

Particulate matter in Belgium

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1. Introduction

This text deals with particulate matter (PM, mostly PM₁₀) in Belgium. Belgium consists of 3 regions (Brussels, Flanders, and the Walloon region). The PM concentration data presented here originate from the Belgian Interregional Environmental Agency (IRCEL - CELINE; <http://www.irceline.be>), which coordinates the monitoring network activities of the 3 regional agencies. The 3 agencies are “Bruxelles Environnement - Leefmilieu Brussel” (<http://www.ibgebim.be>), the “Vlaamse Milieumaatschappij” (VMM; <http://www.vmm.be>), and the “Ministère de la Région wallonne” (<http://mrw.wallonie.be/dgrne/eev>). The 3 agencies perform the PM measurements on a continuous basis using automated instruments with high time resolution, including ESM monitors based on β -absorption and TEOM monitors with the filter at 50°C. For both instruments correction factors (of around 1.4) are applied to convert the PM₁₀ results to data that are equivalent to the European norm EN12341. For the interpretation and discussion of the PM data, I relied heavily on reports (in Dutch) from the VMM and the Brussels region. Besides the PM data and their discussion, also data of chemical composition of the PM are presented in this text. The latter data were all obtained by my own research group in a number of sampling campaigns, some of which were performed by the VMM. The aerosol collection time in these campaigns was 24 hours; a variety of filter devices and filter types were used, the PM mass was determined by gravimetry (at 50% relative humidity and 20°C) and the samples were analysed (using 3 different techniques) for organic and elemental carbon, major anionic and cationic species, and a suite of elements. The chemical data were used to examine differences in composition between summer and winter and between sites and to assess to which extent aerosol chemical mass closure could be obtained (that is to examine to which extent the sum of the measured components added up to the gravimetric PM mass).

2. PM₁₀ concentrations in Belgium and relation with limit values of the EU directives

The networks have currently 46 stations measuring PM₁₀ (5 in Brussels, 31 in Flanders, and 10 in the Walloon region) and 11 stations measuring PM_{2.5} (4 in Brussels and 7 in Flanders). Using the PM₁₀ data from the various stations, maps of the yearly average PM₁₀ concentration over the entire country are produced using an “Inverse Distance Weighting” (IDW) interpolation method [Ircel, 2005]. The maps for the years 2004 and 2005 are shown in Fig. 1. There is clearly a tendency for lower concentrations in 2005 than in 2004. The figure also shows that, for Flanders, the highest levels are noted in the provinces East and West Flanders and in the west of the province of Antwerp; for the Walloon region the highest levels are observed in its northern part, especially in the northern part of the province of Liege. It should be noted that the maps are influenced by the locations of the stations. For example, for East Flanders all stations are in Ghent and to the north of it, where there is a large industrial area, and some stations in the south-eastern part of West Flanders are impacted by nearby industries.

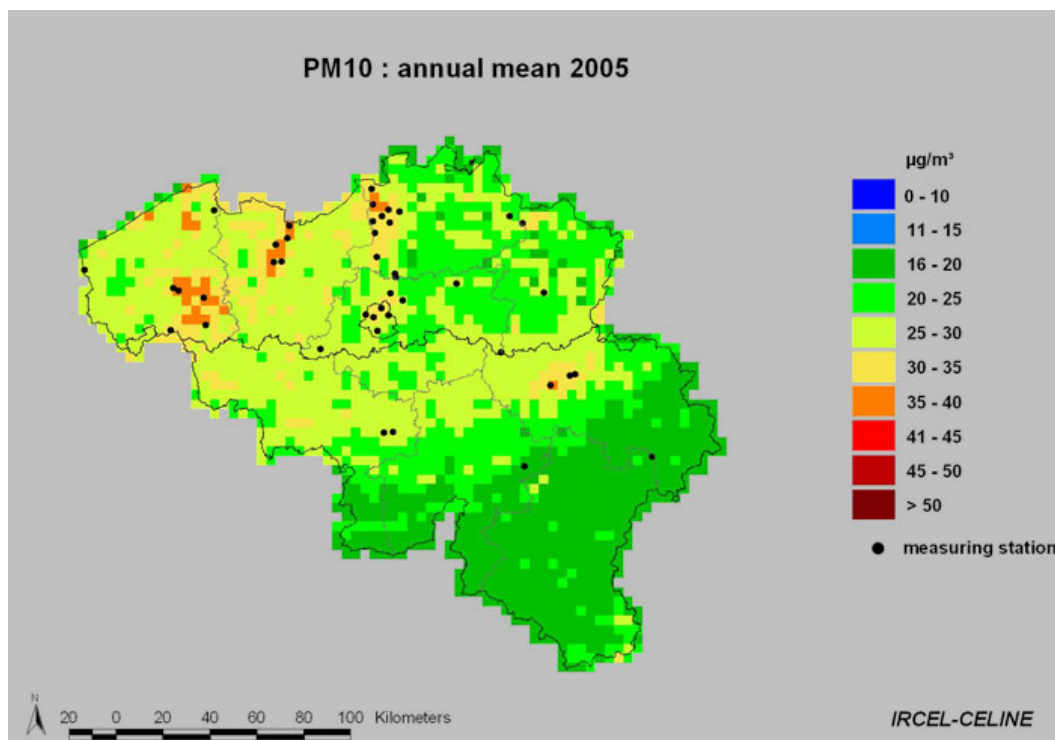
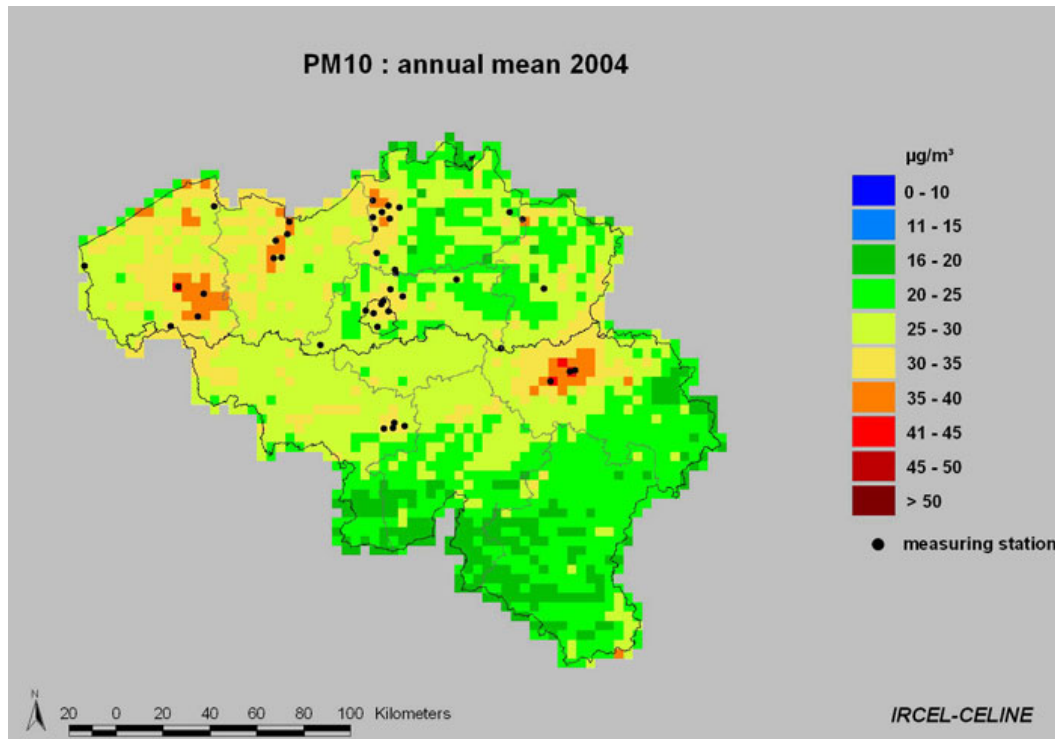


Fig. 1. Annual mean PM10 concentration data for Belgium in the years 2004 and 2005. Flanders is in the north and its provinces are from left to right: West Flanders, East Flanders, Antwerp (top) and Flemish Brabant (around Brussels), and Limburg. The Walloon region is in the south and its provinces are from left to right: Hainaut, Walloon Brabant (top) and Namur (bottom), and Liege (top) and Luxemburg (bottom).

With regard to compliance with the limit values of the EU directive: That there should be no more than 35 exceedances of the daily mean PM₁₀ concentration of 50 µg/m³ was not observed for about half of the stations in 2005, and for 2006 the prospects are even worse (see Fig. 2).

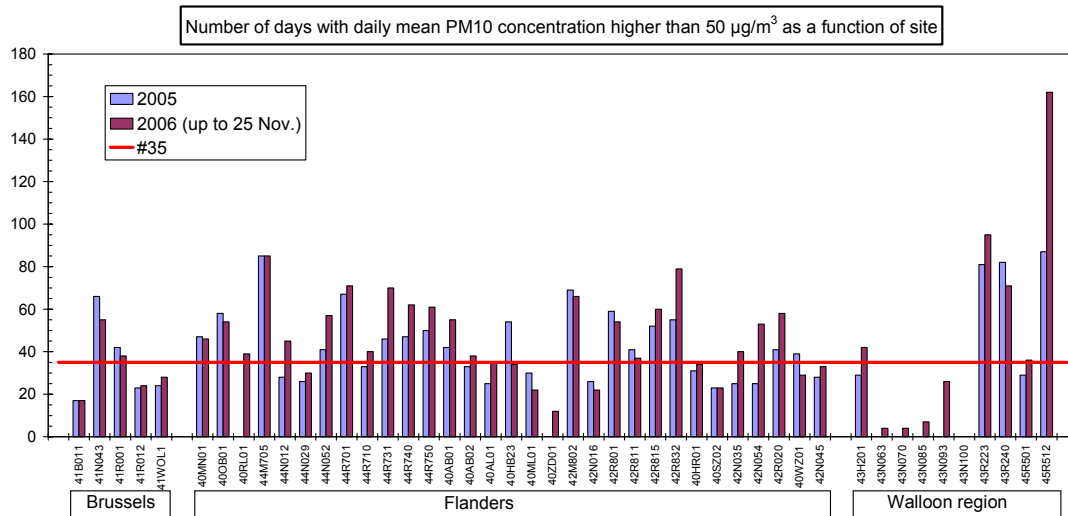


Fig. 2. Compliance with the EU directive of less than 35 days with daily mean PM₁₀ concentration higher than 50 µg/m³ for the stations in the networks.

There are clearly less problems with observing the EU directive of not exceeding the annual mean PM₁₀ concentration of 40 µg/m³. This is illustrated in Fig. 3, which presents the annual mean PM₁₀ data for Flanders as a function of year (since 1996) for different types of sites. As to the 31 individual stations within the Flemish networks, in 2004 there were only exceedances in 2 of them and in 2005 in none of them.

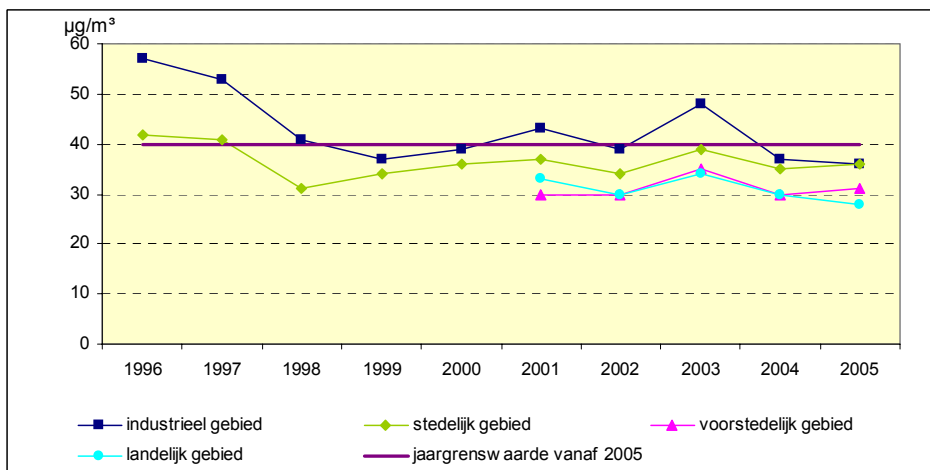


Fig. 3. Annual mean PM₁₀ concentration for different types of stations in the Flemish networks (industrial, urban, urban background, rural); the EU limit of 40 µg/m³ is also indicated.

3. PM concentrations in relation to meteorological conditions

The text in this section 3 (and also in section 4) is entirely based on the Summary of the VMM report “Zwevend stof in Vlaanderen: Periode 2003 en 2004” [2005].

Episodes with elevated PM10 levels (pollution episodes) coincide with mainly dry weather. Important precipitation is only noted at the end of such episode. Pollution episodes are mostly characterised by continental air, lower wind speed or both. There are also frequently temperature inversions during such episodes.

Days with ozone smog coincide with periods of elevated PM levels. The enhanced PM levels may be due to increased photochemical activity. On the other hand, during such days we have mostly meteorological conditions which favour, independently of photochemical activity, enhanced PM levels.

Periods with low PM10 levels coincide mostly with maritime air. Maritime air is mostly accompanied with precipitation and higher daily averaged wind speeds. Temperature inversions are also absent or rare during such periods.

Elevated PM10 levels are mostly noted for low wind speeds. This is even more pronounced for PM2.5. The levels of coarse PM (PM10-2.5) decrease slightly with wind speed or remain rather constant. The mass ratio of PM2.5/PM10 is consequently largest at lower wind speeds. At higher wind speeds a larger fraction of PM10 therefore consists of coarser particles. Apparently, for the coarser aerosol, with increasing wind speed there is not only the dilution effect, but also increased resuspension.

During episodes of PM10 and ozone smog, the wind speed is also lower than average. Possibly, there is an impact from the wind speed also then.

The pollution roses for PM10 and PM2.5 indicated (for most of the stations) elevated levels for winds from the NE to SE. This may be caused by the advection of continental air. Pollution episodes often coincide with winds from that direction. When comparing the pollution roses of 2003 and 2004, there is for nearly all stations in 2003 a NNW component, which is not seen in 2004. During the pollution episode of the first half of August 2003, NNW winds were frequent. Most stations exhibit the highest relative contribution of coarse PM for westerly wind sectors.

4. Variations in PM levels with season, day of the week, and time of day

For PM2.5 lower levels are noted in the summer months than in the winter months. For PM10 the same was true in 2004, but the 2003 summer levels were for many stations higher or equal to the winter 2002-2003 and/or winter 2003-2004 levels. The levels of coarse aerosol show less variation with season.

The mass ratio PM2.5/PM10 is in winter mostly larger than in summer. Consequently, in winter, PM2.5 makes up for a larger fraction of PM10.

During most of the PM10 pollution episodes, the mass ratio PM2.5/PM10 is larger than the average PM2.5/PM10 mass ratio of either winter or summer. The mass ratio PM2.5/PM10 is

mostly also larger during days with elevated ozone levels than during the other days in summer. The wind speed in summer is lower than that in winter.

At nearly all stations lower levels of PM₁₀, PM_{2.5} and coarse PM are observed in the weekend than during workdays. When comparing the 2 weekend days with each other, the lowest levels are noted on Sundays. The Sunday PM₁₀ levels were in 2003 and 2004, on average, 9 and 5 $\mu\text{g}/\text{m}^3$ lower than the average weekday levels. For Saturday, the PM₁₀ levels were, in 2003 and 2004, 6 and 3 $\mu\text{g}/\text{m}^3$ lower than the average weekday levels. In all stations, the contribution from coarse PM to PM₁₀ is larger on workdays than in the weekend and larger on Saturday than on Sunday.

The diurnal pattern for PM₁₀ and PM_{2.5} is mostly as follows: the levels rise in the morning, subsequently they decrease to exhibit a second maximum in the evening. At nearly all stations higher night levels (during 1 or a few hours) are noted in the weekend than during the workdays. The levels of coarse PM are highest during the day and/or in the late evening. The same applies to the relative contribution of coarse PM to PM₁₀.

5. Correlation between PM concentrations of different sites in Flanders in the years 2003 and 2004

Figuur 3.1: Meetplaatsen van PM₁₀ in Vlaanderen (2003-2004)

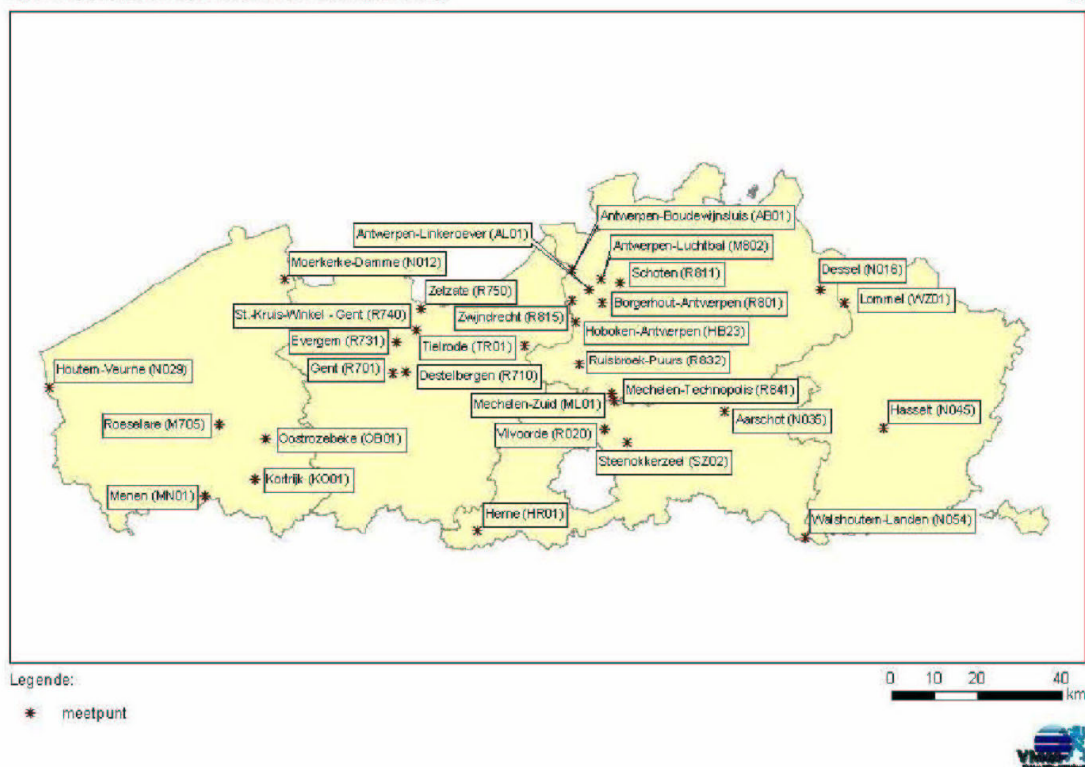


Fig. 4. Location of the PM₁₀ monitoring sites in Flanders.

For both PM₁₀ and PM_{2.5} the data of the different stations in Flanders are fairly well correlated with each other. The correlations are in general better for the year 2003 than for 2004. The correlations between the different PM_{2.5} stations are mostly larger than those between the PM₁₀ stations. In interpreting the latter observation, one has to be cautious,

though, as there are many more PM10 stations than PM2.5 stations. The correlations between the coarse PM data are lower than those between the PM10 or PM2.5 data.

To illustrate the good coherence between the PM data, Table 1 shows the correlation matrix (R^2 values) between the different PM10 sites in Flanders during the year 2004. The location of the sites is shown in Fig. 4.

The total absence of correlation for the site WZ01 (Lommel) in Table 1 is due to soil sanitation works, which converted the surroundings of this site into a bare sand area.

Also the correlations between PM10 and PM2.5 are large. The correlations between PM10 and coarse PM and those between PM2.5 and coarse PM are substantially lower. There is virtually no correlation between PM2.5 and coarse PM.

The correlations between PM10 and PM2.5 are larger in winter than in summer.

6. Sources of PM and contribution from other countries to the PM levels in Flanders

Modelling of the sources of the PM in Flanders is done by the Flemish Institute for Technology (VITO; <http://www.vito.be>) and the VMM, and is incorporated in the MIRA VMM reports (<http://www.milieurapport.be>), e.g., Deutsch et al. [2006].

For a particular site, the PM levels are assumed to be made up by various contributions. For example for a kerbside, these contributions are from (a) local traffic, (b) the city, (c) Flanders, (d) foreign countries (including Wallonia), and (e) other sources. The contributions of (b), (c) and (d) are derived from belEUROS model simulations [Deutsch et al., 2004]. As an example, for a kerbside in Antwerp in 2002, it was derived that the major contribution (43%) came from foreign countries (including Wallonia), followed by Flanders (28%); the contribution from local traffic was 8%, that from the city 6%, and the non-modelled contribution 14% [Deutsch et al., 2006].

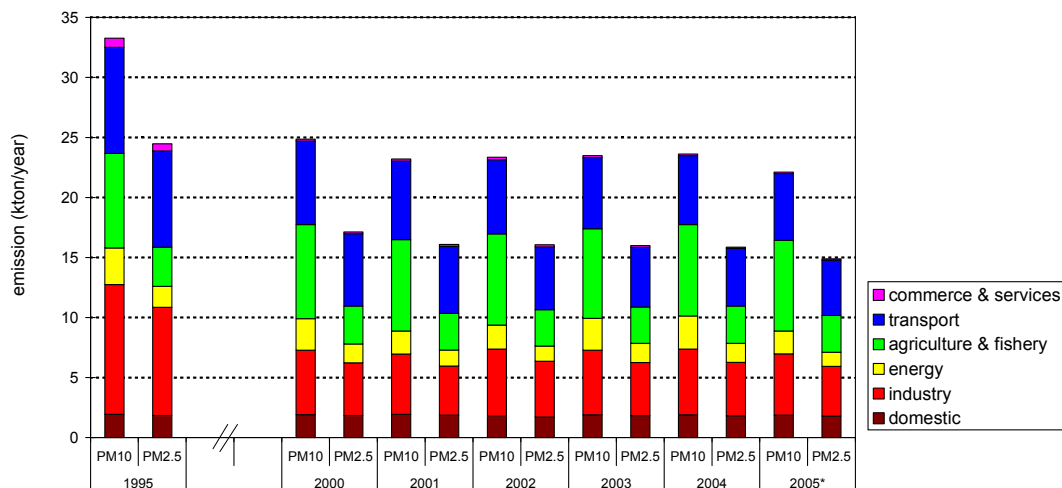


Fig. 5. Emission of primary PM10 and PM2.5 in Flanders. Note that the data for 2005 are preliminary.

With regard to the atmospheric PM, a distinction has to be made between primary PM, which consists of PM that is emitted by the sources in the particulate form, and secondary PM, which is formed within the atmosphere from gaseous precursors (mainly SO₂, NO_x, NH₃ and volatile organic compounds (VOCs)).

The emissions of primary PM from anthropogenic sources in Flanders, as based on emission inventories, are shown in Fig. 5. Since 2001 there is no significant decrease anymore in the emissions. In 2005, agriculture is still the major anthropogenic emitter of primary PM₁₀, followed by transport and industry, whereas for primary PM_{2.5} the major anthropogenic emitter is transport, followed by industry and agriculture.

Using the beEUROS model mentioned above, contributions were not only derived for individual sites, but also for Flanders as a whole. The results obtained for the year 2002 are shown in Fig. 6.

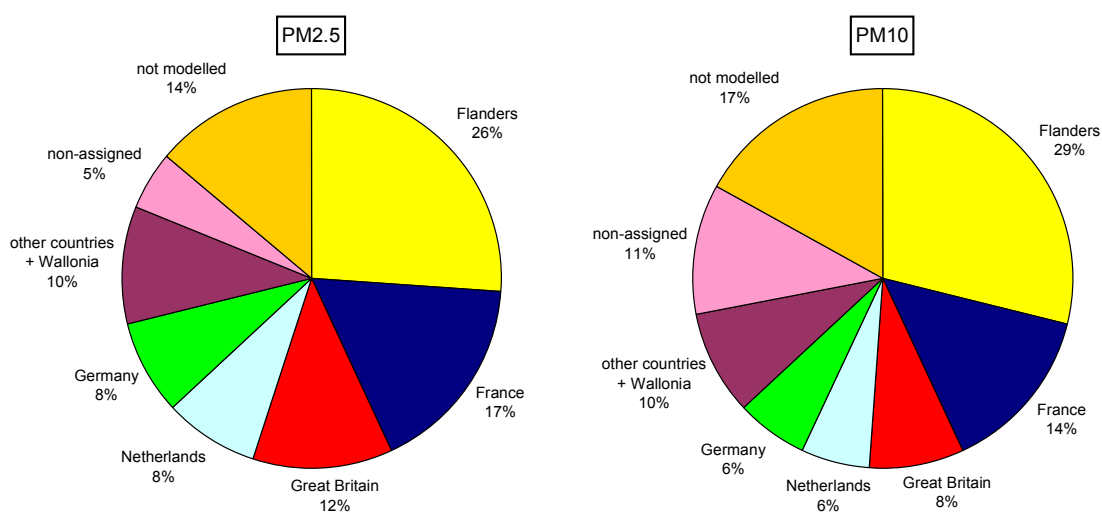


Fig. 6. Modelled contributions to the PM_{2.5} and PM₁₀ levels for Flanders in 2002.

The large contributions from France (17% and 14% for PM_{2.5} and PM₁₀, respectively) are due to the prevailing SW winds and the relatively high emissions in the north of France, close to the border with Belgium. Also the contributions from Great Britain are to a large extent due to the prevailing winds.

However, transboundary transport of PM is not only a matter of transport from other countries to Flanders and Belgium, but also of the latter to other countries. Using beEUROS with emission and meteorological data of 2002, it was estimated what the decrease in annual mean PM_{2.5} concentration would be if there would be no Flemish emissions. The results of this modelling exercise are shown in Fig. 7. For the neighbouring countries or regions, i.e., the Netherlands, Wallonia and the western part of Germany, shutting off the Flemish emissions would result in a decrease in the annual mean PM_{2.5} level of typically between 0.76 and 2 µg/m³. For far away countries, such as the Baltic states and the southern parts of the Scandinavian countries, the Flemish emissions still make a difference of 0.11 to 0.25 µg/m³.

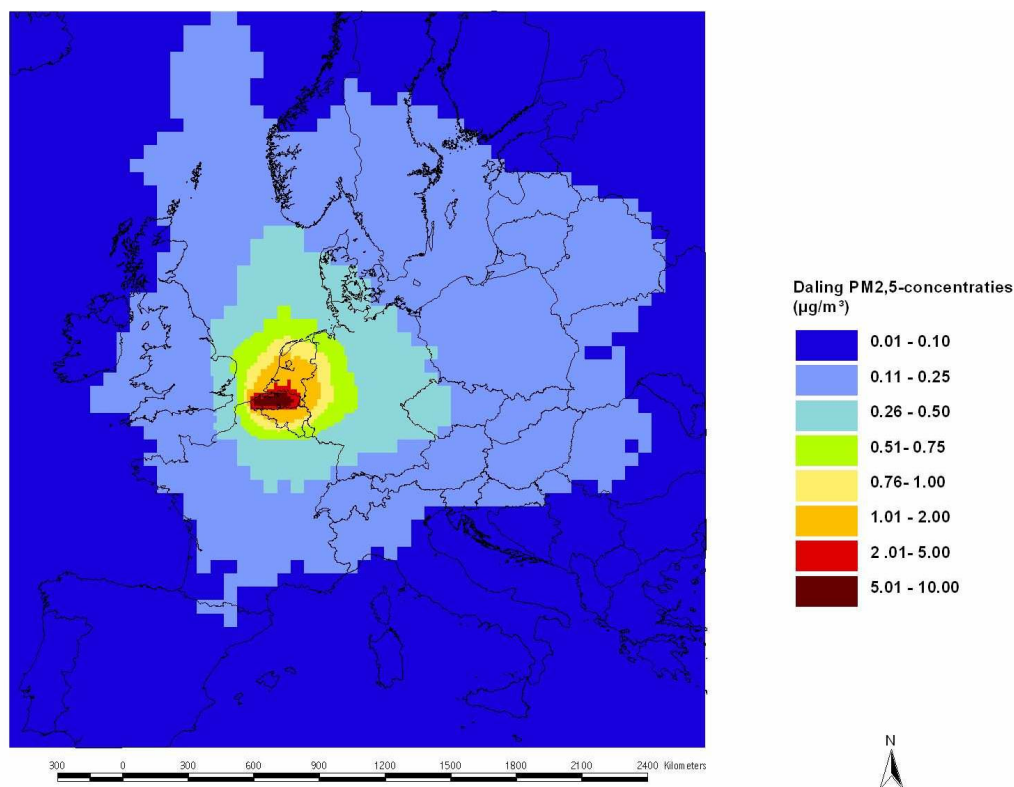


Fig. 7. Modelled decrease of the PM_{2.5} concentrations in Flanders and Europe when the emissions in Flanders are shut off (calculations for the year 2002).

7. Chemical composition and mass closure for PM

Determination of the chemical composition of the PM gives clues on its sources and allows one to assess the relative importance of primary and secondary PM. If the chemical analysis includes a measurement of the various major PM components (or of markers for these components), the data can also be used for aerosol chemical mass closure (that is to examine to which extent the sum of the measured components adds up to the gravimetric PM mass). Within the 3 agencies that take care of monitoring networks in Belgium, chemical analysis of the PM on a routine basis is only done for selected compounds, such as heavy metals and polyaromatic hydrocarbons (PAHs). More detailed analyses are performed by the agencies within special studies. The VMM does the latter typically with cooperation of the VITO and Flemish universities, including the University of Antwerp and Ghent University. On the other hand, within the latter two universities, research groups are involved in studies on atmospheric aerosols and their composition, and these groups have their own research projects (independently of the 3 agencies).

Here, I present recent chemical composition and mass closure data for PM that were obtained by my own research group, whereby some of the projects were done in co-operation with others or were even commissioned by the VMM. The PM samples for this work were obtained in campaigns of typically one month duration and in different seasons (typically summer and winter). The sampling sites for the campaigns were the Institute for Nuclear Sciences (INW) in Ghent, the Royal Meteorological Institute (RMI) of Belgium in Uccle (Brussels), and two sites within the city of Antwerp. The INW and RMI sites can be

classified as urban background sites, and the two sites in Antwerp as kerbsides. For the INW and RMI sites, the PM samplers (and filters) were provided by my research group and the gravimetric PM measurements and chemical analyses were also done by my group [Wang et al., 2005; Maenhaut et al., 2006b]. For the campaigns in Antwerp, which was commissioned by the VMM, the PM samplers (and filters) were provided by VITO, the gravimetric PM measurements were also done by VITO, and the chemical composition measurements by my group [Maenhaut et al., 2006a]. From the chemical composition data, the concentrations of the following 8 aerosol types (components) were derived (on a individual sample basis): (1) organic matter [OM; estimated as 1.4 times particulate organic carbon (OC)], (2) elemental carbon (EC), (3) ammonium, (4) nitrate, (5) non-sea-salt sulphate, (6) sea salt, (7) crustal matter, and (8) other non-sea-salt/non-crustal elements [Maenhaut et al., 2002]. Note that only one of these 8 components (i.e., sea salt) is purely natural; EC, ammonium, nitrate, non-sea-salt sulphate and other elements are in Flanders essentially from anthropogenic origin, and OM and crustal matter are probably also mostly due to anthropogenic sources (For the last component there are contributions from road dust and agriculture). The concentrations of the 8 components were then averaged per campaign, the averages were summed up and compared with the average gravimetric PM of the campaign, and the difference is then referred to as unexplained mass. The results for PM10 in 6 sampling campaigns are shown in Fig. 8.

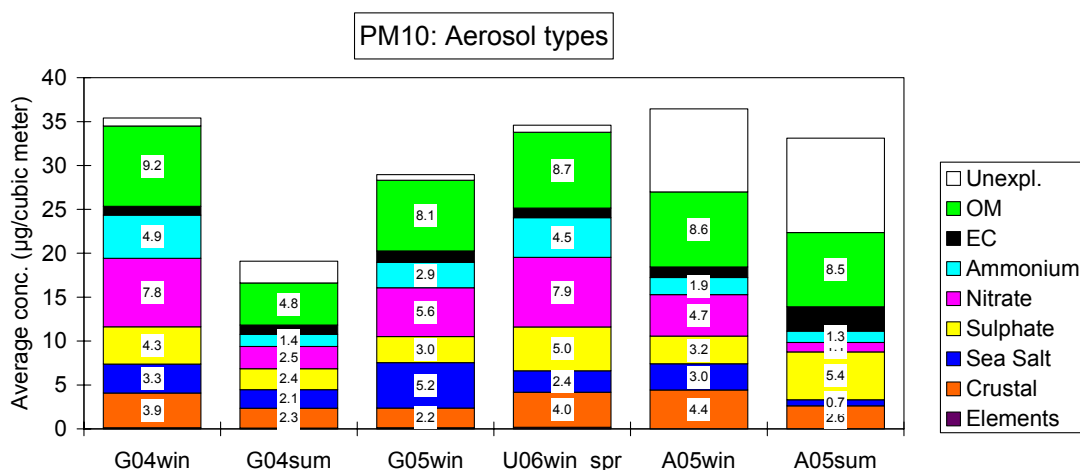


Fig. 8. Average concentrations of 8 aerosol types in PM10 and of the unexplained gravimetric PM10 mass during 6 sampling campaigns [G04win, G04sum, and G05win stand for Ghent 2004 winter, Ghent 2004 summer, and Ghent 2005 winter, resp., and the samplings took place at INW; U06win_spr stands for Uccle 2006 winter-spring, with samplings at RMI; A05win stands for Antwerp 2005 winter (with samplings at the Mechelse steenweg) and A05sum for Antwerp 2005 summer (with samplings in Borgerhout)].

Noteworthy is that the unexplained PM10 mass is much larger for the two campaigns in Antwerp than in the other 4 campaigns. There were difficulties with the gravimetric PM determinations for these two campaigns. Also the gravimetric PM data were derived from low-volume quartz fibre filters for these 2 campaigns and from low-volume Pall Gelman Teflo and/or Nuclepore polycarbonate filters in the other 4 campaigns and substantially higher PM data for low-volume quartz fibre filters than for the other filter types have been observed before [Maenhaut et al., 2002; Hitzenberger et al., 2004]. It is thought that there may be a substantial positive artefact for the PM data of the low low-volume quartz fibre filters, and therefore the PM data from these filters are considered less reliable.

In agreement with what was indicated in section 4 above, there is a tendency for higher PM₁₀ levels in winter than in summer. Particularly for nitrate, much higher concentrations are observed in winter than in summer. This finding is in agreement with the findings of the European Aerosol Phenomenology study [Putaud et al., 2004]. The low nitrate values in summer are due to the fact that the volatile ammonium nitrate is rather in the gas phase than in the particulate phase within the atmosphere during this season (Volatilisation of nitrate from the aerosol collected on the filter may also play a role).

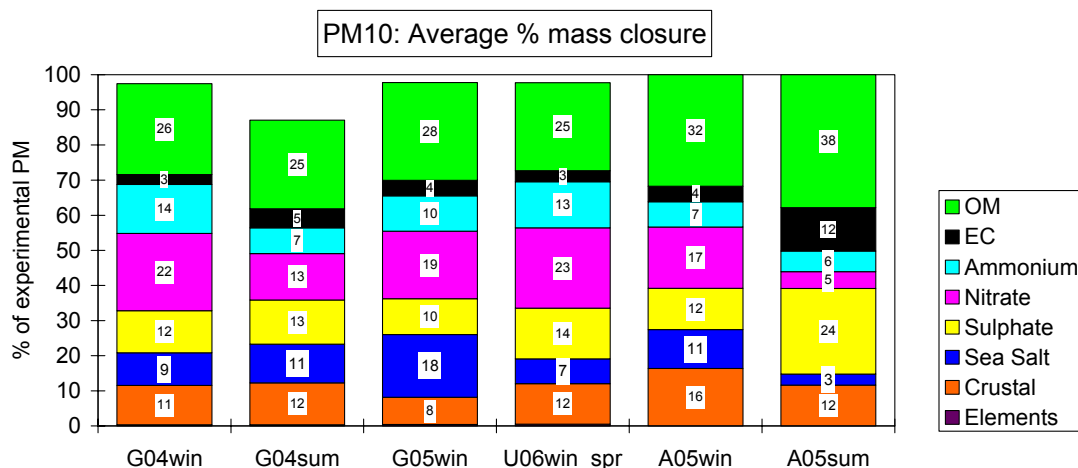


Fig. 9. Average percentages of 8 aerosol types as % of the mean gravimetric PM₁₀ mass (and for the two sites in Antwerp as % of the sum of the components); for additional explanation, see caption of Fig. 8.

In order better examine the differences in PM₁₀ aerosol composition between the various sites and seasons, the data from Figure 8 were expressed as percentage of the gravimetric PM₁₀ mass (and for the two campaigns in Antwerp as percentage of the sum of the components). The results of this calculation are shown in Fig. 9. Even expressed as % instead of as absolute concentrations, there are clearly differences for nitrate between winter and summer, with lower percentages in summer. It further appears that the average PM₁₀ aerosol composition is fairly similar for the 4 winter campaigns. The most noteworthy difference is observed for the sea-salt component, with a percentage as high as 18% for Ghent 2005 winter. In Fig. 8, it can be seen that also the absolute concentration of the sea-salt component was high during this campaign, and that, on the other hand, the average PM₁₀ level was on the low side. Examination of the data for the individual samples of this campaign indicated that the percentage contribution of sea salt to the PM₁₀ mass was especially high for samples with low PM₁₀ mass concentration and was low for samples with high PM₁₀ mass concentration [Viana et al., 2006]. The individual data from the various campaigns clearly indicated that sea salt cannot really be invoked as cause for exceedance of the daily PM₁₀ EU limit value of 50 $\mu\text{g}/\text{m}^3$. When such exceedances occur, both the absolute concentrations of sea salt and the percentages of sea salt in the PM₁₀ aerosol are quite low.

Fig. 9 further indicates that there is a large contribution of secondary aerosol to the PM₁₀ mass. Ammonium, nitrate and non-sea-salt sulphate are all secondary inorganic aerosol (SIA) components. In winter, the sum of these 3 components accounts, on average, for 40% or more of the PM₁₀ mass. For PM_{2.5} even larger percentage contributions of SIA were observed. Also a substantial fraction of the OM may consist of secondary aerosol and thus be secondary

organic aerosol (SOA), especially in summer. The total contribution of secondary aerosol is likely over 50% in all seasons.

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