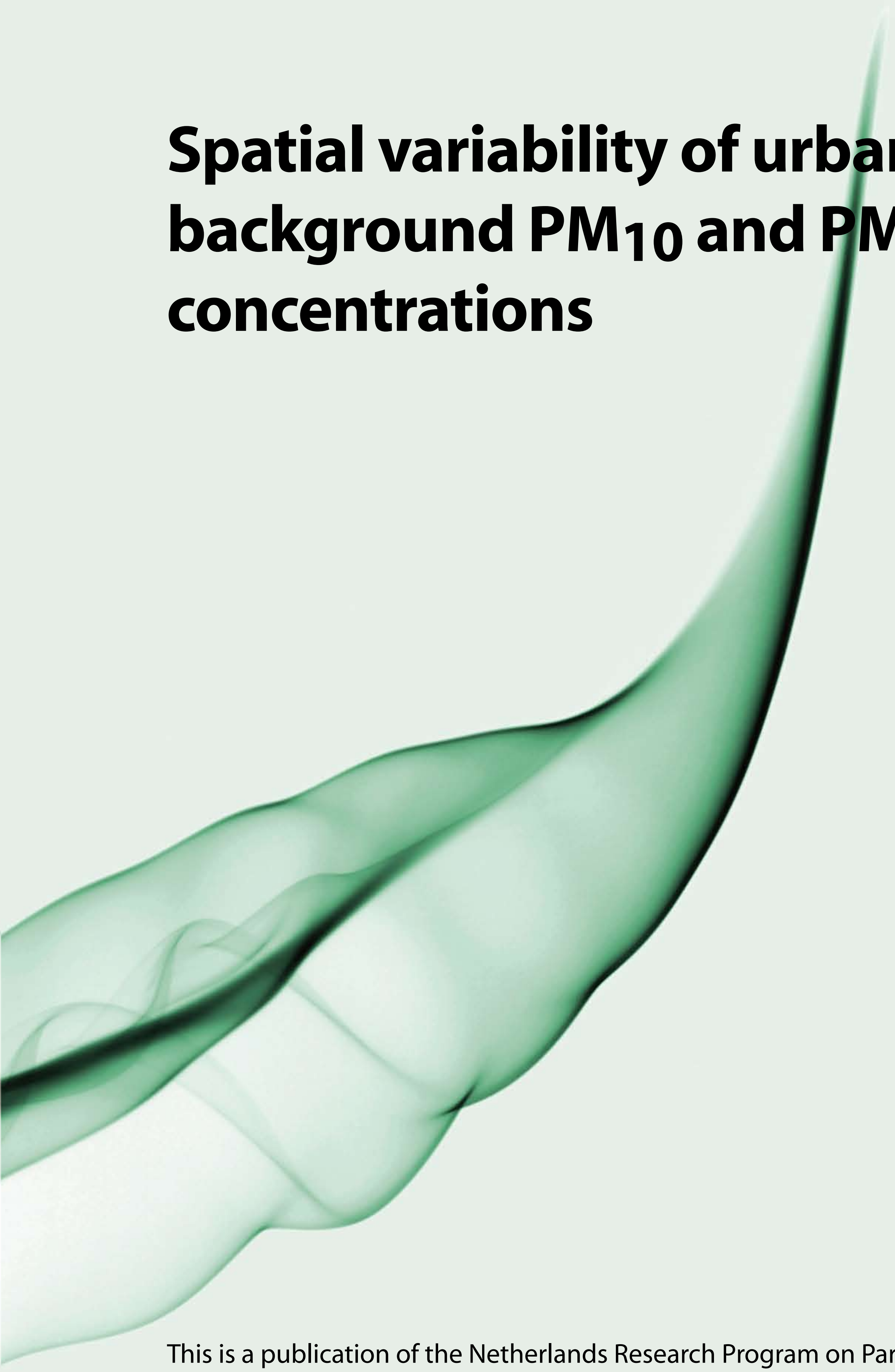
Spatial variability of urban background PM_{10} and $PM_{2.5}$ concentrations



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BOP Report

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Netherlands Environmental Assessment Agency

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Energy research Centre of the Netherlands

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TNO Institute for Applied and Scientific Research
RIVM National Institute for Public Health and the Environment

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Rapport in het kort

Dit rapport beschrijft de studie naar de ruimtelijke variabiliteit, de ruimtelijke representativiteit en temporele variabiliteit van de stadsachtergrondconcentratie van PM_{10} en $PM_{2.5}$ in de stad Rotterdam. De studie is uitgevoerd door TNO en ECN in het kader van het beleidsgeoriënteerd onderzoeksprogramma PM (BOP).

In twee meetcampagnes in september/oktober 2007 en maart 2008, bestaande uit metingen op 11 vaste locaties en aanvullende mobiele metingen, heeft de ruimtelijke variabiliteit van de stadsachtergrond concentratie van PM dezelfde grootteorde als de geschatte meetnauwkeurigheid. Er wordt geconcludeerd dat de ruimtelijke variabiliteit kleiner is dan 10% voor PM_{10} en 5% voor $PM_{2.5}$. De metingen bevestigen de niet significante verschillen tussen de concentraties in de GCN stadsachtergrond grid cellen. Ze suggereren het concept van een PM plateau, waarin een kleine gradiënt van de regionale achtergrondconcentratie leidt tot een constant niveau van de stadsachtergrondconcentratie. Om de onzekerheid in het vaststellen van de stadsachtergrondconcentratie door middel van metingen te verminderen wordt aanbevolen om op meerdere stadsachtergrondlocaties te meten.

Content

Summary

This report describes the study on urban background concentration levels of PM_{10} and $PM_{2.5}$ carried out by TNO and ECN as part of the Netherlands Research Program on particulate matter (BOP). The objectives are to gain insight into:

1. the influence of local shipping, industry, harbour activities and (motorway) traffic on PM concentration levels;
2. the spatial variability of the urban PM background;
3. the urban representativeness of a PM monitoring station (to what extent do measurements collected at a fixed urban background station represent other background areas within the same city?);
4. the local representativeness of a PM monitoring station in an urban background (how large is the area represented in the measurement results from such a fixed urban background station?);
5. the gradient in PM levels from the regional to the urban background;
6. the temporal variability of the urban PM background.

These research objectives are used to address two policy questions:

   a. What is the impact of the urban background variability on the uncertainty in modelling local PM concentrations and exceedances of the EU limit values?
   b. What are the recommendations for an urban background monitoring network?

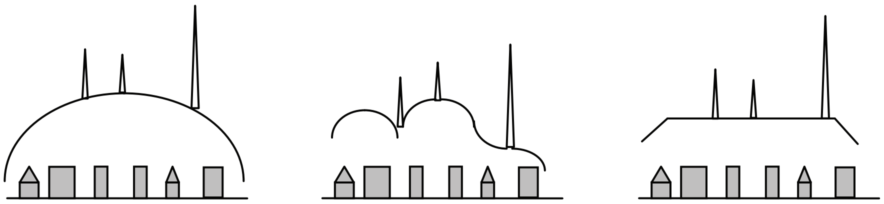
Two monitoring campaigns of one month each were carried out, in which Osiris monitors (optical particle counters) measured PM_{10} and $PM_{2.5}$ concentrations at eleven fixed locations in the city of Rotterdam. The first campaign took place during September/October 2007, the second was held in March 2008. Besides, measurements were taken throughout the studied area, using a mobile unit on ten days during the first campaign. The mobile unit's measurements of PM_{10} and $PM_{2.5}$ were taken with a LAS-x monitor.

The first monitoring campaign, in September/October 2007, was characterised by stable weather conditions, low wind speeds and easterly winds, while the weather during the second campaign, in March 2008, was unstable, windy and wet, and the prevailing winds were west. Due to these different meteorological circumstances, PM concentrations were lower in March 2008 than during the autumn of 2007.

The following conclusions can be drawn with respect to the research objectives:

1. *Local sources;* detection of local influences through Osiris measurements turned out to be difficult, due to the relatively small increments from local sources in the background concentration levels and the measurement uncertainty. As a result, influences from shipping and industry/harbour could not be detected by the measurements. Six of the selected monitoring locations in this study have been regarded as urban background locations that were not influenced by local sources.
2. *Urban spatial variability;* the urban spatial variability is expressed as the relative standard deviation between mean PM concentrations in both monitoring periods (averaged over 16 and 19 days, respectively, of simultaneous monitoring) at the six urban background locations. This was around 5 and 10% for PM_{10} and 5% for $PM_{2.5}$. The estimated measurement uncertainty of the mean PM concentration in both periods was similar to the variability (10% for PM_{10} and 5% for $PM_{2.5}$).
3. *Urban spatial representativeness;* absolute differences between the fixed urban background station in Schiedam and mobile measurements in the urban background were less than 5 $\mu g/m^3$ for PM_{10} and 2.5 $\mu g/m^3$ for $PM_{2.5}$ (average over daytime periods of 6 to 8 hours). The differences were similar to the estimated measurement uncertainty.
4. *Local spatial representativeness;* the study in which mobile measurements are used to assess the local spatial representativeness of measurements at fixed locations is regarded as exploratory research. First results indicated for urban monitoring locations a representative length scale of 3 km, based on measurements collected at half-hourly intervals. More research is required to interpret results, so that the local representativeness of point measurements in an urban environment can be assessed.
5. *Regional-to-urban spatial gradient;* concentration differences between the regional and urban background locations were small: on average 4 to12% for daily mean PM_{10} and $PM_{2.5}$ concentrations. Although measurements were carried out during only two one-month periods and might therefore not be representative for an average year, this result reflects the major influence of large-scale transport and weather conditions. The limited concentration gradient was confirmed by the mobile measuring campaign.
6. *Urban temporal variability;* the temporal variability in PM concentrations at the monitoring locations is high, both during the day and between days. The relative standard deviations between the daily average concentrations during the monitoring periods were from 35 to 50% for

Schematic overview of the urban PM background **Figure 1**



Representation of the urban PM background by three concepts: concentric, spatial variation and the plateau. The peaks on top of the urban background represent the concentration increase by local sources.

PM_{10} and 45 to 70% for $PM_{2.5}$. The temporal variation was very similar for regional background, urban background and traffic sites, indicating that the PM concentration levels in a city are dominated by the regional background contribution.

The following answers to the two policy questions are provided:

Question A. What is the impact of the urban background variability on the uncertainty in modelling local PM concentrations and exceedances of the EU limit values?

The urban background variability is small compared tot the measurement uncertainty. Therefore, the impact on the uncertainty in modelling is presumably small.

The question is related to the GCN[1] background concentration maps used in modelling the local air quality. In 2007, the model resolution of the GCN maps was increased from 5 x 5 km^2 to 1 x 1 km^2. This resulted in a PM_{10} pattern with higher concentrations near local sources, such as motorways. However, the variability in PM_{10} concentration remained small because of the limited range of the scale on the GCN map. For Rotterdam and its surroundings, the range of scale in the GCN PM_{10} map is only 6 $\mu g/m^3$ for both 2006 and 2007. Within the urban background area differences are less than 2 $\mu g/m^3$. The uncertainty (1σ) in the GCN concentration is estimated at 15% (Velders et al., 2008). Taking this uncertainty into account, the differences within the urban background area are not significant. This implies that the higher resolution modelling for the GCN 2007 map does not yield significantly different concentrations and it also does not lessen the uncertainty within the urban background. It must be noted that from a regulatory point of view, the (non-significant) variability is treated as relevant in complying with the yearly averaged PM_{10} limit at locations in inner urban areas.

Measurements carried out during this study confirm the non-significance of the urban background spatial variability of PM_{10}, as well as of $PM_{2.5}$ within the urban background. Whether the change in GCN resolution from 5 x 5 km^2 to 1 x 1 km^2 is an improvement for assessing local PM_{10} concentrations, therefore, cannot be judged by the measurements carried out in this study.

In this study, the representation of the urban background in the GCN maps of 2006 (concentric pattern) and 2007 (spatial variation within the city) could not be confirmed by the measurements. Based on the small spatial variability and small gradients between the regional and urban backgrounds, the measurements suggest a third concept: a small gradient from the regional to urban background leading to an urban background concentration plateau. Figure 1 presents the three concepts.

Due to measurement uncertainty, all three concepts may represent the truth. Dealing with the uncertainty, the plateau concept is the most simplified concept.

With the concept of the urban background plateau in mind, focus with respect to the uncertainty in the GCN maps should not be on the variability within urban areas, but rather on how to reduce the uncertainty in the absolute concentration levels. The present calibration of the GCN model results to measurements from the national monitoring network occurs by adding the same PM_{10} concentration at each location within the Netherlands (14.4 $\mu g/m^3$ in 2007, Velders et al., 2008). It is recommended to assess to what extent city/region-specific calibration may improve the GCN background concentration estimates. Regarding the city/region-specific versus the national calibration procedure, the following is noted. This study focused on the variability of the PM concentration within the city of Rotterdam. Assuming that Rotterdam is the city with the highest variability in the urban background PM concentration levels within the Netherlands due to the relatively large amount of local sources, it is argued that spatial variation in other Dutch cities is also small. This study did not focus on the absolute concentration differences between cities within the Netherlands. With respect to the

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[1] Every year maps are produced showing large-scale concentrations of several air quality components in the Netherlands for which there are European regulations (e.g. Velders et al., 2008). The concentration maps are based on a combination of model calculations and measurements. These maps (called GCN maps) show the large-scale contribution of these components in air in the Netherlands for both past and future years. Local, provincial and other authorities use these maps for reporting exceedances in the framework of the EU Air Quality Directive and for planning.

above mentioned recommendation for local calibration, this issue would be of vital importance.

Another issue following from the small spatial variation in PM background levels, is the focus on PM mass concentration in regulation. The relative increment of local contributions (traffic, shipping, industry/harbour) is limited for both PM_{10} and $PM_{2.5}$, as a result of the large-scale nature of PM_{10} and $PM_{2.5}$. In view of the health effects found near heavy traffic locations, it is recommended to identify a transport-related PM indicator different from PM_{10} and $PM_{2.5}$. More research may be directed towards EC/OC and ultrafine particles: these parameters show larger relative contributions from local sources (e.g. exhaust emissions from road traffic) and are believed to be of relevance in affecting people's health. In the scope of the BOP programme, studies are carried out regarding EC/OC and chemical characterisation. Ultrafine particles represent an area of increasing interest, however this topic is not covered in the BOP programme.

Question B: What are the recommendations on an urban background monitoring network?

From a surveillance and health point of view, the background concentration in a city needs to be known with the highest possible reproducibility. From the rather low spatial variability found in this study, one may argue that one urban background station would sufficiently represent the yearly average urban background in Rotterdam. However, since PM measurement uncertainty is generally high, multiple measurements at urban background locations within one city would decrease the uncertainty in the estimation of the mean urban background concentration. If we assume an uncertainty (1σ) of 10% when using one location, this will reduce to 7% for 2 locations, 6% for 3 locations and 5% for 4 locations.

To be able to quantify the spatial variation within the urban background, the concentration at the locations needs to be measured with higher accuracy than was done in this study. Double (or even triple) the number of measurement recordings at the locations would be needed to reduce the uncertainty below the level of the spatial variability.

Results from this study do not yield straightforward recommendations on the choice of location for an urban background monitoring station. Influence from traffic must be avoided as much as possible. According to the criterion presented by Larssen et al. (1999), there should be no more than 2500 motor vehicles per day within a radius of 50 metres around a monitoring location.

With the concept of the urban background plateau in mind, it is recommended to locate the urban monitoring stations for PM not too close (~1 km) to the edge of a city. More research is needed to assess whether the representation by the plateau is indeed realistic and to define the distance from the edge of a city by which the plateau would be reached.

Due to the small spatial variability for PM within a city, the measurement of PM concentrations will not be very sensitive to the choice of the exact location. However, for other air pollution components, this choice will likely be more critical.

1

Introduction

1.1 Background

This report describes the study on spatial and temporal variability and representativeness of urban background concentration levels of PM_{10} and $PM_{2.5}$, conducted under the auspices of the Netherlands research Program on Particulate Matter (BOP – see Textbox). It is important to have sufficient knowledge on the urban background level, because of several reasons:

- Firstly, this knowledge is required to gain more insight into the degree of exposure to PM for people living in urban areas. In urban areas, high population density coincides with high pollution levels. However, the daily personal exposure to airborne particulate matter is difficult to assess. Usually, epidemiological studies compare results from statistical research on health effects with an average concentration at one station. This is likely to result in significant errors of exposure (Ashmore, 2001). Obviously, a more precise determination of the actual exposure requires detailed knowledge of the concentration levels and variability at locations where people live or frequently pass by.
- Secondly, the new EU Air Quality Directive establishes new air quality standards for $PM_{2.5}$. One of the key elements is a reduction obligation for the $PM_{2.5}$ average exposure levels in major urban agglomerations between 2010 to 2020 (by 15 or 20% depending on the average exposure level). Hence, it is important to adequately assess the urban background through a combination of modelling and monitoring. Consequently, the spatial representativeness of monitoring sites should be as high as possible to establish a cost-effective monitoring network.
- Thirdly, the urban background is important as a basis for modelling local air quality. In these models, the contribution by local sources is added to the urban background concentration.

In view of the above, we need to know (and possibly reduce) the uncertainty in (the determination of) the spatial variation of the urban background. Presently, the urban background in the Netherlands is assessed by a combination of monitoring and modelling, producing large-scale concentration maps (GCN) at a resolution of 1 x 1 km^2 (Velders et al, 2008). Due to the increase in the modelled resolution (from 5 x 5 km^2 to 1 x 1 km^2), in 2008, this study does not aim at assessing how measurement locations are needed to downscale to 1 x 1 km^2. Rather, it assesses whether it is required (in view of assessment objectives) to reduce the uncertainty in the 1 x 1 km^2 PM

Netherlands Research Program on Particulate Matter (BOP)

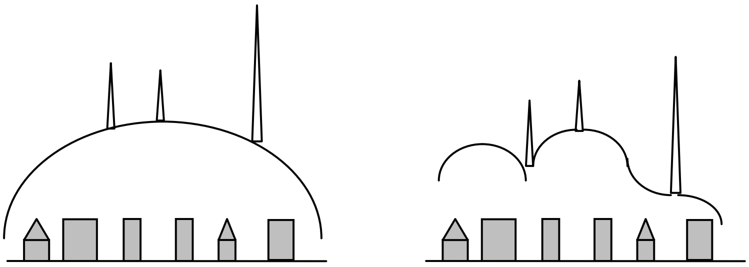
This study is conducted under the auspices of the Netherlands Research Program on Particulate Matter (BOP), a national programme on PM_{10} and $PM_{2.5}$, funded by the Netherlands Ministry of Housing, Spatial planning and the Environment (VROM). The programme is a framework of cooperation, involving four Dutch institutes: 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.

The goal of BOP is to reduce uncertainties about particulate matter (PM) and the number of policy dilemmas, which complicate development and implementation of adequate policy measures. Uncertainties concerning health aspects of PM are not explicitly addressed.

The approach for dealing with these objectives is through the integration of mass and composition measurements of PM_{10} and $PM_{2.5}$, emission studies and model development. In addition, dedicated measurement campaigns have been conducted to research specific PM topics.

The results of the BOP research programme are published in a special series of reports. The subjects in this series, in general terms, are: sea salt, mineral dust, secondary inorganic aerosol, elemental and organic carbon (EC/OC), and mass closure and source apportionment. Some BOP reports concern specific PM topics: urban background (this report), PM trend, shipping emissions, EC/OC emissions from traffic, and attainability of $PM_{2.5}$ standards. Technical details of the research programme are condensed in two background documents; one on measurements and one on model developments. In addition, all results are combined in a special summary for policy makers.

Schematic overview of the urban PM background **Figure 2**



Schematic overview of the urban PM background. Left: urban background level that is increasing towards the city centre. Right: urban background level with more spatial variation. The peaks on top of the urban background are caused by local sources.

Monitoring locations **Figure 3**

The study area Rotterdam situated in the national and European context.

urban background levels in the Netherlands by taking measurements at multiple stations in the urban background. The spatial representativeness of a monitoring station in the urban background area needs to be as high as possible. Although there are recommendations for the locations of rural, urban and 'hotspot' stations (Larssen, 1999), these criteria leave relatively much room for deciding on the exact location. It is difficult to assess the spatial representativeness of a monitoring location. The question to be answered in this perspective is 'What is the rate of change in PM concentrations with increasing distance from a monitoring station?' This is important in urban background areas with a relatively large number of local sources.

The above mentioned aspects of spatial variation and representation are illustrated by the picture on the left in Figure 2. The PM urban background is represented by a concentric pattern with increasing concentration towards the city centre. The background consists of 1) a regional background which depends on the contributions by the large-scale transport of polluting emissions from Europe, the Netherlands and from other sources, such as natural emissions of sea-salt, and 2) contribution from urban sources.

Near local sources, such as urban motorways or inner urban roads, the concentration of PM further increases due to traffic emissions which could be assessed by models, such as a street canyon or a line source model.

The assumption that the PM background concentration gradually increases towards the city centre is questionable, as local sources may cause increases in the local urban background. The spatial pattern of the urban background might be less concentric. This is illustrated by the picture on the right, in Figure 2. Since local sources are considered in GCN maps, the pattern of the urban background in the 1 x 1 km^2 resolution GCN map is more like the picture on the right, in Figure 2. The question is, whether GCN maps 'truly' represent the urban background and what would be the optimal urban background monitoring network for supporting the GCN maps.

This study is carried out by measuring the PM concentrations at several locations within one city. The city of Rotterdam was chosen for this purpose, because of the presence of many different local sources: urban and motorway traffic, industry and harbour activities and shipping (see Figure 3). Rotterdam is considered to be the Dutch city with the highest variability in the urban background PM concentration due to these local sources. This implies that results for Rotterdam can be regarded as worst-case results for other cities.

The study is carried out by ECN and TNO, supported by DCMR Environmental Protection Agency Rijnmond and RIVM.

1.2 Objectives

The study carried out in this part of the BOP programme has the objective to provide insight into:

1. the influence of local shipping, industry, harbour activities and (motorway) traffic on PM concentration levels;
2. the spatial variability of the urban PM background;
3. the urban representativeness of a PM monitoring station (to what extent do measurements collected at a fixed urban background station represent other background areas within the same city?);
4. the local representativeness of a PM monitoring station in the urban background (how large is the area represented in the measurement results from such a fixed urban background station?);
5. the gradient in PM levels from the regional to the urban background;
6. the temporal variability of the urban PM background.

The research objectives are used to address the two following policy questions:

   a. What is the impact of the urban background variability on the uncertainty in modelling local PM concentrations and exceedances of the EU limit values?
   b. What are the recommendations for an urban background monitoring network?

2

Methodology

2.1 Research approach

To study the spatial variation and representativeness and temporal variability of PM in a city, choices were made regarding the monitoring locations, the instruments and the methods of data analysis. Details about the measurement methods and the handling of data are reported in the Annexes of this report, as well as in the *Meettechnisch Rapport (Van Arkel et al, in press)*. The latter presents an overview of all measurements carried out within the scope of the BOP programme and describes the relevant details about measurement protocols and data handling. It supports the database in which measurement data obtained within the BOP programme are collected.

Eleven monitoring locations in and around Rotterdam and the surrounding Rijnmond area were chosen in such a way that (in accordance with EU guidelines) the locations can be expected to represent urban background stations. Secondly, stations were selected to study the gradient from regional to urban locations and a number of urban background locations were selected to study the possible impact from local sources (shipping, industry and traffic). Finally, a typical traffic location was selected as a reference station. An overview of the monitoring locations including their strategic objectives is presented in Paragraph 2.3.

2.1.1 Osiris monitoring campaigns

To meet the objectives 1, 2, 5 and 6 (Paragraph 1.2), two monitoring campaigns were carried out with Osiris Environmental Dust Monitors (‘Osiris’ in short, see Annex 1). Hourly concentrations of PM_{10} and $PM_{2.5}$ were measured during two periods of approximately one month each (during September/October 2007 and March 2008). The Osiris is used because of its reproducibility, cost-efficiency and ease of installing the instrument at different locations during a field campaign. The reproducibility is optimised by a normalisation procedure carried out both before and after the measurement campaign (see Annex 1 for further details and estimation of the reproducibility).

The disadvantage of the Osiris is that the measurements are not conform or equivalent to the reference method (gravimetry). Calibration to the reference method was not carried out because 1) we were more interested in (relative) differences than in absolute concentration levels and 2) available gravimetric measurements from only one location would not yield additional information when evaluating differences between locations. Furthermore, for assessing spatial variation, the reproducibility of the Osiris instruments is more important than measurement of absolute concentrations in accordance with the reference method. The data analyses are thus carried out based on the Osiris measurements, normalised to one reference Osiris level only. Therefore, the Osiris concentrations expressed as ‘μg/m3’ in this report should be regarded as ‘Osiris μg/m3’ rather than ‘reference $\mu g/m^3$’. It is not possible to study absolute levels of PM concentrations and, for example, the fraction of $PM_{2.5}$ /PM_{10}.

The methods of data analysis are presented in Paragraph 2.5.

To identify the influence of traffic, the monthly averaged concentration of NO_2 was measured by passive sampling at the eleven locations. The Palmes tubes used for the measurements were supplied and analysed by Gradko Environmental (UK). The duration of a sampling period depends on the location. The sampling started on the day that the Osiris instrument was installed and ended on the day it was removed.

2.1.2 Mobile measurements

Besides the Osiris monitoring campaign carried out at the eleven locations, measurements were made throughout the studied area with a mobile unit. These measurements were used for objectives 3, 4 and 5 of the BOP programme (Paragraph 1.2).

With respect to the mobile measurements, the following questions were specifically formulated:

1. What is the representativeness of network measurement data within a city? In other words: to what extent are measurements, collected at a fixed urban background station, representative of the quality of the air in other background areas within the same city? This is referred to as the urban representativeness of a monitoring station. (objective 3)
2. Is it possible to quantify a scale length for which measured concentration data are representational? This is referred to as the local representativeness of measurements at a fixed location. (objective 4)
3. What are the changes in PM concentration levels measured when travelling through a city? More specifically, is it possible to estimate an (average) concentration gradient, that is, an absolute concentration change per kilometre? (objective 5)

PM_{10} GCN map for 2006

Figure 4

PM_{10} GCN map for 2006. The monitoring locations from the Osiris campaign are represented by the dots and numbers.

PM_{10} GCN map for 2007

Figure 5

PM_{10} GCN map for 2007. The monitoring locations from the Osiris campaign are represented by the dots and numbers.

The mobile measurements are complementary to the Osiris measurements at the fixed locations. A point measurement at a fixed location is representative of a small area around this point. Mobile measurements can be used to identify the scale of this area, which is referred to as the local representativeness (objective 4). Besides, in the Osiris set-up, to assess the spatial variability, results are in fact compared between those small areas. What is happening in-between the areas is

Description of monitoring locations and specific objectives **Table 1**

Nr	Monitoring location	Monitoring objective	Possible local influence
1	Schiedam Kethel, Educatief Centrum	Regional to urban background	
2	Schiedam West, Tennisvereniging	Urban background	
3	Rotterdam Nieuwe Westen, CBS Mozaiek	Urban background	
4	Rotterdam Provenierswijk, Hildegardis MAVO	Urban background	
5	Rotterdam Crooswijk, Speeltuinvereniging	Urban background	
6	Schipluiden, Zouteveensweg	Regional background	
7	Rotterdam city centre, RIVM Schiedamsevest	Urban background	Inland shipping
8	Vlaardingen, DCMR Vlaardingen	Urban background	Industry/harbour and urban traffic
9	Schiedam, DCMR Schiedam	Urban background	
10	Rotterdam Noord, Bentinckplein	Traffic location	
11	Rotterdam Overschie, DCMR Overschie	Urban background	Motorway traffic

unknown, and for this the mobile measurements give additional information.

Ten daytime periods between 27 September and 16 October were selected. The mobile unit that was used, contained high time-resolution equipment for on-line measuring of ambient levels of mass and particle numbers. The instruments used were a Laser Aerosol Spectrometer (LAS-x), a Condensation Particle Counter (CPC) and an Osiris. From the collected data PM_{10} and $PM_{2.5}$ were calculated. The measurements were taken while driving around different areas of Rotterdam, as well as from stationary positions, that is, near the Osiris locations. At different days, the LAS-x' results were compared to the gravimetric TEOM-SES readings of the DCMR background station at Schiedam. This was necessary to make a sound comparison between the TEOM data and the collected LAS-x data measured in other parts of the city. Further details about applied instruments and methods of data analysis are presented in Paragraphs 2.4 and 2.5.

2.2 GCN 1 x 1 km² background

The large-scale concentration maps (GCN) provided yearly by the Netherlands Environmental Assessment Agency (PBL) serve as the background levels in local air-quality modelling. It is, therefore, important to assess the uncertainty in the GCN maps. The GCN maps are produced by calibrating OPS model calculations to measurements from the Dutch monitoring network (LML) (Velders et al., 2008). For PM_{10}, the current calibration procedure is to add the same value (14.4 µg/m3) to the modelled concentration in each grid cell, in the Netherlands. The spatial resolution of the GCN maps is 1 x 1 km^2. In 2006, the 5 x 5 km^2 model grids were converted to 1 x 1 km^2 grids, by interpolation. In 2007, the modelling resolution was enhanced to 1 x 1 km^2 for most of the Dutch sources. Shipping and foreign sources were still modelled at 5 x 5 km^2 resolution. Consequently, the spatial variability near roads and industry hotspots increased. The PM_{10} GNC maps for 2006 and 2007 are presented in Figure 4 and Figure 5, along with the monitoring locations from the Osiris campaign. It is noted that the GCN map for 2007 was not yet available when the locations in this study were selected.

From Figure 4 and Figure 5, the difference between interpolating from 5 x 5 km^2 (2006) and modelling at 1 x 1 km^2 (2007) is obvious. The 2006 presentation of the GCN concentrations is similar to the picture on the left in Figure 2, with increasing urban background towards the centre of the city. In 2007, the map is similar to the picture on the right in Figure 2, showing an urban area with more spatial variation in the background concentration. Also, the impact by urban motorways is visible on the GCN map, as the urban motorways around Rotterdam are situated in the GCN grid cells with highest concentrations.

However, it should be noted that the range between the lowest en highest concentrations in both GCN maps is only 6 µg/m3. Within the area in which the urban monitoring locations are situated, the range is only 2 µg/m3, at the most. Spatial variability is, thus, very small in both maps.
The spatial variation in the urban PM_{10} background levels from the GCN 2006 and 2007 maps is assessed and compared to the spatial variation derived from the measurement campaigns in September/October 2007 and March 2008. Since the GCN maps present average yearly concentrations, and both campaigns only cover one month each, the comparison is based on differences relative to the concentration levels. This is done by comparing the spatial coefficients of variation (see Paragraph 2.5).

2.3 Measurement locations

As mentioned in Paragraph 2.1, eleven monitoring locations in and around Rotterdam/Rijnmond were chosen and presented on a map in Figure 6. A description of the locations and objectives is given in Table 1.

The locations 2, 3, 4, 5 and 9 are regarded 'true' urban background stations for studying the variability of the urban background. The gradient from regional to urban background is studied from locations 6, 1 and 9. Location 6 is referred to as regional; it is not a rural site, since it is located close to the city of Delft in the north and close to the greenhouse area in the north-west. Location 9 is the DCMR urban background monitoring station at Schiedam, and actually lies in the vicinity of a heavy-traffic crossroad and the motorway A20, but is screened off from this traffic by some multi-storey buildings. The RIVM station Schiedamsevest (7) is characterised as an urban background station; it is located relatively close to the river 'Nieuwe Maas' and the harbour 'Leuvehaven'. Therefore, inland shipping may influence the PM concentrations at this

Monitoring locations in Rotterdam/Rijnmond **Figure 6**

Monitoring locations in Rotterdam/Rijnmond

location, at wind directions from south to east. Location 8 is influenced by traffic from the road that crosses the railway near the railway station at Vlaardingen. Influence from industry and harbour is expected when the wind direction is west to south. Location 10, at Bentinckplein, is a traffic station. The DCMR monitoring station at Overschie (11) is also characterised as a traffic station; influence from motorway traffic (A13) is highest when the wind direction is west to south.

2.4 Mobile measurements

With respect to the mobile measurements, the experimental data presented here were collected with a (small) mobile unit. This unit contained high time-resolution equipment for on-line measuring of ambient levels of mass and particle numbers. The use of mobile laboratories has become a common feature (e.g. Seakins et al., 2002) but recording measurements 'while driving' is a rather new approach. During an extensive campaign, Bukowiecki et al. (2002; 2003) measured on-road concentrations of trace gases and various aerosol parameters in a mountainous Swiss area. In their studies, the suitability of the mobile-measurement approach for short- and long-term air-pollution investigations was shown. Another variant is the 'real-world' measurement of pollutants in exhaust emissions, produced by vehicles driving at a short distance ahead of the mobile laboratory (e.g. Kittelson et al., 2000; Canagaratna et al., 2004).

In our campaign, the mobile measurement technique was used along roads within the city of Rotterdam. Measured aerosol properties were restricted to mass (PM_{10} and $PM_{2.5}$) and particle number. The latter is used for the detection of influence from local traffic nearby, for which data were eliminated. The shortest length scale of interest here is that of an urban street (~30 m). Even with the low vehicle traveling speeds typical in urban agglomerations (~30 km/h), parameters need to be measured with high time resolutions (<20 s).

Measurements were taken on ten days, between 27 September and 16 October, with a mobile unit equipped with registrating measurement equipment, a GPS and a power generator. The measurements were carried out while driving, as well as when parked at certain locations.

For the campaign, three suitable routes were chosen. They are characterised as 'regional to city centre' (going from Schipluiden to Schiedamse Vest, and back), 'urban' (travelling past background locations within the urban centre) and a 'gradient' route from west to east and back (from Vlaardingen to Crooswijk). In general, for all these routes, around 5 locations were visited twice a day; between these locations the shortest possible route was followed, following main roads. At the fixed locations, measurements close to the Osiris were carried out during 15 to 30 minutes. In Annex 8, a detailed scheme of the various routes is given.

For collecting the mobile measurements, different instruments for measuring size distribution and mass concentration of PM_{10} and $PM_{2.5}$ were placed in the mobile unit. The instruments used were a Laser Aerosol Spectrometer (LAS-x (see Annex 2) from PMS Inc.), a Condensation Particle Counter (CPC from TSI inc.) and an Osiris monitor. LAS-x and Osiris are optical instruments. In the campaign the LAS-x and Osiris were compared to a gravimetric instrument, the TEOM-SES of DCMR. The TEOM-SES was used for data analysis, with a standard correction factor of 1.3. This TEOM-SES was used for the comparison instead of FDMS, because most of the standard stations were equipped with the TEOM. In the ten days of measuring, there were no $PM_{2.5}$ measurements by TEOM-SES, so here only the FDMS was used. A description of the instruments is given in Annex 2. The comparison analysis is presented in Annex 3.

The weather conditions for the ten days of our mobile campaign are given in Annex 10, in which data on Rotterdam Airport (from the Royal Netherlands Meteorological Institute (KNMI)) are presented. On most of these days, wind speeds were relatively low, except for the first and last two days. During the campaign, wind direction was predominantly east or west. There was some rainfall on 1 and 3 October.

2.5 Data analysis

This paragraph describes the method of data analysis that was applied in the study. It starts with a description of the analysing method of the two month-long monitoring campaigns, at eleven locations, to assess spatial and temporal variability. Subsequently, the analysing methods are presented that were used for the mobile measurements during specific days.

2.5.1 Osiris monitoring campaigns

The data analyses of the Osiris measurements from the eleven locations are applied after normalisation of the raw data to one reference Osiris level and outlier removal (see Annex 1). Annex 4 presents the normalisation factors.

Spatial variation

For the analysis of the spatial variation, first the locations were identified that represented real urban background locations during both the monitoring periods. This was done by assessing the differences in the mean concentration over both periods. Daily average concentrations of PM_{10} and $PM_{2.5}$ were derived from the hourly data. Afterwards, the average concentration during both monitoring periods was caulated for each location , from the daily averages. Days were only taken into account if the daily average concentrations were available from all eleven locations. Due to power and instrument failure, there were several days without data, for some locations:

- On certain days during the campaign in September/ October 2007, there were problems at locations 7 (Rotterdam city centre) and 9 (Schiedam). As a result, there were only 16 days for which data were available from all eleven locations, during this campaign (3 to 18 October).
- During the campaign in March 2008, severe power problems were encountered at location 2 (Schiedam West). There were so few measurements available from this location that it was decided to exclude it. At location 7 (Rotterdam city centre), the Osiris instrument collapsed after 5 days of monitoring and, consequently, data were lost. And, since there were also two days without data from location 9 (Schiedam), the total number of days for which data were available from all eleven locations, during the second campaign, was 19 (12 to 17 March, 19 to 24 March, 26 March to1 April).

The uncertainty (1σ) in the mean concentration over the monitoring period, per location, was estimated to be 10% for PM_{10} and 5% for $PM_{2.5}$ (see Annex 1). This uncertainty refers to the reproducibility (comparability between instruments). It does not include the bias towards reference concentration values.

The mean concentration at each location was, subsequently, compared to the mean at the predefined urban background locations 2, 3, 4, 5 and 9. The differences (relative to the urban background mean) were plotted in column charts, allowing the identification of real urban background locations and hotspots.

Next, as a measure of spatial variability in urban background PM concentrations, the spatial coefficient of variation (SCV) was calculated. The SCV has been expressed as the standard deviation in measured concentrations divided by the mean measured concentration. The SCV was calculated, based on the locations identified as urban background locations.

The SCV was calculated for the mean concentration per campaign. The uncertainty in the concentration per location was estimated at 10% and 5% for PM_{10} and $PM_{2.5}$, respectively. The SCV needs to be compared to the measurement uncertainty. If SCV values are higher than the uncertainty in the measurements, it can be concluded that the spatial variability was significant. If lower SCV values are found, no significant spatial variability can be determined. In other words, it was less than the measurement uncertainty. Since the uncertainty in the measurements itself is uncertain (it is an estimation), SCV values below the measurement uncertainty do not lead to the conclusion that the spatial variability is zero.

SCVs were calculated for the measurements during both monitoring campaigns and for the 2006 and 2007 GCN maps. A comparison was made between the spatial variability in the measurements and the GCN maps.

Regional to urban background gradient

The regional to urban background gradient analysis was done by wind rose analysis of the gradient between locations 6, 1 and 9, based on daily average concentrations. The gradients were calculated, based on linear regression,using the data from the three locations. This analysis was based on the above mentioned days used in the spatial analysis and on additional days for which the daily mean concentration was available from the locations 6, 1 and 9 (campaign 1: 21 to 29 September + 3 to 21 October 2007; campaign 2: 6 to 17 + 19 to 24 March + 26 March to 1 April 2008).

Temporal variability

A distinction has been made between the regional background, urban background and traffic locations. Apart from the analysis of the temporal coefficient of variation (TCV), based on daily average concentrations (see explanation of SCV), the rate of change on an hourly temporal scale has been investigated.

2.5.2 Mobile measurements

For the mobile measurements, data analysis involved the LAS-x and CPC results. LAS-x data were converted into mass concentrations (PM_{10} and $PM_{2.5}$). The CPC dataset consists of corresponding particle number concentrations, and was used to detect the possible influence of traffic emissions nearby. High values of CPC numbers indicate the presence of vehicles.

When the particle number concentration deviated by more than 3 times the standard deviation, the corresponding LAS-x data were eliminated. This was done for measurements near the Osiris stations, as well as for the data obtained during driving. In addition to this, in the driving route analysis, data were only used when the mobile unit's speed was above 5 km h^{-1} in order to avoid interference from traffic emissions during stop-and-go situations (traffic lights, traffic jams). The unit's speed was calculated from the GPS data.

To explore the urban representativeness of the Schiedam station (location 9), the (mobile) LAS-x data were compared to the TEOM-SES results obtained at this station. To do so, the LAS-x had to be scaled according to synchronised measurements from the TEOM at this station. The time resolution of the TEOM-data is one hour. The measurements with the LAS-x resulted in several periods of about 20 minutes on the Schiedam location. Average LAS-x data were compared with interpolated (hourly) TEOM data. For further details see Annex 3.

In the gradient analysis, the changes in concentrations 'while on the road' were quantified by use of linear regression analysis. Subsequently, the change in the time variable was replaced by the distance covered An average estimate (per day) was then obtained by averaging the absolute results.

When dealing with spatial concentration data in a geographical context, identifying an appropriate scale for analysis is a critical issue. In this study, the mobile monitoring method was applied to collect spatially representative measurements of particulate matter (here only PM_{10} was considered). A geostatistical technique (semivariogram) was applied to characterise the appropriate spatial-analysis scale as defined by the semivariogram range, the maximum distance of spatial dependence within the concentration data. Such a distance may be considered as the maximum distance for which measurements at a fixed location could still be used or would still be 'representative'. This method was used to identify the area size, by characterising the degree of spatial autocorrelation in a dataset. Examples of this technique can be found in Lightowlers et al. (2008) and Larson et al. (2007). For more details see Annex 11.

3

Results and discussion

3.1 Identification of urban background locations based on Osiris data

Annex 5 presents the measured daily average PM concentrations at the eleven monitoring locations, during both Osiris monitoring campaigns. For PM_{10}, it also presents a comparison with TEOM-SES data from measurements by DCMR, simultaneously recorded at locations 9, 10 and 11. The ratios of the measurements collected by both instruments, differed between the locations. However, assuming that the relative uncertainty (1σ) in the mean concentration over the monitoring period of both the Osiris and TEOM-SES were in the order of 10% (see Annex 1 for Osiris), the differences found are within the uncertainty range. The performance of the Osiris instruments, therefore, cannot be evaluated by this comparison. Annex 5 also presents the reference (gravimetric) PM measurements taken by RIVM at locations 9 and 10. Based on the regressions between Osiris and gravimetry, it can be concluded that the Osiris measurements have underestimated the PM concentrations. The underestimation during the first campaign was higher than during the second.

The meteorological variables, as measured by KNMI at Rotterdam Airport, are presented in Annex 10. The prevailing winds during the first monitoring campaign were north-east. The monitoring period was characterised by high pressure systems and little precipitation. The temperature was mostly between 10 and 15 °C, decreasing by the end of the monitoring period.

During the second monitoring campaign, there were few days with high PM concentrations compared to the first monitoring campaign. This can be explained by the large differences in weather conditions. Apart from lower temperatures, the wind direction was 180° different (south-west). Regional PM background concentrations are higher when air is being transported over land rather than over sea. This explains the lower PM concentrations during the second monitoring campaign. Besides, March 2008 was a windy and wet month, causing enhanced dispersion and washout of aerosol particles.

Annex 6 presents the measured NO_2 concentration, indicating the influence from traffic at the locations. The NO_2 concentrations are shown to follow the same pattern in both monitoring periods. Traffic influences can be seen at locations 8, 10 and 11. During the second campaign, with westerly winds, the traffic influence at Overschie (location 11) was indeed higher than during the first campaign. In Vlaardingen (location 8) and at Bentinckplein (location 10), the traffic influence did not seem to depend on the wind direction. It is also apparent, that the NO_2 concentration was the lowest at regional background locations.

An overview of the differences between the average measured concentrations of $PM_{2.5}$ and PM_{10} is presented, allowing the identification of urban background locations. The first monitoring campaign is discussed first, followed by the second.

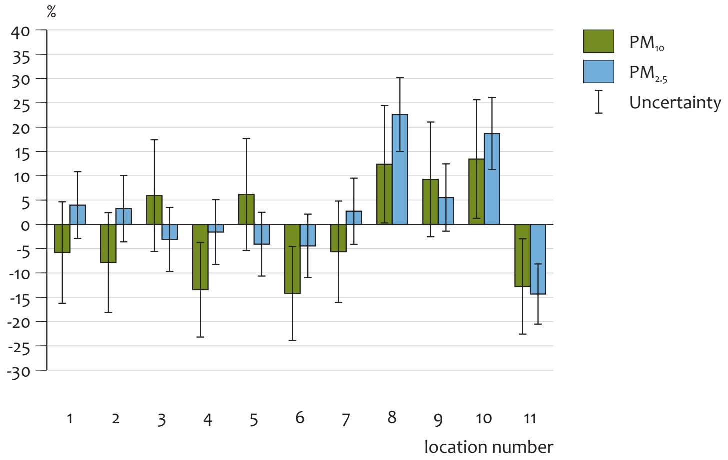
Figure 7 shows the difference between the location specific mean and the urban background mean PM concentration, relative to the urban background mean, for the first monitoring campaign. The urban background mean was derived from the predefined 'true' urban background stations 2, 3, 4, 5 and 9. Uncertainty intervals were based on the estimated reproducibility of 10% (PM_{10}) and 5% ($PM_{2.5}$) in the mean concentration over the monitoring period.

Figure 7 shows concentration differences, ranging from -14 to +13% for PM_{10} and from -15 to +22% for $PM_{2.5}$.As can be seen from the uncertainty intervals, only the largest differences are significant. Several aspects of Figure 7 are discussed:

- *Locations 6 and 10;* As could be expected, the lowest PM_{10} concentration was measured at the regional background location 6, while the highest PM_{10} concentration was measured at the traffic location 10, Bentinckplein. For $PM_{2.5,}$ the concentrations were the second lowest and second highest. These results provide confidence in the reliability of the dataset.
- *Locations 2, 3, 4 and 5;* These locations are the 'true' urban background stations and, as could be expected, had the lowest concentrations (besides the regional location 6 and location 1, which is situated at the edge of the city).
- *Location 9;* The concentration of PM_{10} (and to a lesser extent $PM_{2.5}$) was relatively high. It may be that, with easterly winds, the impact of traffic from the nearby crossroads and the motorway A20 was stronger than expected. However, this did not show up in the NO_2 measurements.
- *Locations 7 and 8;* These locations were urban background stations, potentially influenced by local sources, such as inland shipping, harbour/industry and traffic. At location 7, as was expected in view of the prevailing easterly winds, no impact of shipping was measured for PM_{10} and $PM_{2.5}$. Hence, location 7 was regarded a 'true' urban background location during this monitoring period and has been incorporated in the analysis of spatial variability (Paragraph 3.2).

PM concentration difference — Figure 7

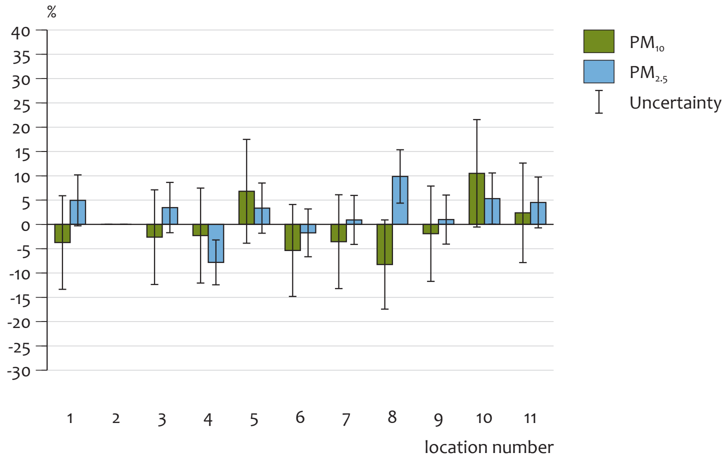
First monitoring campaign



Difference between the location specific mean and the urban background mean PM concentration, relative to the urban background mean, during the first monitoring campaign (%). Here, the urban background mean is the mean at locations 2, 3, 4, 5, and 9.

PM concentration difference — Figure 8

Second monitoring campaign



Difference between the location specific mean and the urban background mean PM concentration, relative to the urban background mean, during the second monitoring campaign (%). Here, the urban background mean is the mean at locations 3, 4, 5, and 9.

The concentrations of PM_{10} and $PM_{2.5}$ were relatively high at location 8, which was attributed to the impact of traffic.

- *Locations 6, 1 and 9;* These locations were selected to determine the gradient from the regional background to the urban background. From these three locations, the lowest PM_{10} and $PM_{2.5}$ concentrations were measured at location 6 and the highest concentrations were measured at location 9, as was . Therefore, the results may be sufficient for analysing this gradient.
- *Location 11;* Since the Overschie station is a 'motorway station' it is remarkable that both PM_{10} and $PM_{2.5}$ concentrations were low (for $PM_{2.5}$ even lower than regional background). In view of the potential influence of the A13 motorway near to station, this was unexpected. Indeed, the NO_2 measurements indicated influence from traffic, even though the prevailing winds were easterly (see Annex 10). From the comparison with TEOM-SES data (Annex 5), it is not possible to conclude whether the Osiris instrument at location 11 has functioned inadequately. In view of this uncertainty, the data from location 11 were not used in further data analyses.

Figure 8 shows the difference between the location specific mean and the urban background mean PM concentration, relative to the urban background mean, during the second campaign. The urban background mean was derived from the predefined 'true' urban background stations 3, 4, 5 and 9.

Measured spatial variability of urban background PM concentration **Table 2**

	PM_{10}			$PM_{2.5}$		
Monitoring campaign	**locations**	**n**	**SCV (%)**	**locations**	**n**	**SCV (%)**
1 (September/October 2007)	2,3,4,5,7,9	6	9.3	2,3,4,5,7,9	6	3.9
2 (March 2008)	3,4,5,7,9	5	4.3	3,4,5,7,9	5	4.6

Spatial coefficients of variation (SCV) in the PM urban background based on the average concentration at the locations over the monitoring periods.

From a comparison between Figure 8 and the results from the first monitoring campaign in Figure 7, is concluded that the range of variability of the concentrations between the locations during the second campaign was far smaller than during the first, both for PM_{10} and $PM_{2.5}$. Also, the overall levels of PM were lower during the second campaign. The latter was against expectations as during the winter period higher concentrations were expected due to less favourable dispersion conditions compared to during the first monitoring campaign. However, the wind direction was mainly south-west with relatively high wind speeds, while during the first campaign easterly winds dominated. In fact, meteorological conditions were reversed, compared to what was expected during the set-up of the monitoring strategy. The prevailing winds during this month were representative of a large part of an average year in the Netherlands. In this respect, the campaign has been very useful.

Both PM_{10} and $PM_{2.5}$ concentration differences ranged from -8 to +10 %. Taking the measurement uncertainty into account, those differences are not significant. Main aspects to discuss are:

- *Location 5;* The PM_{10} concentration at this urban background location was high, compared to the other urban background locations. During the campaign, construction activities took place at the cemetery, located to the north-east. However, the wind from that direction was very light, so it is unlikely that the activities influenced the measurement. Therefore, location 5 was retained in the PM_{10} data analyses.
- Location 7; No influence from shipping was seen. Location 7 was regarded as a 'true' background location during this period and has been incorporated in the analysis of spatial variability (Paragraph 3.2).
- *Location 8*; Typically, the PM_{10} concentration was in the lower range, while the $PM_{2.5}$ concentration was in the higher range. The low PM_{10} concentration is puzzling, especially since the NO_2 measurements indicated a high traffic contribution. Under the westerly wind circumstances, influence would be expected from harbour and industrial activities. It may be that the influence of the harbour and industrial activities were restricted to the finer fraction of PM (due to combustion processes). Care must be taken, however, because the differences are small compared to the measurement uncertainty.
- *Location 11;* The motorway station situated on the east side of the motorway A13, had a higher ranking compared to the first campaign. Due to the south-west wind direction, the impact of the motorway was indeed expected to be higher during the second campaign.

3.2 Spatial variability in urban background

The spatial variability in urban background PM concentrations is measured by the spatial coefficient of variation (SCV). The SCV is expressed as the standard deviation in measured concentrations divided by the mean measured concentration. Locations representing the urban background monitoring stations were: 2 (only available during the first monitoring campaign), 3, 4, 5, 7 and 9. Table 2 presents the SCV values for PM_{10} and $PM_{2.5}$, based on the mean concentration at the mentioned locations during the monitoring periods.

The SCV values were lower than the measurement uncertainty (1σ), which was estimated at 10% for PM_{10} and 5% for $PM_{2.5}$. It must be concluded that, during both monitoring campaigns, no significant spatial variability could be determined in the period's mean concentration at the urban background locations.

Also, a single-factor ANOVA analysis, based on daily average PM_{10} concentrations at the urban background locations during the first monitoring campaign, indicated that the spatial variation was not significant ($p>0.05$). This means that the spatial variation was far less than the temporal variation from day to day (see also Paragraph 3.6).

Under conditions that favour high PM concentrations (such as during the first monitoring campaign in September), the spatial variability in the urban background concentration of $PM_{2.5}$ was less than for PM_{10}. Local sources of PM_{10}, such as non-exhaust emissions from traffic and construction activities, may account for this.

To put the calculated urban background PM_{10} variability in perspective, the variability in GCN maps was assessed for comparison. Keeping in mind that GCN maps are yearly averages and the monitoring campaigns only covered one month each, calculating SCV values allowed a normalised comparison.

The GCN concentrations for the 1 x 1 km^2 grid in which the monitoring locations were located are presented in Annex 7. The concentrations at locations 2, 3, 4, 5, 7 and 9 were used to determine the SCV in the urban background. The calculated SCVs for both monitoring campaigns, GCN 2006 and GCN 2007, are presented in Table 3.

The low SCV values from the GCN reflect the lack of spatial variation in PM, due to long-range transport.

The spatial variability is lower in the GCN maps than in the measurements. The higher, measured SCV values do not automatically mean that the spatial variability was larger than

Spatial variability of urban background PM10 for measurements and GCN **Table 3**

Measurement / GCN map	n	SCV (%)
Monitoring campaign 1	6	9.3
Monitoring campaign 2	5	4.3
GCN 2006	6	1.2
GCN 2007	6	1.8

Spatial coefficients of variation (SCV) in the urban background concentrations for both monitoring campaigns and GCN.

would be expected from the GCN data. Due to the measurement uncertainty, one may only conclude that the spatial variability of PM_{10} was smaller than the measurement uncertainty. In fact, the uncertainty in the GCN concentrations is estimated at 15% (Velders et al., 2008), indicating that the variability of 2% is not significant.

The question now is: how well do the monitoring locations represent the 1 x 1 km^2 grid cells of the GCN? One may expect, in view of the relatively low spatial variability in the urban background cells, that within a grid cell the variability would be low, as well. Hence, a monitoring station located at a sufficient distance from local sources (no more than 2500 motor vehicles per day within a radius of 50 meter; Larssen, 1999) would be representative of the urban background concentration within a 1 x 1 km^2 grid cell. One of the goals of the mobile measurement campaign, related to this question, was to assess the spatial representational scale around the measurement locations. (see Paragraph 3.4).

3.3 Urban representativeness of a measurement station

The first question with respect to the mobile measurements was if point measurements of PM_{10} and $PM_{2.5}$ at an urban background station were comparable with average levels measured in other parts of the city (complying with the definition of urban background). Location 9 in Schiedam is the official DCMR background station at which PM is measured with TEOM instruments (see Annex 40). To investigate the representativeness of this station for other background locations, LAS-x measurements were first levelled with the TEOM readings by using the factors determined in Annex 40 (see the Annex for more details). Also, any possible influence of traffic was considered and if necessary, affected data were removed. The resulting time series were compared with the corresponding time series of the TEOMs from this location.

Figure 9 gives three examples of such time series. These were registrated with the LAS-x and TEOM equipment, on 27 September, 4 and 11 October.. The LAS-x data points represent averages over the stationary, as well as the 'mobile' measurement periods. The first point in the LAS-x graph is always a measurement at a fixed location. On 27 September, the route which was driven was from Schiedam Kethel (location 1) to the city centre of Rotterdam (Schiedamsevest, location 7) and back. The west-to-east route on 4 October from Vlaardingen (location 8) to Crooswijk (location 5), was driven twice. This was also the case for the north-to-south route on 11 October, from Overschie (location 11) to Schiedamsevest (location 7). The figures for the remaining days are given in Annex 6.

Visual inspection learned that the LAS-x recordings (to some extent) resembled the hourly variation observed at Schiedam, for both PM_{10} and $PM_{2.5}$. The TEOM variation appeared quite considerable: changes in the order of 20 µg/m3 or more were observed within a timeframe of a few hours. This pattern was only partially followed by the LAS-x. The LAS-x series further possessed a variation in time that was smaller in magnitude and higher in frequency, due to the shorter averaging time intervals revealing local influences. An example is the relatively large difference for PM_{10} (and less for $PM_{2.5}$) found on 27 September around 13:30 hrs (route from location 7 to location 3). This was caused by road construction, resulting in more resuspended dust and many traffic jams. This route was, therefore, not representative of the urban background.

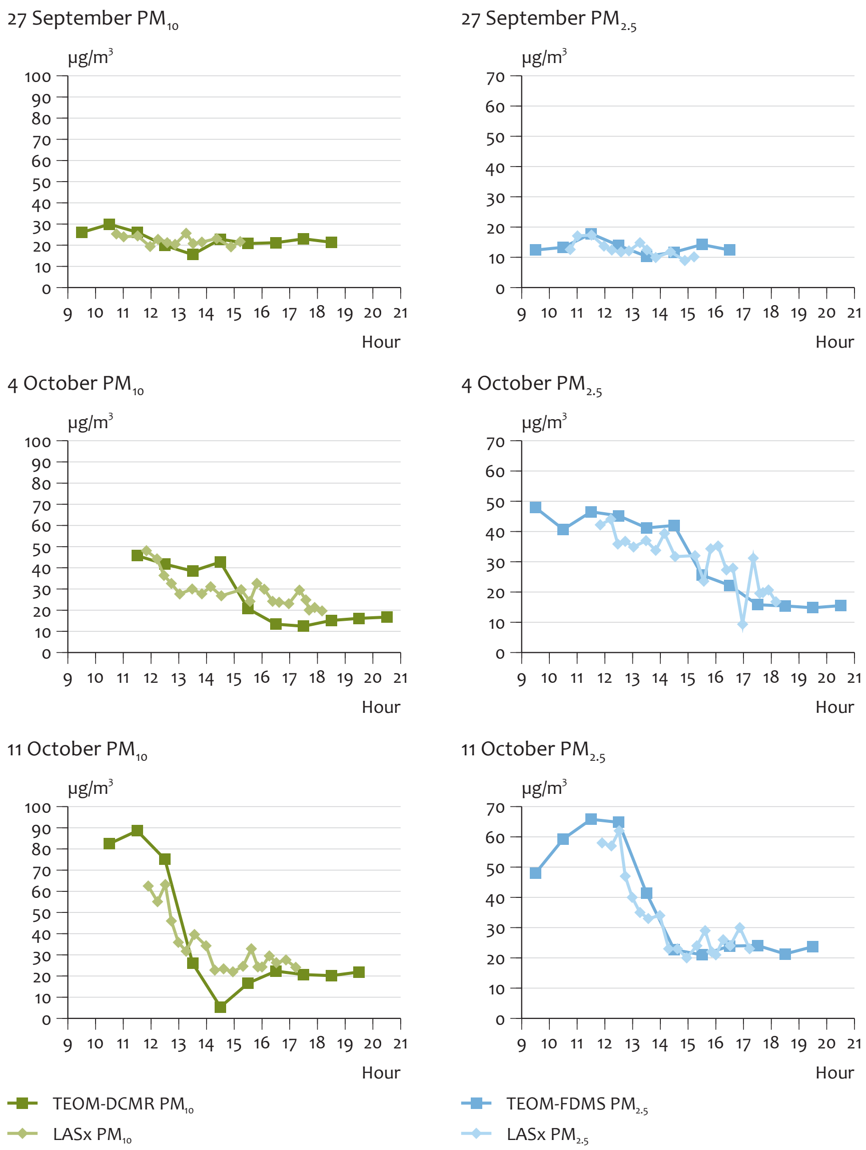
The above findings appear applicable to most recorded measurements. In general, the hourly variation measured at Schiedam was recognisable in the LAS-x measurements, supporting the idea of a large-scale 'blanket' of PM lying over the entire city, with variations caused by both regional transport of PM and weather conditions. However, large deviations occurred within some of the series, which could have been due to local influences on the LAS-x measurements.

Table 4 gives the average difference in concentrations between LAS-x and TEOM-SES (in case of PM_{10}) and TEOM-FDMS ($PM_{2.5}$) measurements, for each day. These are presented, separately, for the mobile and stationary measuring periods, as well as for both modes together. A positive sign means that the LAS-x measured a higher (average) concentration than the TEOM at Schiedam.

Averaged over all measurements days, the difference for PM_{10} was -1.5 µg/m³, and for $PM_{2.5}$ -1.9 µg m⁻³. On most of the days (8 out of 10) the (average) absolute difference between the LAS-x and TEOM time series at Schiedam was less than 5 µg/m³. On the remaining two days (15 and 16 October) rather large differences were found which appear systematic throughout both days (see Annex 9). A possible explanation would be that on these days no representative calibration factor for the TEOM was determined. Instead, the average value was used. It may well be that this average is not representative of these two particular days. However, studying the weather conditions for these days, no apparent explanation was found.

In terms of percentages, the daily differences measured for PM_{10} were between -36% and +26%. Without 15 and 16 October, the range would become -24% to +17%. Taking into account that 1) the direct influence of vehicular emissions could still have been present in the dataset and 2) measurements

Measured PM concentration **Figure 9**



Synchronised time series of LAS-x (representing the measurements recorded at different fixed locations, as well as while driving between them) and TEOM (at the fixed urban background location at Schiedam) on 27 September, 4 and 11 October for PM_{10} and $PM_{2.5}$. During some measurements intervals, $PM_{2.5}$ concentrations are higher than PM_{10} concentrations. This is due to the different TEOM monitoring system for $PM_{2.5}$ (TEOM FDMS) and PM10 (TEOM SES), to wich the LAS-x data were calibrated.

were executed during the daytime (the smaller night-time variation has not been included), one might say that, when measurements were recorded over the entire day (including the night), the above values would be upper limits. Other marked results were the relatively small differences between the averages for the mobile measurements and those for the fixed measurements, suggesting that both are equally representative.

The technical uncertainty of the LAS-x (according to the manufacturer) is about 5%. Additional uncertainty influencing the accuracy of these measurements was introduced by the determination of the calibration factor for comparison with TEOM, by taking a short period of the day and applying it to other parts of the day. The calibration factor was influenced by meteorological conditions and was variable during the daytime. Other uncertainties were caused by the use of an average calibration factor on days for which no factor was determined, and by the accuracy of the TEOM (partly due to the varying chemical composition in ambient air). Here, these uncertainty factors will not be discussed any further. The (relative) standard deviation of the average ratios per day between TEOM and LAS-x (see Table 10 in Annex 3) gives the possible variation in the calibration factors. These deviations were found to lie in the range of 5 to 27%, indicating that an estimate of the accuracy in the average measurements (which have similar time scales) would be at least 10% (but might be as high as 35%). It seems, therefore, reasonable to

PM concentration differences between mobile and fixed measurements **Table 4**

Date	LASX-TEOM (µg m^{-3})	percentage	Routes only (mobile)	Locations only (fixed)
27-Sep	0.7	3%	0.7	0.7
01-Oct	-4.9	-24%	-4.8	-5.1
03-Oct	-3.1	-8%	-1.9	-4.6
04-Oct	2.1	8%	3.3	0.5
05-Oct	-2.4	-11%	-1.2	-3.2
10-Oct	0.5	2%	-1.1	1.6
11-Oct	4.3	17%	4.5	4.1
12-Oct	-0.6	-6%	-0.3	-0.9
15-Oct	-19.0	-36%	-17.1	-21.3
16-Oct	7.7	26%	4.9	12.3
Average	-1.5	-3%	-1.3	-1.6
stdev	7.2	18%	6.3	8.6

Date	LASX-FDMS (µg m^{-3})	percentage	Routes only (mobile)	Locations only (fixed)
27-Sep	-0.8	-6%	-0.7	-0.9
01-Oct	1.4	12%	1.9	0.8
03-Oct	-2.5	-6%	-1.9	-3.5
04-Oct	-0.3	-1%	0.0	-0.6
05-Oct	2.4	17%	1.3	3.0
10-Oct	-1.0	-2%	-2.3	0.1
11-Oct	-4.4	-14%	-3.3	-5.7
12-Oct	-1.5	-14%	-1.7	-1.3
15-Oct	-10.1	-21%	-10.4	-9.7
16-Oct	-2.7	-10%	-1.3	-4.1
Average	-1.9	-4%	-1.8	-2.2
Stdev	3.5	12%	3.4	3.6

Average concentration difference (absolute and relative) between LAS-x and TEOM, for each measurement day. PM$_{10}$ upper, PM$_{2.5}$ lower.

conclude that the measured average (relative) differences between LAS-x and TEOM in Table 4 are of the same order as the estimated experimental accuracy and, therefore, are not significant.

Results from the mobile measurements were based on comparison between the PM concentration at two locations (the fixed urban background location at Schiedam and the location of the mobile unit). They are expressed as (relative) concentration differences, averaged over monitoring periods of 20 to 30 minutes.

Results from the Osiris monitoring campaign were based on comparison between the PM concentrations at six urban background locations. They are expressed as the relative standard deviation in the concentration at those locations, averaged over the monitoring period.

When comparing both methods, these differences must be taken into account. The results were similar regarding the finding that no significant spatial variability could be measured.

3.4 Local representativeness of an urban measurement station

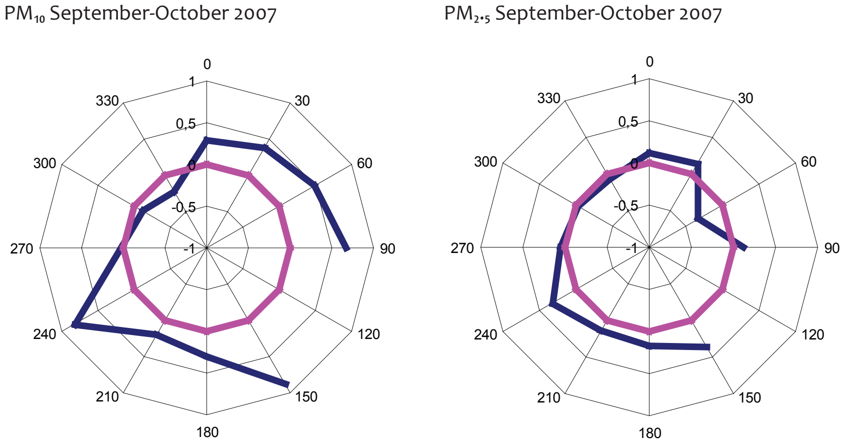
When dealing with spatially-dependent data in a geographical context (such as the Osiris measurements at fixed locations), identifying an appropriate scale for analysis (measurement or modelling) is a critical issue. It stands to reason, that measurements taken at fixed locations correlate better when collected at short distances from each other. For example, the relationship between (instantaneous) measurements over a distance of 10 kilometres in an urban environment would be (very) weak when sites or the area between those sites are influenced by dominant highly variable local sources. The relationship is enhanced by calculating averages over large time periods (hour, day, year). Taking the averages diminishes the short-time effects of local and highly variable contributions and, in fact, favours the role of gradually changing and large-scale processes (weather conditions, regional background contribution). The process of calculating averages, in fact, increases the urban representativeness of the measurements.

This still leaves the question of local representativeness of a single measurement site and its dependence on the averaging time interval. It would be ideal if the local (geograpical) representativeness of one single measurement could be determined for its immediate surroundings. Clearly, the representativeness of a distance for a particular site becomes increasingly dependent on the presence of sources nearby and their emission characteristics. The shorter the averaging time interval, the smaller the representative distances. The mobile monitoring method allows for (sequential) collection of spatially distributed measurements, performed at relatively short distances from each other, due to a high resolution. A

Mean daily regional-to-urban gradient **Table 5**

	PM_{10}		$PM_{2.5}$	
Monitoring campaign	**Osiris µg/m³/km**	**% /km**	**Osiris µg/m³/km**	**% /km**
1 (September-October 2007)	0.38 ± 0.56	1.6 ± 2.4	0.12 ± 0.24	1.4 ± 2.0
2 (March 2008)	0.08 ± 0.19	0.6 ± 1.4	0.04 ± 0.06	0.5 ± 0.8

Wind rose diagrams of the daily PM regional-to-urban background gradient **Figure 10**


PM_{10} September-October 2007

$PM_{2.5}$ September-October 2007


PM_{10} March 2008

$PM_{2.5}$ March 2008

Wind rose diagrams of the daily PM regional-to-urban background gradient in blue (Osiris µg/m³/km). The zero gradient line is presented in purple.

geostatistical technique (semivariogram) is applied to determine a spatial-analysis scale, the so-called semivariogram range, based on the degree of spatial autocorrelation within a dataset. Such a range may be considered as the maximum distance for which measurements at a fixed location could still be used (for the considered time scale) or would still be 'representative'. This also motivates the choice of scale for the modelling and the measurements. Examples of this technique, as applied on a ('mobile') air quality dataset, are described by Lightowlers et al. (2008) and Larson et al. (2007). The current study which deals with the semivariogram technique and corresponding use of the mobile measurements can be considered as exploratory research. Therefore, results are presented separately (see Annex 11 Semivariogram: details and results), along with a further explanation of the statistics involved.

3.5 PM gradients

3.5.1 Average regional-to-urban background gradient

The gradient from the regional to the urban background is studied from locations 6, 1 and 9. The gradient in the PM concentrations between the locations was calculated for each day within the monitoring periods, for which concentrations at the three locations were available, based on a linear regression taking into account the distance between them. The distance between locations 6 and 9 was ca 7.5 kilometres. It is noted that this distance is critical when expressing differences in concentrations as a gradient. If the locations were selected along another, more perpendicular line, the distance would be different. Therefore, and to compare the gradient with values from the GCN maps, the difference in concentrations between region and urban background is also presented.

The mean gradients for the monitoring period and their standard deviation are presented in Table 5. Since the Osiris mass concentration was not equivalent or calibrated to the reference level, the slope is also expressed as the relative change per kilometre.

Apparently, the gradient was very variable during the monitoring period and did not deviate, significantly, from zero. It is expected that the wind direction explains (part of) this variability. Due to the situation at the regional background locations, close to city of Delft in the north and close to the greenhouse area in the north-west, largest gradients could be expected with winds from the ENE (main wind direction during the first campaign) to winds from the WSW (main wind direction during the second campaign). In these cases, any influence from the cities of Delft and Rotterdam on the regional monitoring location would be absent. To investigate this, wind rose diagrams of the gradient were constructed, based on daily average wind directions.

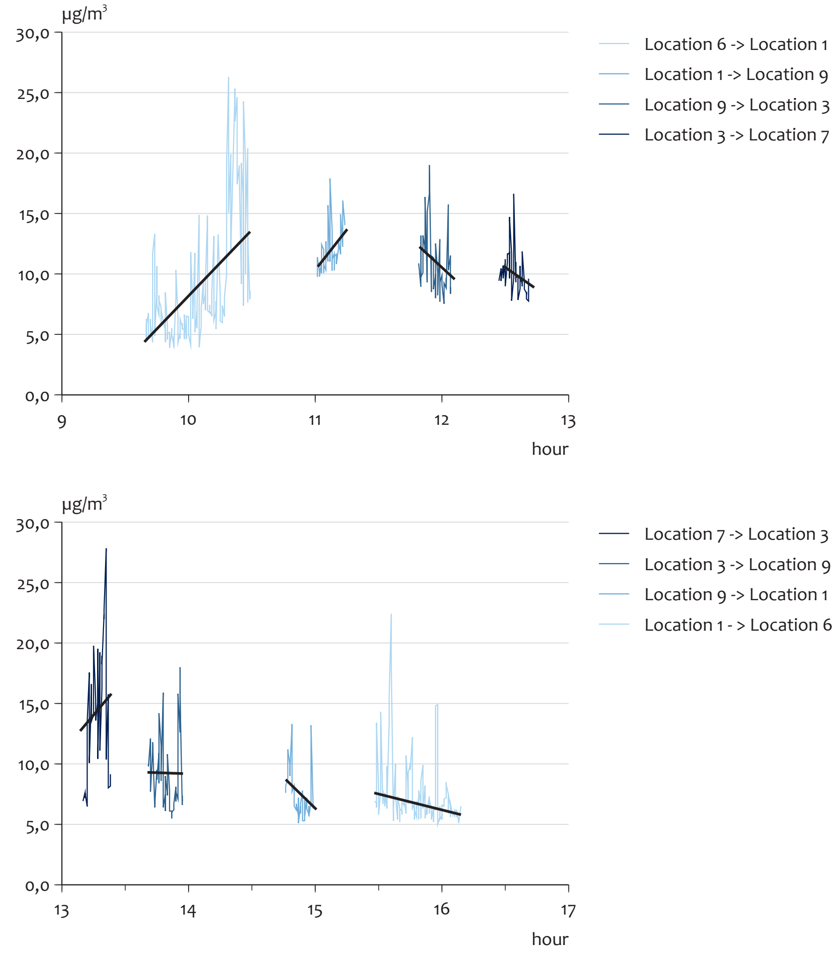
In Figure 10 can be seen that, for $PM_{2.5,}$ the gradient was zero during most wind directions. $PM_{2.5}$ gradients of 0.2 to 0.3 µg/m3/km occurred during the first monitoring period under winds from the S-WSW. When the wind direction was north-east, gradients were found to be both positive and negative. Under winds from the WSW, during the second campaign, no $PM_{2.5}$ gradient could be detected.

For PM_{10}, the gradient seemed more pronounced. Under ENE winds during the first campaign, the gradient ranged from 0.4 to 0.7 µg/m^3/km. Largest gradients in this period were found for WSW (0.8) and SSE (0.9) wind directions. The latter is against expectations. Apparently, the coarse fraction of PM (2.5-10 µm), resuspended in the city, was not transported far into the region. The second campaign showed smaller PM_{10} gradients.

The difference between region and urban background concentration, expressed as a percentage derived from the average gradients found for both campaigns by using linear regression, was 4 to 12%, for both PM_{10} and $PM_{2.5}$. The GCN 2007 value for the difference in the PM_{10} concentration between locations 6 (regional background) and 9 (urban background) was 2 µg/m3, which corresponds with 7%. The order of magnitude is comparable to the range for $PM_{10,}$ based on both monitoring campaigns.

Examples of $PM_{2.5}$ gradients measured while driving **Figure 11**

27th of September



Examples of $PM_{2.5}$ gradients measured while driving, on 27 September 2007

Rates of change in concentrations for PM_{10} and $PM_{2.5}$. **Table 6**

	PM_{10} trend (µg/m³/km)		$PM_{2.5}$ trend (µg/m³/km)	
Date	**Average (absolute)**	**stdev**	**Average (absolute)**	**stdev**
27-Sep	1.6	1.2	1.2	1.0
3-Oct	2.5	1.6	1.8	1.5
4-Oct	5.0	6.4	2.5	4.5
5-Oct	1.8	1.1	0.6	0.5
10 Oct	1.7	3.3	1.7	3.2
11 Oct	4.3	6.5	1.5	1.6
12-Oct	1.0	0.8	0.7	0.7
15 Oct	3.6	2.0	1.3	0.6
16 Oct	2.2	2.4	0.9	0.5
Average	2.6	2.8	1.3	1.6

e small differences in concentration and gradients confirm that PM concentrations at regional and urban background locations were mainly determined by large-scale transport. Contributions from local sources were limited.

3.5.2 Instantaneous gradients measured while driving

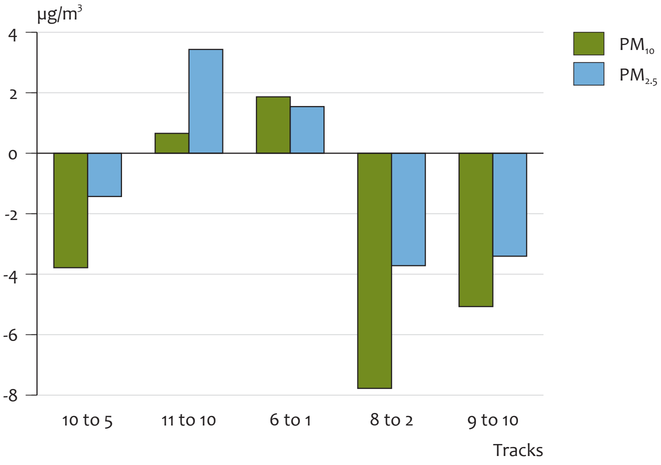
In the previous sub-paragraph, the gradient between regional and urban background has been determined via linear regression through three data points. The number of data points could be increased by collecting mobile measurements while driving between locations. In this way, information collected between locations could be taken into account and mobile measurements could be regarded as complementary to the fixed measurements in the Osiris monitoring network. In this sub-paragraph, an estimation is given of typical changes in concentration levels of PM_{10} and $PM_{2.5}$, measured while travelling through an urban (background) area. Also, average gradients between fixed (Osiris) locations are compared to gradients in the GCN map.

The estimations were carried out by considering the linear trend in concentrations over each 'mobile' measuring period and the corresponding distance between the fixed (Osiris) stations. Figure 11 gives examples of such trends, as measured on 27 September 2007. Clearly recognisable are the trends, both upward and downward, when going from region to city centre (and back). Also within the urban region, distinct gradients were observed, going from one place to another. Obviously, in such mobile measurements, the background concentrations change with time. However, due to the short time scale of the mobile measurements (15 to 30 minutes for one route), these changes were assumed to be negligible.

In Table 6, the averages in (absolute values of) trends, measured per day (and hence for different routes), are given for PM_{10} and $PM_{2.5}$. The overall average over the 10 measurement days was 2.6 (± 2.8) µg/m³/km for PM_{10} and 1.3 (±1.6) µg/m³/km for $PM_{2.5}$. The standard deviations indicate the large (and expected) variability within these gradients. The magnitude of such gradients is expected to be influenced by local conditions, time of day and weather conditions.

Considering the gradient of regional-to-urban background, such as in the previous sub-paragraph (averaging over the routes 6 to 1 and 1 to 9, and back), but this time using the data information between the locations, results were found of 0.3 ± 1.0 µg/m3/km for PM_{10}, and 0.4 ± 1.1 µg/m3/km for $PM_{2.5}$. The gradient for the reverse route (urban to regional) was nega-

Average gradients of different routes **Figure 12**



Average gradients of different routes, expressed as absolute concentration differences. The figure shows only the routes which were driven 5 or 6 times.

Measured temporal variability of PM concentrations **Table 7**

		PM_{10}			$PM_{2.5}$		
Monitoring campaign	**Number of days (n)**	**regional bg**	**urban bg**	**traffic**	**regional bg**	**urban bg**	**traffic**
		TCV (%)			***TCV (%)***		
1 (Sept/Oct 2007)	16	38.7	37.1	36.9	70.1	59.5	60.3
2 (March 2008)	19	48.9	43.5	38.6	49.9	48.8	45.0
		Standard deviation (µg/m³)			**Standard deviation (µg/m³)**		
1 (Sept/Oct 2007)	16	7.4	8.2	9.3	8.4	7.5	9.1
2 (March 2008)	19	7.3	6.6	6.7	4.1	4.0	3.9

Temporal coefficients of variation (TCV) in the PM concentrations at regional background, urban background and traffic locations. Also, the standard deviation in the daily average concentration is presented (in Osiris µg/m³)

tive: -1.2 ± 2.3 µg/m3/km for PM_{10} and -0.4 ± 0.4 µg/m3/km for $PM_{2.5}$. The gradients found did not deviate, significantly, from zero. This confirms the results from the Osiris monitoring campaign described in the previous sub-paragraph.

With the Osiris data, an average of the regional-to-urban background was calculated for the entire monitoring period in September/October 2007: 0.38 ± 0.56 µg PM_{10}/m³/km and 0.12 ± 0.24 µg $PM_{2.5}$/m³/km. Given that the Osiris readings have underestimated the concentration to some extent (see Annex 5), the Osiris and LAS-x results can be considered to be in the same range.

In Figure 12 the average gradients per route are given, expressed as an absolute difference in concentration, over the distance covered. All routes were driven 5 times or more. This figure shows a gradient of -8. to 2 µg/m3 for PM_{10} and -1.5 to 1.5 for $PM_{2.5}$. The standard deviations are of the same order as the gradients. For most of these routes, the GCN map visually indicates gradients of the same sign (positive or negative). For example, the GCN map for 2007 (Figure 5) shows an average yearly increase of 1 µg/m³, going from location 6 to 1. Here, while driving, a change of +2 µg/m³ was measured. A contradiction emerged only for the route from location 8 to 2. The GCN map indicates a positive gradient, whereas the measurements indicated a decrease in PM_{10} concentration. The reason for this might be that location 8 (Vlaardingen) was influenced by industry/shipping, as well as by nearby traffic.

The GCN map and the data from the mobile measurements in this study can only be compared to a certain extent. GCN concentrations are yearly averages, whereas the mobile measurements are averages calculated from half-hour data, over 5 or 6 days (within one month).

3.6 Temporal variability

The temporal variability can be assessed under the same terms as the spatial variability, by using the Temporal Coefficient of Variation (TCV). The SCV analysis was based on concentration averages at different urban background locations,over the monitoring period. To analyse the temporal variation during this period, the variation in daily mean concentrations has been assessed. To make a fair comparison with the spatial variation analysis, the same days were taken into account.

A distinction was made between the regional background, urban background and traffic locations. The regional background was represented by location 6. For the urban background, the average was used from the measured concentrations at locations 2, 3, 4, 5, 7 and 9 (during the second campaign, data from location 2 were not available). For the traffic locations during the first campaign, the average was used from the concentrations at locations 8 (Vlaardingen) and 10 (Bentinckplein). During the second campaign, traffic was only represented by location Bentinckplein, due to the unlikely low PM_{10} concentration measured at location Vlaardingen.

Results of the analysis of the TCV values for the different types of monitoring locations are presented in Table 7.

The regional background showed the highest TCV values, followed by urban background. Traffic locations had the smallest TCVs. This could be explained by the average concentration level, used to normalise the standard deviation. The standard deviations themselves showed a different order, but did not differ much between the categories.

The high TCVs for $PM_{2.5}$ during the first monitoring campaign, were caused by relatively high standard deviations, similar to those for PM_{10}. This indicates that large-scale transport, causing the variation from day tot day, consisted almost totally of particles smaller than 2.5 µm. During the second monitoring campaign, this was not the case. This could be explained by the prevailing winds. Easterly winds in the first campaign were responsible for the transport over land. Smaller particles can be transported over larger distances, causing large-scale transport to consist of the finer PM fraction. Transport during the second campaign was over sea. Sea salt exists of particles in the coarser fraction (2.5-10 µm).

The TCV values for urban background were high, compared to the spatial equivalent SCV values (Table 2). This confirms the outcome of the single-factor ANOVA analysis, presented in Paragraph 3.2. As could be expected, for a pollutant for which the concentrations are strongly dependent on large-scale emissions, the temporal variability from day to day was much higher than the spatial variability.

The temporal variation on an hourly basis was assessed by calculating the rate of change in concentration. This was done by calculating the relative increase or decrease in the concentra-

Standard deviations in the hourly rate of change of PM concentrations

Table 8

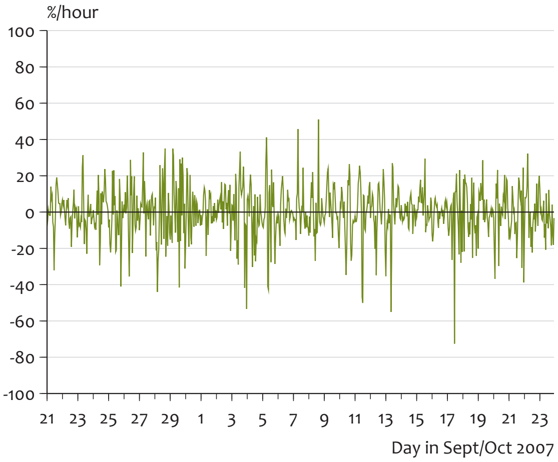
Monitoring campaign	PM_{10}			$PM_{2.5}$		
	regional bg	urban bg	traffic	regional bg	urban bg	traffic
1 (September-October 2007)	3.3	3.0	3.7	2.5	2.0	2.4
2 (March 2008)	4.0	4.0	4.0	2.4	2.3	2.3

Temporal coefficients of variation (TCV) in the PM concentrations at regional background, urban background and traffic locations. Also, the standard deviation in the daily average concentration is presented (in Osiris µg/m³)

Hourly rate of change in PM_{10} urban background

Figure 13

First monitoring campaign



Hourly rate of change in PM_{10} (%/hour) for the urban background during the first monitoring campaign.

tion between two consecutive hours. For example, the time series of the hourly rate of change in urban background PM_{10} during the first campaign, is presented in Figure 13.

The hourly rate of change for most of the hours was between -20 and +20 %, but higher values did occur, once in a while. Since the average rate of change over the entire period was very close to zero, the standard deviation in the rate of change was a good measure for comparison of the different types of monitoring locations (regional, urban and traffic). The standard deviation in the rate of change could be regarded as the rate of change that was exceeded in 32% of the time. For a fair comparison between types of locations, the rate of change is expressed in absolute concentration values. It is noted that the concentration is expressed as 'Osiris µg/m³', which was not equivalent or calibrated to the reference level. Results for PM_{10} and $PM_{2.5}$ in both campaigns are presented in Table 8.

Differences in hourly standard deviation between regional background, urban background and traffic locations were small. This implies that the additional local sources in the city and the streets were far less variable during the day and between days than in the regional background level. Results from the hourly assessment were in line with those from the analysis of TCV, based on daily average concentrations. At both temporal scales, temporal variability is mainly governed by large-scale transport and weather conditions, due to the long lifetime of PM.

4

Conclusions

The first monitoring campaign, in September/October 2007, was characterised by stable weather conditions, low wind speeds and easterly winds, while the weather during the second campaign, in March 2008, was unstable, windy and wet, and the prevailing winds were west. Due to these different meteorological circumstances, PM concentrations were lower in March 2008 than during the autumn of 2007.

The following conclusions can be drawn with respect to the research objectives:

1. *Local sources;* detection of local influences through Osiris measurements turned out to be difficult, due to the relatively small increments from local sources in the background concentration levels and the measurement uncertainty. As a result, influences from shipping and industry/harbour could not be detected by the measurements. Six of the selected monitoring locations in this study have been regarded as urban background locations that were not influenced by local sources.
2. *Urban spatial variability;* the urban spatial variability is expressed as the relative standard deviation between mean PM concentrations in both monitoring periods (averaged over 16 and 19 days, respectively, of simultaneous monitoring) at the six urban background locations. This was around 5 and 10% for PM_{10} and 5% for $PM_{2.5}$. The estimated measurement uncertainty of the mean PM concentration in both periods was similar to the variability (10% for PM_{10} and 5% for $PM_{2.5}$).
3. *Urban spatial representativeness;* absolute differences between the fixed urban background station in Schiedam and mobile measurements in the urban background were less than 5 $\mu g/m^3$ for PM_{10} and 2.5 $\mu g/m^3$ for $PM_{2.5}$ (average over daytime periods of 6 to 8 hours). The differences were similar to the estimated measurement uncertainty.
4. *Local spatial representativeness;* the study in which mobile measurements are used to assess the local spatial representativeness of measurements at fixed locations is regarded as exploratory research. First results indicated for urban monitoring locations a representative length scale of 3 km, based on measurements collected at half-hourly intervals. More research is required to interpret results, so that the local representativeness of point measurements in an urban environment can be assessed.
5. *Regional-to-urban spatial gradient;* concentration differences between the regional and urban background locations were small: on average 4 to12% for daily mean PM_{10} and $PM_{2.5}$ concentrations. Although measurements were carried out during only two one-month periods and might therefore not be representative for an average year, this result reflects the major influence of large-scale transport and weather conditions. The limited concentration gradient was confirmed by the mobile measuring campaign.
6. *Urban temporal variability;* the temporal variability in PM concentrations at the monitoring locations is high, both during the day and between days. The relative standard deviations between the daily average concentrations during the monitoring periods were from 35 to 50% for PM_{10} and 45 to 70% for $PM_{2.5}$. The temporal variation was very similar for regional background, urban background and traffic sites, indicating that the PM concentration levels in a city are dominated by the regional background contribution.

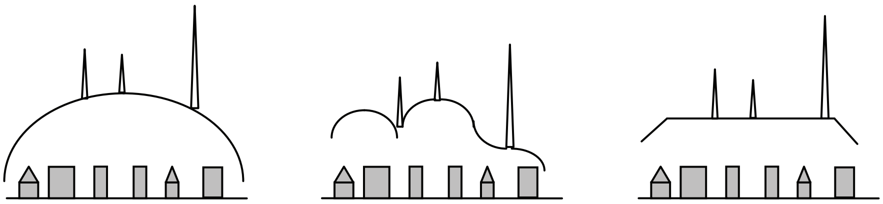
Two policy questions are addressed:

Question A. What is the impact of the urban background variability on the uncertainty in modelling local PM concentrations and exceedances of the EU limit values?

The urban background variability is small compared tot the measurement uncertainty. Therefore, the impact on the uncertainty in modelling is presumably small.

The question is related to the GCN background concentration maps used in modelling the local air quality. In 2007, the model resolution of the GCN maps was increased from 5 x 5 km^2 to 1 x 1 km^2. This resulted in a PM_{10} pattern with higher concentrations near local sources, such as motorways. However, the variability in PM_{10} concentration remained small because of the limited range of the scale on the GCN map. For Rotterdam and its surroundings, the range of scale in the GCN PM_{10} map is only 6 μg/m3 for both 2006 and 2007. Within the urban background area differences are less than 2 μg/m3. The uncertainty (1σ) in the GCN concentration is estimated at 15% (Velders et al., 2008). Taking this uncertainty into account, the differences within the urban background area are not significant. This implies that the higher resolution modelling for the GCN 2007 map does not yield significantly different concentrations and it also does not lessen the uncertainty within the urban background. It must be noted that from a regulatory point of view, the (non-significant) variability is treated as relevant in complying with the yearly averaged PM_{10} limit at locations in inner urban areas.

Schematic overview of the urban PM background **Figure 14**



Representation of the urban PM background by three concepts: concentric, spatial variation and the plateau. The peaks on top of the urban background represent the concentration increase by local sources.

Measurements carried out during this study confirm the non-significance of the urban background spatial variability of PM_{10}, as well as of $PM_{2.5}$ within the urban background. Whether the change in GCN resolution from 5 x 5 km^2 to 1 x 1 km^2 is an improvement for assessing local PM_{10} concentrations, therefore, cannot be judged by the measurements carried out in this study.

In this study, the representation of the urban background in the GCN maps of 2006 (concentric pattern) and 2007 (spatial variation within the city) could not be confirmed by the measurements. Based on the small spatial variability and small gradients between the regional and urban backgrounds, the measurements suggest a third concept: a small gradient from the regional to urban background leading to an urban background concentration plateau. Figure 14 presents the three concepts.

Due to measurement uncertainty, all three concepts may represent the truth. Dealing with the uncertainty, the plateau concept is the most simplified concept.

With the concept of the urban background plateau in mind, focus with respect to the uncertainty in the GCN maps should not be on the variability within urban areas, but rather on how to reduce the uncertainty in the absolute concentration levels. The present calibration of the GCN model results to measurements from the national monitoring network occurs by adding the same PM_{10} concentration at each location within the Netherlands (14.4 µg/m^3 in 2007, Velders et al., 2008). It is recommended to assess to what extent city/region-specific calibration may improve the GCN background concentration estimates. Regarding the city/region-specific versus the national calibration procedure, the following is noted. This study focused on the variability of the PM concentration within the city of Rotterdam. Assuming that Rotterdam is the city with the highest variability in the urban background PM concentration levels within the Netherlands due to the relatively large amount of local sources, it is argued that spatial variation in other Dutch cities is also small. This study did not focus on the absolute concentration differences between cities within the Netherlands. With respect to the above mentioned recommendation for local calibration, this issue would be of vital importance.

Another issue following from the small spatial variation in PM background levels, is the focus on PM mass concentration in regulation. The relative increment of local contributions (traffic, shipping, industry/harbour) is limited for both PM_{10} and $PM_{2.5}$, as a result of the large-scale nature of PM_{10} and $PM_{2.5}$. In view of the health effects found near heavy traffic locations, it is recommended to identify a transport-related PM indicator different from PM_{10} and $PM_{2.5}$. More research may be directed towards EC/OC and ultrafine particles: these parameters show larger relative contributions from local sources (e.g. exhaust emissions from road traffic) and are believed to be of relevance in affecting people's health. In the scope of the BOP programme, studies are carried out regarding EC/OC and chemical characterisation. Ultrafine particles represent an area of increasing interest, however this topic is not covered in the BOP programme.

Question B: What are the recommendations on an urban background monitoring network?

From a surveillance and health point of view, the background concentration in a city needs to be known with the highest possible accuracy. From the rather low spatial variability found in this study, one may argue that one urban background station would sufficiently represent the yearly average urban background in Rotterdam. However, since PM measurement uncertainty is generally high, multiple measurements at urban background locations within one city would decrease the uncertainty in the estimation of the mean urban background concentration. If we assume an uncertainty (1σ) of 10% when using one location, this will reduce to 7% for 2 locations, 6% for 3 locations and 5% for 4 locations.

To be able to quantify the spatial variation within the urban background, the concentration at the locations needs to be measured with higher accuracy than was done in this study. Double (or even triple) the number of measurement recordings at the locations would be needed to reduce the uncertainty below the level of the spatial variability.

Results from this study do not yield straightforward recommendations on the choice of location for an urban background monitoring station. Influence from traffic must be avoided as much as possible. According to the criterion presented by Larssen et al. (1999), there should be no more than 2500 motor vehicles per day within a radius of 50 metres around a monitoring location.

With the concept of the urban background plateau in mind, it is recommended to locate the urban monitoring stations for PM not too close (~1 km) to the edge of a city. More research is needed to assess whether the representation by the plateau is indeed realistic and to define the distance from the edge of a city by which the plateau would be reached.

Due to the small spatial variability for PM within a city, the measurement of PM concentrations will not be very sensitive to the choice of the exact location. However, for other air pollution components, this choice will likely be more critical.

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Annex 1 Osiris environmental dust monitor

Method

The Osiris environmental dust monitor is developed and provided by Turnkey Instruments Ltd. The instrument is fitted with a heated TSP inlet and measures mass concentrations of TSP, PM_{10}, $PM_{2.5}$ and PM_1. It uses a light scattering technique to determine the concentration of airborne particles and dust in the size range from about 0.4 to about 20 µm in diameter. The air sample is continuously drawn into the instrument by a pump with a flow rate set by the microprocessor at 600 cm3/min. The incoming dusty air passes through a laser beam in a photometer and then through a filter to remove the particles before reaching the pump.

The photometer only measures light scattered through very narrow angles. This narrow angle scatter is virtually the same for black or white particles of the same size. That is, it doesn't depend on the material composition of the particle. The intensity of the light pulse is therefore an indicator of particle size. The particle size is converted into particle mass by assuming the particle is perfectly spherical and the material density is 1.5 g/cm^3.

Having evaluated the mass of the particle, the microprocessor then evaluates the likely chance of collection of the particle according to the sampling convention being used. For PM_{10} this is Table A.1 from European Standard EN 12341. For $PM_{2.5}$ (and PM_1) it is assumed that the collection efficiency is 100% for diameters less than 2.5 µm (or 1 µm) and 0% for larger diameters. The particle mass, corrected for the collection efficiency, is accumulated over the course of the sampling integration period.

Strategy

The Osiris dust monitor is not equivalent to the European reference method (gravimetry). In order to improve the trueness of the Osiris measurements, calibration towards the reference method is needed. However, during the BOP study, this was not carried out because 1) we are more interested in (relative) differences than in absolute concentration levels and 2) available gravimetric measurements from only one location would not yield additional information when evaluating differences (variability) between locations.

The absolute values of concentrations measured by different Osiris monitors may differ to some extent. In order to improve the reproducibility of Osiris measurements, normalisation between different instruments is carried out by a monitoring campaign of around 4-6 days, in which the instruments are put together at the same location. Based on these days, an average normalisation factor to one of the instruments, the so-called reference Osiris, is determined for all the Osiris instruments:

$$(normalisation\_factor)_{Osiris"x"} = \frac{(mean\_concentration)_{Osiris"2507"}}{(mean\_concentration)_{Osiris"x"}}$$

The reproducibility after this normalisation, expressed as the relative standard deviation of the mean concentrations of the different instruments (n = 8 to 11), is circa 5% for hourly averaged PM_{10} measurements and 3% for hourly averaged $PM_{2.5}$ measurements. The normalisation is carried out both before and after the actual monitoring campaign, because it is known from previous studies that the normalisation factors may change during the campaign. A linear interpolation is then applied to obtain the normalisation factor during the monitoring campaign.

The reproducibility determined during the normalisation procedure will in general not be achieved in the field during the measurement campaign. Duplicate measurements during a study carried out in 2008 (not published), show that the 'field' reproducibility expressed as the between instrument uncertainty (1σ) in that study is estimated to be 10% for PM_{10} and 5% for $PM_{2.5}$ for daily averaged concentrations. Also, it turned out that the duplicate differences do not show a random distribution. In general, the duplicate monitor always measured a higher (or lower) concentration than the reference monitor. For this reason, it is assumed that the uncertainty does not decrease when the concentration is averaged over a period of multiple days.

In the present monitoring campaigns, the reproducibility (1σ) in the period mean concentration at one location is estimated at 10% for PM_{10} and 5% for $PM_{2.5}$

Outliers

After the normalisation, the data set of hourly averaged values was screened for outliers. First, individual concentration values that differed from the concentration average for all locations by more than two times the standard deviation were identified. Afterwards, the ratio between the standard deviation for the instruments without the assumed outlier and the standard deviation for all instruments was calculated and evaluated against the critical value corresponding to the 95% confidence level. A final check up was done to prevent the removal of hourly concentrations that are in fact no outliers (such as low or high concentrations during a period of several successive hours). Removed outliers have been filled by linear interpolation.

Annex 2 Mobile measuring instruments

The Laser Aerosol Spectrometer (LAS-x) from Particle Measuring Systems, Inc (PMS) is an optical-scattering laser-based high sensitive spectrometer for sampling and counting extremely small airborne particles. The LAS-x counts the number of particles while classifying them in up to 100 different diameter classes (in this case 32 channels were used) from 0.09 to 7.5 µm and up to 10 µm with an oversized channel.

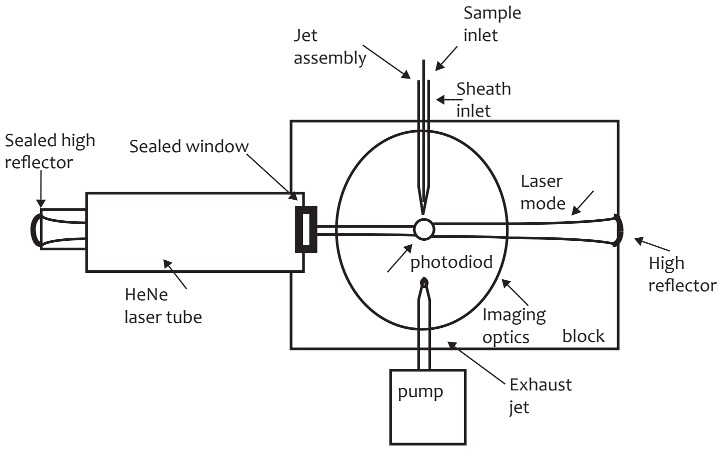
Assuming spherical particles and by using a standard density, the number particle distribution was first recalculated to volume distribution and then to mass distribution. The density that was used in this study, is 1.65 g cm^{-3}, this was based on earlier comparison experiments between calculated mass concentrations derived from LAS-x data and filter measurements along a busy provincial road near the ring of Alkmaar. This is also in accordance with Tuch et al. (2000). The instrument, allowing a time resolution of 1 second, instead was used with a 20-second time resolution, which reduces the noise in the calculated mass.

In Figure 14, a schematic view of the optical system of the LAS-x is given where the laser and detection optics are situated. The particles are confined to a space that is smaller than the laser beam diameter, which gives the instrument the high particle sizing resolution (Operator's manual, HSLASII and LAS-x II, PMS Inc.).

A CPC can count the total particle concentration, but these data cannot be used for calculating the mass concentrations, since the particles remain unsized. However with its high time resolution of one second, it can be used to follow outbreaks of particle numbers in time and location. The time resolution that was used in this campaign was 20 seconds for the LAS-x and one minute for the Osiris.

The different aerosol instruments used the same inlet system: this is a stainless steel pipe with a diameter of 100 mm attached at the front of the mobile unit, at a height of 3 metres. At the front end of the pipe, a hood was mounted to avoid intake of all kinds of light weight - but huge sized - material, actual aerosol with a physical diameter larger than 10 µm but with a density much smaller than 1. At the end of the tube a ventilator was connected to force the air in through the pipe. In the pipe small metal tubes are inserted to get a by-pass flow to the instruments. For the aerosol instruments the inserts were mounted with the opening facing the incoming airstreams. The different instruments tap their own flow from the inserts with copper tubing or carbon filled

Schematic view of the optical system of the LAS-x **Figure 15**



Schematic view of the optical system of the LAS-x.

silicone tubing (specially for aerosol, from TSI Inc. USA). Only the Osiris has a different type of inlet. This inlet was connected to the end of the pipe and fan, via a wide PVC flexible hose flushing the air around the inlet.

In the campaigns, the LAS-x and Osiris were compared to a gravimetric instrument: a TEOM-SES, of DCMR. The difference between these instruments is the temperature setting. The temperature inside the LAS-x is about 10 degrees above ambient temperature. The final temperature of the air stream entering the LAS-x is uncertain, since the time in which it travels through the metal tubing is rather short. Therefore, it is not certain that the LAS-x measures completely dry aerosol at high relative humidity of ambient air. A standard TEOM (Tapered Element Oscillating Microbalance) has a sensor temperature of 50°C. This instrument underestimates the semivolatile compounds and this is why a TEOM-SES (Sample Equilibration System) is built. This system has a lower temperature of 30°C and uses a Naflon dryer. A TEOM-FDMS (Filter Dynamics Measurement System) measures both the volatile and non-volatile fractions of particulate matter (and no correction factor needs to be applied).

Annex 3 Comparison of different instruments

The urban background station of interest here is the official DCMR site 'Alphons Ariensstraat' in Schiedam (location 9, here denoted by AA; see Figure 6 for the geographical location). This station is situated at a car park and surrounded by apartment blocks. Behind these apartments, busy streets and a crossing are located to the north-east of the station. Hence, vehicular influence is anticipated at certain wind directions. At AA various air quality measurements are performed on a continuous basis. Hourly data of particulate mass is collected by a TEOM-SES ($PM_{10,}$ corrected with 1.3) and a TEOM-FDMS (PM_{10} and $PM_{2.5}$). The mobile measurements involved the use of LAS-x and Osiris.

To compare the variability within the concentration sets measured with different instruments, a comparison was performed to estimate (absolute) differences in the measured concentrations. This data comparison between LAS-x (in the mobile unit) and Osiris (in mobile unit and at AA) with the DCMR TEOMs, was done for selected periods on five days, with coinciding measurements at AA. The average results for the various periods are displayed in Figure 16 and Figure 17, and in Table 9 [2].

In general, the time variation for both PM_{10} and $PM_{2.5}$ indicate similar behaviour for all instruments which is also reflected in the correlation coefficients in Table 9. In most cases, the TEOM instruments measured the highest concentrations, with the FDMS measuring larger values (of PM_{10}) than the SES. Sometimes, large differences were measured between SES and FDMS, for instance, on 12 October, at 14:00 hrs, the SES measured 8.5 μg/m3 (corrected with 1.3), while the FDMS indicated 23 μg/m3. The reason might be that the volatile compounds were measured more precisely by the FDMS and are expected to be underestimated by the SES (in spite of the correction factor). On an annual basis, however, no clear difference is apparent: the hourly averaged ratio between FDMS and (corrected) SES for 2007 was almost exactly 1.0. In absolute value, the SES average was negligibly higher: -0.9 $\mu g/m^3$.

As noticed, the LAS-x and Osiris mostly measured lower values than the TEOMs, with the exception of 4 October (17:00 hrs) and 12 October (14:00 and 17:00 hrs), with respect to the TEOM-SES. There are various reasons for over- or underestimations. Both LAS-x and Osiris are based on optical measuring principles. The TEOM weighs the presence of all particles arriving at a filter in the (oscillating) microbalance (with aerodynamic diameters of less than 10 or 2.5 μm). Results of optical devices appear sensitive to wavelength and (assumed) mass densities and, therefore, differ in measuring particles of within certain (aerodynamic) diameter ranges. For example, the upper diameter that the LAS-x can measure with 100% efficiency is around 4 µm, towards the larger diameters this declines, slowly (hence, missing part of the heaviest particles); the Osiris is known to underestimate mass contribution below 1 μm (counting particles larger than 0.5 µm). The measurements by TEOM are affected by loss of volatile material (leading to lower values) and the presence of water (leading to higher values), depending on the operational temperature.

The observed variability in Figure 16 and Figure 17 and the difference between TEOM and LAS-x can (to some extent) be understood by looking at the air transport and wind direction. The higher PM_{10} concentrations on 3 and 10 October can be ascribed to the relatively stable atmospheric conditions and transport of air from the (north-)eastern direction. At AA, this happens to be the sector for which traffic influence may be present. The increase at 17:00 hrs might have been due to the traffic rush hour. During 4 October, concentration levels appear to decrease. Indeed, in the course of this day, the wind direction veered from south-south-west to north-west, leading to the transport of clean maritime air. For the same reason, levels on 12 October were relatively low: the main wind direction on this day was north-west. This condition may well explain the reversal of the ratio between TEOM-SES and LAS-x (Osiris). After transport over sea, the air contains more sea salt. Due to the hydrophilic nature of sea salt, the water content of aerosols increase. If the drying procedure within the TEOM operates more efficiently than is the case

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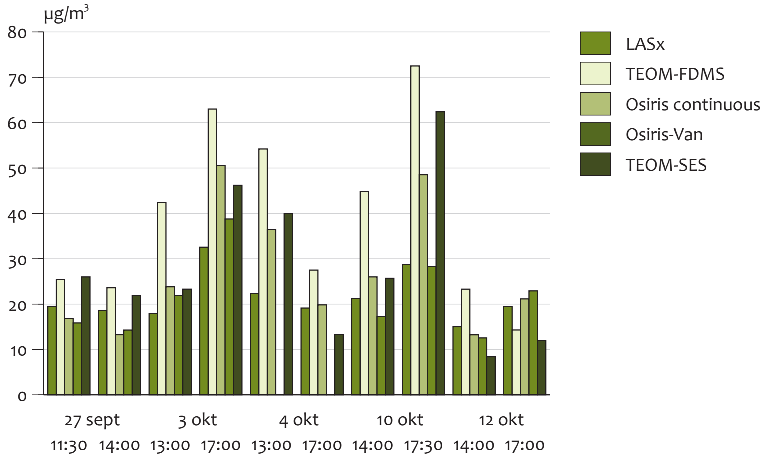
[2] For the sake of completeness, the results from 10 October, 17:30 hrs, have been added to these figures. However, in the further data processing and -interpretation the afternoon results have been omitted, due to a wrongly adjusted inlet flow.

PM_{10} concentration for different instruments Figure 16

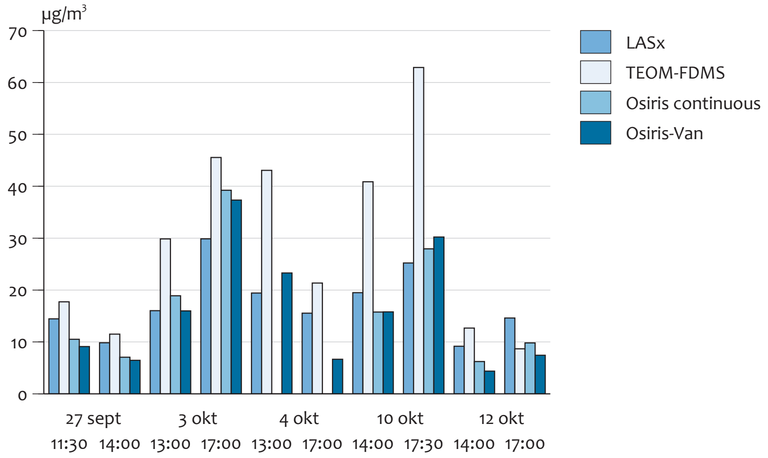
Location Alphons Ariensstraat, Schiedam, Location 9



Comparison of LAS-x with TEOM-SES, TEOM-FDMS and Osiris at Alphons Ariensstraat for PM_{10}

$PM_{2.5}$ concentration for different instruments Figure 17

Location Alphons Ariensstraat, Schiedam, Location 9



Comparison of LAS-x with TEOM-SES, TEOM-FDMS and Osiris at Alphons Ariensstraat for $PM_{2.5}$

for the LAS-x (Osiris) instrument, the latter will overestimate the aerosol mass by inadvertently measuring a higher water contribution[3].

The correlation (R2) between the various instruments has been summarised in Table 9 for PM_{10} and $PM_{2.5}$. These coefficients have been determined by using the concentrations measured when positioned at the AA station.

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[3] No silicagel dryer was used upward of the aerosol stream into the LAS-x.

Correlation coefficients between measurement methods

Table 9

PM_{10}	LAS-x	SES	FDMS	Osiris
LAS-x	1			
SES	0.75	1		
FDMS	0.65	0.82	1	
Osiris	0.86	0.65	0.64	1

PM_{10}	LAS-x	SES	FDMS	Osiris
LAS-x	1			
SES	0.75	1		
FDMS	0.65	0.82	1	
Osiris	0.86	0.65	0.64	1

Average ratios for LAS-x to TEOM (PM_{10}) or FDMS ($PM_{2.5}$)

Table 10

	PM_{10} TEOM/LAS-x	$PM_{2.5}$ FDMS/LAS-x
27-Sep	1.25	1.20
03-Oct	1.36	1.69
04-Oct	1.24	1.79
10-Oct	1.21	2.10
12-Oct	0.59	0.99
Average	1.13 (± 0.3)	1.55 (± 0.5)

The highest correspondence is found between LAS-x and Osiris, which is due to the common measurement technique (optical). The weakest correlation is always observed with the FDMS.

To level the LAS-x measurements to the TEOM readings, average ratios per day have been calculated. Results are given in Table 10. The overall average factor is used for the five remaining days on which no instrumental comparison occurred.

Regarding the PM_{10} estimates, ratios were within comparable range on most days (4 out of 5). Only on 12 October the ratio deviated for reasons explained above. In case of the $PM_{2.5}$ values, ratios increased, with respect to those found for PM_{10}. The reason probably was that the (relative) contribution of volatile compounds to $PM_{2.5}$ was larger than to PM_{10}.

Annex 4 Osiris normalisation factors

Normalisation between Osiris instruments is carried out by a monitoring campaign of around 4-6 days, in which the instruments are put together at the same location. Based on these days, an average normalisation factor to one of the instruments, the so-called reference Osiris, is determined for all the Osiris instruments:

$$(normalisation\_factor)_{Osiris"x"} = \frac{(mean\_concentration)_{Osiris"2507"}}{(mean\_concentration)_{Osiris"x"}}$$

The factors used to normalise the Osiris PM_{10} and $PM_{2.5}$ data to one reference Osiris level (2507) are tabular and graphically presented below. 1σ uncertainty intervals, based on hourly calculated factors are plotted in the graph. During the BOP monitoring campaign, a linear interpolation was applied between the before and after normalisation factor.

Monitoring campaign 1: September–October 2007
For the Osiris instruments 2529, 2563 and 2487, either the before or the after normalisation factor was not known, due to technical or planning difficulties. For these instruments, a constant factor was assumed.

The instrument 2532 broke down during the campaign and was replaced by instrument 2563. Data from the 2532 instrument were not taken into account in the spatial variability analysis.

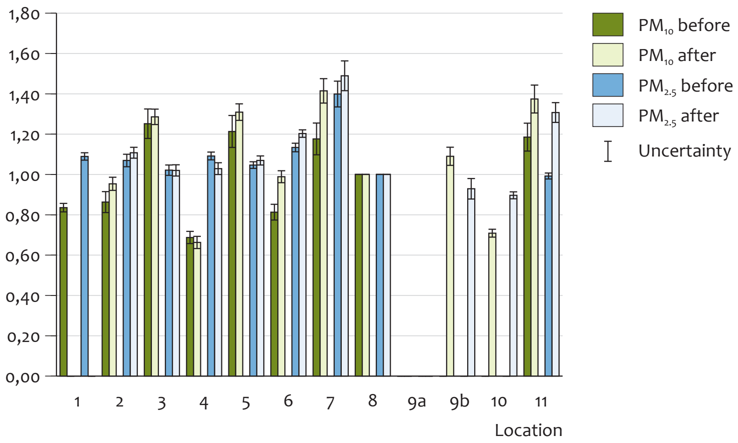
Osiris normalisation factors for the first monitoring campaign **Table 11**

	Location	Osiris	PM_{10} before	PM_{10} after	$PM_{2.5}$ before	$PM_{2.5}$ after
1	Schiedam Kethel, Educatief Centrum	2529	0.84	-	1.09	-
2	Schiedam West, Tennisvereniging	2531	0.86	0.95	1.07	1.11
3	Rotterdam Nieuwe Westen, CBS Mozaiek	2564	1.25	1.29	1.02	1.02
4	Rotterdam Provenierswijk, Hildegardis MAVO	2340	0.69	0.66	1.09	1.03
5	Rotterdam Crooswijk, Speeltuinvereniging	2566	1.21	1.31	1.05	1.07
6	Schipluiden, Zouteveensweg	2530	0.81	0.99	1.13	1.20
7	Rotterdam city centre, RIVM Schiedamsevest	2525	1.18	1.41	1.40	1.49
8	Vlaardingen, DCMR Vlaardingen	2507	1.00	1.00	1.00	1.00
9a	Schiedam, DCMR Schiedam until 30/9	2532	-	-	-	-
9b	Schiedam, DCMR Schiedam from 1/10	2563	-	1.09	-	0.93
10	Rotterdam Noord, Bentinckplein	2487	-	0.71	-	0.90
11	Rotterdam Overschie, DCMR Overschie	2565	1.19	1.37	0.99	1.31

Osiris normalization factors including 1 σ uncertainty intervals

Figure 18

First monitoring campaign; September-October 2007



Osiris normalisation factors including 1σ uncertainty intervals for the first monitoring campaign.

Monitoring campaign 2: March 2008

Osiris normalisation factors for the second monitoring campaign

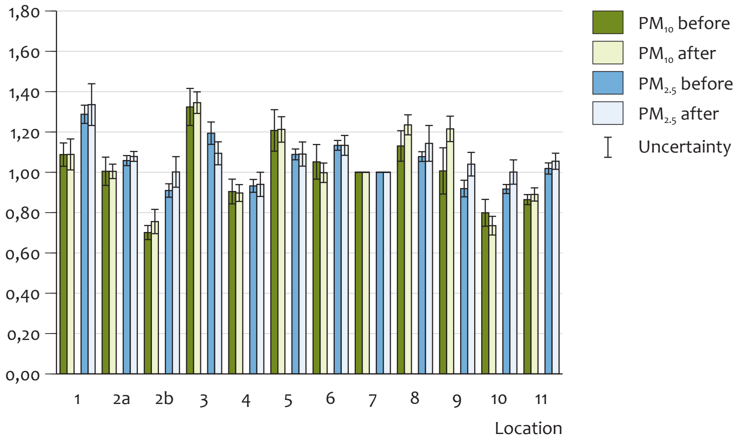
Table 12

	Location	Osiris	PM_{10} before	PM_{10} after	$PM_{2.5}$ before	$PM_{2.5}$ after
1	Schiedam Kethel, Educatief Centrum	2530	1.09	1.09	1.29	1.34
2a	Schiedam West, Tennisvereniging until 8/3	2531	1.00	1.00	1.06	1.08
2b	Schiedam West, Tennisvereniging from 12/3	2339	0.70	0.76	0.91	1.00
3	Rotterdam Nieuwe Westen, CBS Mozaiek	2564	1.32	1.35	1.19	1.09
4	Rotterdam Provenierswijk, Hildegardis MAVO	2525	0.90	0.90	0.93	0.94
5	Rotterdam Crooswijk, Speeltuinvereniging	2566	1.21	1.21	1.09	1.09
6	Schipluiden, Zouteveensweg	2527	1.05	1.00	1.13	1.13
7	Rotterdam city centre, RIVM Schiedamsevest	2507	1.00	1.00	1.00	1.00
8	Vlaardingen, DCMR Vlaardingen	2532	1.13	1.24	1.08	1.14
9	Schiedam, DCMR Schiedam	2563	1.01	1.22	0.92	1.04
10	Rotterdam Noord, Bentinckplein	2487	0.80	0.74	0.92	1.00
11	Rotterdam Overschie, DCMR Overschie	2488	0.86	0.89	1.02	1.05

Osiris normalization factors including 1 σ uncertainty intervals

Figure 19

Second monitoring campaign; March 2008



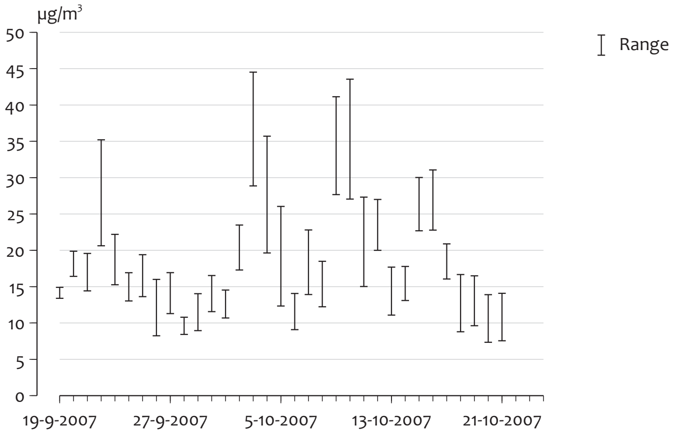
Osiris normalisation factors including 1σ uncertainty intervals for the second monitoring campaign.

The Osiris instrument 2563, located at Schiedam (9), showed unrealistic low concentration levels, both for PM_{10} and $PM_{2.5}$, directly from the beginning of the monitoring campaign. The normalisation factor after the campaign turned out to be far higher than the factor before. We decided to apply the after normalisation factor during the whole monitoring campaign.

Annex 5 Osiris data

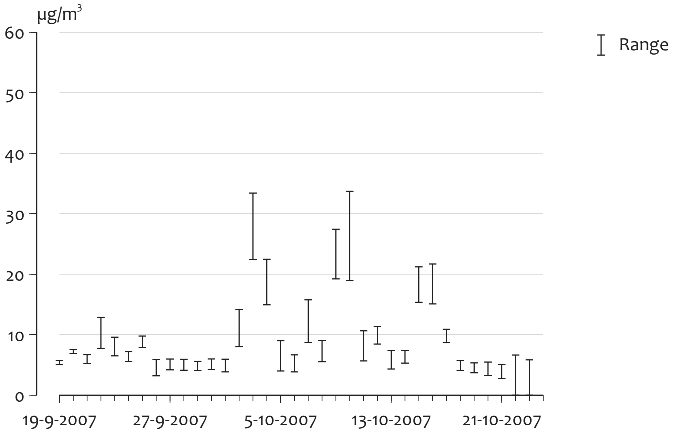
Daily average concentrations for PM_{10} and $PM_{2.5}$ during both monitoring campaigns, are presented in Figure 20-Figure 23. The concentration values are not absolutely correct, since the data were not calibrated to the reference method. Therefore, figures only reveal relative variability in time and space.

Daily average PM_{10} concentration **Figure 20**



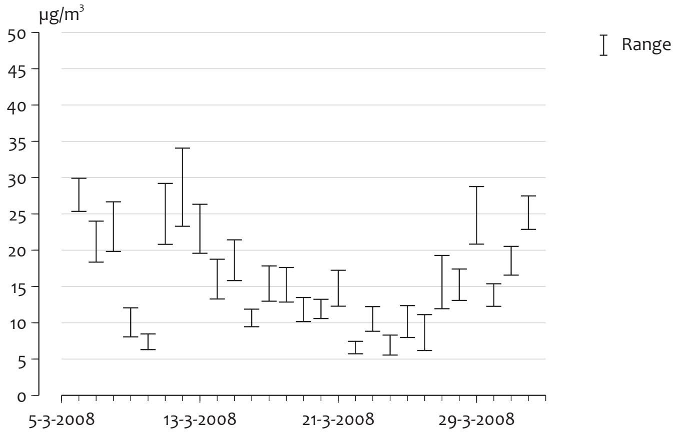
Daily average PM_{10} concentrations during the first monitoring campaign. The data were normalised to one reference Osiris level. The data were not calibrated to the reference method.

Daily average $PM_{2.5}$ concentration **Figure 21**



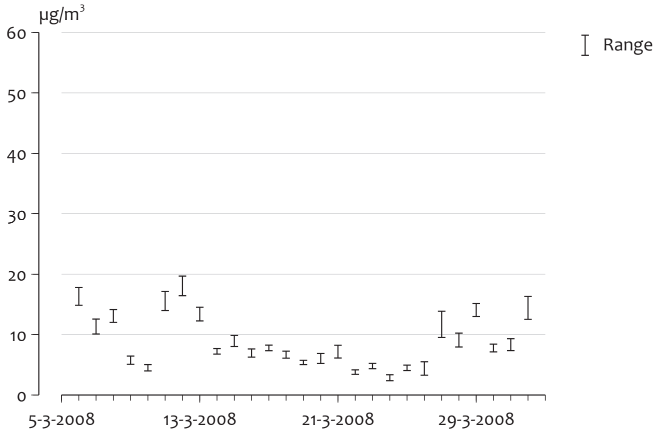
Daily average $PM_{2.5}$ concentrations during the first monitoring campaign. The data were normalised to one reference Osiris level. The data were not calibrated to the reference method.

Daily average PM_{10} concentration **Figure 22**



Daily average PM_{10} concentrations during the second monitoring campaign. The data were normalised to one reference Osiris level. The data were not calibrated to the reference method.

Daily average $PM_{2.5}$ concentration **Figure 23**



Daily average $PM_{2.5}$ concentrations during the second monitoring campaign. The data were normalised to one reference Osiris level. The data were not calibrated to the reference method.

Comparison of PM_{10} with TEOM-SES data from DCMR

During both the monitoring campaigns, at three monitoring stations, simultaneous measurements of PM_{10} by TEOM-SES instruments were carried out by DCMR: Overschie, Schiedam and Bentinckplein. To evaluate the relative differences between the locations measured by the Osiris instruments a comparison was made with the TEOM-SES data. The ratio between the TEOM-SES and normalised Osiris average concentrations were calculated for each of the three locations. Days were only taken into account if both Osiris and TEOM-SES data were available. The uncertainty (1σ) in the ratio was based on the assumption that the uncertainty in the measured mean concentration was 10% (1σ), for both methods.

During the first campaign, the ratios for Bentinckplein and Overschie were very similar, but the Schiedam ratio was substantially lower. Assuming that the TEOM-SES measurements were 'correct', this could mean that either the Osiris instrument at Schiedam overestimated the PM_{10} concentration or the Osiris instruments at Overschie and Bentinckplein underestimated the PM_{10} concentration.

During the second campaign, the ratios differed less. Due to the uncertainty in both methods, conclusions about malfunction of Osiris instruments could not be drawn.

Comparison with gravimetric data

At Schiedam and Bentinckplein, gravimetric measurements were carried out by RIVM, during the BOP monitoring period. A regression between Osiris and gravimetry was forced through zero, with the Osiris on the X-axis to make the slope of the regression indicate an average correction factor needed for calibration towards the reference method. Results for PM_{10} and $PM_{2.5}$ are presented in Figure 24 (first monitoring campaign) and Figure 25 (second monitoring campaign).

TEOM-SES average concentration divided by normalised Osiris average concentration.

Table 13

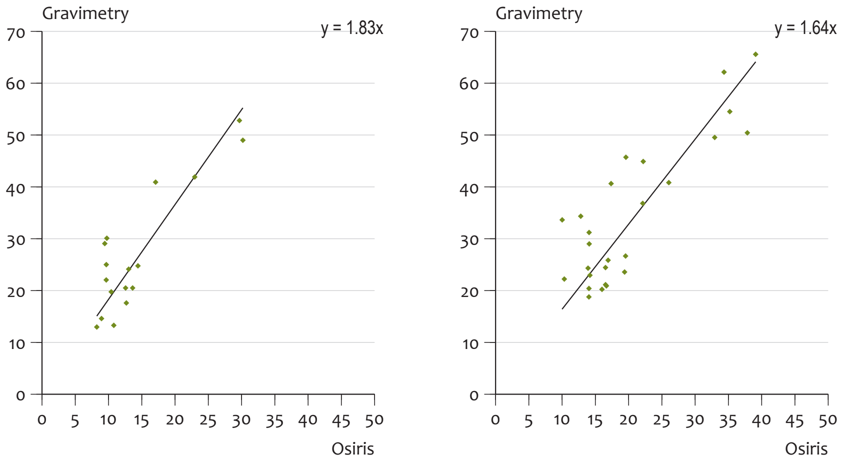
	First campaign (3-7; 12-21 October)	**Second campaign (7-17; 19-24 March)**
Schiedam	1.33 ± 0.19	1.33 ± 0.19
Overschie	1.61 ± 0.23	1.44 ± 0.20
Bentickplein	1.56 ± 0.22	1.30 ± 0.18

Regression between daily average PM concentrations

Figure 24

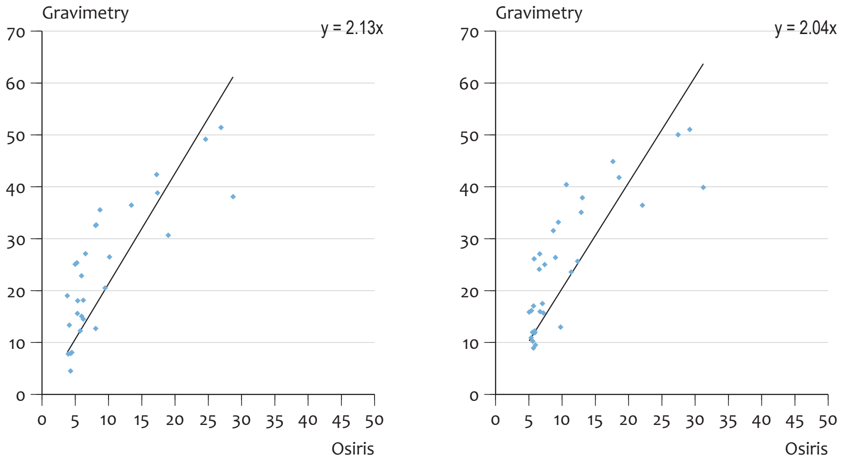
Schiedam DCMR, Location 9 PM_{10}

Bentinckplein PM_{10}



Schiedam DCMR, Location 9 $PM_{2.5}$

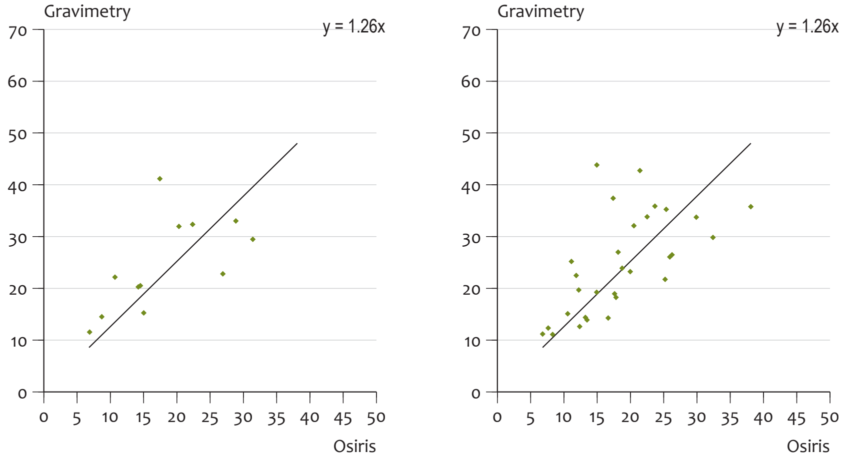
Bentinckplein $PM_{2.5}$



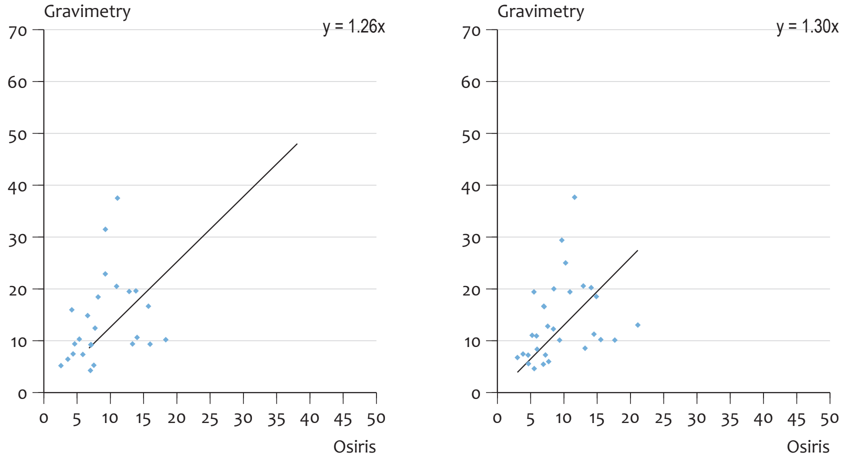
Regression between daily average PM concentrations measured by Osiris and gravimetry during the first monitoring campaign (September-October 2007)

Regression between daily average PM concentrations **Figure 25**

Schiedam, location 9 PM $_{10}$ Bentinckplein, location 10 PM $_{10}$



Schiedam, location 9 PM $_{2.5}$ Bentinckplein, location 10 PM $_{2.5}$



Regression between daily average PM concentrations measured by Osiris and gravimetry during the second monitoring campaign (March 2008)

During both campaigns, the Osiris underestimated the PM concentrations, in comparison with the gravimetric measurements. During the first campaign, differences in the regression coefficient between Schiedam and Bentinckplein and between PM_{10} and $PM_{2.5}$ were observed. The underestimation was highest for $PM_{2.5}$ (with a regression factor exceeding 2).

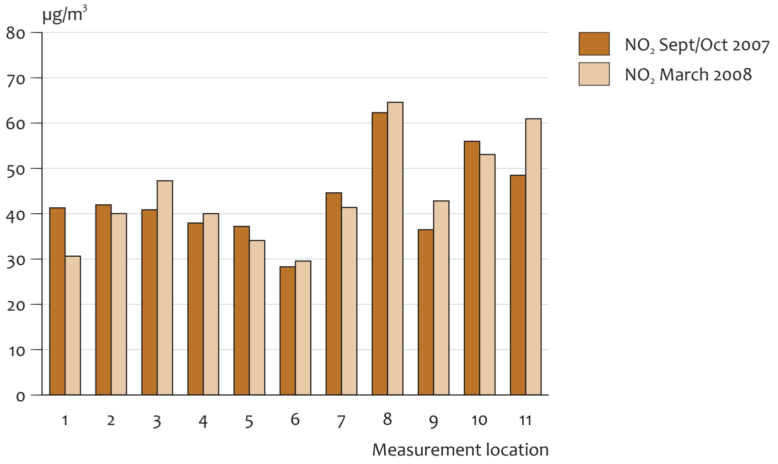
During the second campaign, the regression coefficient was similar for both locations and PM_{10}/$PM_{2.5}$: around 1.3

Annex 6 Passive sampling data (NO_2)

Three locations show up as influenced by traffic: Vlaardingen (8), Bentinkcplein (10) and Overschie (11).

Compared to the PM analyses described in this report, the monitoring period for the passive sampling was longer. For the first monitoring campaign, this meant that more days with south-westerly winds in September 2007 were taken into account. This might partly explain the elevated NO_2 concentration, compared to PM, at the Overschie location (11), and the decreased NO_2 concentration at the Schiedam location (9).

NO_2 concentration measured by diffusive sampling **Figure 26**

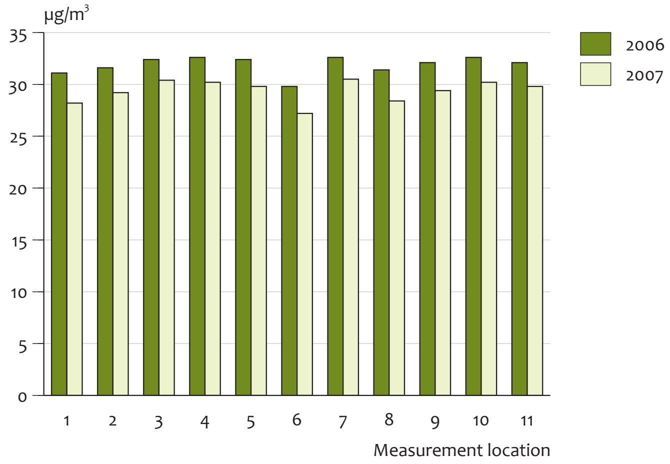


NO_2 concentrations (µg/m³) measured by passive sampling

Annex 7 GCN data

The GCN PM_{10} concentrations for the 1 x 1 km^2 grid in which the monitoring locations in this study were located are presented in Figure 27. The concentrations at the monitoring locations were gathered using a GIS application including the GCN data for 2006 and 2007.

PM_{10} concentration in 2006 & 2007 for GCN grid cells **Figure 27**



PM_{10} concentration values in the 2006 and 2007 GCN grid cells

Annex 8 Scheme of the different routes during the mobile measurements

The scheme of the different routes driven by the mobile unit during the campaign in 2007 is presented in Table 14.

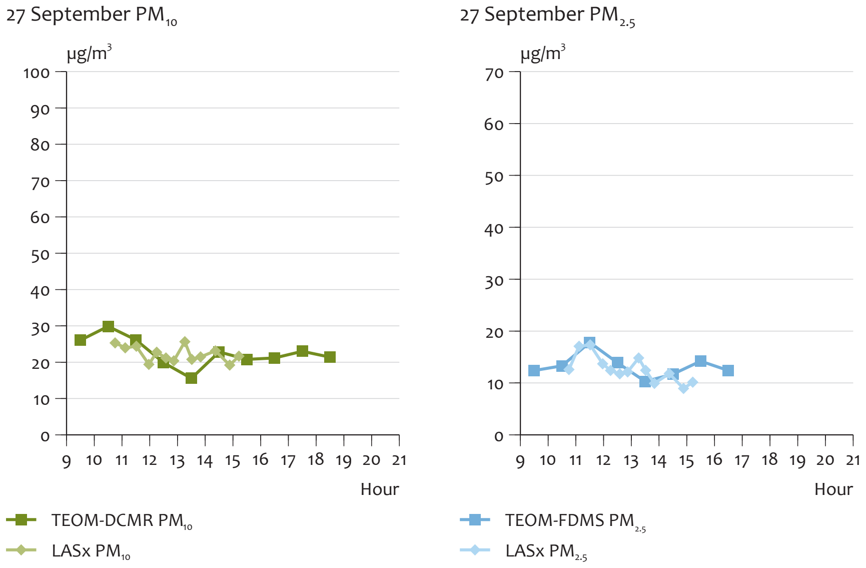
Scheme of the routes of the mobile measurements **Table 14**

Date	Route	Location	Arrival time	Departure time
27-Sep-07	1	6	09:07	09:40
		1	10:30	11:00
		9	11:15	11:48
		3	12:05	12:26
		7	12:42	13:04
		3	13:24	13:40
		9	13:58	14:45
		1	15:00	15:27
		6	16:10	16:45
01-Oct-07	2	11	11:18	11:43
		10	11:52	12:13
		4	12:18	12:37
		5	12:47	13:05
		7	13:24	13:49
		3	13:49	14:20
		10	14:36	14:55
		4	14:57	15:13
		5	15:23	15:44
		7	16:00	16:20
		3	16:32	16:58
		11	17:13	18:02
03-Oct-07	3	8	11:12	11:45
		2	12:00	12:31
		9	12:41	13:20
		10	14:18	14:41
		5	15:00	15:21
		8	15:44	16:00
		2	16:13	16:30
		9	16:45	17:02
		10	17:21	17:41
		5	17:52	18:14

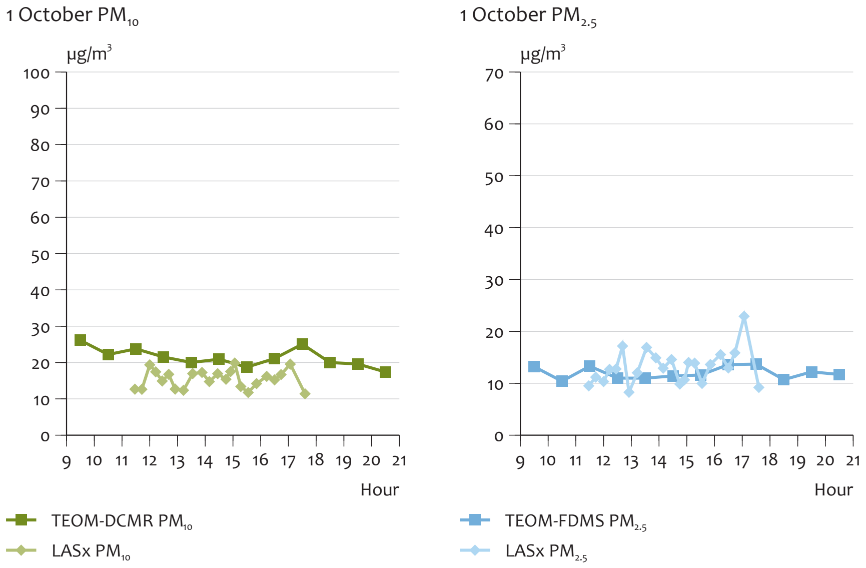
Date	Route	Location	Arrival time	Departure time
04-Oct-07	3	8	11:34	12:07
		2	12:19	12:39
		9	12:48	13:21
		10	13:39	14:02
		5	14:15	15:00
		8	15:27	15:43
		2	15:55	16:17
		9	16:28	16:48
		10	17:12	17:32
		4	17:37	17:48
		5	18:00	18:22
05-Oct-07	1	6	11:35	12:02
		1	12:30	13:00
		3	13:34	13:55
		7	14:19	14:41
		6	15:33	15:54
		1	16:36	16:57
		3	17:26	17:40
		7	18:03	18:30
10-Oct-07	1	7	12:00	12:42
		3	13:00	13:14
		9	13:35	14:05
		1	14:15	14:25
		6	15:05	15:39
		1	16:23	16:50
		9	17:20	17:37
		3	17:56	18:10
		7	18:29	18:45
11-Oct-07	2	11	11:45	12:05
		10	12:22	12:42
		4	12:47	13:12
		5	13:20	13:50
		7	14:09	14:28
		11	14:46	15:10
		10	15:26	15:49
		4	15:54	16:07
		5	16:22	16:42
		7	17:02	17:25
12-Oct-07	3	11	13:00	13:30
		10	13:49	14:05
		9	14:20	14:35
		2	14:43	15:05
		8	15:19	15:41
		2	15:55	16:12
		9	16:25	16:45
		10	17:02	17:20
15-Oct-07	2	11	12:02	12:35
		10	12:47	13:16
		5	13:32	13:55
		7	14:19	14:38
		11	14:56	15:16
		10	15:27	15:47
		5	16:03	16:19
		7	16:36	16:55
16-Oct-07	1	6	11:54	12:18
		1	12:58	13:15
		2	13:30	13:50
		8	14:02	14:19
		2	14:31	14:46
		6	16:00	16:21

Annex 9 Data mobile monitoring campaign

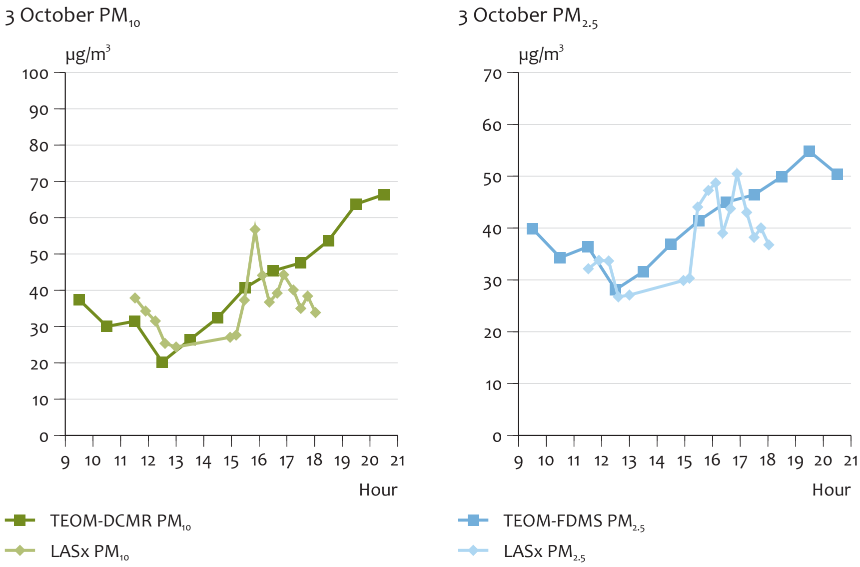
Measured PM concentration **Figure 28**



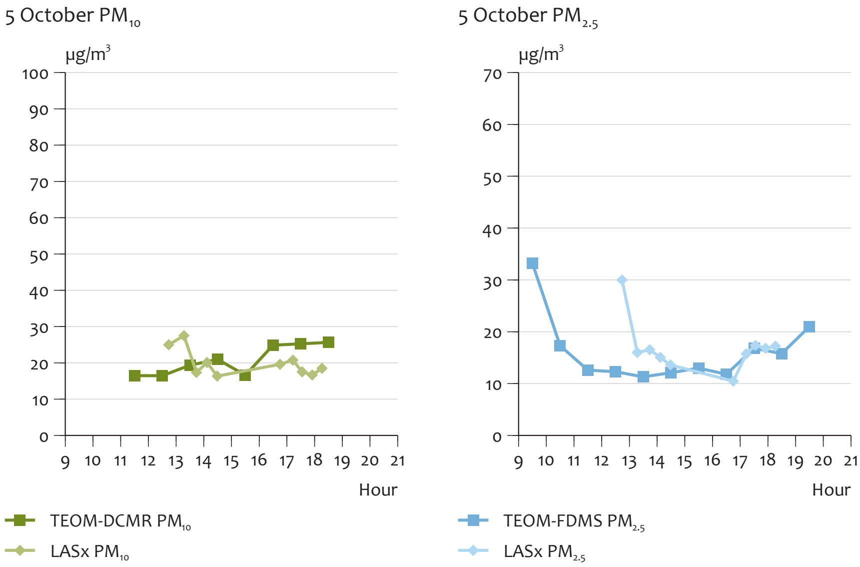
Measured PM concentration **Figure 29**



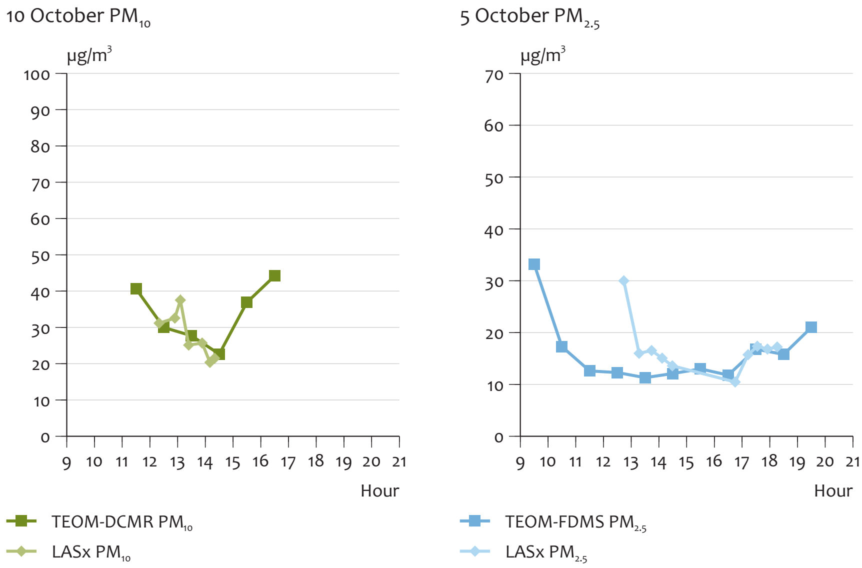
Measured PM concentration **Figure 30**


3 October PM_{10} 3 October $PM_{2.5}$

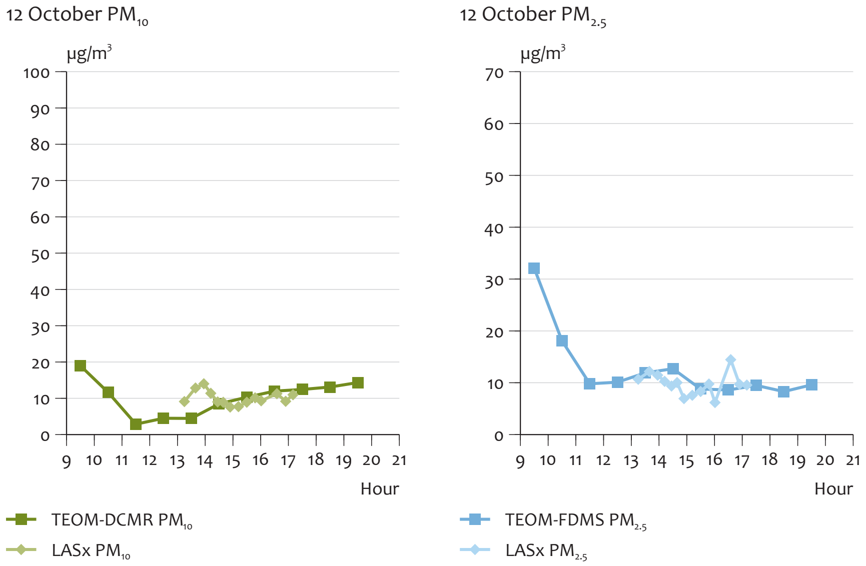
Measured PM concentration **Figure 31**


5 October PM_{10} 5 October $PM_{2.5}$

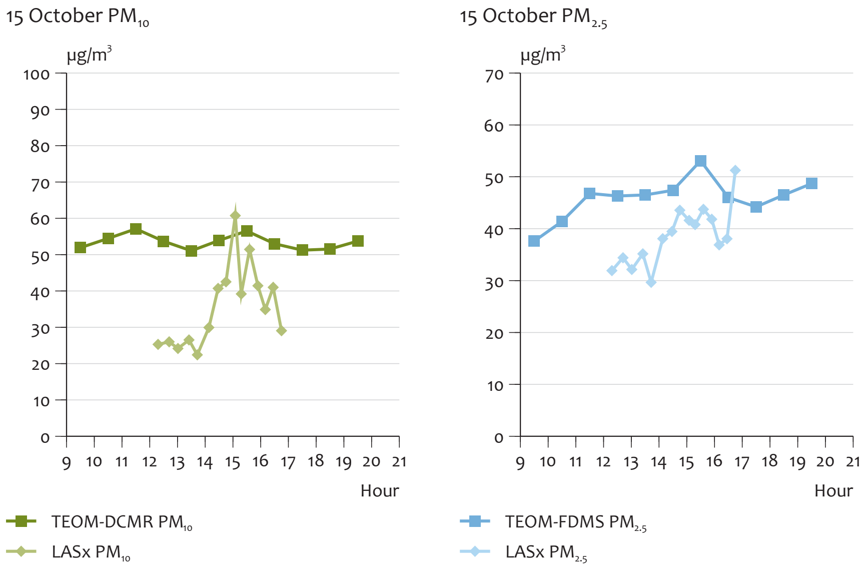
Measured PM concentration **Figure 32**


10 October PM_{10} 5 October $PM_{2.5}$

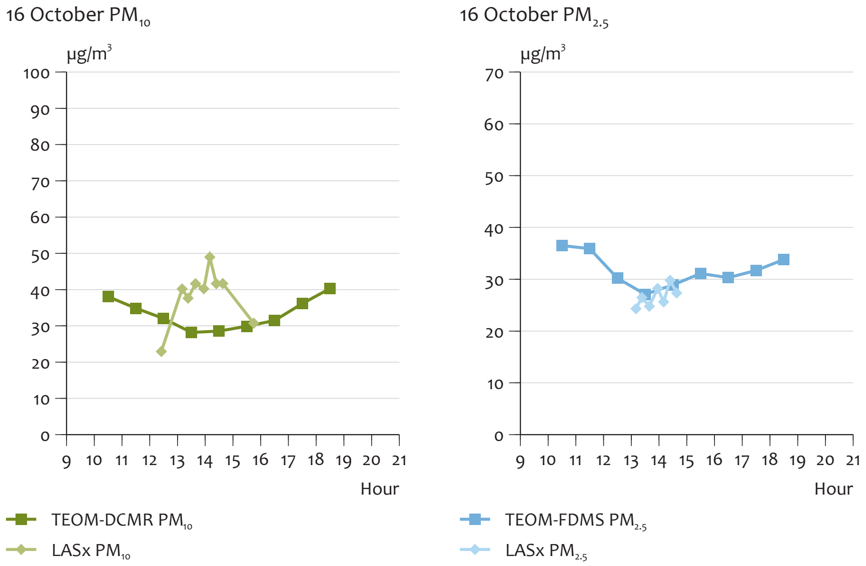
Measured PM concentration **Figure 33**


12 October PM_{10} 12 October $PM_{2.5}$

Measured PM concentration **Figure 34**


15 October PM_{10} 15 October $PM_{2.5}$

Measured PM concentration **Figure 35**

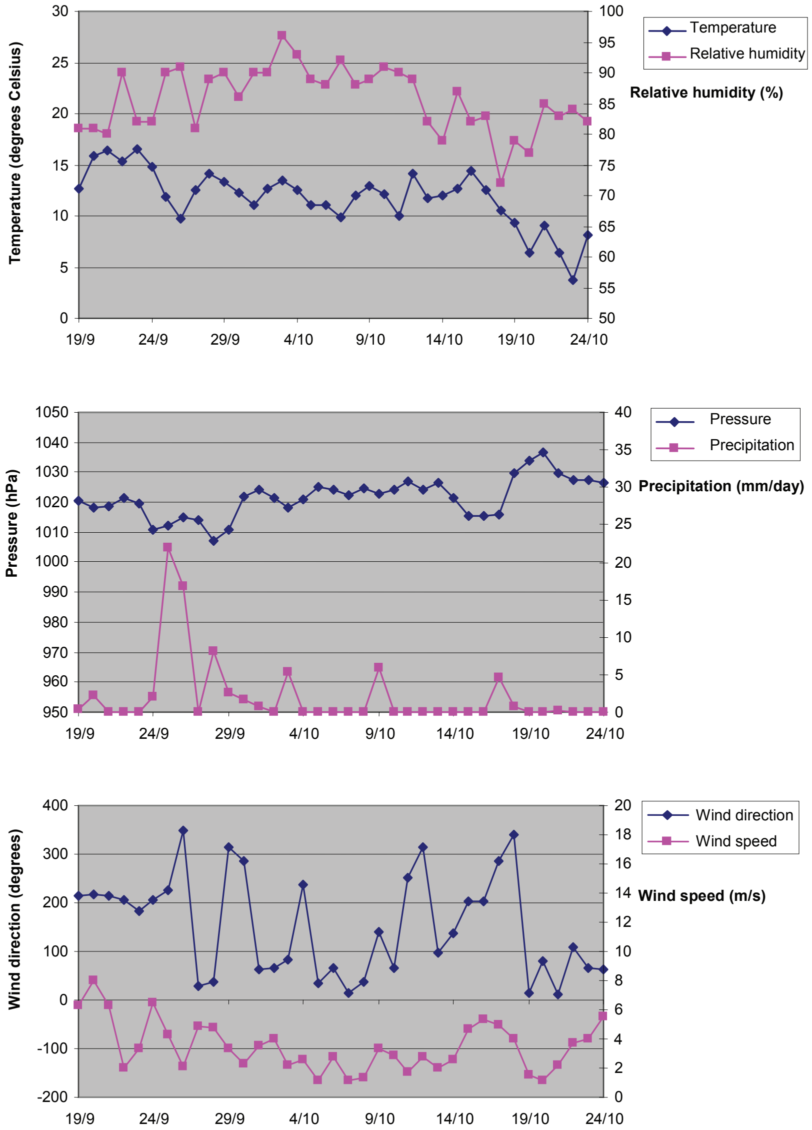

16 October PM_{10} 16 October $PM_{2.5}$

Annex 10 Weather conditions during monitoring campains

KNMI daily average measurements at Rotterdam Airport during the first monitoring campaign (September-October 2007).

KNMI daily average measurements at Rotterdam Airport **Figure 36**

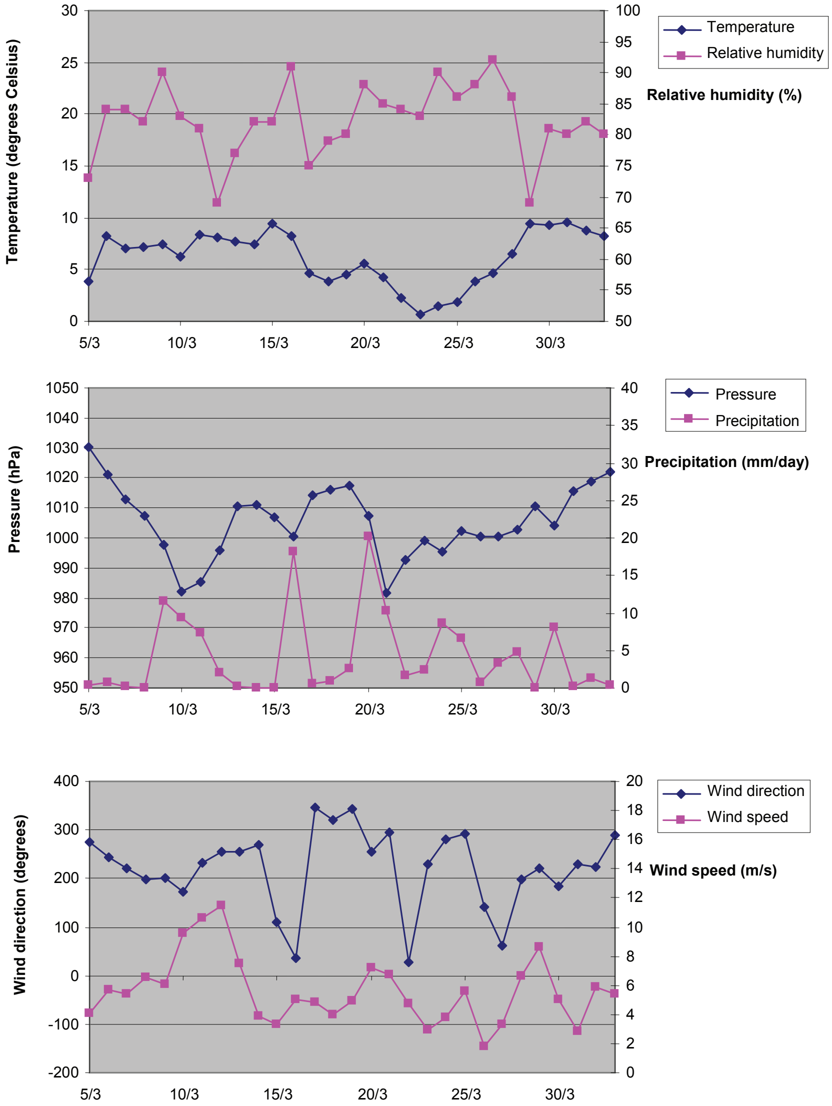
First monitoring campaign (September-October 2007)



KNMI daily average measurements at Rotterdam Airport during the first monitoring campaign (September-October 2007). A. Temperature and relative humidity, B Pressure and precipitation, C Wind direction and wind speed.

KNMI daily average measurements at Rotterdam Airport Figure 37

Second monitoring campaign (March 2008)



KNMI daily average measurements at Rotterdam Airport during the second monitoring campaign (March 2008). A. Temperature and relative humidity, B Pressure and precipitation, C Wind direction and wind speed.

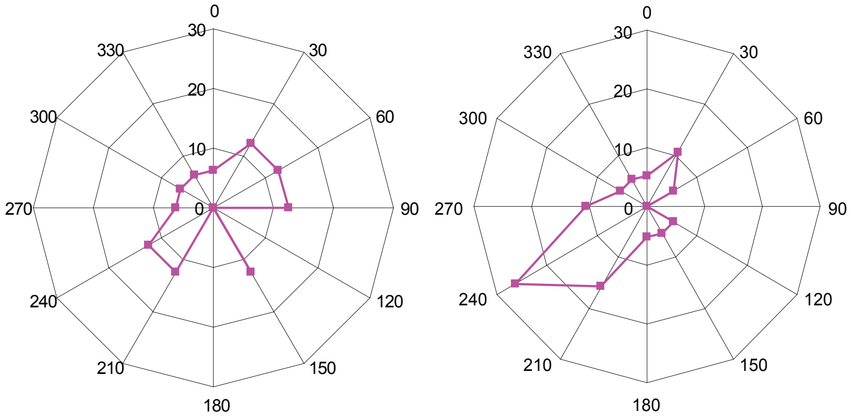
Wind roses

Frequency distribution (%) of wind direction

Figure 38

First monitoring campaign

Second monitoring campaign



Frequency distribution (%) of wind direction for the days during the first and second monitoring period, taken into account in the spatial variability analysis

Weather conditions for the mobile measurements

Weather conditions at mobile measurement days

Table 15

Date	Time (local time)	Wind direction (degrees)	Wind speed (m/s)	Rain	Cloud fraction
27-Sep	9:00-17:00	40 (20-40)	6.3 ± 1.1	no	3/8
01-Oct	11:00-18:00	70 (50-80)	4.6 ± 0.5	yes	8/8
03-Oct	11:00-18:30	90 (60-140)	1.9 ± 0.6	yes	8/8
04-Oct	11:30-18:30	260 (210-310)	2.3 ± 1.2	no	4/8
05-Oct	11:30-18:30	30 (350-70)	1.8 ± 0.5	no	7/8
10-Oct	12:00-19:00	50 (30-90)	3.0 ± 0.0	no	3/8
11-Oct	11:30-17:30	270 (230-290)	2.3 ± 0.8	no	3/8
12-Oct	13:00-17:30	330 (320-350)	4.2 ± 0.4	no	6/8
15-Oct	12:00-17:00	210 (210-220)	5.7 ± 1.0	no	5/8
16-Oct	11:30-16:30	210 (210-220)	7.2 ± 0.8	no	1/8

Meteorological data from the KNMI station at Rotterdam airport. The data for the 10 days are the averages of the measurement over time.

Annex 11 Semivariogram: details and results

Details about method

To obtain more insight into the local representativeness of point measurements in an urban environment, the geostatistical technique of spatial variograms has been applied. With this technique it is possible to determine the spatial variation within observational datasets. Semivariograms identify the distance where data are no longer spatially autocorrelated. An example of how this technique can be applied on data acquired with a mobile laboratory, is described by Lightowlers et al. (2008) and Larson et al. (2007). In fact, a semivariogram gives a better understanding of spatial continuity of data. It is a function of distance and is based on the average sum of squared differences in attribute values for all pairs of points (see Equation 1 and Figure 41).

$$\gamma(h) = \frac{1}{2n(h)} \sum_{s_i - s_j = h} (z_i - z_j)^2 \qquad (1)$$

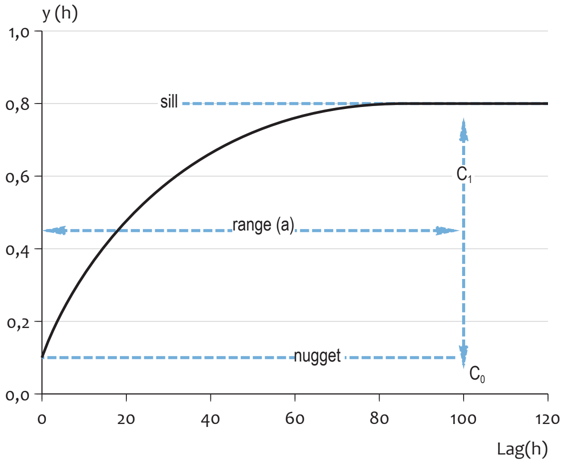
The γ is the symbol for a semivariogram and zi and zj are the attribute values of points si and sj. The summation is over all pairs of points that are separated by a distance h, and n (h) is the number of pairs. The semivraiogram is commonly represented as a graph that shows the variance in measure with distance between all pairs of sampled locations. To reduce the number of points on the semivariogram, pairs of locations are binned based on their distance from each other. The number of pairs in each lag was never < 40 to ensure statistical reliability.

At relatively short distances, the semi-variance is small, but increases with the distance between the measurement points. At the distance referred to as range, the semi-variance levels off to a relatively constant value (referred to as sill). This implies that beyond this ‘range’, the variation is no longer spatially correlated. The semivariogram is usually modeled by fitting a theoretical function. Such a graph is helpful to build a mathematical model that describes the variability of a parameter with distance. The special geostatistical analyst toolbox of Geographic Information Systems (GIS) is used, to quantify the spatial relationships by semivariogram analysis.

Results from mobile measurements

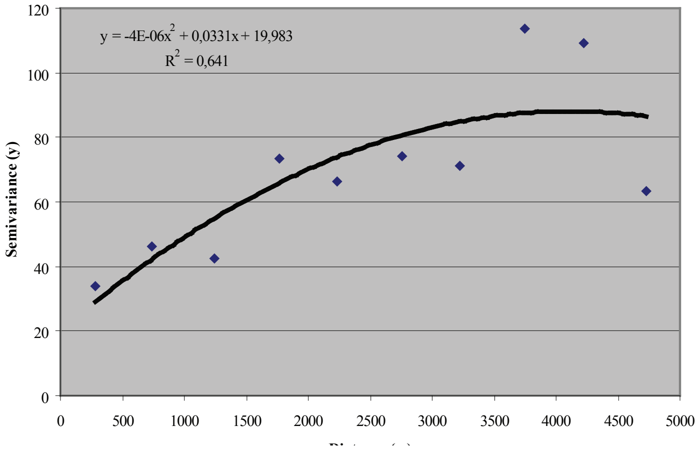
Three routes were discerned, in particular. The first route was from regional (location 6) to urban background (location 1), and back; the second route was within the urban area between locations 3 and 9; the third route was from traffic location 10 to the nearby urban background location 4 and back.

Example of a semivariogram **Figure 39**



Semivariogram from location 1 to 6

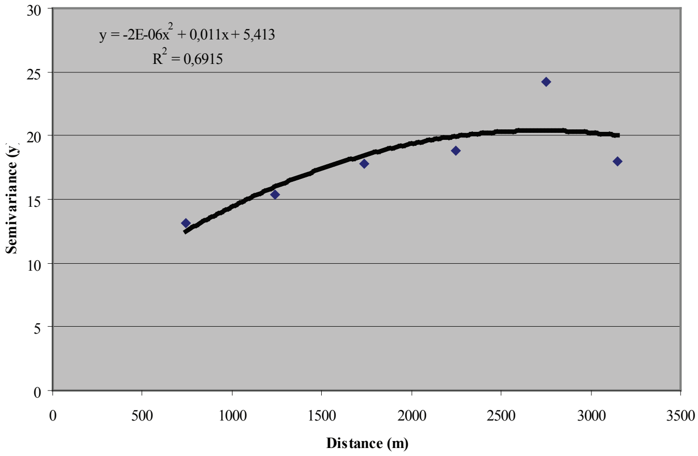
Figure 40



Semivariogram from location 1 (urban background) to 6 (regional background) (distance = 4000m)

Semivariogram from location 3 to 9

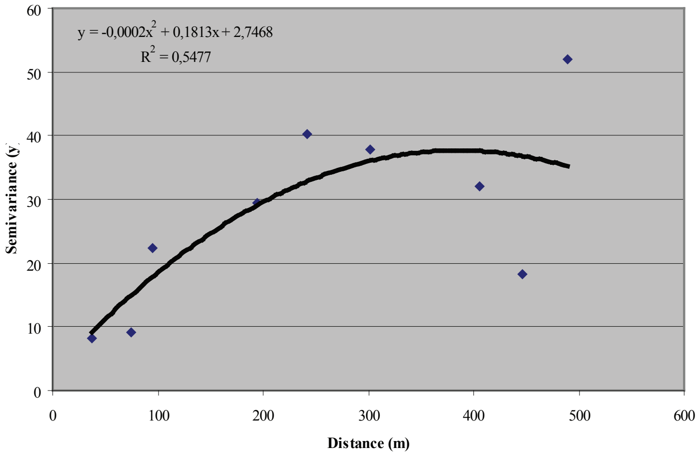
Figure 41



Semivariogram from location 3 to 9 (both urban background) (distance = 2750m).

Semivariogram from location 10 to location 4

Figure 42



Semivariogram from location 10 (traffic) to location 4 (urban background) (distance = 450m).

Results are given in Figure 40, Figure 41 and Figure 42. As an example of how concentration levels change during the ride, the concentration variation going from the regional background location 6 to urban background location 1, is given in Figure 42. Figure 43 shows the corresponding semivariogram. Using all data from the seven times that this route was covered, the estimated distance was around 4000 m.

Following the same procedure, but now for the route within the urban area, gives a distance of 2750 m. The correlation of data acquired within the urban area appeared less than in the regional area, which was not unexpected. Further proof for this comes from the calculation for the route between Bentinckplein (traffic location 10) and the urban background location 4. Here, the semivariogram gives a range of 450 m, which is most likely due to a steep downward gradient when going from the street location to the urban background area.

The finding that within the urban background area the range was smaller than in the regional area, was expected: due to a smaller surface roughness (with less building infrastructure) in the regional area the relationship between (nearby) measurements will be stronger.

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DCMR and RIVM are acknowledged for providing part of the measurement locations and measurement data from their monitoring network.

This report describes the study on the spatial variability, spatial representativeness and temporal variability of urban background concentrations of PM_{10} and $PM_{2.5}$ in the city of Rotterdam. The study was carried out by TNO and ECN as part of the Netherlands Research Program on Particulate Matter (BOP).
Two monitoring campaigns in September/October 2007 and March 2008, including measurements at 11 fixed locations and additional mobile measurements, showed that the spatial variability of urban background PM concentrations is similar to the estimated measurement accuracy. We concluded that the spatial variability is less than 10% for PM_{10} and less than 5% for $PM_{2.5}$. Measurements confirm the non-significant differences between concentrations in different GCN urban background grid cells. They suggest the concept of an urban PM plateau, in which a small gradient from the regional background concentration leads to a constant level of urban background PM concentrations. To reduce the uncertainty in the assessment of the monitored urban background level, multiple urban background monitoring locations are recommended.

This study is a BOP publication produced under the auspices of TNO and ECN.

The Netherlands Research Program on Particular 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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