosol. Because the SIA concentrations and the trend in these concentrations were found to be uncertain, we considered two SIA cases (see below). The modelled concentration differences between 1993 and 2007 due to anthropogenic emission changes of SO_2, NO_x, NH_3 and PM_{10} are viewed with respect to the observed trends in PM_{10}.

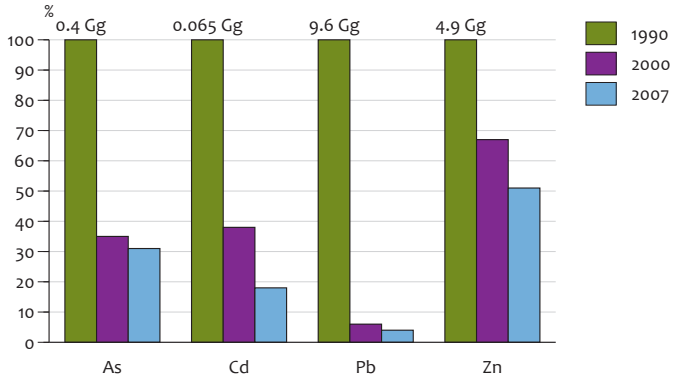
Two SIA trend cases

SIA concentration were found to have been underestimated in the 1993-2007 by the measurements performed in the Dutch national monitoring network (LML), as well as in results from the air quality model used for the assessment of PM_{10}. Therefore, the following two cases were investigated for the analysis of the PM_{10} trend:

Approach 1

SIA 1993-2007: We interpreted the routine measurements as being representative of PM_{10} and used the same model results that have been used in the yearly air quality reports. Although these model results, on average, underestimate SIA levels by about 40%, the trend may be correct.

Heavy metals emissions 1990, 2000, 2007 relative to the 1990 emission **Figure 5.8**



Heavy metal emissions in 1990, 2000, and 2007, relative to 1990 emission levels. The 1990 emissions are shown in kilotonnes, and represent the total emissions in Belgium, Germany, France, the Netherlands and the United Kingdom (ER, 2009; EMEP, 2009b).

Approach 2
High SIA trend: To analyse an upper-limit case, the measured and modelled time series according to approach 1 were scaled to fit the measured PM_{10} SIA concentrations in the BOP campaign of 2007 to 2008.

For this scaling, we applied constant factors per SIA component: 1.4 for NH_4, 1.6 for NO_3 and 1.7 for SO_4. These factors were derived from simultaneous, collocated SIA measurements from monitoring network instruments and the updated, more accurate (with respect to the PM_{10} reference method) monitoring instruments used during the BOP measurement campaign.

5.4.2 Model calculations
We used the OPS model for calculating average annual (1993-2007) concentration levels of secondary and inorganic aerosol (SO_4, NO_3, NH_4) and primary PM_{10} on a 5x5 km² grid for the Netherlands. The modelled concentrations were calculated from anthropogenic emissions of primary PM_{10}, SO_2, NO_x and NH_3 in the Netherlands (ER, 2009) and in the whole of Europe (EMEP, 2009a). For an overview of the changes of the main relevant emissions used, see Subsection 5.3. Additional information on the model runs, the OPS model and its performance can be found in Annex A and references.

5.4.3 Model results and measurements of PM_{10} at rural locations
In order to compare the average annual PM_{10} concentrations for the rural locations with the model results for PM_{10} fractions, all measurements and model results were determined for these locations. To do so, we used modelled concentrations for those 5x5 km² grid cells that correspond with the rural measurement locations of PM_{10} (Figure 5.1). Furthermore, SIA was not measured at every PM_{10} measurement location, therefore, where SIA measurements were missing, concentrations were extrapolated from the measurements, by using modelled spatial gradients for each year.

SIA concentration measurements and model results
Figure 5.9 shows the measured and modelled SIA concentrations as an average of the levels at the rural PM_{10} locations.

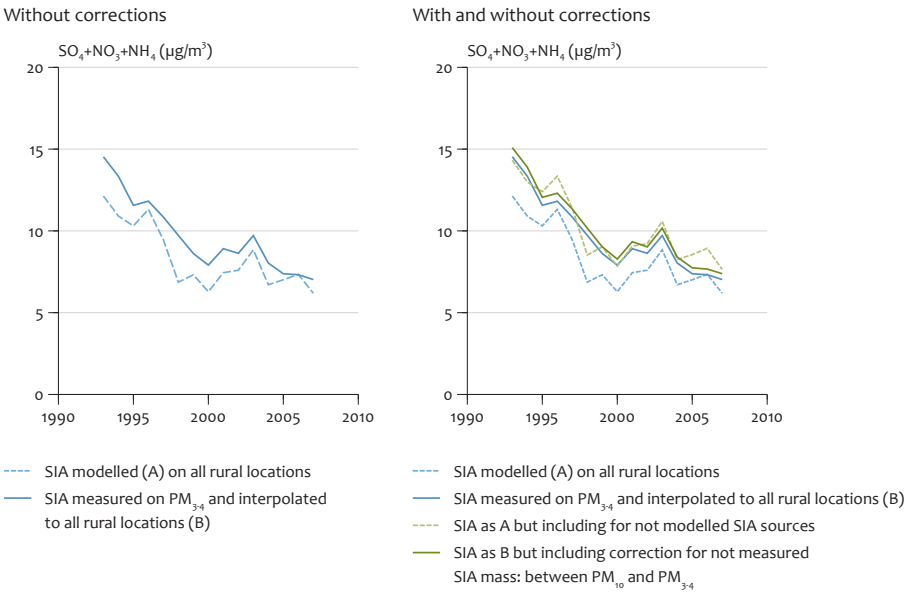
Both SIA measurements and model results for 1993 to 2007 contain several biases.
- Nitrate measurements were multiplied by 1.1, to correct for a particle size cut-off of the measurement instrument, which was smaller than 10 microns (for details, see Annex B).
- Modelled SIA levels would not properly describe all NH_3 sources, and some other sources of SIA were missing. Altogether, modelled SIA concentrations would have been about 2.2 µg/m³ higher in 1993, and 1.5 µg/m³ in 2007 (for details, see Annex B).

In approach 1, the modelled and measured trend and annual SIA concentration levels were in very good agreement, especially, when the proper corrections were taken into account (Figure 5.9).

Approaches 1 and 2 and contribution of primary PM_{10} and water
From the SIA model results (approach 1) the high SIA trend case (approach 2) was calculated, simply by scaling of the SIA components (see Subsection 5.4.1 under *Two SIA trend cases*). Subsequently, the contribution of two other modelled fractions to PM_{10} was added to both SIA trend cases: 1) Primary PM_{10} due to registered emissions in Europe of primary PM_{10} and 2) the particle bound water associated to SIA (about 15% of the modelled SIA mass). Annex C presents the reason for, and approach to, estimate the magnitude of particle bound water associated to SIA. Figure 5.10 allows a comparison of measured PM_{10} concentrations (Chapter 2) and modelled average PM_{10} fractions for the PM_{10} rural locations.

There are large differences between both modelled trend cases, the SIA 1993-2007 trend case and high SIA trend case. The trends observed in the PM_{10} measurement range (dark

Modelled and measured data for SO_4+NO_3+NH_4 **Figure 5.9**



The left-hand panel shows SO_4+NO_3+NH_4 observed (solid line) and modelled (dashed line). The right-hand panel shows the same, but also includes estimates of SIA mass between PM_{3-4} and PM_{10}, and model results include estimates on the contribution from missing sources (see text in this subsection). For comparison, the uncorrected concentrations shown in the left panel are indicated with grey lines.

blue solid lines in Figure 5.10) fit between the two model results for the anthropogenic fraction of PM_{10} (light blue solid and dashed lines in Figure 5.10). Both modelled trend cases are probably not fully representative of the 'real' trend. The scaling of the modelled trend, in order to fit the more accurate SIA observations in the BOP campaign in 2007/2008, was done with factors constant in time, although these factors used per SIA component, in fact, were only known for 2007. For the other years, the factors may very well have been significantly different. In the following section, under 'discussion', the observed trends are further explored. To zoom in on the contribution of anthropogenic emission changes in the past, we compared modelled and measured differences between the 1993-1995 and 2005-2007 periods.

5.4.4 Contribution of secondary organic aerosol to PM_{10}

The formation of secondary organic aerosol (SOA) due to anthropogenic NMVOC emissions was not included in the model. Therefore, we estimated the contribution of SOA concentrations to the PM_{10} trend between 1993 and 2007. A SOA trend estimate of about 0.08 µg/m³ per year was derived based on the 60% NMVOC emission decrease between 1990 and 2007, and the following two assumptions:

- In 1990, the average anthropogenic SOA concentration was about 1.5 µg/m³.
- Anthropogenic SOA concentrations in the Netherlands reacted in a linear way to the overall anthropogenic NMVOC emission changes in the Netherlands, Belgium, France, Germany and the United Kingdom. A decline of about 60% was used, based on EMEP (2009a) and ER (2009) (see also Table 5.3).

The contribution of primary organic aerosol to PM_{10}, due to anthropogenic emissions, is implicitly described by PPM_{10}, because the primary PM_{10} emissions include primary organic aerosol.

5.5 Discussion

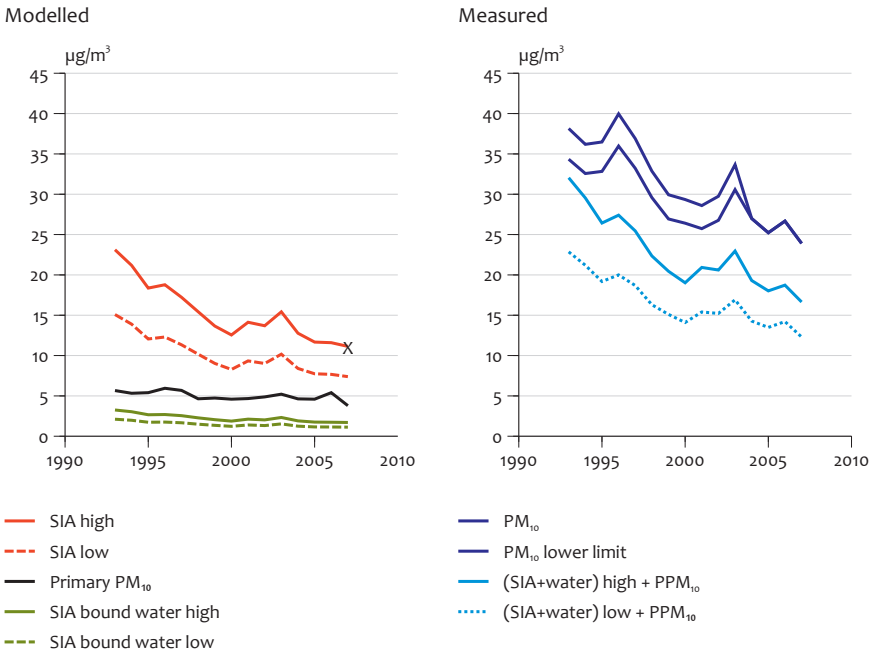
In the preceding subsections different pieces of information have been presented on the trends of different fractions of PM_{10} from observations and from measurements and model calculations. In this section, a further exploration is presented of the trend in PM_{10} in the 1993-2007 period, which was carried out by integrating the information above.

What is the trend in PM_{10}, derived from the observations?

Section 2.1 describes the exploration of two possible PM_{10} concentration trends. The linear trend in PM_{10} at rural locations, between 1993 and 2007, was between 0.7 and 1.0 µg/m³ per year. Here, we argue that the lower trend (according to the lower estimate in Figure 2.1) is more consistent with most other observations.

- The lower estimate is more in accordance with the trend of other particulate substances, such as heavy metals, in the Netherlands, and with PM_{10} trends measured by other networks within and outside the Netherlands (see also Figure 5.7). Mol et al. (2007 and 2008) show trends in PM_{10} and $PM_{2.5}$. Since 2000, no significant trend has been observed in the Netherlands, or for PM measurements in the 27 EU Member States.
- The lower estimate is also a better option from a dispersion point of view. It reflects a more or less constant urban increment, instead of an increase in the urban increment between 1993 and 2003, on the one hand, and 2004 and 2007, on the other. A decrease in urban increment from 1993 to 2007 would result in an even lower

Average annual modelled and measured concentrations of PM_{10} **Figure 5.10**



Average annual concentrations of modelled PM_{10} fractions for 1993 to 2007: SIA, SIA-bound water, and primary PM_{10}, for two SIA trend cases, per fraction (left panel). The cross (x) indicates the SIA concentration in 2007 according to the BOP programme. The right-hand panel shows summed modelled PM_{10} fractions and the measured average annual PM_{10} concentrations, as in Figure 2.1. Average concentrations were determined from measurements at rural locations in the Netherlands.

estimate of PM_{10} for the years before 2003. Emissions related to traffic are the main sources for the urban increment. An increase in urban increment, on average for 1993 to 2007, did not seem plausible, given the decline in national traffic emissions during that period.

- The lower trend is consistent with the low SIA estimate. The lower PM_{10} estimate is inconsistent with the high SIA estimate.

Can we explain why the PM_{10} trend has levelled off after 2000?

The trends in concentrations of heavy metals, arsenic, cadmium, lead and zinc reflect the relevant emission changes over the period of 1990 to 2006. The trends in heavy metal concentrations show a levelling off after 2000. Although, emission changes in EC (soot) have not been recorded for 1990 to 2006, the trend in black smoke appears plausible. The long time series of SIA observations, although probably biased low with respect to SIA measured in PM_{10} according the reference method, do also show a levelling off of the trend, after 2000. The levelling off of the trend in PM_{10} is in line with these findings.

In the Netherlands the sea salt contribution correlates negatively with PM_{10} concentrations (Manders et al., 2009). High PM_{10} concentrations are associated with air masses from continental origin, which have logically low sea salt concentrations. When air comes from south western to north western directions the contribution of sea salt is higher and the anthropogenic contribution is generally low. Sea salt generally constitutes a significant portion of several µg/m³ to the average annual PM_{10} concentration in the Netherlands. A trend in the anthropogenic contribution will therefore will be less explicit due to the antagonistic behaviour of the sea salt contribution.

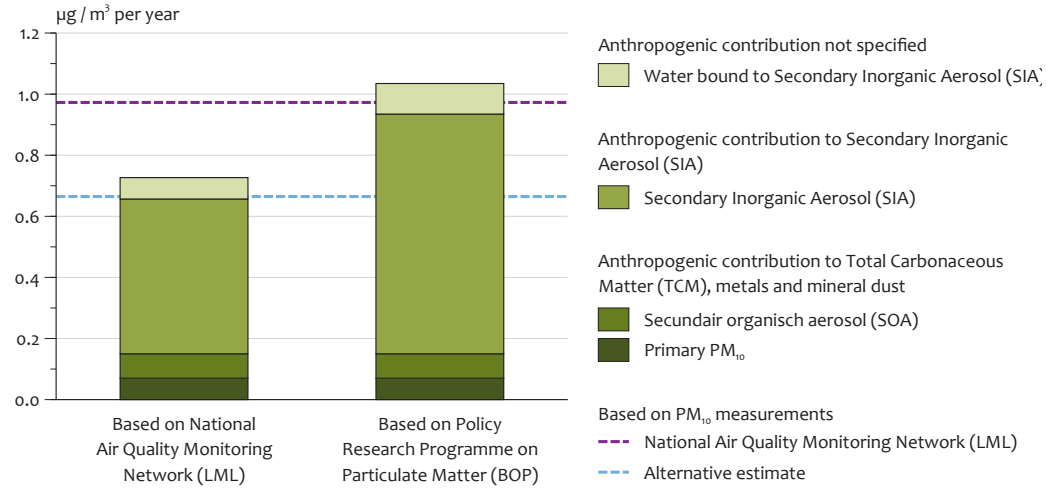
Furthermore, averaged annual secondary inorganic aerosol concentration levels in the Netherlands, in particular of NH_4, show saturation effects, which are due to low SO_2, and relatively high NH_3 and NO_x concentrations. Emission reductions in SIA precursor gases have a smaller effect, today, than they had in the 1990s.

Is the observed PM_{10} trend in line with the relevant anthropogenic emission changes?

The trend in anthropogenic PM_{10}, the bottom-up approach, and the trend in observed PM_{10}, cannot be compared, straightforwardly, as a relatively large part of PM_{10} could not be accounted for (see Figure 5.10; the difference between the light and dark blue lines). This unaccounted part represents natural sources and anthropogenic sources that were not included in the model. Therefore, in stead of a trend comparison, we compared three-year averaged PM_{10} concentration differences between 1994 and 2006.

Figure 5.11 shows estimated concentration decreases in µg/m³ per year, for the anthropogenic components, between 1994 and 2006, in comparison to the observed range of concentration decreases in PM_{10} concentrations. The concentrations for 1994 (2006) were averaged over the years

Change in background concentration PM_{10}, 1993 – 2007 **Figure 5.11**



PM_{10} concentration change (µg/m³ per year) between 1993 and 2007, dotted lines based on the PM_{10} measurements (see Figure 2.1) and anthropogenic components: secondary inorganic aerosol, primary PM_{10}, secondary organic aerosol and water bound to secondary inorganic aerosol.

1993, 1994 and 1995 (2005, 2006 and 2007), to eliminate disturbing effects due to fluctuating weather conditions.

Two SIA trend cases were considered: Approach 1, SIA 1993-2007, and approach 2, high SIA trend. It should be noted that the contribution to the PM_{10} trend due to changes in secondary organic aerosol (SOA) is a simple estimate and not part of the model calculations.

Figure 5.11 represents our current view of the PM_{10} concentration changes induced by changes in anthropogenic emissions, over the 1993-2007 period. The modelled concentration changes for the SIA 1993-2007 trend case explain the observed changes in PM_{10} concentrations, and are close to the PM_{10} concentrations in the lower estimate. The modelled concentration changes for the high SIA trend is close to the high end of the observed PM_{10} changes.

Therefore, although SIA levels may have been biased low, the trend according to the long time series of SIA may very well be roughly correct. As a consequence, the high SIA trend case, using constant scaling factors in time, gave an unrealistic upper limit estimate of the PM_{10} trend due anthropogenic emission changes. A comparison with trends from SIA measurements abroad may confirm this.

The major contribution to a modelled PM_{10} concentration difference (delta PM_{10}) was caused by changes in SIA concentrations (of about two thirds) that were directly related to emission reductions in SO_2, NO_x and NH_3. As much as 4% of SIA concentration changes in the Netherlands, between 1993 and 2007, may be attributed to emission reductions in SIA precursor gases from across the Atlantic Ocean.

The last third of the modelled delta PM_{10} was about equally distributed between the anthropogenic concentration changes in primary PM_{10}, water associated with SIA, and secondary organic aerosol. It should be noted that water significantly magnifies the SIA trend when considering its contribution to the trend in PM_{10} concentrations.

Consequences for the assessment of future PM_{10} concentrations

Uncertainty about SIA concentration levels causes future projections of PM_{10} concentrations to be more uncertain than previously thought. Moreover, projections of the PM_{10} concentrations are likely to be biased high. SIA concentration levels are currently thought to be about 55% higher than was reported before. In case the downward SIA trend is underestimated, this would mean that its effect on PM_{10} concentration in the Netherlands, currently, is also underestimated. For the high SIA trend case, which is likely to indicate an upper limit for the SIA trend, preliminary model results indicate that PM_{10} may be up to 1 µg/m³ lower in 2010, than was previously calculated, and possibly up to 2 µg/m³ lower in 2020. These figures are to be regarded as uncertain, since the actual SIA trend is unknown. More efforts are needed for a more accurate assessment of the historical SIA trends and their relation to precursor gas emissions.

PM_{10} projections for the Netherlands are made by using estimates of the future PM_{10} fraction from unregistered and natural sources, to fill in the missing mass. Particle-bound water associated with SIA forms a natural fraction of PM_{10}. For this study, we have newly explored its contribution to PM_{10} and the PM_{10} trend. The uncertainty concerning our model estimates of this fraction are to be regarded as large; there were no measurements available to verify our model results for particle-bound water associated with SIA. In addition, the current uncertainty about SIA concentration levels directly translates to the amount of water associated with them.

From Figure 5.10 can be observed that the difference between the modelled (anthropogenic) and the observed PM_{10} concentration is about 4 µg/m³ smaller in the high SIA trend case, compared to the SIA 1993-2007 case. This difference which is due to other, 'natural', sources is therefore also not very well understood. The difference, in 2007, in the high SIA trend case constituted about 8 µg/m³, whereas in the SIA 1993-2007 case, this was about 12 µg/m³. Note that the contribution of particle-bound water associated with SIA (1-2 µg/m³) is not included in this amount. Mass closure analysis based on the BOP measurements further corroborated our findings (Schaap *et al*., 2010).

Sensitivity analysis of PM_{10} measurements and the PM_{10} reconstruction from its major components, for the 1994-2007 period, showed similar ranges in trends of 0.7 to 1.0 µg/m³/year.

5.6 Conclusion

This chapter presents a trend analysis of PM_{10} and secondary inorganic aerosol - ammonium nitrate and ammonium sulphate – for the Netherlands, over the 1993-2007 period. Over the 1990-2007 period, concentration changes of black smoke and Heavy Metals on particles were investigated. The main goal was to obtain more insight into the effect of past emission reductions on PM_{10}. A better understanding of how PM_{10} concentrations were formed from different sources may help us to improve our assessments of future levels of PM_{10} and $PM_{2.5}$.

Sensitivity analysis of PM_{10} measurements and the PM_{10} reconstruction from its major components, for the 1993-2007 period, showed similar ranges in trends of 0.7 to 1 µg/m³/year.

The levelling off of the trend in PM_{10} after the year 2000 is in line with other observations and model calculations based on anthropogenic emissions. In addition, the contribution of sea salt to PM_{10} in the Netherlands tends to counteract the anthropogenic contribution.

The analysis in this chapter carries major uncertainties, which are largely due to a bias in the (interpretation of the) current observations and model results of secondary inorganic aerosol. As a consequence, the trend in SIA is very uncertain. This uncertainty means that past PM_{10} projections for 2010 and 2020 may have been biased high, by up to 1 µg/m³ for 2010, and up to 2 µg/m³ for 2020. $PM_{2.5}$ projections may contain similar biases, although these are probably somewhat lower. Model results indicate that the trend in SIA concentrations that was based on the same long time series of observations in the national monitoring network (LML), may nevertheless represent the actual trend in SIA concentrations.

Regardless of this bias in SIA measurements, we concluded that emission changes in SO_2, NO_x and NH_3 contributed about two thirds to the changes in PM_{10} concentrations of 1993 to 2007. The other third could be about equally attributed to the three other sources of anthropogenically induced change: changes in primary PM_{10} emissions, NMVOC emission changes leading to secondary organic aerosol, and particle-bound water associated with the changes in secondary inorganic aerosol.

Recommendations

Urgent action should be taken to verify the trend in secondary inorganic aerosol concentrations (SIA), by means of model analyses and comparison with SIA trends abroad. Currently used models should be evaluated and possibly adapted, to correctly describe historical and future SIA concentration levels in the Netherlands.

The assessment of carbon should be further extended. To date, this has been limited due to a lack in good measurements, emissions and models.

6

Emission and air quality trends from national to European scales

Air quality in the Netherlands and Western Europe has been improving since the 1990s. However, recent investigations have shown that PM concentrations in cities across Europe have not further improved, over the 2000-2006 period. Examples of this fact have been published, for instance, for Amsterdam (Woudenberg *et al*., 2008) and London (Harrison *et al*., 2008). Furthermore, Beijk et al. (2007a) concluded, based on air quality monitoring data, that in Netherlands, since the year 2000, such a general declining trend could no longer be observed. The European Environment Agency reached a similar conclusion, on European scale (EEA, 2007). The remarkable aspect of the fact is not the stagnating trend itself, but that at the same time emissions are continuing to decline, as is evident from country-reported emission data (EEA, 2008a). The combination of these two observations seems a paradox, as when emissions decrease, concentrations follow suit – unless other factors become prevalent and temper or counteract the decrease. To better understand the causes for stagnating trends, a number of research questions were formulated;

1. Are (reported) emissions indeed declining across Europe, and does this apply to both primary PM emissions and precursor emissions?
2. Is the stagnating trend observed mostly within cities, or does it also apply to rural background concentrations?
3. To what extent are increases in unreported emissions, such as those from resuspension by traffic, capable of counteracting decreases in reported emissions?
4. Are there changes in urban traffic flows in the Netherlands that possibly explain stagnating PM trends? (see Chapter 7 and Annex D Goudappel-Coffeng study)
5. To what extent could meteorological variability be playing a role in obscuring trends? (see Chapter 3)

This chapter examines 1) official reported emission data (available per country and as a time series), in order to identify the spatial and temporal variation in emission trends, and 2) official reported ambient air quality data (available per country, monitoring location and as a time series), in order to identify the spatial and temporal variation in ambient air quality trends. To structure the data and limit the number of unnecessary details, the analyses were made for the 1990-2006 period, for four groups of countries:

- The Netherlands (NL)
- Belgium, Germany, The United Kingdom, France, Denmark (BE+DE+UK+FR+DK)
- Other EU Member States (Other EU)
- Other UNECE[1] and non-EU Member States (UNECE+non-EU)

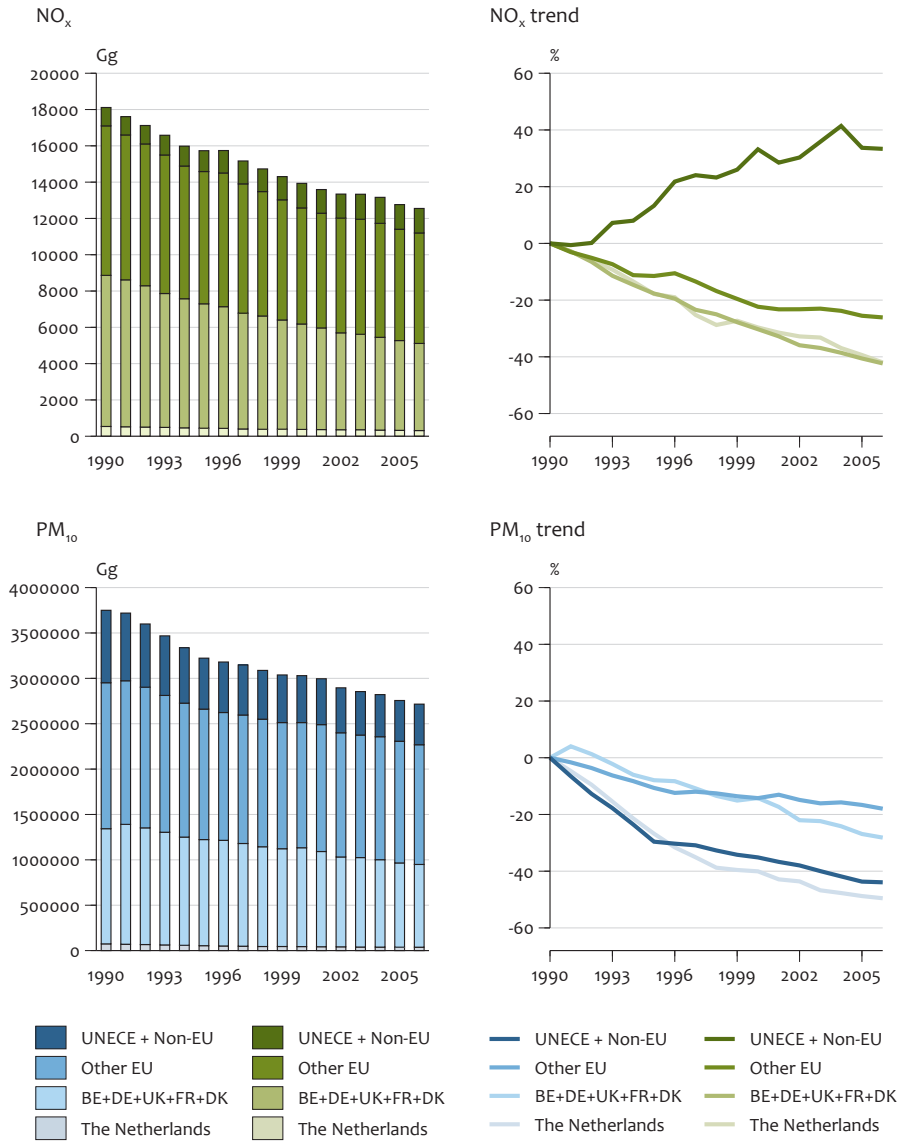
6.1 Country Emission Trends

The UNECE implemented the LRTAP[2] Convention under which member countries (EU + non EU) are required to report air quality related emissions. The EU and its Member States ratified the LRTAP convention and thereby committed to report their national emissions annually to EMEP[3]. The European Environment Agency (EEA) collates the EU data submitted under the LRTAP and Climate Conventions and implements gap-filling procedures, resulting in the EEA 'aggregated and gap filled air emission data'. These data are annually updated and available from the EEA website: http://dataservice.eea.europa.eu/dataservice/metadetails.asp?id=1006.

Total UNECE Europe NO_x and PM_{10} emissions have declined substantially, since 1990 (Figure 6.1). Trend analysis confirmed a significant decline between 1990 and 2000 (Figure 6.1), although the decline was generally less steep over the 2000-2006 period. In non-EU European countries, NO_x emissions increased between 1990 and 2006 (Figure 6.1, upperright). These gradients indicate that emission reductions, since the year 2000, have significantly slowed down in *Other EU* and *UNECE+non-EU* countries, compared with those in the Netherlands and its neighbouring countries, and relative to the entire study period between 1990 and 2006, thereby complementing the observations of Harrison et al. (2008) in the United Kingdom. The relative change (%)

1 UNECE = United Nations Economic Commission for Europe
2 LRTAP = Convention on Long-Range Transboundary Air Pollution
3 EMEP = European Monitoring and Evaluation Programme

Total official country-reported NO_x and PM_{10} emissions arising from all sources **Figure 6.1**



Left) Total of officially reported country data on NO_x and PM_{10} emissions, from all sources; right) Normalised NO_x and PM_{10} emission trends. Negative values represent emission reductions relative to 1990 values; positive values represent emission increments relative to the 1990 values. (Source: EEA, 2008a).

in total pollutant emissions for the periods between 1990 and 2006 (A), 1993 and 1999 (B), and 2000 and 2006 (C), is presented for NO_x, PM_{10}, $PM_{2.5}$, NMVOC, CO, NH_3, and PFP (particulate forming potential) (Table 6.1). Negative values represent emission reductions relative to the starting year of a particular period, while positive values refer to emission increments. Although between 2000 and 2006 the decline in emissions was less steep than reported for the 1990-2000 period, the reported emission reductions suggest a decline of around 10% for most relevant substances, for the Netherlands and nearby countries. The emissions in *UNECE+non-EU* countries behaved slightly different, but, given the distance from western Europe and the still relatively small contribution to total emissions (Figure 6.1, upperleft), this has certainly not caused a stagnation in trends in ambient concentrations in western Europe. So, based on reported emissions, and assuming a directly coupled system, we would expect a around 10% decrease in ambient air concentrations between 2000 and 2006, for priority pollutants (Figure 6.1). The exception may be PM_{10} for which decreases in PM_{10} concentrations would have been more likely to be in the -5% range, because not all PM_{10} would have been due to anthropogenic activity.

6.2 National Ambient Air Quality Trends

In the Netherlands, air quality measurements on a city scale provide similar findings to those of Harrison et al. (2008) and the EEA (2007a). In Amsterdam, concentrations of PM_{10} and NO_2 have not declined since 1999 (Van der Zee and Woudenberg, 2006). The same is true on a greater

Annual change, in percentages, in total pollutant emissions for the period between 1990 and 2006 (A), 1993 and 1999 (B), and 2000 and 2006 (C) Table 6.1

COUNTRY GROUP	POLLUTANT/ INDICATOR (% CHANGE/YEAR)																				
	NO_x			PM_{10}			$PM_{2.5}$			NMVOC			CO			NH_3			PFP		
	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C
Netherlands	-2.5	-2.7	-1.7	-3.0	-3.7	-1.3	-3.3	-3.7	-1.6	-3.8	-4.0	-1.9	-3.0	-2.7	-1.9	-2.8	-2.7	-1.1	-2.8	-3.1	-1.4
BE+DE+UK+FR+DK	-2.5	-2.6	-1.4	-1.8	-2.1	-2.0	-2.0	-2.6	-2.0	-3.5	-3.4	-2.1	-3.6	-3.4	-2.1	-0.9	0.0	-0.7	-3.5	-4.0	-1.3
Other EU	-1.8	-2.3	-0.4	-1.4	-1.3	-0.7	-1.1	-1.1	-0.7	-1.3	-1.0	-0.6	-2.5	-3.7	-1.9	-1.5	-0.7	-0.4	-2.5	-2.7	-0.9
UNECE+non-EU	2.2	3.4	0.6	-2.6	-2.1	-1.3	-2.2	-1.9	-1.6	1.6	6.3	-2.6	-1.2	0.6	-3.1	-0.4	-0.6	-0.3	-0.1	3.3	-3.7

*A = 1990-2006, B = 1993-1999, C = 2000-2006.
Note: Negative values represent emission *reductions* relative to the starting year of a particular period; positive values represent emission *increments* relative to the starting year of a particular period. PFP refers to 'particulate forming potential'. Data from the *EEA (2008a)*.

Percentage change in average annual concentrations of ambient air pollutants, for the period between 1990 and 2006 (A), 1993 and 1999 (B), and 2000 and 2006 (C) (source: EEA, 2008b) Table 6.2

COUNTRY GROUP	Annual change in pollutants (%)					
	NO_2			PM_{10}		
	1990-2006	1993-1999	2000-2006	1990-2006	1993-1999	2000-2006
Netherlands	-0.8	-1.9	1.1	-1.5	-2.6	-0.1
BE+DE+UK+FR+DK	-1.6	-2.6	0.1	-1.2	-1.9	-0.4
Other EU	-2.6	-3.1	-0.1	3.8	6.3	-0.6
UNECE+non-EU	-2.1	-3.3	0.1	-1.2	-3.6	4.6

Note: Negative values represent concentration *reductions* relative to the starting year per particular period; positive values represent concentration *increments* relative to the starting year per particular period.

urban scale, at which city background levels are determined (Beijk *et al*., 2007b). Measurements at rural background and traffic locations in the Netherlands have not shown a significant trend in NO_2, since 2000 (Van der Zee and Woudenberg, 2006; RIVM, 2008b). For a discussion on the PM_{10} trend in the Netherlands we refer to Chapter 5. The trend in measurements in the Netherlands is comparable to those in neighbouring and other EU countries. The change in concentrations in Table 6.2 is reported in a similar way as the emission data in Table 6.1. The reported emission data show a declining trend, whereas the data on ambient air concentrations for the Netherlands and neighbouring countries, between 2000 and 2006, show no (significant) trend (Table 6.2).

We have investigated air quality trends in the aforementioned four groups of selected countries, using the AirBase[4] database. AirBase is the air quality information system maintained by the EEA through the European Topic Centre on Air and Climate Change (ETC/ACC). The database comprises reported measurements, per country, of up to 137 ambient air pollutants and air quality indicators. For some selected pollutants, data are available from 1969 onwards. However, widespread observations, at European level and for an extended number of pollutants, are mainly available from 1996 onwards.

This report only presents data on PM_{10} from background and traffic locations (Figure 6.2), and on NO_2 from traffic locations (Figure 6.3). The data from the AirBase database tend to fluctuate, because over time monitoring locations are added or removed from the total set of locations. For this reason, data for *UNECE+non-EU* countries are not shown in Figure 6.2 and Figure 6.3. Data for the *Other EU* countries appear to have been less robust roughly before 1997, presumably for the same reason. Nevertheless, the figures illustrate that the trend in the Netherlands is similar to that in its neighbouring countries. In general, the data illustrate the point made in Table 6.2; there appears to have been no significant trend over the period between 2000 and 2006, in the Netherlands and neighbouring countries. Concentration levels in the Netherlands at traffic locations were similar to those in neighbouring countries, but Dutch background concentrations were relatively high (Figure 6.2), making the concentration increment between (urban) traffic locations and background locations rather small.

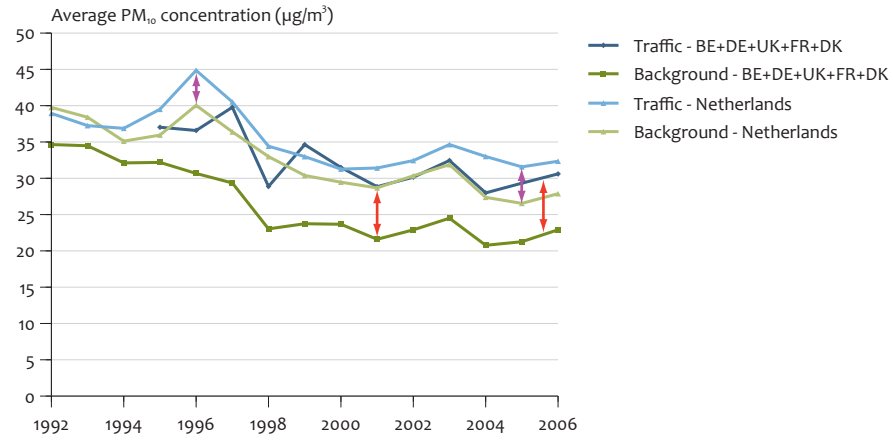
6.3 Conclusion

This study has explored various sources of information on emissions and ambient air quality in Europe. A stabilisation of concentration levels of PM and NO_2 was seen across Europe. Nevertheless, overall emissions were reported to have decreased since 2000. This means that either we do not properly understand all components of the system, or that uncertainties surrounding emission reporting, air quality measurements and modelling are such, that they are not sensitive enough to pick up on trends that are relatively small, compared to the earlier period of 1990 to 2000. Some of the uncertainties are listed below and several of them have been further explored.

- Incomplete coverage of emission sources in official reports by nations. However, if these incomplete sources would be stable they would not influence the trends – hence, to

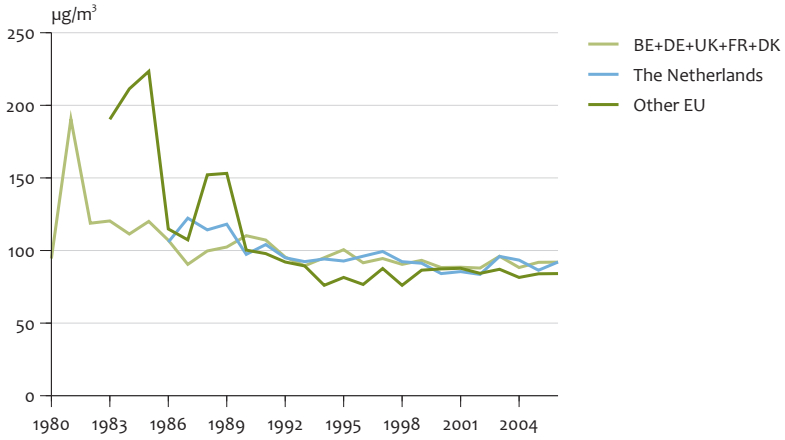
4 Available at http://dataservice.eea.europa.eu/dataservice/metadetails.asp?id=1029

Average annual PM_{10} concentration at background stations and street stations Figure 6.2



Average annual PM_{10} concentrations at background and traffic locations in the Netherlands and surrounding countries (data source AirBase, 2008). Purple and red arrows show the increment at background and traffic locations for the Netherlands and its surrounding countries (Belgium, Germany, UK, France and Denmark), respectively.

Country-reported ambient air concentrations of NO_2 for traffic stations Figure 6.3



Data on ambient air concentrations of NO_2 at traffic locations, per country group (data source AirBase, 2008).

counter balance the decrease in reported emissions these sources would not have been stable but increasing over the years. This issue is partly addressed in Chapter 7, under resuspension of dust by traffic.

- Potential errors in emission calculations. For example, if the efficiency of emission control technologies is overestimated, the calculated (and reported) emissions will show a steeper decline than is really occurring.
- Complications and/or adjustments in monitoring techniques. This topic is outside of the scope of this study, but may especially play a role when comparing data of 2000 to 2006 with previously monitored air quality data.

Finally, the (declining) trend may have been obscured by meteorological variability during the period of study. The impact of meteorological variability is quantified in Chapter 3 and leads to the uncomfortable conclusion that we seem to be exactly on the limit of trends that we are able to identify.

Chapter 2 presents the independent conclusion that changes during the 2000-2006 period needed to be larger than 5 to 10% for any trend to have been significantly identifiable, despite meteorological variability. The present chapter shows that the reported emission data indicate a declining trend exactly in the aforementioned range of 5 to 10%. Hence, the amount of change was too small for a trend to be detected with any certainty. Change would need to continue for a larger number of years to be unambiguously identified (or falsified, based on observational data).

7 Trends in traffic emissions

As discussed earlier in the text, there appears to be a mismatch or paradox between declining (reported) PM emissions and observed stabilisation of PM concentrations in ambient air, since around 2000. This phenomenon is omnipresent in western Europe and, therefore, unlikely to be related to a minor source or local situation. Road transport is one of the key sources of PM in western Europe and has received much attention, in recent years, particularly related to the implementation of policy measures for reducing emissions of air pollutants. This raises the question if uncertainty in the estimation of traffic emissions could (partly) explain the aforementioned paradox. Moreover, within urban areas and city centres the relative importance of road transport in relation to PM concentrations will continue to increase, since other major sources of air pollution, such as power plants, are generally located away from urbanisation. Therefore, the trends in road traffic emissions were explored further, to obtain a better idea of 1) how technology advances have influenced urban air quality, and 2) how uncertainties in these assessments might explain the paradox between stabilisation of trends in ambient air quality while reported emissions show a decrease (as shown in the previous chapter). Since analysis of source contributions per sector has shown that, for the Netherlands and surrounding countries, road transport is the major source of NO_x, in this study we looked not only at PM but also at NO_x, as it is an important precursor for Secondary Inorganic Aerosol (SIA) and a good indicator of combustion-related traffic emissions.

7.1 Differences in emission trends for road transport

To be able to compare road transport emission trends in different countries in a similar and consistent way, we used the results from an EU-wide study by Pulles et al. (2008) conducted on behalf of the European Environment Agency (EEA). However, it should be realised that the results from this study are not necessary fully consistent with officially reported data, especially on years before 2000. For example, for $PM_{2.5}$ (Figure 7.1) a trend can be seen in the EEA study that is consistent with the trend in officially reported data, when looking at the entire domain. The small consistent gap between the estimate of the EEA study and official reported data (Figure 7.1) can be explained by structural differences and, for example, by the fact that wear emissions were not included in the EEA study (as it focuses on exhaust emission reduction due to policy measures). However, for the Netherlands, the trend is really different from that in the other EU countries, up to 1996 (Figure 7.1). Therefore, when we discuss the trend from 1990 to 2005, the starting year is important. However, for the period between 2000 and 2005, both data sources are fully comparable and show the same trend. The difference between them for the years 1990 to 1995 is thought to be due to differences in assumed emission factors for pre-EURO[1] vehicles. The Dutch data are likely to have a sound basis, which may than also have consequences for emission data from other countries – since the same types of vehicles are also being driven in other countries. This still needs to be further explored. For NO_x (Figure 7.2) the story is different, in the the Dutch reported data the major emission reduction occurs a few years earlier than the EEA study suggests. Again, after 2000, the trends are exactly the same.

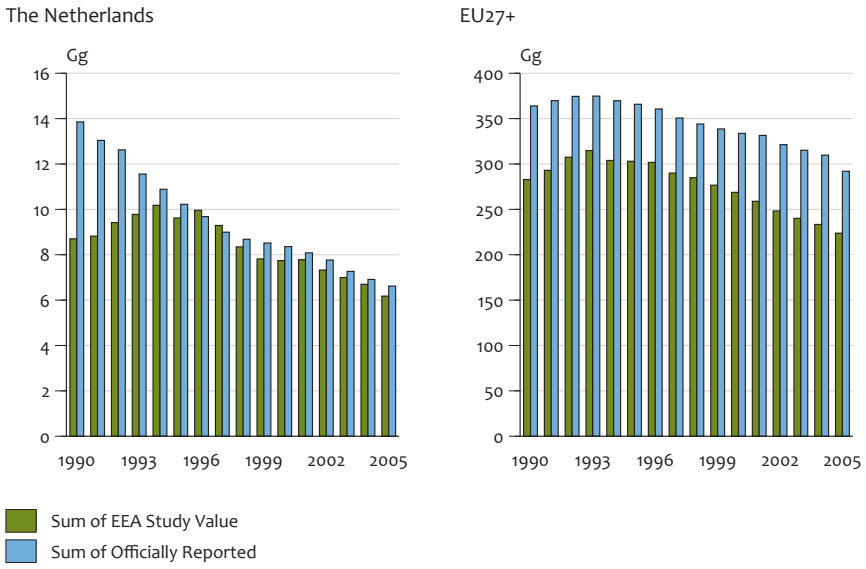
It is important to note that the absolute emission values, as used for identifying European trends (e.g., Figure 7.3 and Figure 7.4), are not fully in line with data from the Dutch Pollutant Release & Transfer Register (PRTR), and there are clear explanations for why this is so (the PRTR includes more detailed data on pre –EURO vehicles.)

7.2 Impact of technology advances in road transport on emissions

Beginning in 1992, the European Commission implemented a series of requirements (European Emission Standards or Euro standards), defining the acceptable limits for exhaust emissions from new vehicles sold within the EU. Non-compliant vehicles cannot be sold in the EU, but new standards do not apply to vehicles already on the roads. The Euro standards have been and continue to be introduced in phases, with the introduction times and actual standards varying according to pollutant, vehicle category, vehicle weight class, or engine volume, and fuel type. As such, the implementation of the Euro standards, to date, has not addressed all pollutants and vehicle categories in an equal manner.

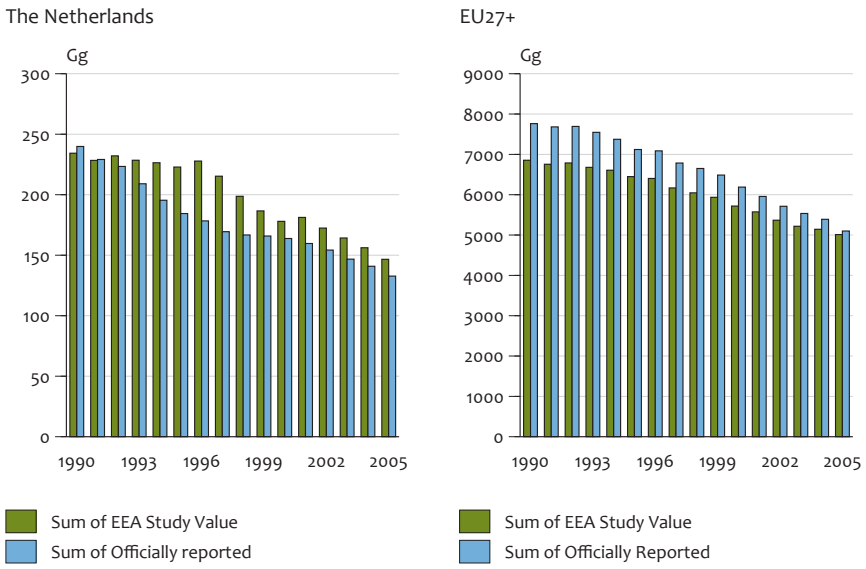
1 EURO here refers to *European emission standards* that define the acceptable limits for exhaust emissions of new vehicles sold in EU member states. The emission standards are defined in a series of European Union directives staging the progressive introduction of increasingly stringent standards.

Official reported and EEA study $PM_{2.5}$ emissions from road transport for 1990-2005 **Figure 7.1**



$PM_{2.5}$ emissions from road transport in the Netherlands (left) and EU27+ (right), for 1990 to 2005, according to officially reported data and a study by the EEA.

Official reported and EEA study NO_x emissions from road transport **Figure 7.2**

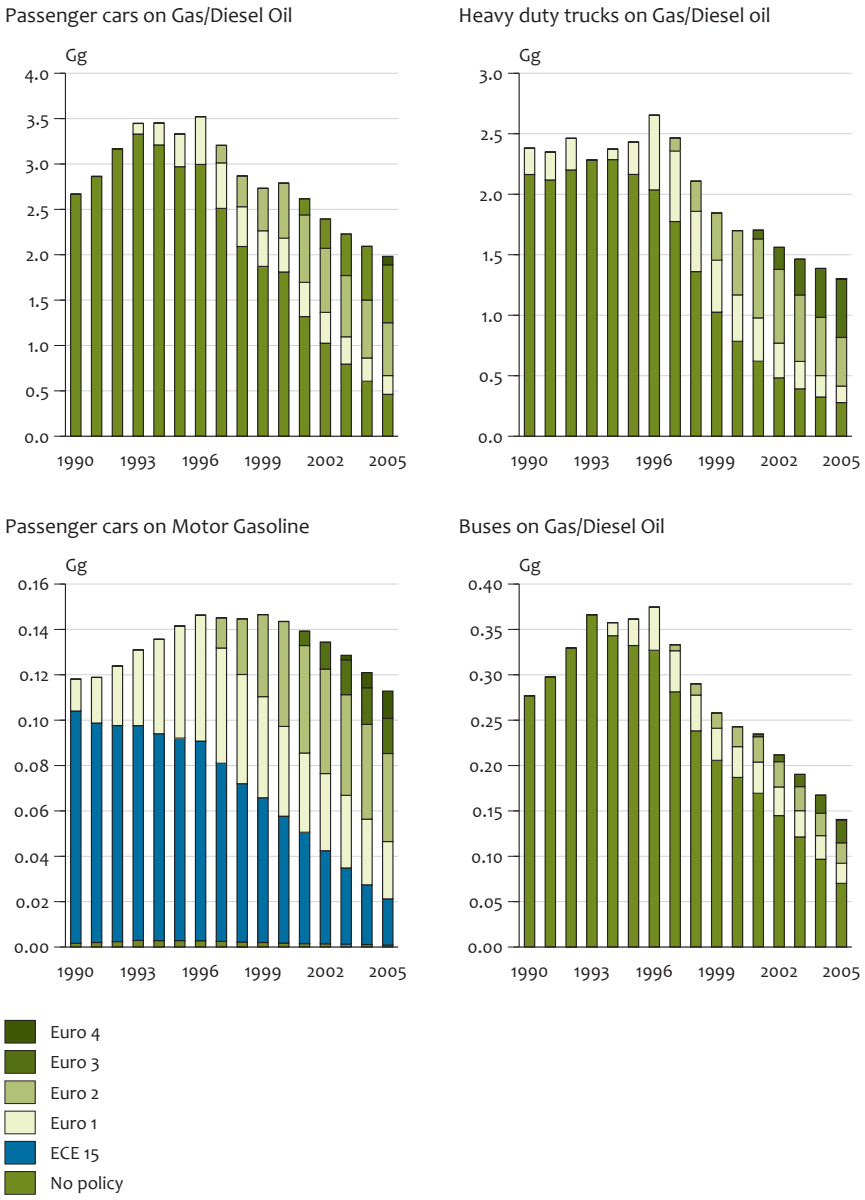


NO_x emissions from road transport in the Netherlands (left) and EU27+ (right) for 1990 to 2005, according to officially reported data and a study by the EEA.

Pulles et al. (2008) have analysed the effectiveness of ECE and Euro Standards in reducing emissions in the entire EU, over a 15-year period (1990-2005). That study made use of Eurostat road transport energy statistics and COPERT 4 vehicle fleet data and emission factors. The TNO Emission Assessment Model (TEAM) was used to calculate emissions for the analysis on a European scale, see Pulles et al. (2008). Here, the calculations by Pulles et al. have been repeated for NO_x and $PM_{2.5}$ in the Netherlands.

The implementation of Euro standards for diesel-fuelled vehicles focused on PM and resulted in substantial emission reductions in primary PM (Figure 7.3, note the different scales of the Y-axis in the sub-figures). For $PM_{2.5}$, emission reductions appeared most significant for heavy-duty trucks and buses. For diesel-fuelled passenger vehicles, the reduction may seem less dramatic, but has been realised despite a tremendous growth in kilometres driven by these vehicles. Detailed analyses for other pollutants and for the rest of Europe, for

Exhaust emissions of $PM_{2.5}$ in the Netherlands **Figure 7.3**



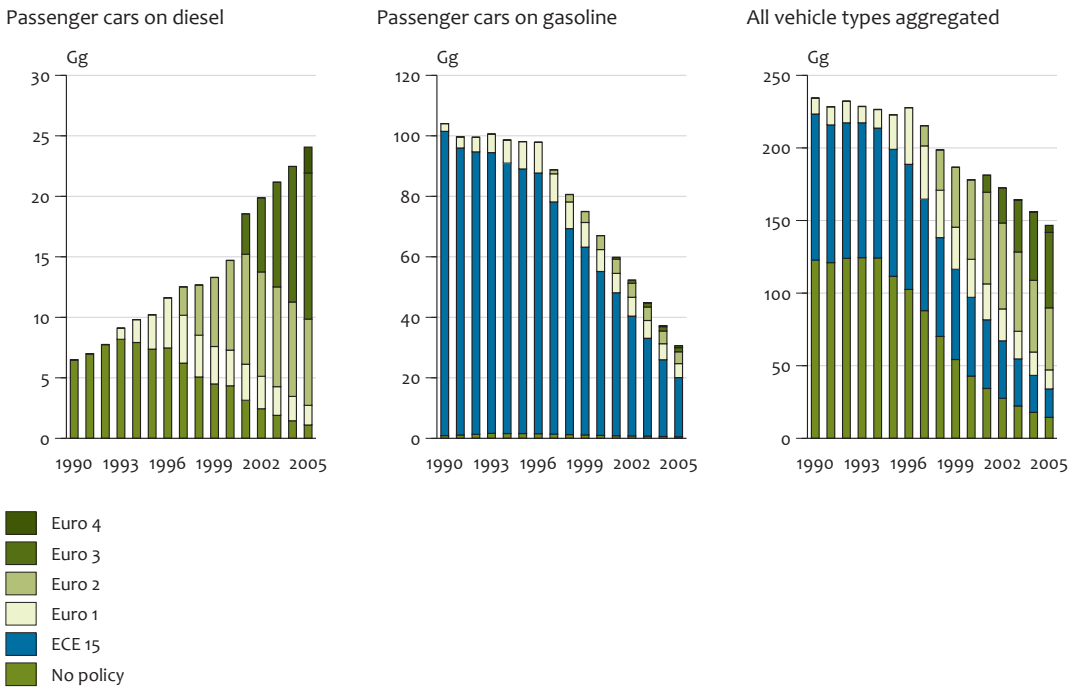
Exhaust emissions of $PM_{2.5}$ in the Netherlands: trends and contribution per vehicle class and vehicle technology type, following the approach described in Pulles et al. (2008).

various scenarios, can be found in the report by Pulles *et al.* (2008).

For NO_x, the emissions are presented only for passenger vehicles on diesel and petrol (Figure 7.4), and the aggregated emission changes over time for all vehicle types and fuel types combined (Figure 7.4). Remarkably, NO_x emissions from passenger vehicles on diesel increased sharply from 1990 to 2006. For diesel-fuelled heavy-duty trucks, NO_x emissions remained stable (data not shown). Therefore, implementation of the Euro standards has not led to reductions in NO_x emissions for these categories. This conclusion is congruent with the type (or lack) of technological adjustments made to these vehicle types to address NO_x emissions under the Euro standards, and the increase in traffic volumes for these categories. The Euro standard for diesel vehicles focuses on PM reductions and disregards the consequences for NO_x. However, the Euro standards did have tremendous effect on reducing NO_x emissions from passenger vehicles on petrol (Figure 7.4) and LPG-fuelled vehicles (data not shown).

Overall, the reduction in NO_x emissions from the road transport sector was only limited (Figure 7.4), because the share of diesel-fuelled vehicles increased over time and NO_x emission reduction in these types of vehicles is limited, and the increasing traffic volumes have counteracted the reduction in emission per vehicle. Based on these data we conclude that the amount of NO_x available to form SIA has not changed much, following the implementation of the

Exhaust emissions in The Netherlands for NO_x **Figure 7.4**



Exhaust emissions in the Netherlands, between 1990 and 2005, for NO_x, from passenger vehicles on diesel (a) and petrol (b), and for all vehicle types aggregated (c): trends and contribution per vehicle class and vehicle technology type (source: Pulles et al., 2008).

Euro standards, as much larger reductions in NO_x would be required to achieve this (see also Chapter 6).

The results depicted in Figure 7.3 and Figure 7.4 illustrate that several impacts were less dramatic than generally expected. However, without the implementation of any Euro standard, the air quality today would have been much worse. Nevertheless, the improvements were less positive than expected. In the 1990s, it was expected that vehicles would become more fuel efficient in the future. However, this expectation has not materialised. Although engines have become more efficient, their fuel-efficiency has been counteracted, for example, by the increased weight of vehicles and their air conditioning systems. As a result, today, basically the same amount of fuel is consumed per kilometre as was the case several decades ago. Moreover, the focus on NO_x emission reductions from petrol-fuelled vehicles partly has been counterbalanced by increasing NO_x emissions from diesel engines.

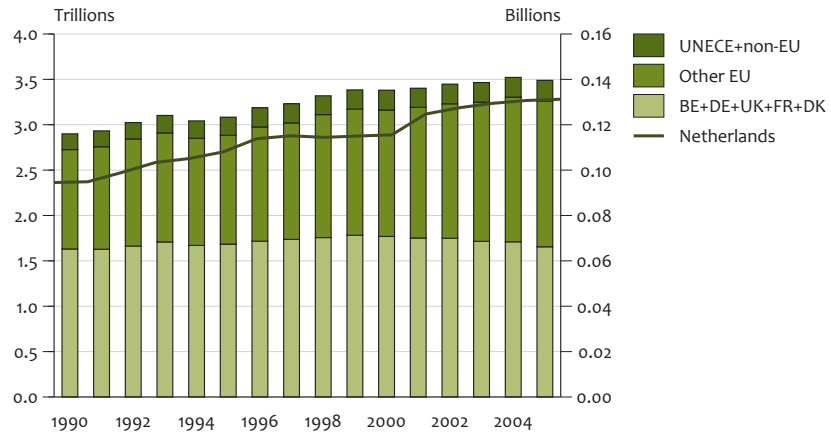
7.2.1 Diesel particulate filters

It should be noted that diesel particulate filters (DPF) were not compulsory under the Euro standards implemented during the period covered by this study (1990-2006). However, retrofitting of certain emission abatement technologies can occur in later stages of implementation of a standard, even years after a particular standard first came into force (*i.e.* phased application). For instance, the upcoming Euro 5 (2009) standard requires that particle filters be retrofitted on existing diesel passenger vehicles and light-duty vehicles already in use. Therefore, most new diesel-fuelled vehicle models already have been fitted with particle filters, in anticipation of the Euro 5 standard (Senternovem, 2008a, b). Furthermore, special subsidies and programmes exist to promote the installation of DPFs. The effect of retrofitting a certain amount of all Dutch vehicles has not been included in the results presented in Figure 7.1. However, including them would not have affected the results, as the introduction of DPFs mostly took place after 2006. In the Netherlands, the number of diesel vehicles that are currently on the roads (2008) and are retrofitted with soot filters is 70,000 for passenger vehicles; 30,000 for light-duty vehicles; and 22,000 for heavy-duty vehicles (Senternovem, 2008). This share has steadily grown over the past couple of years, as the government in 2006 started providing subsidies for retrofitting especially in public transport vehicles. Currently, however, there are no data available from authorities, such as Statistics Netherlands (CBS), on the number of new vehicles that are currently on the road and are already fitted with particle filters. Moreover, it is unclear how efficient the retrofitted filters are in the real world. To analyse the effectiveness of DPFs, the share of kilometres driven with these vehicles, and the locations where they are used (urban vs motorway), would require separate studies, and is outside the scope of the present study.

7.3 Evidence that PM exhaust emissions from road transport are declining

PM exhaust emissions from traffic are mostly $PM_{2.5}$ and consist for a large part of elemental carbon (EC). EC in the Netherlands is closely correlated with black smoke (Schaap and Denier van der Gon, 2007), which is another indicator

Total vehicle kilometres travelled in the road transport sector **Figure 7.5**



Total vehicle kilometres travelled in the road transport sector (Source: Eurostat and COPERT 4).

for air pollution that has been monitored over longer periods than EC. For a discussion on the suitability of black smoke measurements to derive EC concentrations in the Netherlands, we refer to Schaap and Denier van der Gon (2007). Keuken et al. (2008a, b) have shown that in order to track changes in exhaust-related emissions, elemental carbon (EC) is a better indicator than $PM_{2.5}$ or PM_{10}. Keuken et al. (2008a, b) analysed measurements in Rotterdam and found that, while PM concentrations remained stable between 2000 and 2005, black smoke concentrations (and thus EC concentrations) decreased. Keuken et al. argued that, apparently, PM_{10} concentrations stabilised while PM exhaust emissions declined (as indicated by the lower EC concentrations). If this was the case, then gradients within cities could become less sharp. However, for now, this conclusion seems premature, as the overall compilation of Dutch black smoke data (Beijk *et al.*, 2009) shows a stabilising trend for both rural and traffic locations. The hypothesis from Keuken et al. should be further explored by finding matching rural, urban background and traffic locations where the trend in EC or black smoke could be monitored and used for determining a decrease in exhaust emissions.

7.4 Change in non-exhaust emissions

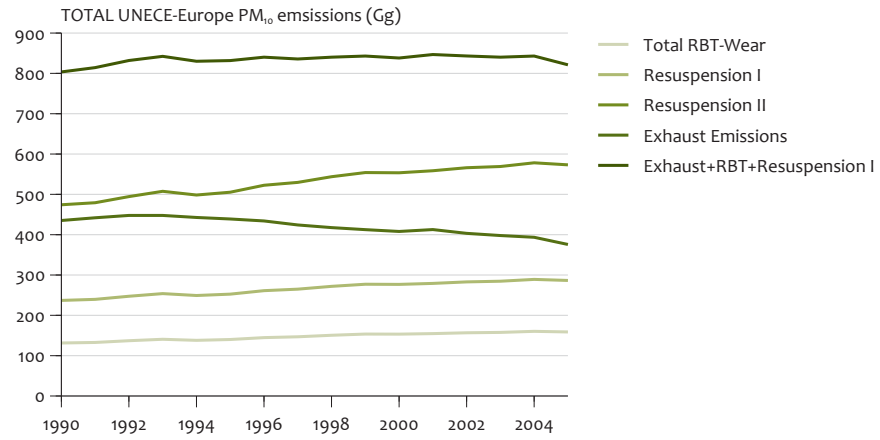
Although PM exhaust emissions have declined over the past few decades due to cleaner engines and exhaust system technologies, non-exhaust emissions have not necessarily declined and, therefore, are sensitive and responsive to growth in traffic volumes. Road wear, brake wear and tyre wear (RBT wear) are all PM emission sources. Moreover, traffic causes local scale atmospheric turbulence, which, in turn, evokes resuspension of dust. For an impression of changes in these emission sources over time, we calculated the emissions from RBT wear and resuspension, by considering the total travelled kilometres in the four groups[2] of countries identified earlier (see Chapter 2).

The total travelled kilometres (tVKMs) per vehicle and fuel type have been calculated by dividing the amount of energy consumed (TJ) per vehicle and fuel type (Figure 7.5) by the typical energy consumption rate per vehicle and fuel type combination (expressed in MJ km^{-1}). The results presented have been aggregated according to technology type (technology = ECE, conventional, Euro 1,2,3… standards). Between 1990 and 2005, the annual total travelled vehicle kilometres increased for all transport and fuel type combinations, in each group of countries. Of the major vehicle categories, most kilometres were driven in heavy-duty vehicles, followed by petrol- and diesel-fuelled passenger vehicles. When considering differences between fuel types, the growth in tVKMs between 1990 and 2006 was most significant for diesel-fuelled vehicles (data not shown).

The calculated total travelled vehicle kilometres were used for calculating the growth in PM_{10} emissions from resuspension, road wear, tyre wear and brake wear in Europe. Emission factors for road, tyre and brake wear (RBT wear) have been sourced from the EEA Revised Guidebook (2008) while those for resuspension are based on TNO estimates (Denier van der Gon *et al.*, 2007; Schaap *et al.*, 2009). The TNO data on resuspension emission factors were provided for the categories of heavy- and light-duty vehicles only. In our analyses, buses and heavy-duty vehicles were assigned the same emission factor, as were passenger vehicles and light-duty vehicles; motorcycles and mopeds were assigned half of the emission amount of light-duty vehicles. In order to conduct a sensitivity analysis, a total of three scenarios were considered for resuspension emissions of PM_{10}. In the scenario 'Resuspension I', the original emission factors from Denier van der Gon et al. (2007) and Schaap et al. (2009) were used. These emission factors were then doubled for the 'Resuspension II' scenario. Analyses have shown that emissions from RBT wear and resuspension were substantial, compared to exhaust emissions (Figure 7.6). PM_{10} emissions from resuspension, in the Resuspension I scenario, were more than half of the exhaust emissions. Emissions from RBT wear were more than one third of the exhaust emissions.

2 I) Netherlands (NL); II) Belgium, Germany, United Kingdom, France, Denmark (BE+DE+UK+FR+DK); III) Other EU Member States (Other EU); IV) Other UNECE and non-EU Member States (UNECE+non-EU)

Trends in exhaust, resuspension, road war, brake wear and tyre wear emissions **Figure 7.6**



Trends in emissions from vehicle exhausts, resuspension, road wear, brake wear and tyre wear, for UNECE Europe.

Furthermore, we found that PM_{10} emissions from both resuspension and RBT wear increased over time (by 20%, between 1990 and 2006). In comparison, PM_{10} exhaust emissions decreased by 14% during the same period. The results indicated that the rise in emissions from RBT wear and resuspension could have contributed to the cancelling out of the PM_{10} exhaust emission reductions.

The resuspension II scenario (Figure 7.6) was added to show that, although resuspension is an uncertain source, in some cases, it might exceed PM_{10} exhaust emissions. The amount of emissions from RBT wear, as calculated in this study, has been validated for the Netherlands, using figures provided in fact sheets for the Dutch Pollutant Release & Transfer Register (Denier van der Gon *et al.*, 2008; Ten Broeke *et al.*, 2008; Rijkswaterstaat *et al.*, 2008). For the Netherlands, emissions from RBT wear made up around 25% of the exhaust emissions from road traffic. The fact that the amount of emissions from RBT wear and resuspension were substantial, relative to exhaust emissions, has consequences for policies aimed at reducing vehicle exhaust emissions.

The implication is that such policies alone would be insufficient to reduce ambient air concentrations of particulate matter. Although future vehicles may emit less particulates, resuspension and RBT wear potentially could neutralise these gains.

7.5 Uncertainty in primary organic aerosol emissions from road transport

The above presented analysis of exhaust emissions from traffic was based on accepted activity data and emission factors. However, recently, a scientific discussion has been started concerning the nature of exhaust emissions and on whether their chemical characteristics may be at the root of some of the 'unexplainable' observations, such as a relatively small $PM_{2.5}$ increment at traffic locations. Most emissions of primary organic particulates are semi-volatile; thus, they partially evaporate with atmospheric dilution, creating substantial amounts of low-volatility gas-phase material. Robinson et al. (2007) showed that photo-oxidation of diesel emissions rapidly generates organic aerosol, greatly exceeding the contribution from known secondary organic aerosol precursors. They attributed this unexplained secondary organic aerosol production to the oxidation of low-volatility gas-phase material. Accounting for partitioning and photochemical processing of primary emissions creates a more regionally distributed aerosol, and brings model predictions into better agreement with observations. The work by Robinson et al. suggests that the semivolatile character of primary emissions requires that, instead of measuring fixed primary organic aerosol emission factors, we should measure the volatility distribution of emissions. Models and inventories must take account of these distributions and their evolution according to photochemical age. A major implication of this work may be that the well-known increments at traffic locations, which are caused by the close proximity to traffic, in reality, may become less sharp, because much of the emitted volatile organic aerosol only forms particulates further away from the source, upon cooling and dilution. This issue could not be further addressed in the present study, but may well become a major issue in the near future, for the proper understanding of the origin of PM concentrations within cities, as well as the trend in these concentrations. This will have an impact on one of our major 'instinctive' interpretations or checks of measurement data, which are that we expect higher concentrations close to the source.

7.6 Model estimate of PM_{10} increment due to traffic at LML locations

In order to estimate the contribution of traffic emissions to PM_{10} concentrations, measured at the LML traffic locations, calculations have been performed using several versions of the Dutch CAR-II model. Emission factors for PM_{10} emissions from urban traffic are available for the period from 2001 to 2020. Several data sets of emission factors haven been published since 2001. Unfortunately, not all available data sets

Modeled traffic contributions to PM_{10} for a number of LML traffic locations Table 7.1

Station	Street	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
136	Heerlen-Looierstraat	5.5	5.3	5.1	4.8	4.6	4.4	4.2	3.9	3.7	3.5	3.3
236	Eindhoven-Genovevalaan	3.8	3.7	3.5	3.3	3.2	3.0	2.9	2.7	2.6	2.4	2.3
237	Eindhoven-Noord Brabantlaan	4.7	4.5	4.3	4.1	3.9	3.7	3.5	3.3	3.2	3.0	2.8
240	Breda-Tilburgseweg	3.0	2.8	2.7	2.6	2.5	2.4	2.2	2.1	2.0	1.9	1.8
433	Vlaardingen-Marathonweg	3.1	3.0	2.9	2.7	2.6	2.5	2.4	2.2	2.1	2.0	1.8
447	Leiden-Willem de Zwijgerlaan	3.1	3.0	2.9	2.7	2.6	2.5	2.4	2.2	2.1	2.0	1.8
448	Rotterdam- Bentinckplein	4.8	4.6	4.4	4.2	4.0	3.8	3.6	3.4	3.2	3.1	2.9
537	Haarlem-Amsterdamsevaart	5.4	5.1	4.9	4.7	4.5	4.3	4.1	3.8	3.6	3.4	3.2
544	Amsterdam-Bernhardplein	3.5	3.4	3.2	3.1	3.0	2.8	2.7	2.5	2.4	2.2	2.1
636	Utrecht-De Jongweg	3.5	3.4	3.2	3.1	3.0	2.8	2.7	2.5	2.4	2.2	2.1
639	Utrecht-Erzeijstraat	4.3	4.1	4.0	3.8	3.6	3.4	3.3	3.1	2.9	2.7	2.6
741	Nijmegen-Graafseweg	5.0	4.8	4.6	4.4	4.2	4.0	3.8	3.6	3.4	3.2	3.0
937	Groningen-Europaweg	3.5	3.4	3.2	3.1	3.0	2.8	2.7	2.5	2.4	2.2	2.1

provide emission factors for all relevant years. Therefore, the calculations for 2001 have been performed using emission factors published in 2006, with CAR-II, version 5. Calculations for 2007 have been performed using emission factors published in 2008, with CAR-II, version 7. Results obtained with the older version of the CAR-II model have been corrected according to a recalibration of the CAR-II model that was performed in 2007 in order to achieve consistent results.

As input for the present calculations, the traffic conditions were used that were provided by municipalities for a study by the RIVM in 2006. It was assumed that traffic volumes per street would increase (on average) by 2.5%, annually.

Comparing results calculated for 2001 with those for 2007 showed that the contribution from emissions by traffic may have decreased by 28%. This translates to a decrease of 4.6% for every year. Assuming this variation to be constant over the years 1998 to 2008, traffic contributions have been calculated for every year of this period. As a random check, the results obtained for the year 2005 were compared with those from an additional CAR-II calculation. The results are within 15% of the estimates shown in Table 7.1. Therefore, it was concluded that the estimates are sufficiently accurate.

7.7 Recent developments in urban traffic volumes

An important uncertainty around the understanding of trends in urban air quality in Dutch cities is the development in urban traffic volumes. As has been indicated (e.g., Figure 7.5), total vehicle kilometres driven have steadily increased over the years, but it is not known if this increase was equally spread over rural, urban and motorway traffic. This is questionable, especially considering that, in recent years, various policies have been implemented to redirect or discourage traffic from entering the urban centres of major Dutch cities, for example, by increasing parking tariffs. As part of the research done within the framework of this BOP programme, Goudappel Coffeng, a consulting firm specialised in traffic, transport and spatial planning, was asked to make an analysis of the trend in inner city traffic, based on data available from recent studies. The Dutch text of the full report on their findings has been attached to this report (Annex D). The main results have been summarised below.

An analysis of traffic data for three major Dutch cities (Rotterdam, Amsterdam and Utrecht) illustrates that traffic developments in these cities differ. This may seem obvious, but the important notion is that we cannot take one city as an 'example' or 'representative' city; conclusions for Rotterdam are not necessarily valid for Amsterdam and vice versa.

The inner-city traffic in Utrecht has declined in recent years. Utrecht's urban traffic in general has shown a mixed trend, apparently depending on local situations, with traffic on the ring roads and city outskirts still increasing. Developments in Amsterdam are comparable, with the inner-city traffic also declining over recent years, but traffic on the ring roads has shown to be stable or increasing.

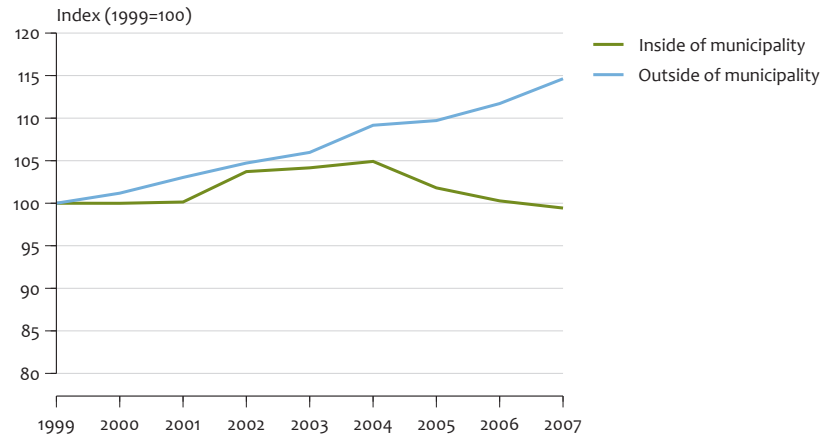
For Rotterdam, the picture is altogether different, with no or very limited stagnation of road transport. An explanation for this may be the more 'open' character of the city of Rotterdam, which still provides room for growth in road transport, whereas the old city centres of Utrecht and Amsterdam no longer have the capacity to absorb more traffic.

Based on data from Mobility Research Netherlands (MobiliteitsOnderzoek Nederland), Goudappel Coffeng concluded (see Annex D) that the findings for Utrecht seemed representative of many similar municipalities that consist of an old city centre (pre-World War II or older), with limited space to accommodate more traffic. This hypothesis was supported by an analysis of the Mobility Research Netherlands data. The findings are summarised in Figure 7.7 and indicate that since 1999:

- Road transport within city limits has slightly decreased after an initial growth.
- Growth in road transport occurs mostly outside the city limits where it steadily continues.

The main conclusion is that, in most centres of major Dutch cities and municipalities, road transport within city limits showed a declining trend, Rotterdam being an important exception. The growth in road transport concerned especially

Trend in vehicle kilometres **Figure 7.7**



Trend in vehicle kilometres remaining within municipality limits (bibeko) and exceeding the municipality (bubeko) (Source: MON 1999-2007, recalculated by Goudappel Coffeng – see Annex D for details).

traffic to and from cities, which is most remarkably visible on the ring roads and outskirts of city centres where, for example, business parks are located. This conclusion implies that, for these cities, the contrast or increment in PM concentrations between urban background and traffic locations has decreased over time since 2000.

Annex A OPS model

The OPS model is used for calculating time averaged concentrations and depositions, on a local to regional scale, of European emissions to the atmosphere (Van Jaarsveld, 2004). OPS model results are used in combination with air quality measurements for assessment of air pollution in the Netherlands (e.g. Velders *et al.*, 2009; Velders and Diederen, 2009). The model describes the processes of emission, transport, chemical conversion and dry and wet deposition.

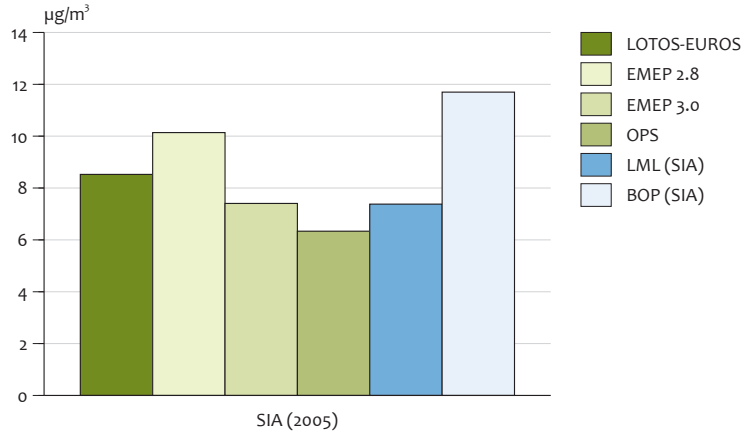
The OPS model is a universal air quality dispersion model and is a fit for those substances for which the atmospheric loss processes can be described as first-order loss reactions. Ozone is therefore excluded. The OPS model can be characterised as a Lagrangian model in which transport equations are solved analytically. Contributions from the various sources are calculated, independently of each other, by using backward trajectories; local dispersion is introduced via a Gaussian plume formulation. Dry deposition, wet deposition and chemical conversion are incorporated as first-order processes and are independent of concentrations of other substances. The only exceptions are the NH_3 and SO_2 dry depositions, which are mutually dependent. The basic meteorological data needed as input for the model (wind direction and velocity, temperature, solar radiation and precipitation) are taken from 16 stations in the national meteorological network in the Netherlands. This also includes data from the 200-metre-high meteorological tower at Cabauw. On the basis of these data, meteorological statistics are derived with a specific pre-processor for six regions within the Netherlands.

For every receptor point, the model determines specific dispersion properties by interpolation of the regional data and depending on terrain roughness. The operational OPS model calculates long-term average (annual) concentrations and depositions. A short-term version of the model is available (e.g. Van Jaarsveld and Klimov, 2009). For a comparison of the OPS model results for SIA and primary $PM_{2.5}$ with results from the LOTOS-EUROS model and EMEP model, see Schaap et al. (2009).

SIA concentrations in the Netherlands, in 2005, according to different model calculations

Although uncertainties about European emissions of SIA precursor gases are not very large (typically 10 to 15%), different models lead to important differences in the SIA concentration levels (see Figure A.1). All modelled concentration levels appear to underestimate the new SIA measurements (BOP). Note that SIA concentrations calculated with the EMEP model 3.0 version are considered superior to those calculated with the 2.8 version. The 3.0

Average annual SIA concentrations on average for the Netherlands, 2005 **Figure A.1**



Average annual SIA (SO_4, NO_3, NH_4) concentrations, for the Netherlands, in 2005, resulting from model calculations with the LOTOS-EUROS, EMEP v2.8, EMEP v3.0 and OPS.

version includes updated SIA chemistry and dispersion (Tarrasón et al., 2008).

The green and red bars indicate average SIA concentrations for all PM_{10} rural measurement locations in the Netherlands, in 2005, derived from routine SIA measurements (LML), and from the same measurements but scaled to fit the new measurements (BOP). For scaling, constant factors were applied per SIA component; 1.4 for NH_4, 1.6 for NO_3 and 1.7 for SO_4. These factors were derived from simultaneous, collocated SIA measurements, by using the (outdated) monitoring network instruments and the updated more accurate monitoring instruments that were also used during the BOP measurement campaign (2007/2008).

Annex B Bias corrections of SIA measurements and model results; approach 1

When comparing modelled and measured concentrations of secondary inorganic aerosol, it should be noted that both concentration levels, for several reasons, are biased. For a proper comparison, these biases should be corrected for. Below, the bias corrections have been done for the comparison of OPS model results with the routine measurements of SIA concentrations. This is according to the historical practice of SIA modelling and measurements (1993-2007).

Nitrate measurements

Nitrate measurements are multiplied by 1.1 to correct for the particle size cut-off of the measurement instrument, which was smaller than 10 microns.

- The routine SIA measurements do not have a clear cut-off size for particles at 10 microns, but the inlet selects particles with a 50% cut-off of around 3 to 4 microns. As a consequence, particles that are larger than 4 microns and still contain SIA components are not effectively measured. Therefore, we applied a correction to the measured SIA components derived from the ratio between SIA components in $PM_{2.5}$ and PM_{10}, which was about 0.8 for NO_3, 0.9 for SO_4 and 1.0 for NH_4 (Weijers *et al.*, 2010). Arguably, significant amounts of nitrate may be present only in the particle fraction between 3 to 4 and 10 microns. To obtain NO_3 in PM_{10}, the measured $PM_{3\text{-}4}$ concentrations were multiplied by 1.1 (=1/0.9).

Modelled SIA levels

Modelled SIA levels did not properly describe all sources of NH_3, and some other sources of SIA were missing. Altogether, modelled SIA concentrations should have been about 2.2 µg/m³ and 1.5 µg/m³ higher, in 1993 and 2007, respectively.

- Sources outside Europe were not included in the model. A significant contribution to the SIA concentration in the Netherlands from across the Atlantic Ocean was derived from estimates on Mace Head, Ireland (Savoie *et al.*, 2002) and by Weijers et al. (2001). A trend in cross-Atlantic SIA contributions is based on the US emission reductions between 1990 and 2006, for NO_x -30%, and for SO_2 -40% (US EPA, 2008). We derived an amount of 0.3 µg/m³ SIA, being about constant in time, from natural sources. The anthropogenic cross-Atlantic contribution to NO_3 was 0.4 µg/m³ in 1990 and 0.3 µg/m³ in 2006; to SO_4 this was 0.6 µg/m³ in 1990 and 0.4 µg/m³ in 2006. The anthropogenic contribution of NH_4 to SIA in the Netherlands was insignificant. The total cross-Atlantic contribution was estimated at 1.3 µg/m³ in 1993 and 0.9 µg/m³ in 2007, with a reduction between 1993 and 2007 of about 0.3 µg/m³.
- Ammonia (NH_3) is underestimated by models in the Netherlands. Ammonia emission density in the Netherlands is among the highest of Europe and uncertainties are very large. NH_4 concentrations are also underestimated by models (see Van Pul *et al.*, 2008; Wichink Kruit *et al.*, 2010). To correct for this bias, modelled ammonium concentrations were multiplied by 1.4. As a consequence, modelled NH_4 concentrations should have been higher; in 1990 by about 0.8 µg/m³ and in 2007 by about 0.4 µg/m³. This leads to an additional trend in NH_4 of about 0.4 µg/m³.
- Sea salt contains about 8% of sulphate, sea salt sulphate, which is not accounted for by the model. Small amounts (for the Netherlands, 0.1 µg/m³, on average) were added to the modelled anthropogenic SO_4 concentration levels to correct for the contribution of sea salt to sulphate. The correction for sea salt sulphate was derived from sea salt measurements and model calculations for the Netherlands (Van Jaarsveld and Klimov, 2009; Manders *et al.*, 2009).

Annex C Particle-bound water

Water is a natural component of PM_{10}. Hygroscopic salts on particles, such as secondary inorganic and organic aerosols, attract water. We included estimates of particle-bound water associated with SIA.

Water bound to particles contributes to PM_{10} mass measurements. Ideally, this water is removed by pre-heating the air sample or by conditioning of filters under dry (50% RH) circumstances, following the reference method (NEN, 12341). However, heating or drying only partly removes the particle-bound water, due to hysteresis (see, e.g., Speer *et al.*, 2003). Therefore, PM_{10}, which for official purposes is determined by the reference method, consists partly of water. So, when comparing the trend in modelled anthropogenic particle concentrations to the trend in PM_{10}, the effect of particle-bound water should be included.

We estimated that the amount of water in PM_{10} due to the presence of SIA, would be about 15% of the SIA mass. The difference in particle-bound water for SIA between 1990 and 2007 was:

- SIA 1993-2007 (approach 1): 2.1 µg/m³ in 1993 and 1.1 µg/m³ in 2007; 1.0 µg/m³
- high SIA trend (approach 2): 3.3 µg/m³ in 1993 and 1.7 µg/m³ in 2007; 1.5 µg/m³

The estimate of particle-bound water was based on the work by Speer et al. (2003). They reported molar ratios of between 0.5 and 1.0 for water attached to ammonium nitrate and ammonium sulphate particles, at a RH of 50% and 75%, respectively. PM_{10} reference measurements are to be performed at a 20 °C temperature and a relative humidity of 50%. Given the hysteresis of the drying process, we considered a molar ratio of 0.75 for both water attached to ammonium nitrate and ammonium sulphate particles to be a conservative estimate.

For a trend analysis, the amount of water attached to anthropogenic organic carbon (OC) may also play a role. However, the amount is probably of minor importance to the trend, because the contribution of hygroscopic OC to ambient PM_{10} levels in the Netherlands is much smaller than the contribution of SIA. Hygroscopic OC is a fraction of the total OC amount. In 2007/2008 OC was about 2 µg/m³ (Brink *et al.*, 2009) and SIA levels were of the order of 10 µg/m³ (Weijers *et al.*, 2010). In addition, the amount of water bound to OC was less than to SIA (Speer *et al.*, 2003).

Annex D Developments of urban traffic (in Dutch)

Bijdrage Goudappel-Coffeng
Wim Korver en Nico Aardoom

Inleiding

Achtergrond en vraagstelling

Binnen Nederland en Europa wordt gesuggereerd dat er een mismatch is tussen onze emissiekennis en de buitenluchtconcentraties. Preciezer; volgens rapportages dalen de emissies maar over de laatste jaren stabiliseren de concentraties. Echter - indien alles verder gelijk / constant blijft - moeten dalende emissies resulteren in dalende concentraties. Een van de hypotheses is dat mogelijk het stedelijk verkeer sterker is toegenomen dan eerder aangenomen. Het aantal verreden kilometers in de stad is een belangrijke variabele en bepalend voor de totale uitstoot in de stad. De kwaliteit van de huidige verkeersprestatiecijfers is onduidelijk. Er zouden verschuivingen opgetreden kunnen zijn in het aandeel diesel binnen de stad, het type voertuig (meer bestelbusjes) en het rijgedrag (professioneler/ agressiever rijden) rijden. Om hier een antwoord op te geven heeft TNO Bouw & Ondergrond Goudappel Coffeng gevraagd een nadere analyse te doen van de ontwikkelingen in het binnenstedelijk verkeer van de afgelopen jaren.

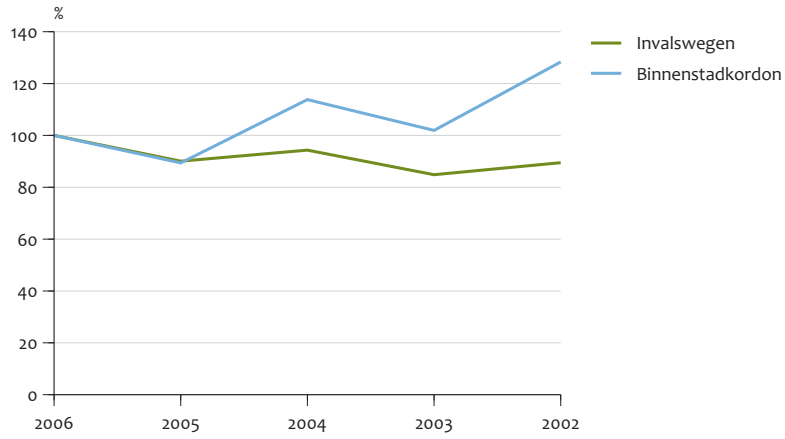
Aanpak

Cijfers over de omvang en ontwikkelingen van het stedelijk wegverkeer worden niet systematisch verzameld. Daarom dat er voor gekozen is enkele steden te selecteren waarvan Goudappel Coffeng zelf data beschikbaar heeft en/of waarvan bekend is dat er goede tijdsreeksen aanwezig zijn. Voor een aantal gemeenten zijn de trends in de ontwikkeling van de verkeersintensiteiten geanalyseerd (zie hoofdstuk 3). Het betreft de steden Utrecht, Rotterdam en Amsterdam. Ten tweede zijn (voor de personenautomobiliteit) enkele geaggregeerde analyses gedaan voor heel Nederland gedaan met het MobiliteitsOnderzoek Nederland (MON).

Trends voorbeeld gemeenten

Beleidsmonitor verkeer 2006, Gemeente Utrecht
De Beleidsmonitor verkeer 2006 rapporteerd vrij uitgebreid de telcijfers voor de stedelijke locaties. Hierbij kan onderscheid worden gemaakt naar drie soorten locaties/ meetpunten:
invalswegen;
binnenstad kordon;
stad (niet centrum).

Gemeten trend aantal motorvoertuigen **Figure D.1**



Gemeten trend aantal motorvoertuigen

Voor invalswegen en binnenstadkordon wordt in figuur 3 een samenvatting van de trend getoond. Hieruit blijkt dat voor de invalswegen de trend stijgend is en voor het binnenstadkordon dalend.

Daarnaast wordt het basismateriaal uit de betreffende rapportage gepresenteerd. Hieruit blijkt dat voor de overige meetpunten (stedelijk) een mixed beeld geldt.

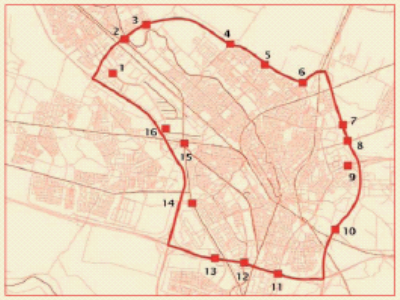
Kordon rond Utrecht: invalswegen: totaal lichtstijgend

Kordon rond Utrecht **Figure D.2**

Motorvoertuigen

Aantal motorvoertuigen	In	Uit	Totaal 2006	Totaal 2005	Totaal 2004*	Totaal 2003	Totaal 2002**
Ochtendspits (7-9 uur)	34.510	24.546	59.056	55.771	57.108	49.553	51.759
Dalperiode (9-16 uur)	80.138	79.145	159.283	140.328	147.380	133.445	143.250
Avondspits (16-18 uur)	27.152	38.961	66.113	60.129	63.796	58.331	59.536
Subtotaal 7-18 uur	141.800	142.652	284.452	256.228	268.284	241.329	254.545
Avond (18-19 uur)	13.456	14.176	27.634	26.249	26.866	24.453	onbekend***
Totaal 7-19 uur	155.256	156.828	312.086	282.477	295.152	265.782	onbekend***

Tabel 3.1. Aantal motorvoertuigen van en naar Utrecht op werkdagen tussen 7-19 uur, gebaseerd op visuele tellingen op één werkdag in het najaar.
* Een aantal tellocaties is sinds 2004 gewijzigd.
dit heeft invloed gehad op de resultaten van de tellingen.
** Het totaal van 2002 was exclusief de Papendorpseweg.
*** In 2002 is geteld van 7-18 uur.



Kaart 3.1. Kordon rond Utrecht.

Binnenstadkordon: daling van aantal motor voertuigen

Binnenstadskordon **Figure D.3**

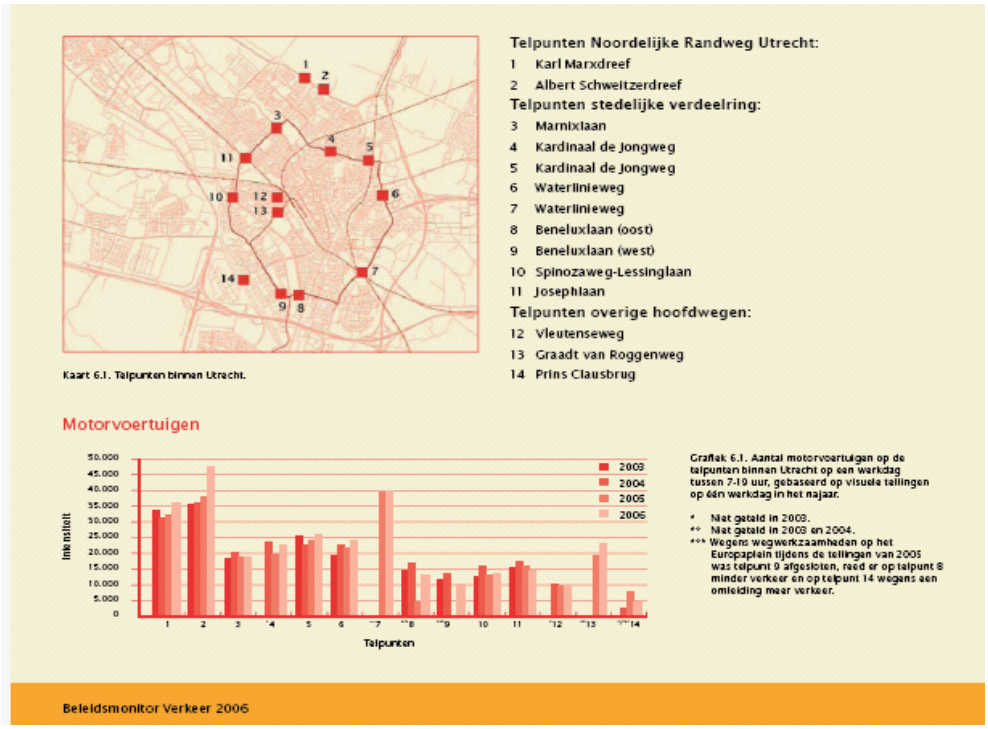
Motorvoertuigen

Aantal motorvoertuigen	In	Uit	Totaal 2006	Totaal 2005	Totaal 2004	Totaal 2003	Totaal 2002
Ochtendspits (7-9 uur)	2.324	2.180	4.504	4.115	4.516	4.182	5.261
Dalperiode (9-16 uur)	10.293	10.478	20.771	18.821	23.657	21.308	26.154
Avondspits (16-18 uur)	2.656	3.712	6.368	5.350	7.842	6.762	9.191
Subtotaal 7-18 uur	15.273	16.370	31.643	28.286	36.015	32.252	40.606
Avond (18-19 uur)	1.300	1.427	2.727	2.469	3.754	2.893	onbekend**
Totaal 7-19 uur	16.573	17.797	34.370	30.755	39.769*	35.145	onbekend**

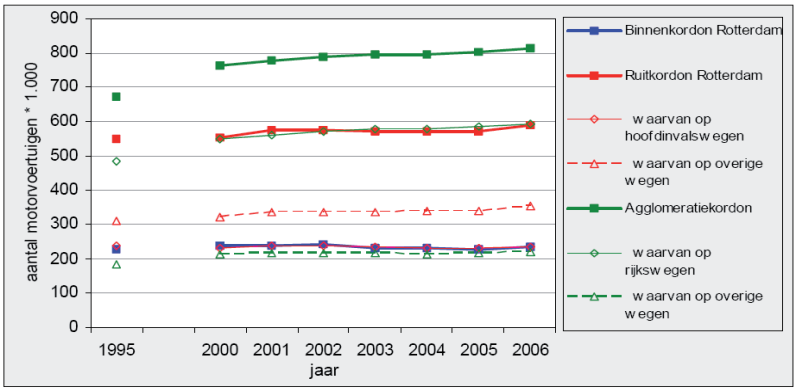
Tabel 4.1. Aantal motorvoertuigen van en naar de binnenstad op werkdagen tussen 7-19 uur, gebaseerd op visuele tellingen op één werkdag in het najaar.
* In 2004 is geteld op een donderdag, waardoor er in verhouding meer motorvoertuigen zijn geteld in de avond(spits).
**In 2002 is geteld van 7-18 uur.

Kaart 4.1. Binnenstadskordon.

Verkeer binnen Utrecht: mixed beeld **Figure D.4**



Ontwikkeling van het aantal motorvoertuigen dat de kordons per werkdag passeert **Figure D.5**



Intensiteiten

De verkeersdrukte in Rotterdam wordt bepaald door tellingen op de drie kordons in en om de stad. De gestage stijging op het agglomeratiekordon heeft zich voortgezet in het afgelopen jaar. Deze stijging vindt plaats op zowel de Rijkswegen als de overige wegen welke onderdeel zijn van het agglomeratiekordon. De stagnatie van de afgelopen twee jaar voor de ruit- en binnenkordon heeft zich omgezet in een stijging. De grootste stijging is zichtbaar voor de verkeersdrukte op de overige 'belangrijke' wegen (niet zijnde de hoofdinvalswegen) welke de ruit van Rotterdam kruisen. Een afbeelding van de verschillende kordons is opgenomen in bijlage A.

In tegenstelling tot Utrecht laat Rotterdamse tellingen zien dat het centrum gerelateerde verkeer nog maar beperkt stagneert. Een verklaring hiervoor kan zijn dat door het open karakter van ' autostad' Rotterdam er in het centrum toch nog enige ruimte is voor groei van het autoverkeer.

Factsheet februari 2005, Genmod infosysteem gemeente Amsterdam

Onderstaande uitsnede van een factsheet uit het GENMOD informatiesysteem (februari 2005) laats zien dat voor het binnenstadkordon de hoeveelheid gemotoriseerd verkeer gedaald is. Door een woordvoerder van DIVV, Amsterdam wordt bevestigd dat daling van het binnenstedelijke verkeer een algemene trend is.

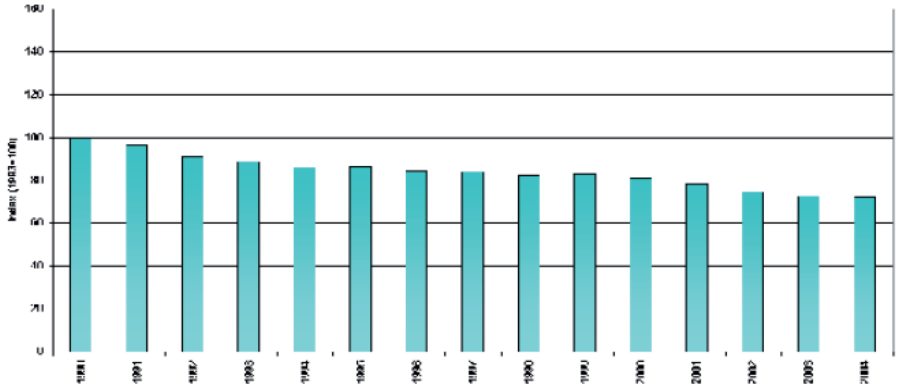
Factsheet februari 2005, hoeveelheid gemotoriseerd verkeer in de binnenstad van Amsterdam **Figure D.6**

Meer fietsers en minder auto's in de Binnenstad

De hoeveelheid autoverkeer in de Amsterdamse Binnenstad is sinds 1990 gedaald. Ten opzichte van 1990 passeerde in 2004 ongeveer 25% minder gemotoriseerd verkeer de Singelgracht.

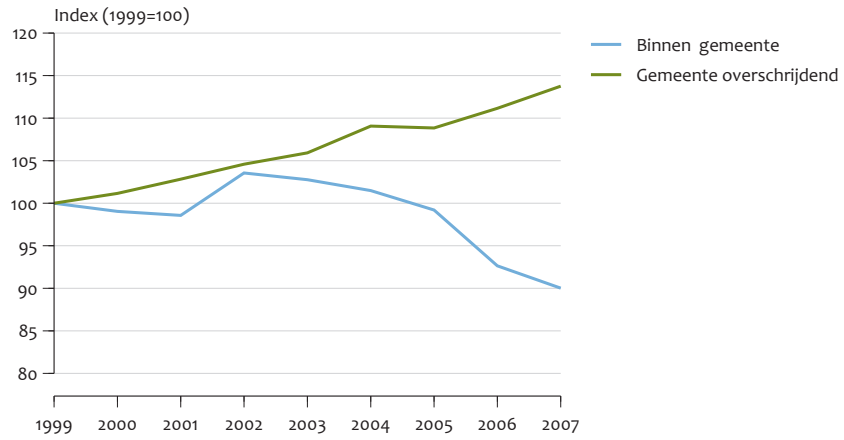
Singelgrachtpasseringen gemotoriseerd verkeer 1990-2004

van 7.00 uur tot 19.00 uur



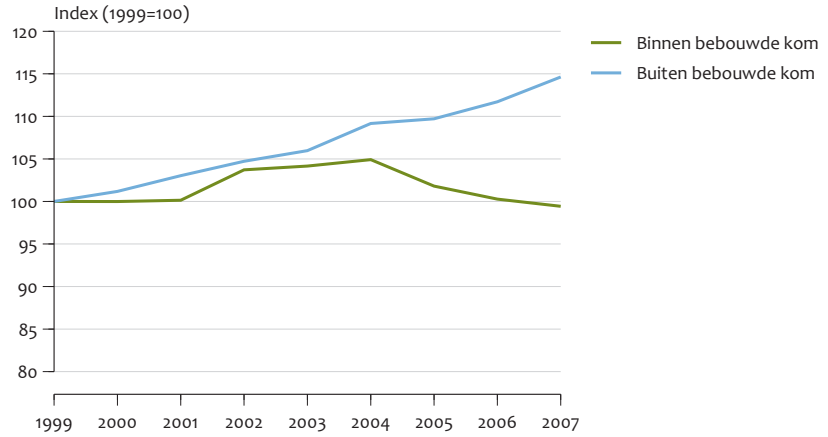
In dezelfde periode is het aantal fietsbewegingen van en naar het Centrum sterk gestegen. Vanaf 1990 is het aantal fietspasseringen toegenomen met meer dan 40%. Vooral de laatste jaren laten een fikse groei zien.

Jaarkilometrage personenautos (MON) **Figure D.7**



Jaarkilometrages personenauto's (bron: Mobiltiteits Onderzoek Nederland voor de jaren 1999 t/m 2007).

Trend kilometrages bibeko en bubeko **Figure D.8**



Trend kilometrages bibeko en bubeko (Bron: MON 1999-2007, bewerking Goudappel Coffeng)

Overzicht voor heel Nederland: Analyses met het MON

In figuur D7 wordt de ontwikkeling van de jaarkilometrage binnen Nederland van de Nederlandse autobestuurders geschetst (personenauto's). Er is gesegmenteerd naar autobestuurder verplaatsingen die binnen de gemeente blijven en gemeente grens overschrijdende verplaatsingen. Hierbij is gecorrigeerd voor gemeentelijke herindelingen die in de beschouwde periode (1999-2007) hebben plaats gevonden. Duidelijk is dat het intergemeentelijke verkeer qua kilometrage toeneemt en het binnen gemeentelijk verkeer afneemt. Dit ondersteunt het eerder geschetste beeld op basis van de gemeentelijke tellinggegevens.

De op basis van het MON geschetste ontwikkeling van de jaarkilometrages komen niet direct overeen met het aantal verreden personen autokilometers binnen gemeentelijke centrumgebieden, de gemeentelijke randen en intergemeentelijk. Om een betere benadering te verkrijgen zijn de verplaatsingsafstanden bewerkt. De binnen gemeentelijke verplaatsingen zijn in zijn geheel gedefinieerd als bibeko-kilometers. De gemeentegrens overschrijdende verplaatsingen zijn als volgt opgesplitst:

verplaatsingen korter dan 5 km: half bibeko, half bibuko;
verplaatsingen groter dan 5 km: 5 km bibeko, overig bubeko.

In figuur D8 worden de ontwikkeling voor de ingeschatte bibeko en bubeko kilometers getoond. Het eerder geschetste beeld wordt bevestigd: de binnen gemeentelijke kilometrages dalen, de buitengemeentelijke kilometrages blijven stijgen.

Locatie typering RIVM meetstations **Table D.1**

Locatie type	Aantal meetstations
Straat	15
Voorstad	4
stedelijk	4
regionaal	16
totaal	39

Locatie meetstations

Wat zijn grosso modo de locatie van de meetstations? Bevinden deze zich op (centraal) stedelijke locaties, aan de stadsranden (invalswegen) of buitenstedelijke.

Van de landelijk meetstations van het RIVM worden de locatietyperingen in onderstaande tabel weergegeven.

Daarnaast worden veel meetstations door gemeenten beheerd. Sommige gemeenten meten vrij intensief (bijvoorbeeld Den Haag), ander gemeenten niet. In het algemeen kan gesteld worden dat het gros van de gemeentelijke locatie gericht is op het centraal stedelijk verkeer en veel minder op de periferie en de invalswegen. Dit betekent dat op deze locaties veelal een daling van de intensiteiten heeft plaats gevonden. Op basis daarvan zouden bij die meetstations lagere waarden gemeten moeten worden dan bij de meetstations die meer aan de periferie liggen. Een mogelijk aanvullende analyses is de meetstations op te delen in twee groepen: meetstations in het binnenstedelijk gebied en meetstations in de periferie.

Conclusies:

Op basis van de beleidsmonitor verkeer 2006 van de gemeente Utrecht kan geconcludeerd worden dat het centrum gerelateerde verkeer de afgelopen jaren gedaald is, het stedelijk verkeer in zijn algemeen een mixed beeld laat zien en dat het verkeer aan de randen (stadsin/uitvalswegen) nog steeds stijgt.

In tegenstelling tot Utrecht laat Rotterdamse tellingen zien dat het centrum gerelateerde verkeer nog maar beperkt stagneert. Een verklaring hiervoor kan zijn dat door het open karakter van ‘ autostad’ Rotterdam er in het centrum toch nog enige ruimte is voor groei van het autoverkeer.

In Amsterdam daarentegen is het beeld voor het centrum heel duidelijk: de intensiteiten van het binnenstadcordon dalen al jaren gestaag.

Het algemene beeld wat voor Utrecht geschetst is zeer waarschijnlijk van toepassing voor tal van gemeenten met een vooroorlogse kern met weinig verkeersruimte. Om dit algemene beeld te toetsen zijn op nationaal niveau enkele trends bepaald op basis van het MobiliteitsOnderzoek Nederland (MON). Uit de analyse komt naar voren dat:

De omvang van het wegverkeer binnen de bebouwde kom is de afgelopen jaren licht gedaald en

De goei van het wegverkeer vindt met name plaats buiten de bebouwde kom.

Op basis van het voorgaande kan geconcludeerd worden dat:
- In de centra van de grote Nederlandse steden in de meeste gevallen een afname is waar te nemen van de omvang van het wegverkeer.
- de groei van het wegverkeer van en naar steden met name plaatsvindt aan de randen van de stad en

References

Amann, M., Cofala, J., Gzella, A., Heyes, C., Klimont, Z., Schöpp, W., 2007, Estimating Concentrations of fine particulate matter in urban background air of European cities, IIASA Interim report IR-07-001, Laxenburg.

Andersson, C., J. Langner, R. Bergstrom, 2007. Interannual variation and trends in air pollution over Europe due to climate variability during 1958–2001 simulated with a regional CTM coupled to the ERA40 reanalysis. Tellus 59B, 77–98.

AQEG, 2006. Primary nitrogen dioxide in the United Kingdom. Report prepared by the Air Quality Expert Group for the Department for Environment, Food and Rural Affairs; Scottish Executive; Welsh Assembly Government; and Department of the Environment in Northern Ireland, Draft, August 2006. Report available from http://www.defra. gov.uk/ENVIRONMENT/airquality/aqeg/index.html

Ayala, A., Kado, N.Y., Okamoto, R.A., Homen, B.A., Kuzmicky, P.A., Kobayashi, R., Stiglitz, K.E., 2002. Diesel and CNG heavy-duty transit bus emissions over multiple driving schedules: regulated pollutants and project overview. SAE 2002-01-1722.

Beijk R., R. Hoogerbrugge, T.L. Hafkenscheid, F.Th. van Arkel, G.C. Stefess, A. van der Meulen, J.P. Wesseling, F.J. Sauter and R.A.W. Albers, 2007a. PM_{10}: Validatie en equivalentie 2006. RIVM Rapport 680708001 (in Dutch), Bilthoven, the Netherlands.

Beijk R, Mooibroek D, Hoogerbrugge R. 2007b. Jaaroverzicht Luchtkwaliteit 2003-2006, RIVM Rapport 680704002 (in Dutch), Bilthoven, the Netherlands.

Beijk R, Mooibroek D, Hoogerbrugge R., 2009. Jaaroverzicht Luchtkwaliteit 2008, RIVM rapport 680704008 (in Dutch), Bilthoven, the Netherlands.

Brink ten, H.M., E.P. Weijers, F.Th. van Arkel FT; D. de Jonge, 2009. Carbon (EC/OC) concentrations as derived from routine PM measurements in the Netherlands, Netherlands Environmental Assessment Agency, PBL Report 500099005, Bilthoven, the Netherlands.

Broeke ten, H., Hulskotte, J., Denier van der Gon, H., 2008. Factsheet: Bandenslijtage wegverkeer, http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx?ROOT=\Water\Factsheets

Carslaw, D.C., Beevers, S.D., Bell, M.C., 2007. Risks of exceeding the hourly EU limit value for nitrogen dioxide resulting from increased road transport emissions of primary nitrogen dioxide, Atmos. Environ., 41, 10, 2073-2082.

Carslaw, D.C. and S.D. Beevers, 2004. Investigating the potential importance of primary NO_2 emissions in a street canyon. Atmos. Environ., 38, 3585–3594.

Carslaw, D.C. and S.D. Beevers, 2005a. Estimations of road vehicle primary NO_2 exhaust emission fractions using monitoring data in London. Atmos. Environ., 39, 1, 167–177.

Carslaw, D.C., Beevers, S.D., 2005b. Development of an urban inventory for road transport emissions of NO_2 and comparison with estimates derived from ambient measurements. Atmos. Environ., 39, 11, 2049–2059.

Cofala, J., Klimont, Z. and Amann, M., 2006, The potential for further control of emissions of fine particulate matter in Europe, IIASA Interim Report IR-06-011, Laxenburg.

Denier van der Gon, Broeke ten, H., Hulskotte, J., 2008. Factsheet: wegdekslijtage wegverkeer, http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx?ROOT=\Water\Factsheets

Denier van der Gon H., Visschedijk A., Boersen G. en Schaap M., Bijdrage Nederlandse landbouw aan secundair fijn stof, TNO-034-UT-2009-00062_RPT-ML.

ECMWF (European Centre for Medium-Range Weather Forecasts), meteorogical data; http://www.ecmwf.int/products/data/software/mars.html

EMEP (Co-operative Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe), 2007, Transboundary Particulate Matter in Europe, Status Report 4/2007, Chemical Co-ordinating Centre of EMEP, Kjeller.

EMEP, 2007. H. Klein, A. Benedictow and H. Fagerli: MSC-W Data Note 1/07, Transboundary data by main pollutants (S, N, O3) and PM, The Netherlands. ISSN 1890-0003. (http://www.emep.int/publ/reports/2007/Country_Reports/report_NL.pdf)

EMEP, 2009a. Centre on Emission Inventories and Projections: emissions used in EMEP models. (See http://www.ceip.at/emission-data-webdab/emissions-used-in-emep-models/)

EMEP, 2009b. Centre on Emission Inventories and Projections: Officially Reported Emission Data. (See http://www.ceip.at/emission-data-webdab/emission-as-reported-by-parties/)

EEA, 2007a, Air pollution in Europe 1990-2004, EEA Report, No. 2/2007, Copenhagen, ISSN 1725-9177.

EEA, 2008a, Aggregated annual country-reported gap-filled emission data, version 6, http://dataservice.eea.europa.eu/dataservice/metadetails.asp?id=1006.

EEA, 2008b, Annual country-reported measurements of ambient air pollutant concentrations (Airbase Data), http://dataservice.eea.europa.eu/dataservice/metadetails.asp?id=1029.

EEA, 2008c, Climate for a transport change: TERM 2007 – Indicators tracking transport and environment in the European Union, No. 1/2008, Copenhagen, ISSN 1725-9177.

EU, 2001, 'Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on national emission ceilings for certain atmospheric pollutants', Official Journal of the European Union 309, 27.11.2001, p. 2.

EU, 2008, 'Directive 2008/50/EC of the European Parliament and the Council of 21 May 2008, on ambient air quality and cleaner air for Europe', Official Journal of the European Union, 152/1.

Fagerli, H. and W. Aas, 2008. Trends of nitrogen in air and precipitation: Model results and observations at EMEP sites in Europe, 1980–2003. Environmental Pollution. 154, 3, 448-461.

Gouw, J.A., Welsh-Bon, D., Warneke, C., Kuster, W.C., Alexander, L., Baker, A.K., Beyersdorf, A.J., Blake, D.R., Canagaratna, M., Huey, L.G., Junkermann, W., Onasch, T.B., Sjostedt, S.J., Sullivan, A.P., Tanner, D.J., Vargas, O., Weber, R.J., Worsnop, D.R., Yu, X.Y., Zaveri, R., 2008. Emission and chemistry of organic carbon in the gas phase and aerosol phase at a sub-urban site near Mexico City in March 2006 during the MILAGRO study, Atmos. Chem. Phys. Discuss., 8, 21265-21312.

Guo, G., Xu, N., Laing, P.M., Maricq, M.M., Hammerle, R.H., 2003. Performance of a catalyzed diesel particulate filter system during soot accumulation and regeneration. SAE 2003-01-0047.

Harrison, R.M., Stedman, J., Derwent, D., 2008, Why are PM_{10} concentrations in Europe not falling? Atmos. Environ., 42, 603-606.

Katz, J.L. and Mirabel, P., 1974, The generation of aerosols by binary homogeneous nucleation, Water, Air and Soil Pollution, 3, 4, 507-513.

Keuken, M. and Voogt, M., 2008a, De fijnstof paradox: tijd voor een nieuwe norm?, Lucht, 5, 11-14.

Keuken, M., Voogt M., Elshout van den, S. and Jonge de, D., 2008b, Meer aandacht voor gezondheid door luchtkwaliteit van fijn stof, Lucht, 4, 13-17.

Kulmala, M., 2003, How particles nucleate and grow, Science, 302, 5647, 1000-1001.

Latham, S., Kollamthodi, S., Boulter, P.G., Nelson, P.M., Hickman, A.J., 2001. Assessment of primary NO_2 emissions, hydrocarbon speciation and particulate sizing on a range of road vehicles. Transport Research Laboratory (TRL): PR/SE/353/2001. Report available from http://www.dft.gov.uk/stellent/groups/dft_roads/documents/downloadable/dft_roads_031921.pdf.

Manders, A.M.M., M. Schaap, M. Jozwicka, F. van Arkel, E.P. Weijers and J. Matthijsen, 2009. The contribution of sea salt to PM_{10} and $PM_{2,5}$ in the Netherlands, Report 500099004, Bilthoven: Netherlands Environmental Assessment Agency.

Matthijsen J. and H. Visser, 2006. PM_{10} in Nederland. Rekenmethodiek, concentraties en onzekerheden, MNP Rapport 500093005 (in Dutch), Bilthoven, The Netherlands.

Mirabel, P.J. and Jaecker-Voirol, A., 1988, Part I: Formation and characteristics of atmospheric aerosols, in Atmospheric Aerosols and Nucleation: Lecture notes in Physics, 309, DOI 10.1007/3-540-50108-8_1004.

Mobiltiteits Onderzoek Nederland (MON), 2007. Voor documenten en gegevens zie: http://www.rws.nl/wegen/innovatie_en_onderzoek/betere_doorstroming/mobiliteitsonderzoek_nederland/.

Planbureau voor de Leefomgeving (PBL), 2008, http://www.milieuennatuurcompendium.nl/indicatoren/nl0081-Relatie-ontwikkelingen-emissies-en-milieukwaliteit.html?i=14-66

Pulles, T., Gondwe, M., Kuenen J., 2008. Effectiveness of European air emission reduction measures, EEA Report, (submitted).

Putten, E.M. van and M.G. Mennen, 1998. Laboratory study of five methods for measuring ambient ammonium aerosol, RIVM-rapport 722601004, Bilthoven: RIVM.

Rijkswaterstaat, Deltares, TNO, 2008, Factsheet: Remslijtage wegverkeer, http://www.emissieregistratie.nl/erpubliek/misc/Documenten.aspx?ROOT=\Water\Factsheets

RIVM, 2008, http://www.lml.rivm.nl/meetnet/lml_ovz.html

RIVM, 2008b, Luchtkwaliteit 2008: NO_2-concentraties niet gedaald, wel daling van fijnstofniveaus, RIVM webbericht, 16 februari 2009, R. Beijk, J. Wesseling, R. Hoogerbrugge, http://www.rivm.nl/milieuportaal/images/PM10_NO2_2008rapportage.pdf

Robinson, A.L., Donahue, N.M., Shrivastava, M.K., Weitkamp, E.A., Sage, A.M., Grieshop, A.P., Lane, T.E., Pandis, S.N., Pierce, J.R., 2007, Rethinking organic aerosols: Semi volatile emissions and photochemical aging, Science, 315, 1259.

Rost J., T. Holst, E. Sahn, M. Klingner, K. Anke, D. Ahrens and H. Mayer, 2009. Variability of PM_{10} concentrations dependent on meteorological conditions. Int. J. of Environment and Pollution, 36, 1/2/3, 3-18

Ruiter, J.F. de, W.A.J. van Pul, J.A. van Jaarsveld and E. Buijsman, 2006. Zuur- en stikstofdepositie in Nederland in de periode 1981–2002, Netherlands Environmental Assessment Agency, PBL report (in Dutch) 500037005/2006, Bilthoven, the Netherlands.

Savoie, D.L., R. Arimoto, W.C. Keene, J.M. Prospero, R.A. Duce and J.N. Galloway, 2002, Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean, J. of Geophys. Res. Atmospheres, 107, D18, pp 4356.

Schaap M. and Denier van der Gon H.A.C., 2007. On the variability of Black Smoke and carbonaceous aerosols in the Netherlands, Atmos. Environ., 41, 5908 – 5920.

Schaap, M., A.M.M. Manders, E.C.J. Hendriks, J.M. Cnossen, A.J.S. Segers, H.A.C. Denier van der Gon, M. Jozwicka, F. Sauter, G.J.M. Velders, J. Matthijsen, P.J.H. Builtjes, 2009. Regional Modelling of Particulate Matter for the Netherlands, Netherlands Environmental Assessment Agency, PBL Report 500099008/2009, Bilthoven, The Netherlands.

Schaap, M., van Loon, M., ten Brink, H.M., Dentener, F.D., Builtjes, P.J.H., 2004a, Secondary inorganic aerosol simulations for Europe with special attention to nitrate, Atmos. Phys. Chem., 4, 857-874

Schaap, M., H.A.C. Denier Van Der Gon, F.J. Dentener, A.J.H. Visschedijk, M. Van Loon, H.M. Ten Brink, J-P Putaud, B. Guillaume, C. Liousse and P.J.H. Builtjes, 2004b, Anthropogenic Black Carbon and Fine Aerosol Distribution over Europe, J. Geophys. Res., 109, D18201, doi: 10.1029/2003JD004330

Schaap, M., Timmermans, R.M.A., Sauter, F.J., Roemer, M., Velders, G.J.M., Boersen, G.A.C., Beck, J.P., and Builtjes, P.J.H., 2008. The LOTOS-EUROS model: description, validation and latest developments. Int. J. of Environment and Pollution, 32, 270-290.

Schaap, M., A.M.M. Manders, E.C.J. Hendriks, J.M. Cnossen, A.J.S. Segers, H.A.C. Denier van der Gon, M. Jozwicka, F.J. Sauter, G.J.M. Velders, J. Matthijsen and P.J.H. Builtjes, 2009. Regional modelling of particulate matter for the Netherlands, PBL-rapport 500099008, Den Haag/Bilthoven: PBL.

Schaap, M., E.P. Weijers, D. Mooibroek and L. Nguyen, 2010. Composition and origin of particulate matter in the Netherlands. Results of the Dutch Research Programme on Particulate Matter, PBL-rapport 500099011, Den Haag/Bilthoven: PBL (in press).

Senternovem, 2008a, Retro-fitting of soot filters in the Netherlands, http://www.senternovem.nl/srp/

Senternovem, 2008b, Retro-fitting of soot filters in The Netherlands http://www.senternovem.nl/srv/

Sokal R.R. and F.J. Rohlf, 1995. Biometry. W.H. Freeman and Company, New York, p 887.

Speer, R. E., Edney, E. O. and Kleindienst, T. E., 2003. Impact of organic compounds on the concentrations of liquid water in ambient $PM_{2.5}$, J. Aerosol Sci., 34, 63–77.

Tarrasón, L., Fagerli, H., et al., 2008. Transboundary acidification, eutrophication and ground level ozone in Europe in 2006, status report 1/2008, The Norwegian Meteorological Institute.

UNECE, 1998. Protocol to the 1979 Convention on Long-range Transboundary Air Pollution on Heavy Metals, Aarhus, 24 June 1998. (http://www.unece.org/env/lrtap/hm_h1.htm)

UN-ECE 1999. The 1999 Protocol to Abate Acidification, Eutrophication and Groundlevel Ozone; 23 Parties. Entered into force on 17 May 2005, (http://www.unece.org/env/lrtap/status/lrtap_s.htm).

USEPA, 2008. Latest Findings on National Air Quality, status and trends through 2006, Office of Air Quality Planning and Standards EPA Publication No. EPA-454/R-07-007, Air Quality Assessment Division, Research Triangle Park, NC, USA

Van Pul W.A.J., Van den Broek M.M.P., Volten H., Van der Meulen A., Berkhout S., Van der Hoek K.W., Wichink Kruit R., Huijsmans J.F.M., Van Jaarsveld J.A., De Haan B., Koelemeijer R., 2008. Het ammoniakgat: onderzoek en duiding. RIVM Rapport 680150002 (in Dutch), Bilthoven, the Netherlands.

Van Jaarsveld, J.A., 2004. The operational priority substances model, RIVM-rapport 500045001, Bilthoven: RIVM.

Van Jaarsveld, J.A. and D. Klimov, 2009. Modelling the impact of sea-salt particles on the exceedances of daily PM_{10} air quality standards in the Netherlands, Int. J. of Environment and Pollution.

Velders, G.J.M. and H.S.M.A. Diederen, 2009. Likelihood of meeting the EU limit values for NO_2 and PM_{10} concentrations in the Netherlands, Atmos. Environ. 43, pp 3060 3069, doi:10.1016/j.atmosenv.2009.03.029.

Velders, G.J.M. and J. Matthijsen, 2009. Meteorological variability in NO_2 and PM_{10} concentrations in the Netherlands and its relation with EU limit values, Atmos. Environ. 43, pp 3858–3866, doi:10.1016/j.atmosenv.2009.05.009.

Visschedijk, A.H.J. and H.A.C. Denier van der Gon, Gridded European anthropogenic emission data for NO_x, SO_2, NMVOC, NH_3, CO, PM_{10}, $PM_{2.5}$ and CH_4 for the year 2000, TNO B&O-A Rapport 2005/106, 2nd version November, 2005.

Visser H. and H. Noordijk, 2002. Correcting air pollution time series for meteorological variability. RIVM report 722601007, Bilthoven, The Netherlands.

Wesseling, Mooibroek, D., van Pul, A., 2007. Trends in jaargemiddelde stikstofdioxide, Milieu, 13, 3, 28-30.

Wesseling, J and R. Beijk, 2008. Korte-termijn trend in NO_2 en PM_{10} concentraties op straatstations van het LML (in Dutch), RIVM Report 680705007, Bilthoven, the Netherlands.

Weijers, E.P., E. Sahan, H.M. ten Brink, M. Schaap, J. Matthijsen, R.P. Otjes, F. Th. Van Arkel, 2010. Contribution of secondary inorganic aerosol to PM_{10} and $PM_{2.5}$ in the Netherlands; measurement and modelling results, Netherlands Environmental Assessment Agency, PBL Report 500099006, Bilthoven, the Netherlands.

Wichink Kruit, R.J., Van Pul W.A.J., Sauter F.J., Van den Broek M., Nemitz E., Sutton M.A., Krol M., Holtslag A.A.M., 2010. Modeling the surface-atmosphere exchange of ammonia. Atmospheric Environment 44, 945-957.

Woudenberg, F., van der Zee, S., Dijkema, M., 2008, Trends in Concentraties PM_{10} en NO_2 in Amsterdam periode 1999-2007, GGD Amsterdam - Leefomgeving, http://www.gezond.amsterdam.nl/client/1/?websiteid=1&contentid=156&hoofdid=5&pagetitle=Milieu_-_bodem_&_lucht

Zee, S.C. van der, H. Helmink and D. de Jonge, 2007. No decrease in PM_{10} concentrations in Amsterdam in the periode 1999-2005, GGD Amsterdam. Medische Milieukunde. - In: Newsletter WHO Collaborating Centre for Air Quality Management and Air Pollution Control; 39, 9-11.

Trends in Particulate Matter

The trend in particulate matter (PM_{10}) and its most important constituents does not contradict the developments in registered emissions. At first glance there appears a contradiction; 'concentrations stable and emissions (slightly) decreasing', however, after a detailed study the statistical power appears to be insufficient to prove the contradiction.
The uncertainty about the trend in the measured results is relatively large due to metrological influences and also due to measurement uncertainties. In this study, several methods are presented for normalisation of metrological effects that reduce uncertainties.
For both emissions and measurements, the decreasing trends over the 1993-2000 period were obviously larger (typically 2 to 4%, annually) than over the period between 2000 and 2008 (0 to 2%, annually). A detailed study on traffic emissions, important with respect to health effects, has shown that any reductions due to cleaner car engines are cancelled out by the increase in total distance driven and the increasing weight of vehicles.
The study has also shown that the uncertainty in the estimation of PM trends over short periods of time (<10 years) is too large for an unambiguous interpretation of the developments. After normalisation for meteorological influences, decreasing trends can be proven more early.

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

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