QUANTIFICATION OF DIFFUSE SOURCES OF PM10 IN REGULATORY HOT SPOT REGIONS IN FLANDERS

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INTRODUCTION

Belgium and many other European countries are faced with episodes of high concentrations of PM_{10} and $PM_{2.5}$. These lead to exceedances of the limit values introduced by the air quality directive (1999/30/EC) in order to protect human health. More specifically, exceedances for 2002 were observed in three PM_{10} monitoring stations in Flanders (northern part of Belgium). In 2003 there were more than 10 episodes of high particle concentrations (PM_{10} > 100 µg/m³) resulting in several exceedances of the limit values (6 stations). A comprehensive study was carried out in which we looked at the causes of the exceedances. The objectives were i) to identify and quantify the sources that have contributed to these exceedances; and ii) to propose emission reduction measures and calculate their impact with respect to compliance with the current (2005) and future air quality standards (2010). In order to detect these local sources and propose abatement measures, we followed an integrated multi-disciplinary approach consisting of a detailed analysis of the PM_{10} data series, specific air quality measuring campaigns, air quality modelling and a thorough exploitation of the available expertise in emission reduction measures (including current BAT and BREF studies).

This paper focuses on the methodology that was used to quantify the local emission sources that lead or contribute to exceedances of the PM_{10} limit values. The methodology is further discussed in section 2. In section 3, results for one station (Oostrozebeke) are presented in more detail to illustrate the individual steps in the methodology.

METHODOLOGY

An innovative methodology (called hereafter the "hot-spot" methodology) was designed based on 5 steps that helped to reveal the identity of the sources and finally allowed to quantify these sources.

Analysis using pollution roses

By plotting the observed concentrations in function of the wind direction, a pollution rose is obtained. This was carried out as a first step in the analysis for each of the individual monitoring stations. By subtracting the background concentration, a much sharper pollution rose is obtained, highlighting the directions in which the main local emission sources are to be found. Typical pollution roses for observed and peak (peak = observed – background) PM_{10} concentrations are shown in Figure 1 for the monitoring station Sint-Kruis-Winkel. From this figure it is clear that local sources are probably active in the directions north-northwest, southeast and southwest of this station.

Wind speed analysis

In a next step the variation of the PM_{10} concentration in function of wind speed was assessed. In terms of dispersion, fine particles, - i.e. particles in the accumulation mode with an aerodynamic particle diameter < 2 µm -, behave more like a gas. Their dispersion characteristics can be described by a Gaussian type formula:

$$C = \alpha \cdot Q/U \qquad (1)$$

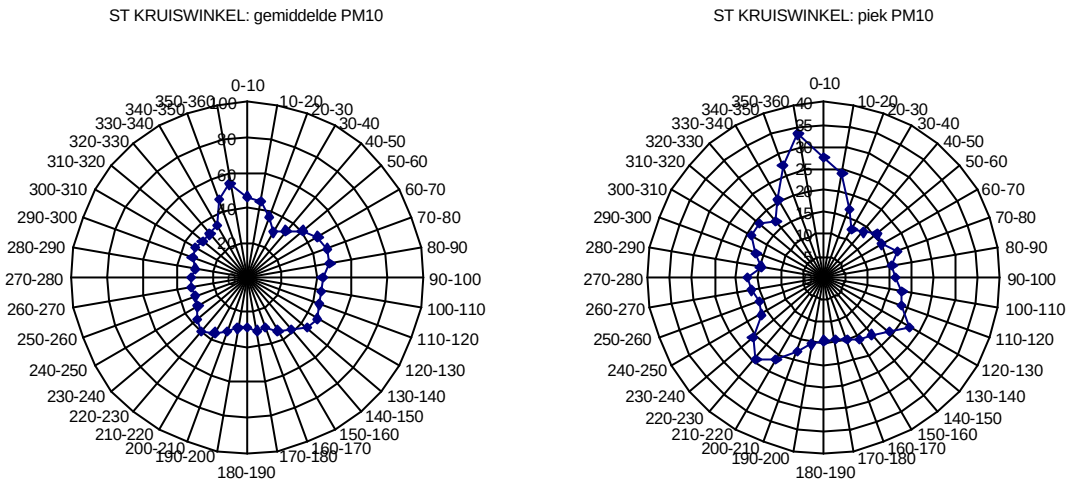

Fig. 1; Pollution roses for the monitoring station Sint-Kruis-Winkel in 2002 as based on observed PM_{10} concentrations (left panel) and on peak PM_{10} concentrations (right panel).

where α contains the dispersion parameters σy and σz and the Gaussian distribution functions in horizontal and vertical direction as a function of the distance from the source. If source term Q in equation (1) is expected to be constant, then concentration C will diminish with increasing wind speed U: $C \approx 1/U$. However when particles are bigger than 2 µm, gravity will become important and the main transport phenomenon of these larger particles will be wind driven (*Venkatram, A.*,1992). In general the source term of larger particles is proportional to the third power of the wind speed (*Greeley R. and J.D. Iversen*, 1985; *Sørensen, M.*, 2004):

$$Q = K \cdot U^3 \qquad (2)$$

Substitution of (2) in (1) yields that the concentration will increase with wind speed: $C \approx U^2$. This distinction in dispersion behaviour is used to analyze the type of local sources. If the wind speed analysis shows a decrease of concentrations with wind speed, it is very likely that the source contains small particles, typically originating from combustion processes, nucleation and coagulation processes (secondary particles), or from condensation (droplets). If, in a certain wind sector, the PM_{10} concentration is increasing with wind speed, it is very likely to find sources of mechanically generated aerosols, like wind blown dust (mineral dust, sand, cement, pollen, road dust, etc.). Figure 2 shows the difference in dispersion behaviour for a stationary source in the case when particles are small (< 2 µm) and behave like a gas and in the case when particles are bigger and the source strength is wind driven.

Analysis of temporal patterns

In a third step a time analysis of the data was made, revealing the temporal patterns of the emission source. These are easily revealed by splitting the data into different time intervals. We used 5 periods, i.e. from 00h00 – 06h30; 06h30 – 12h30; 12h30 – 17h30; 17h30 – 20h30; and 20h30 – 24h00. In this way a distinction could be made between continuous sources and/or sources with a diurnal pattern. The first type of behaviour is typical for (continuous) industrial processes, whereas the latter can be typical for storage and handling activities during daytime. Typical traffic profiles were found as well, with peak values during morning and late afternoon rush hours. Note that the diurnal differences in wind speed can also contribute to differences in emission patterns, especially when the sources are fugitive and their dispersion is wind driven.

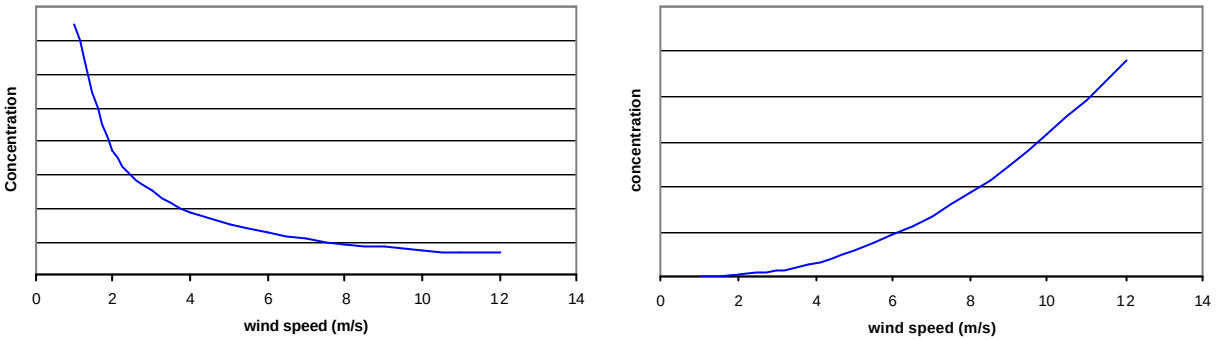

Fig. 2; Dispersion behaviour (concentration versus wind speed) for a stationary source in the case when particles are small (< 2 µm) and behave like a gas (left panel) and in the case when particles are bigger and the source strength is wind driven (right panel).

Local measurement campaigns

In an additional step, and based on the results obtained so far, specific measurement campaigns were organized in the neighbourhood of the monitoring station and the potential sources. Among others, this included mapping of PM_{10}, $PM_{2.5}$ and PM_1 concentrations using mobile dust monitoring and the GRIMM cartography. Gravitair and PARTiSOL equipment was used for reference measurements. Specific component analyses were carried out in order to specify the type of aerosol. The whitening of the aerosols was found to be a crucial element in detecting the type of source (e.g. mineral dust and sand).

Reversed modelling

The last step concerned the quantification of the (fugitive) PM_{10} sources by means of reversed modelling. This technique can be summarized as follows. Under varying meteorological conditions M, an unknown source X among known sources Q_i can be identified by minimizing the difference between the measured concentration and the calculated concentration C:

$$C_{difference} = C_{measured} - C\,(Q_i,X,M) \rightarrow 0 \qquad (3)$$

We used Gaussian dispersion modelling in combination with triangularisation and multiple regression techniques (*Cosemans, G. and J.G. Kretzschmar*, 2004) in order to solve a redundant system for unknown sources X_i under various meteorological conditions. The solution technique can be optimized by adding information obtained from the specific measurement campaigns and by adding knowledge based on user skills and user experience. The latter can be formalised by imposing the following constraints:

- reject negative values as a solution for the source strengths,
- reject or suppress a solution that systematically overestimates the concentration in one receptor and systematically underestimates the concentrations in an other receptor,
- avoid giving too much attention to high peak values,
- observe and learn from the differences between predicted and measured pollution roses,
- use moving averages of the time series to further exploit the meteorological dependence of the emissions sources.

Uncertainties are mainly introduced by the wind field. In most cases we are dealing with a wind field over complex terrain, which means that there is a large variation in wind speed and wind direction.

RESULTS

Figure 3 shows the pollution rose of hourly peak concentrations for 2002 in the monitoring station Oostrozebeke. The concentration peaks are clearly pointing to the south west, i.e. in the direction of a chipboard factory which is located southwest of the monitoring station.

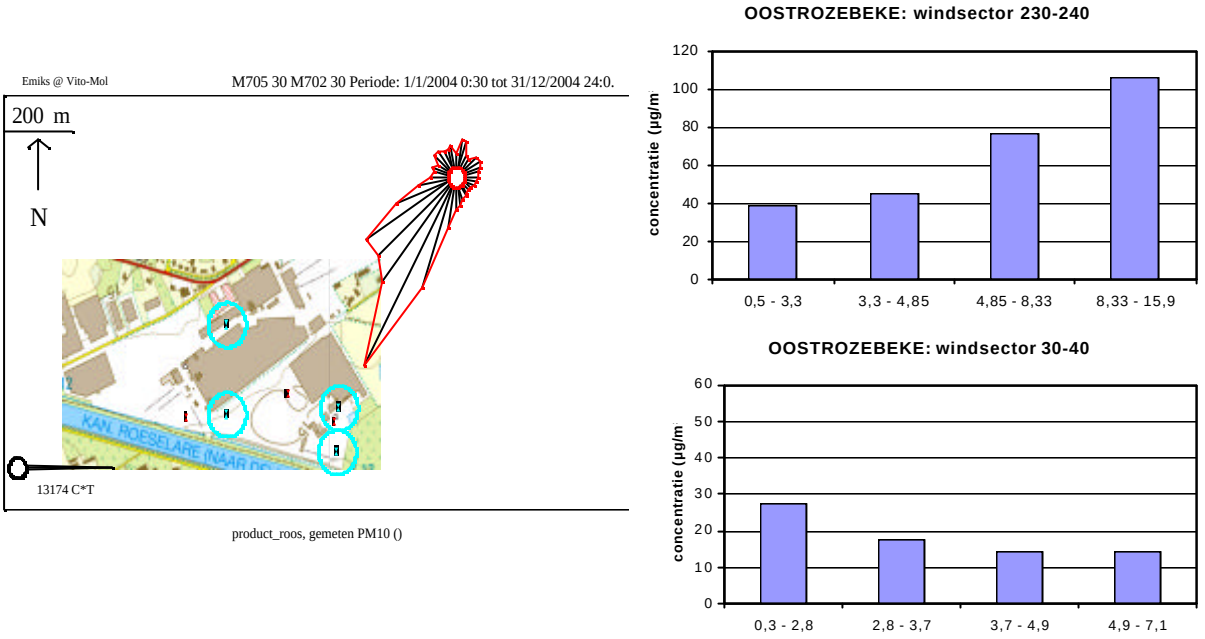

Fig. 3; Left: Pollution rose of hourly peak concentrations for 2002 observed in Oostrozebeke; Right: Observed PM_{10} peak concentrations (µg/m³) in function of wind speed (m/s) in wind sector 230°- 240° (top) and in wind sector 30°-40° (bottom).

When we analyse the concentration in function of the wind speed in this direction (230° - 240°) we clearly observe an increase of the PM_{10} concentration with wind speed as can be observed in Figure 3. In the opposite direction (30° - 40°) we observe that the concentration is decreasing with wind speed, which is typical for a gas like dispersion of very small particles. From the temporal analysis we learned that the source is continuous. The wind speed analysis thus reveals that we are dealing with (a) wind driven or fugitive source(s).

If we calculate the impact of the registered point sources, we find a contribution which is not able to explain the observed concentrations, as shown by the time series of the observed peak concentrations (top panel) and the concentration contributions from the known point sources (central panel) in Figure 4. Furthermore the annual average contribution from the known point sources was found to be 5 µg/m³ in 2002, whereas the observed annual average peak concentration was 13,0 µg/m³. Thus a contribution of 8 µg/m³ is still missing. It is only when we allow for diffuse contributions with a source term that is varying with wind speed, that we can to a large extend simulate correctly the observed concentrations. This is shown in the lower panel in Figure 4. Applying reversed modelling for the diffuse sources resulted in an estimated source term of 92 tons/year. For these fugitive emissions, the contribution to the annual averaged peak concentrations was found to be 7,3 µg/m³. So in total we can explain a contribution of 12,3 µg/m³ of the observed peak concentrations of 13,0 µg/m³. 5 µg/m³ (or 38%) is attributed to the known point sources, 7,3 µg/m³ (or 56%) is attributed to fugitive sources and 0,7 µg/m³ (or 5%) could not be attributed. The contribution of the diffuse sources to the observed annual average PM_{10} concentration was calculated to be 16%. In order to judge the over all result of the regression analysis, the right panel of Figure 4 shows the cumulative frequency distributions of the observed and calculated concentrations.

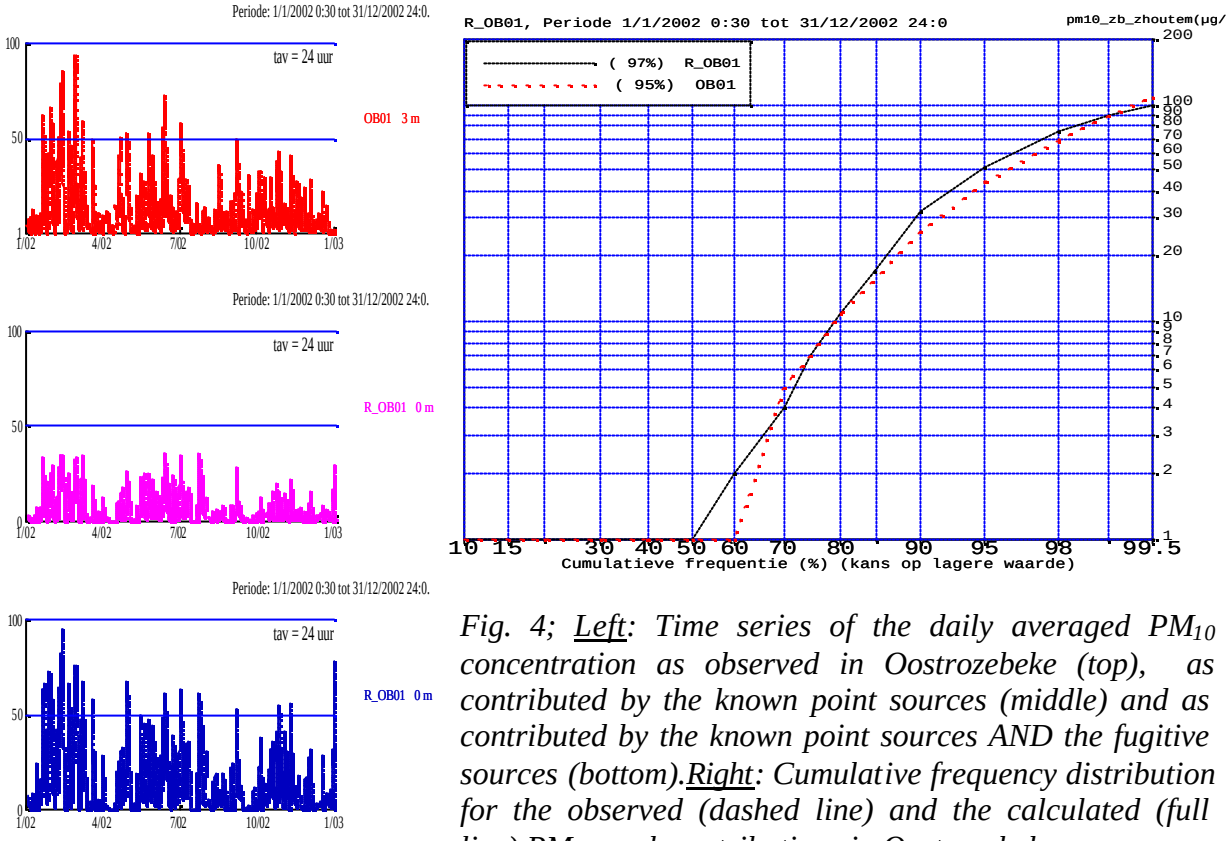

Fig. 4; <u>Left</u>: Time series of the daily averaged PM_{10} concentration as observed in Oostrozebeke (top), as contributed by the known point sources (middle) and as contributed by the known point sources AND the fugitive sources (bottom).<u>Right</u>: Cumulative frequency distribution for the observed (dashed line) and the calculated (full line) PM_{10} peak contributions in Oostrozebeke.

CONCLUSIONS

The study showed that it is possible to quantify PM sources contributing to exceedances of the daily limit value of PM_{10}. Local fugitive emissions on top of high background concentrations were found to be responsible in many cases. Large contributions were detected from local diffuse sources related to storage and handling activities. For the case of Oostrozebeke, the overall contribution of diffuse sources was found to be 16%. It was found that many of the diffuse PM sources are not (yet) registered or reported, because so far they were not known. Additional specific (mobile) measurements in combination with reversed modelling may help to identify these sources.

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