# The origin of high PM10 and $NO_2$ concentrations in an highly industrialized area: model calculations and analyses of source contributions.

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**Abstract**

PM10 and $NO_2$ concentrations in industrialized parts of the Netherlands are close to or beyond the threshold values of EU directive 1999/30/EC. Monitoring air quality is carried out to achieve insight in the origin and relative contribution of different sources. An effective air quality improvement plan is founded on an in depth analysis of all contributions to PM10, $NO_2$ and other air pollutants. Computer models play an important role in this monitoring and improvement process. In this paper we discuss the individual contribution of large industrial sources, ship emissions and fugitive dust sources to the PM10 and $NO_2$ concentrations, calculated with a local scale computer model STACKS for both industries and traffic. STACKS is the KEMA version of the Dutch National Dispersion model. The basics of this model are explained. This model is applied to the Rotterdam area: a highly industrialized area with the world largest harbour and the presence of several major oil industries, intensive truck and ship transport routes. Both from model calculations it appears that the local contributions of these large industrial sources to PM10 or $NO_2$ concentrations is rather low. Measurements confirm this conclusion. High PM10 concentrations are found to correlate with specific weather conditions. A strong relation was found between meteorological large scale effects and episodes of high PM10 concentrations. Large scale weather systems in western Europe seem to be highly related with episodes of elevated PM10 concentrations: period's of high pressure systems without rainfall cause elevated PM10 concentrations. For $NO_2$ concentrations the contribution of ship and road emissions are significant on a local scale, however, back ground concentrations are dominant, too. Large (tall) sources have small impacts, although the latter have a major contribution to the total amount of emissions. Air pollution reduction strategies should focus first on reduction of the back ground concentrations by introducing structural and generic measures. Hot spots can be much better combated by local measures, such as speed reduction, noise screens and additional local emission requirements for the low industrial sources.

## 1 INTRODUCTION

Air quality issues have become of great importance in decision-making processes. Since the new EU limit values for $NO_2$ and PM10 became in to effect, The Netherlands struggle with the air quality standards. In many places in the country the limit values are approached or exceeded. People who are sensitive to irritation of the bronchial tubes may suffer during episodes of air pollution. Although these episodes may occur in both summer and winter and their origin and composition may be different, they have two things in common. At first, large scale airborne transport is important; the origin of the episodes is not primarily found in local emissions causing local problems, but in large areas that are affected (typically a trans-boundary problem). Secondly, both summer and winter smog cause problems for people who are asthmatic. Individual stacks can sometimes contribute to this type of episodes and are therefore subject to regulations.

The Rijnmond area is a large business area with the worlds largest harbour and many large and heavy industries. Due to the transit and commercial activities there is an intensive flow of traffic and ships

both outside and inside the harbour area. At the western point of the Rijnmond area a large land reclamation has been planned by the Dutch, called the Maasvlakte ('Plain of river Maas'). This Maasvlakte is gradually filled up with new industries. Within a period of 10 years a strong need is expected to extend the area to house new industries. An area of 1000 ha of newly reclaimed land is planned in the period until 2015. As a result of these new developments a larger impact on the environment is expected. Part of this impact is air pollution. Especially the surplus of (road and water) traffic will cause extra emissions of $NO_x$ and PM10. In this area the EU limit values for PM10 and $NO_2$ are already exceeded. Due to Dutch legislation, realization of the new activities is only permitted when the extra exceedings are compensated with additional measures. For $NO_2$ this means measures such as noise screens, speed limits for ships and cars and introducing incentives for cleaner cars. PM10 is more difficult to address. In this area the exceedings are assumed to be caused by: 1) high back ground concentrations and 2) local emissions of transhipment companies and of seagoing ships. In order to get insight in the possibilities to reduce the local PM10 concentrations and especially the number of days with PM10 concentrations above 50 ug/m3 (the EU limit value) it is necessary to estimate the local contributions of different contributors to the total concentrations. Moreover, it is interesting to answer the question under what circumstances the daily PM10 concentrations are high.

In this paper we report the results of the analysis of measured PM10 values at specific locations in the Rijnmond area and model calculations with source emissions and meteorology as input. Moreover, $NO_2$ calculations are done to explore the difference in local contributions for PM10 and $NO_2$.

## 2 PM10 AND $NO_2$ CONCENTRATIONS IN THE NETHERLANDS

Existing and future levels of PM10 concentrations are made public by the Institute of Environment and public health, the RIVM and its related organisation MNP. The regional environmental institute, DCMR, carries out additional ambient measurements on a regular basis. In figure 1 the estimations of the PM10 concentrations are given for 2010. This picture gives the calculated PM10 concentration of MNP, this figure 1 is constructed on the basis of both modelled and measured values. It is clear from these pictures that the PM10 concentrations exceed the critical level of 32 ug/m3, being the level whereat the number of days with concentrations higher than 50 ug/m3 will be exceeded.

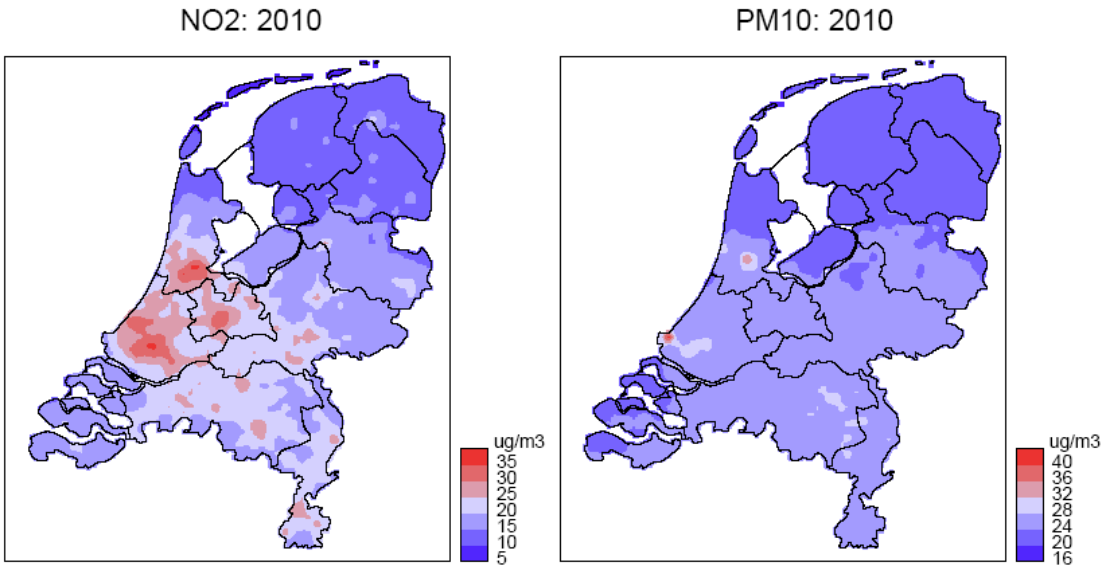


Figure 1. Predicted averaged $NO_2$ and PM10 concentrations in 2010 (source: MNP, 2006).

**Rijnmond situation**

As can be concluded from figure 1 the highest concentrations are calculated in the Rijnmond area. Especially in the west section of the area, the Maasvlakte (see figure 3), high concentrations are predicted to occur. To answer the question what causes the higher concentrations, one has to look at the emissions in this region. The relative contributions of all sectors to the total PM10 emissions is given in figure 2.

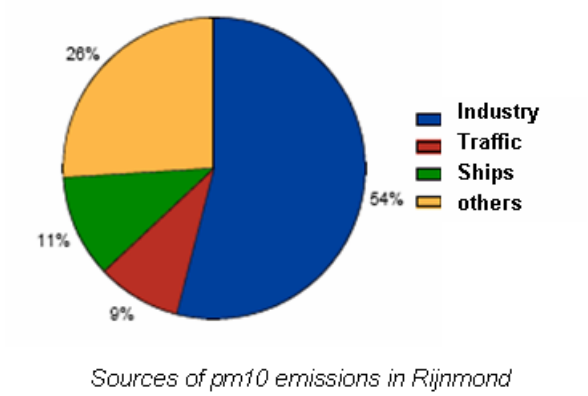


Figure 2. PM10 sources in the Rijnmond area (DCMR, Regionaal actieprogramma Rijnmond, 2005)

The majority of the PM10 emissions is coming from the large industries, mainly emitted by tall stacks. In the Maasvlakte area, diffuse emissions from coal and other bulk storages in the region also contribute to the PM10 emissions. The quantity of those emissions is rather uncertain and subject to ongoing discussions. A main reason for these higher projected local/regional PM10 concentrations is found in the emissions of the storage and transhipment companies, which were thought to be rather high. After a critical review these emissions were corrected downward with a factor of about two. The total industrial emission of PM10 is about 1200 tons/year and emissions of traffic (ships + cars) account for several hundreds of tons, other sectors are minor contributors. The levels of PM10 concentrations exceed the critical limit values for daily averaged PM10 concentrations, set by the EU and implemented by the Dutch authorities. Therefore, a more detailed study to the reasons of these exceedings is done.

## 3 ANALYSIS OF PM10 MEASUREMENTS

To gain more insight of the contribution of local PM10 sources, measured data from DCMR continuous measuring stations are examined (for locations, see figure 4). Data from 2002 and 2003 were available; station numbers 3 (village Hoek van Holland), 4(village Hoogvliet), 5 (village Maassluis) and 9 (town of Vlaardingen) were used. These measurements were extended by TSP (total suspended particles) measurements at Oostvoorne, Rotterdam centre and Maasvlakte (2). TSP and PM10 are not the same, because their size distributions are different, but for our purpose they can be used in a relative manor. From the measurements we made pollution roses to show the contributions of significant local sources. Whenever present local sources become clearly visible in those wind-rose diagrams. Figure 5 shows the concentration distributions (over two years: 2003, 2004) as a function of wind direction at 4 locations (a pollution rose).

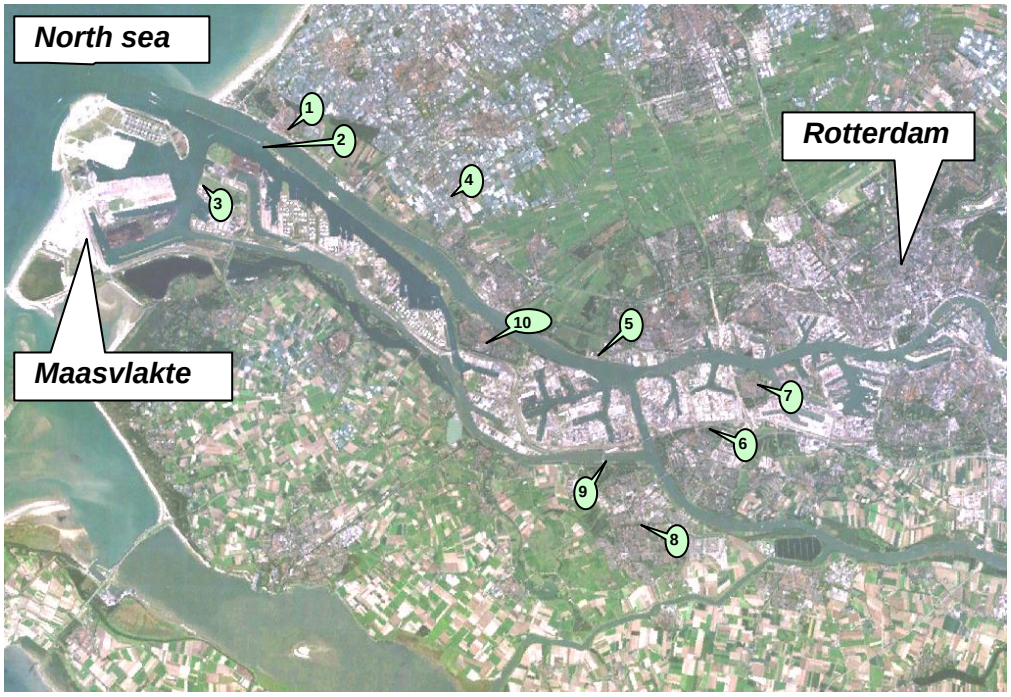

Figure 3. The Rijnmond area. In the western part the Maasvlakte is located. Green points refer to locations for the computer runs (see section 4.3).

The hourly measured concentrations were filtered by taking the moving average over 50 data points (each of 1 degree in wind direction) in order to obtain a reasonably readable picture. Figure 5 shows us that the concentration distributions over the wind rose are quite the same although the orientation relative to Rotterdam and other large sources are very different. The main features in the picture make us to conclude that the local contribution is of minor importance. Especially noticeable is the absence of a clear peak in the plot for Hoek van Holland (site 3). This monitoring station is within a distance of not more than 3 km from large bulk storage companies at the other side of the river. The dominating wind direction at this site is south west (see figure 6, the wind rose); hence at wind directions of about 240 degrees there should be a clear peak in the graph; this is not obvious present. Taking into consideration that the frequency of this wind direction is about 15% (30 degrees sector) the yearly average contribution of the such a source can not exceed a level of 0.15*5ug/m3 (maximum concentration contribution of potential sources in this sector) which is not higher than 1 ug/m3. A clear characteristic of the picture is that at wind directions of 60-150 there is a smooth peak noticeable, indicating that eastern flows towards Rijnmond give rise to higher concentrations, which is a well-known phenomena. A narrow peak at 300 degrees is apparent; probably caused by ship emissions on the river.

Theoretically is it possible that the open bulk storages in the Maasvlakte area have their largest emissions during strong wind periods. Therefore we made the same diagram for the two most nearby stations (2 and 3 in figure 4, both within 3 km of the diffuse sources), but only for days with mean wind speeds of more than 7 m/s, see figure 7. In this picture the peak in the wind sector 220-320 degrees is a bit more pronounced, but even now, the contribution is not very large.

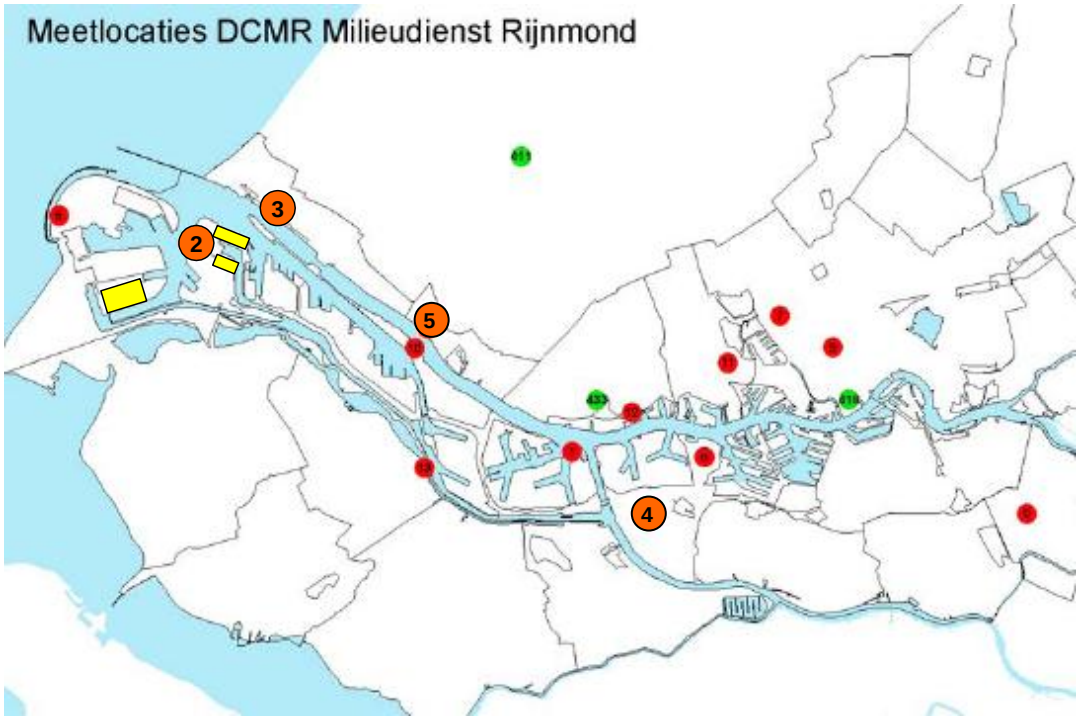


Figure 4. Monitoring sites in the Rijnmond Area (numbers are referred to in the text, yellow blocks are open bulk storages).

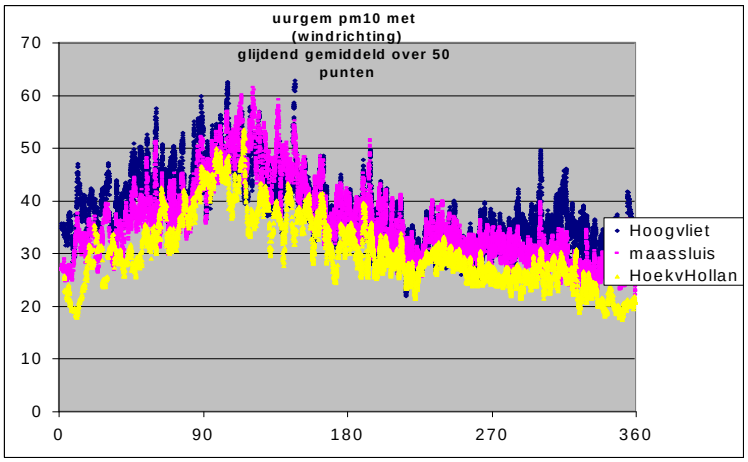


Figure 5. Measured fine particle concentrations (two years) at three locations (3, 4 and 5 in figure 4) as function of wind direction (filtered by taking the moving average over 50 (1 degree) data points).

The next question deals with the origin of the higher PM10 concentrations. It is obvious that wind direction is one explaining parameter. But, we have tried to correlate the daily averaged PM10 concentrations over the two years with all type of meteorological parameters, but strong simple correlations were not found. From literature it is known that weather systems and trajectories may be of strong influence on the contribution of long range air pollution to the local concentrations. From winter smog analysis in the eighties we know that large scale weather systems might be a principal factor in describing episodes of air pollution.

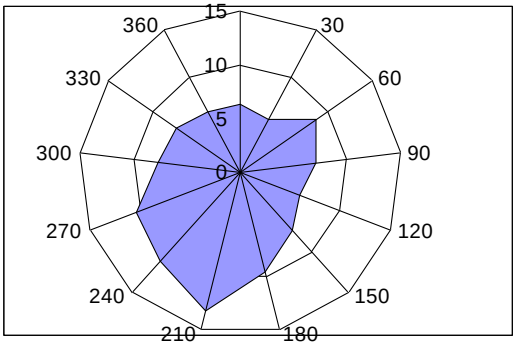

Figure 6. Wind rose (30 degrees sectors).

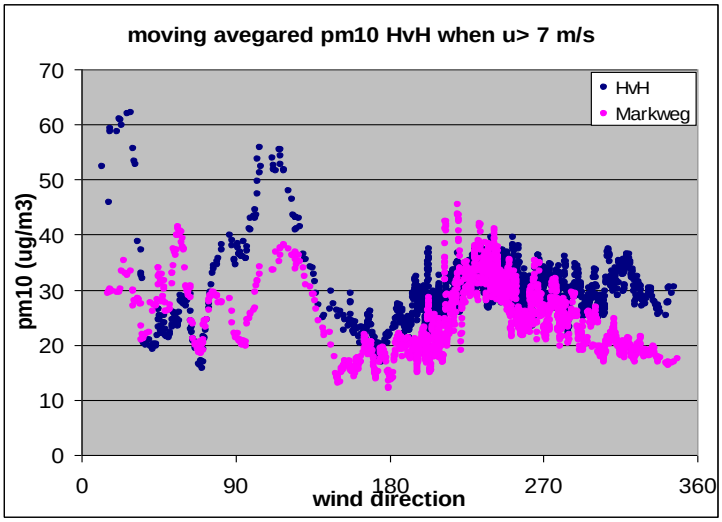

Figure 7. Measured fine particle concentrations (two years) at two locations as function of wind direction). HvH is Hoek van Holland (site number 3); Markweg is site number 2.

We gathered information from the German Weather Service, that made available data sets on historical Grosswetterlagen (GWL) for Western Europe, available on the internet. For each day in the year an analysis of the large scale circulation pattern in Europe is made and translated into one of the GWL codes[1] From the measured PM10/TSP concentrations at the monitoring sites we selected the periods when the daily averaged concentrations at 4 out of 6 stations were beyond the limit value (50 ug/m3). About 2/3 of the days with PM10 levels higher that 50 ug/m3 form together such episodes of more than two days. It is striking that on the average both the beginning and the ending of these episodes interfere more or less with a change in circulation type. Analyzing the main types of weather systems, we see that the episodes all fall into the group of anticyclonal weather systems, see figure 8. These anticyclonal weather systems are characterized by high pressure and dry conditions during more than one day. This means in terms of air pollution modelling that there are no (or slight) (wet) deposition processes in effect and – because of the dominating air subsidence – the exchange of air between the lower parts of the atmospheric boundary layer and the free atmosphere is quenched because of the presence of a temperature inversion layer that acts like a cap on the lower atmospheric layers. Hence, during episodes of high PM10 concentrations almost always we see dry and high pressure situations; but not in all periods of high pressure elevated PM10 concentrations occur: these high concentrations are also the result of trajectories in Europe, determining which parts of Europe are affected. This means that large parts of Europe are dry and dusty during these conditions; transport directions determine where the higher concentrations will occur. This is also recently concluded in

[1] (see www.dwd.de/de/wir/Geschaeftsfelder/Medien/Leistungen/GWL).

German studies (see e.g. Ebel et al., 2004). In an analysis of Environmental Services Rijnmond the existence of this correlation is also reported (DCMR, 2006).

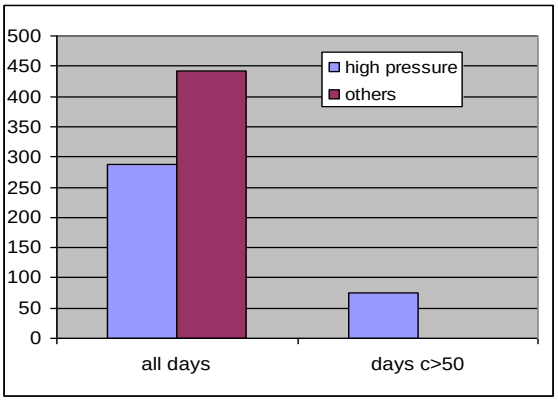


Figure 8: Frequency of anticyclonal weather systems (high pressure) and other systems during all days of the years 2003 and 2004 (left) and only during the days with high PM10 concentrations (right).

# 4 MODEL CALCULATIONS

## 4.1 General

Not only analysis of measurements can be used to see what causes high PM10 concentrations, also model calculations scan be helpful is answering the question about what can be the contribution of local sources to the total PM10 levels. In addition, $NO_2$ calculations are carried out to conclude about the local contribution to the forecasted $NO_2$ levels. In order to do such an analysis we received all information on emissions from local sources (industries) from DCMR, typical for 2004 with a few extrapolations to 2020, using known plans for emission abatements. With our advanced Gaussian dispersion model STACKS we made several runs to calculate the contribution of the large (mostly high) sources, traffic, ships and diffuse sources (mainly being the bulk storage and transit companies[2]). Meteorology was available on a hour-to-hour basis and back ground concentrations as well (MNP, 2006). This database of Generic Concentration in the Netherlands (GCN) supplies the user with detailed spatial and temporal information of all relevant atmospheric pollutants. First we shall give a brief description of the STACKS model is given, followed by the model results and the discussion.

## 4.2 The Model KEMA STACKS+

The model KEMA STACKS is an advanced gaussian model in which scaling parameters are implemented and adjusted to many measurements. Plume rise, transport speed and dispersion parameters are continuous functions of turbulence parameters and, in addition, they are height dependent. Also, special attention has been paid to formulate plume rise in vertically structured atmospheres. The model and its validation has been described in detail by Erbrink (1995).

[2] Open bulk storages contribute tot the PM10 emissions, not to NOx emissions.

The main features of KEMA STACKS are:

- Dispersion of both $\sigma_y$ and $\sigma_z$ using Taylor's statistical theory; the necessary turbulence parameters are functions of boundary-layer scaling parameters, which also applies for the time-scale $T_l$.

- The boundary-layer height during stable and neutral conditions is calculated with calibrated formulae (assuming stationariness) with scaling parameters as input. For the unstable periods during the daytime a growth model is adapted. The model needs vertical temperature profiles as input.

- Plume rise is computed for stable, neutral and unstable with the possibility to pass complexly structured atmospheres. Moreover, stack tip downwash is calculated for situations with relatively lower wind speeds compared to the exit velocity.

- Deposition is calculated using the resistance method for $NO_2$, $SO_2$ and fine particles as well.

- Percentiles are calculated by determining the frequency distribution of concentrations over thousands of hours (brusque force method). This method is the most reliable one available; no assumptions are necessary for the concentration distributions.

KEMA STACKS has been developed in the KEMA laboratories and has been selected as the reference model to up-date the Dutch National Model. In the Netherlands STACKS has the status of accepted and recommended model (by the Dutch EPA, see van Ham, et al., 1998). The following parameters are of special interest:

- Transport speed (wind speed at plume level)
- Height of the boundary (mixing) layer
- Degree of turbulence (Stability) of the atmosphere, input for both $\sigma_y$ and $\sigma_z$

All these model parameters are derived from the determining scaling parameters L (Obukhov length) and surface roughness (u*), see the scheme in figure 9.

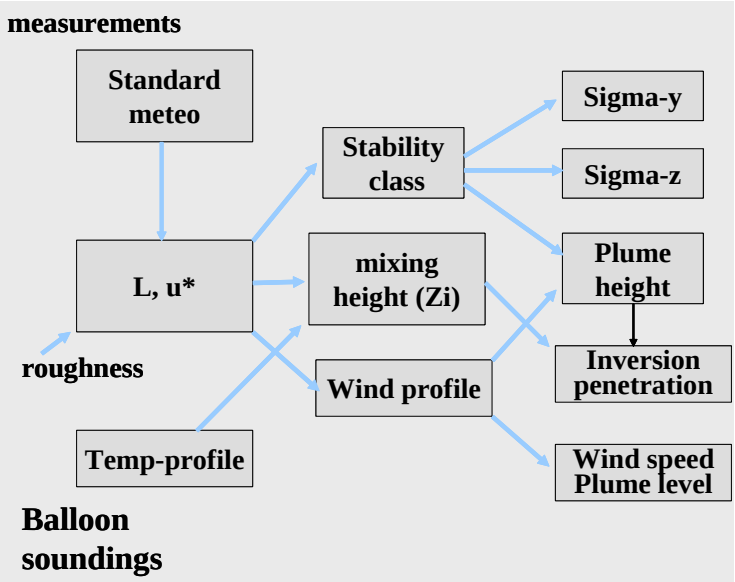


Figure 9. Scheme for pre-processing of the meteorological data to calculate stability and mixing heights, necessary for dispersion calculations.

The gaussian formula can simply be deduced from the mathematical formula for the bi-gaussian expression for dispersion in both the y and z directions for a continuous source:

$$c(x,y,z) = constant \frac{1}{\sqrt{(2\pi)}\,\sigma_y} e^{-\frac{1}{2}\frac{y^2}{\sigma_y^2}} \frac{1}{\sqrt{(2\pi)}\,\sigma_z} e^{-\frac{1}{2}\frac{(h_e-z)^2}{\sigma_z^2}}$$

The height of the plume (stack height plus plume rise) is denoted as $h_e$ (effective stack height).
In STACKS $\sigma_y$ and $\sigma_z$ are continuous functions of the turbulence parameters $\sigma_v$, $\sigma_w$ and $T_l$ instead of using stability classes. A long time series of hourly mean values of all relevant meteorological parameters is used as input for the model. Hence, the model calculates basically hourly averaged concentrations; long-term averages are obtained by summarizing all individual 1-hour concentrations (brusque force method). The dispersion parameters $\sigma_y$ and $\sigma_z$ are calculated using Taylor's theorem for continuous sources:

$$\sigma_{yf,z}^{\ 2} = 2\,\sigma_{vf,w}^{\ 2}\,T_{l_{y,z}}^{\ 2}\,(t/T_{l_{y,z}} + e^{-t/T_{l_{y,z}}} - 1)$$

The turbulence intensities in horizontal and vertical directions and related time scales are described by assuming two dispersive terms (see Erbrink, 1995):

$$\sigma_y^{\ 2} = \sigma_{y_f}^{\ 2} + \sigma_{y_s}^{\ 2}$$

$$\sigma_{y_s}^{\ 2} = (\sigma_{v_s}\,t)^2$$

The first term (with subscript f) reflects the dispersion due to turbulence (mechanical and convective) with time scales up to 10 - 30 minutes; the second term (with subscript s) is the contribution of slower wind direction fluctuations, resulting from medium and large scale motions. The model for point sources is conform the description of the Dutch National Model (van Ham et al, 1998).

To calculate the impact of ships or traffic there is no consensus model available in The Netherlands. An extension of the STACKS model is made for this purpose. For traffic and ships the well-known segmented line model is used. This line source model takes the “normal” Gaussian formula, but with a modified (initial) source term Q1 to Q2:

$$Q_2 = Q_1 \frac{\sigma_y\sqrt{2\pi}\cdot\left(erf\left(\frac{b-y}{\sigma_y\sqrt{2}}\right)+erf\left(\frac{b+y}{\sigma_y\sqrt{2}}\right)\right)}{4b\,\exp\left(\frac{-y^2}{2\sigma_y^2}\right)}$$

In this formula b is the length of the line source, y the distance from plume axis to receptor. For traffic in more or less open area’s this is completed with modules for traffic induced turbulence and effects of noise screens etcetera. For ships the line source model needs no further extensions; source height is assumed to be ground level (1,5m) without ship induced turbulence. For cars the traffic induced turbulence increases the vertical dispersion parameter:

$$\sigma_z^{\ 2} = \sigma_{z0}^{\ 2} + ((1-F_{tr})*(1.5+(P/10)) + F_{Tr}*(4+P/10))^2$$

The traffic induced contribution to the dispersion parameter $\sigma_z$ is taken from Benson et al., 1989 (used in the US Caline4 model) and is a function of the residence time of the exhaust gases in the zone above the road-pavement. P= W/(u*sin(φ)), u being the wind speed, φ the angle between road direction and wind direction, w being the width of the road and $F_{Tr}$ is the fraction of trucks (trucks induce more turbulence than private cars). For street canyons the OSPM submodel (Berkowicz, et al.,

1997, Berkowicz, 2003) is implemented; but for the roads in this study it is not used (there are no relevant street canyons present in this model study).

### 4.3 Model results

The model has been applied to the all main sources in the region. For cars the emissions are taken from the data of the CAR model (see www.infomil.nl), which is widely accepted as a appropriate dataset for car emissions. For ships the emissions are taken from CBS, (2004). The concentrations are calculated for 10 specific points (for locations, see figure 3). The selection of those points is such that all critical locations are represented (traffic, industry, back ground, ships).

Figures 10 and 11 show the results for the yearly averaged concentrations; .

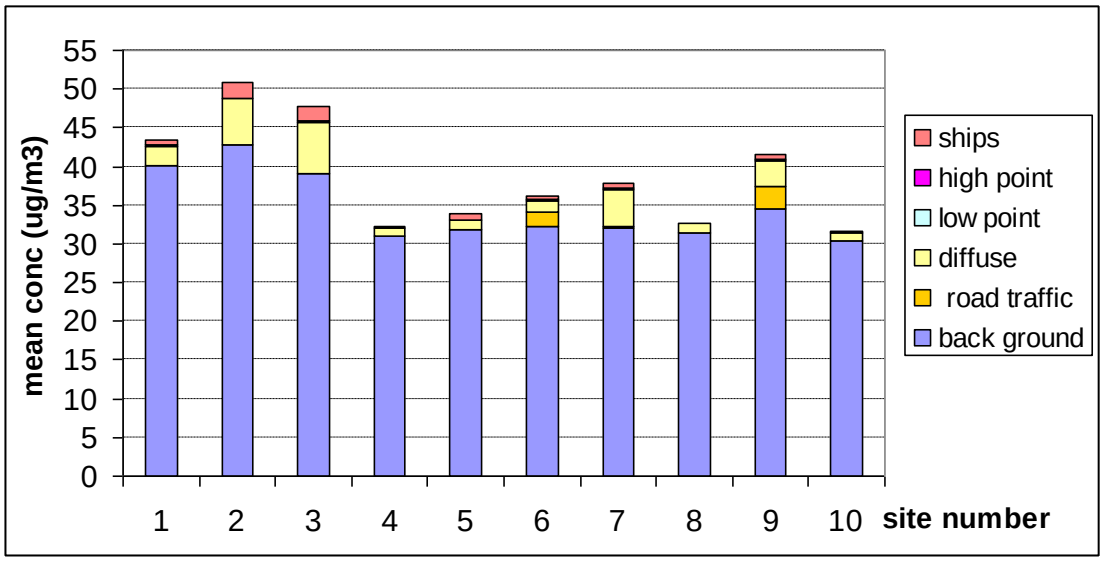


Figure 10: Calculated PM10 contributions at 10 specific points (see figure 3).

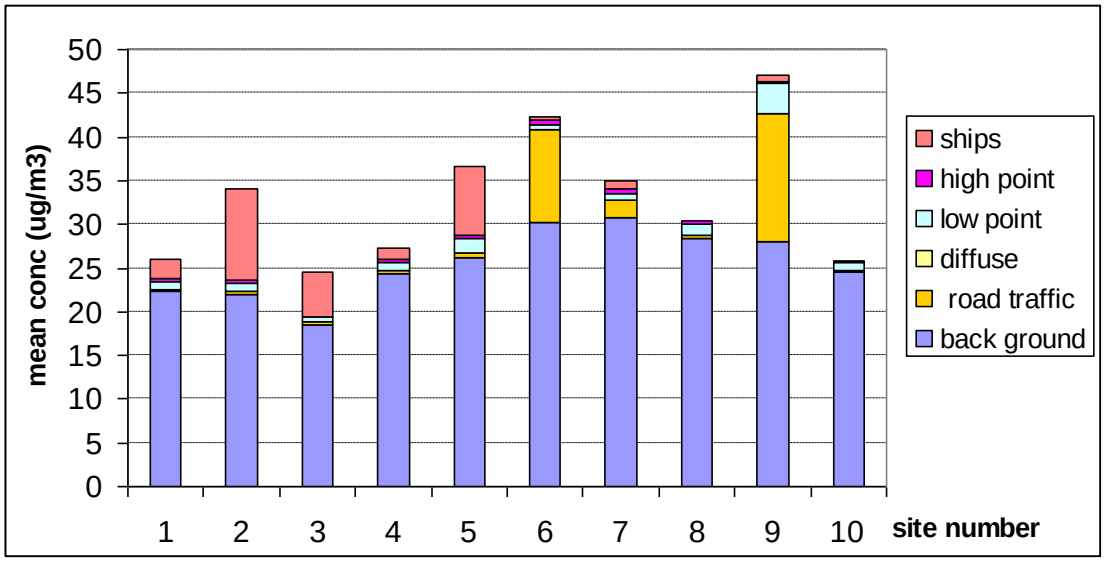


Figure 11: Calculated $NO_2$ contributions at 10 specific points (see figure 3).

As can be concluded from figure 10, neither the low nor the high sources can be recognized in the bars: their contributions to the PM10 levels is near zero at all ten locations. At places close to roads (point 6) or near waterways (point 1,2,3,5 and 9) the contribution is visible, although relatively small. The relative calculated contributions of the diffuse sources (bulk storage locations) is the biggest, although never higher than 10 ug/m3. In section 3 no evidence was found that those sources can contribute more than 1 ug/m3 at distances of 2 to 3 km from the sources. This means that the emissions in the databases might be a obsolete and needs to be updated. Also it is clear from the calculations that the exceeding of the limit values is not caused by increased contributions from local sources, but by an increased inflow of PM10 from other regions. Probably the exceedings are caused by unfavourable meteorological conditions in large parts of Europe. In areas where the pollution is not confined within built-up areas, emission abatement measures on a local scale may not be expected to have considerable effects.

For $NO_2$ the situation is quite different (figure 11): the local contribution can be significant, especially because of intensive traffic and ship movements in the harbour. The back ground concentrations in this area are also dominating the total levels, but the relative contribution of local sources is much more pronounced compared to PM10. Sites 6 and 9 are located about 30 m from the road axes, sites 2, 3 and 5 are located at the border of important waterways. As can be seen, not only traffic but also the ships in this area contribute significantly to the $NO_2$ concentrations. While emissions of road traffic are constantly subject to strict emission control strategies, ships (especially seagoing ships) are hardly restricted in their emissions. On the largest waterways in this areas, over 100.000 ships per year can be counted and thereby causing a significant part of the PM10 and $NO_x$ emissions in this area (see also figure 2).

## 5 CONCLUSIONS

From the case study for the Rijnmond area in The Netherlands we conclude that high PM10 concentrations are in general not caused by local effects. Only open bulk storages, when not equipped with effective emission reduction measurements, PM10 contributions can be significant. It is likely that high PM10 concentrations occur in unfavourable meteorological conditions in large parts of Europe. The reason for these unfavourable conditions should not be found in local meteorological parameters (such as low wind speed, stable conditions) but in large scale weather systems. In the case study a strong correlation was found between high daily PM10 concentrations and the occurrence of specific “Grosswetterlagen”. When high pressure weather systems without rainfall dominate the weather in Europe, large parts of Europe are dusty and suffer from high PM10 levels. Local sources do not contribute significantly to these high PM10 concentrations. Specifically for the Rijnmond area strong evidence was found in two years of measuring that the bulk storage companies in the vicinity of the Maasvlakte contribute less to the local PM10 levels than can be concluded from the present and officially used background concentrations (de so called gcn’s).

$NO_2$ concentrations are much more caused by local emissions. Especially at locations close to roads and waterways the local contribution can approach half of the total concentration. Tall stacks, although their contribution to the tonnages of emissions is the largest of all sources, do not contribute to local concentrations, nor for PM10 neither for $NO_2$. The plumes of such sources are spread over a large area with minor impact.

**Acknowledgments**

We wish to thank the DCMR Environmental Services Rijnmond for making available the measured data from the monitoring network and the emission data for the industries. The work has been partly funded by the Ministry of Transport, Public Works and Water Management.

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