JOAQUIN - JOINT AIR QUALITY INITIATIVE

INTERRIM REPORT

SOURCE APPORTIONMENT OF POLLUTANTS OVER THE JOAQUIN DOMAIN USING MODEL SIMULATIONS

September 2015

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1 Introduction

Figure 1 The LOTOS-EUROS domain as used in this report. The JOAQUIN domain is the red box over Northwest Europe.

In the framework of the JOAQUIN project, model simulations were performed for Northwest Europe (see red rectangle in Figure 1) at a resolution of 7 kilometres with four different models: AURORA, BEL-EUROS, CHIMERE, and LOTOS-EUROS. The focus of these calculations was on determining the concentration distribution of the air pollutants Elemental Carbon (EC), NO_2, PM_{10} and $PM_{2.5}$ for the years 2009-2011. An extensive model inter-comparison including measurements was performed by Adriaenssens et al. (2015).

Apart for the production of concentration maps, calculations were also performed in order to determine a blame matrix, based on the modelled results. In a blame matrix, one can see how much countries are contributing to concentrations of air pollutants in a specified region. In the JOAQUIN project, two of the four models were used in order to make a blame matrix for the year 2009 using the methods of source apportionment. The BEL-EUROS model was used for this, which is reported in Deutsch et al. (2014), and in this report we present results from the source apportionment calculations with the LOTOS-EUROS model. At the end of the report we make a concise comparison between the outcomes of the LOTOS-EUROS and the BEL-EUROS model.

The 3D chemistry transport model LOTOS-EUROS (for details see Schaap et al., 2008) is used in this study to calculate a blame matrix for the Northwest European domain, as defined within the JOAQUIN project (see Figure 1). A model simulation was performed for this with LOTOS-EUROS version 1.9 for the year 2009.

The LOTOS-EUROS model grid is longitude-latitude with a domain bound at 35^0 and 70^0 North and 15^0 West, 35^0 East, with a resolution of 0.50^0 longitude × 0.25^0 latitude. Subsequently, a zoom run is performed on the JOAQUIN domain (see Figure 1), at a resolution of 0.125^o longitude × 0.0625^o latitude, approximately 7 × 7 km^2.

The vertical direction in LOTOS-EUROS extends up to 3.5 km above sea level. The first layer is a surface layer of 25 meter height, the second layer is a dynamic layer being the mixing layer. The 3^{rd} and 4^{th} layer of the model are reservoir layers of equal depth lying above the mixing layer.

Detailed emissions of 2009 have been supplied by TNO for the JOAQUIN project (Denier van der Gon et al., 2014). The meteorological parameters are obtained from the ECMWF model. Finally, the source apportionment module in LOTOS-EUROS was used to calculate the blame matrix of the three components considered here, i.e. EC, $PM_{2.5}$ and PM_{10}.

2 Method of constructing a blame matrix

In the LOTOS-EUROS model, a source apportionment module calculates the contribution from labelled emission categories in order to track the origin of air pollutants (Kranenburg et al., 2012). Therefore, apart from concentrations which are calculated for each grid cell, additionally, also fractions of these air pollutants are calculated, which indicate which fraction of the concentration in a grid cell comes from a labelled source. One of the applications of source apportionment in the LOTOS-EUROS model was to determine the origin of ambient particulate matter concentrations in the Netherlands (Hendriks et al. 2013).

The source apportionment module in LOTOS-EUROS has been extensively tested and compared with more traditional methods of source apportionment, in which the sources of interest are reduced by a small percentage. The current method is expected to be more reliable since no differences are occurring due to reduced emissions which obviously influence the chemistry. Indeed, test runs performed with the source apportionment module in LOTOS-EUROS show that a labelling technique like used in the LOTOS-EUROS model yields more reliable results than source apportionment studies based on (reduction) scenario calculations, especially for reactive species.

In this study we label the following regions of the JOAQUIN domain:

1. Brussels;
2. Flanders;
3. Wallonia;
4. The Netherlands;
5. Paris;
6. France;
7. Germany;
8. London;
9. England;
10. Luxembourg;
11. Seas (North Sea and Atlantic);
12. Natural sources (for example sea salt);
13. Rest term.

It should be noted that for France, Germany and England, only sources from the regions inside the JOAQUIN domain have been labelled. Furthermore, we also labelled a rest-term, which includes beside non-labelled sources from countries outside the JOAQUIN domain, natural sources from outside of the boundary, initial conditions and from aloft.

3 Results of the blame matrix for elemental carbon (EC)

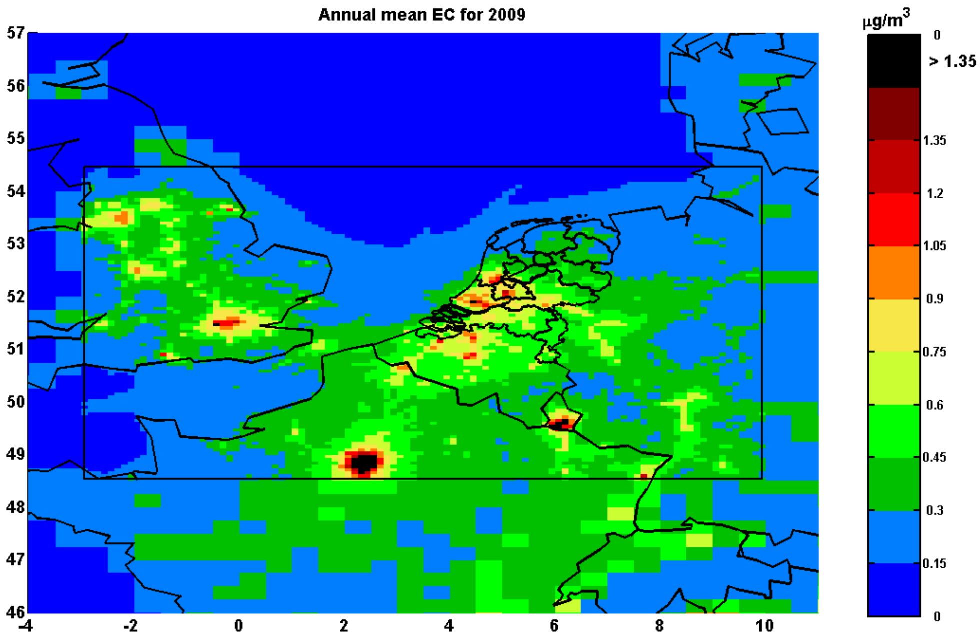

Figure 2 The EC concentration over the JOAQUIN domain for the year 2009.

Figure 2 shows the annual mean value for concentrations of EC (in $PM_{2.5}$) for the year 2009 as calculated with the LOTOS-EUROS model. One can see the cities as hot spots in this map, because most EC emissions originate from traffic. Highways are not observed in the map, due to the resolution of 7 km of the model, which is too coarse to see these local emission patterns.

The performed source apportionment calculations yield the fractions of the EC concentration of the 13 considered labels. As an example Figure 3 (top panel) shows the fraction of EC over the Netherlands, due to Dutch emissions of this component. The maximum value of this fraction is up to 0.8 in big cities like Amsterdam, Utrecht and Rotterdam. Outside of the borders of the Netherlands, the fraction decreases substantially, such that Dutch EC emissions typically contribute 5-10% to Belgium. Figure 3 (bottom panel) shows the blame matrix for the whole of the Netherlands, one can see that 54% of the EC concentrations in the Netherlands is due to Dutch emissions. This percentage is lower than the ~80% for cities, because an average over the whole of the Netherlands is seen in the blame matrix.

The example as given in the bottom panel of Figure 3, can be extended for all regions considered in the performed simulation, such that one can construct a blame matrix for the JOAQUIN domain with the 13 labels as mentioned in the previous section. Figure 4 shows the blame matrix for EC. Notice that big cities like London and Paris with clear hot spots in the EC concentration map (Figure 2) contribute up to 50% to their local EC concentrations. Furthermore, notice that ship traffic also contributes significantly to countries like the Netherlands, Belgium, and the United Kingdom. Finally the rest term for EC is in the order of 10-20% for most countries. This rather small percentage is due to the fact that EC emissions are quite local, as compared to for example particulate matter and secondary inorganic aerosols like ammonium nitrate and ammonium sulphate.

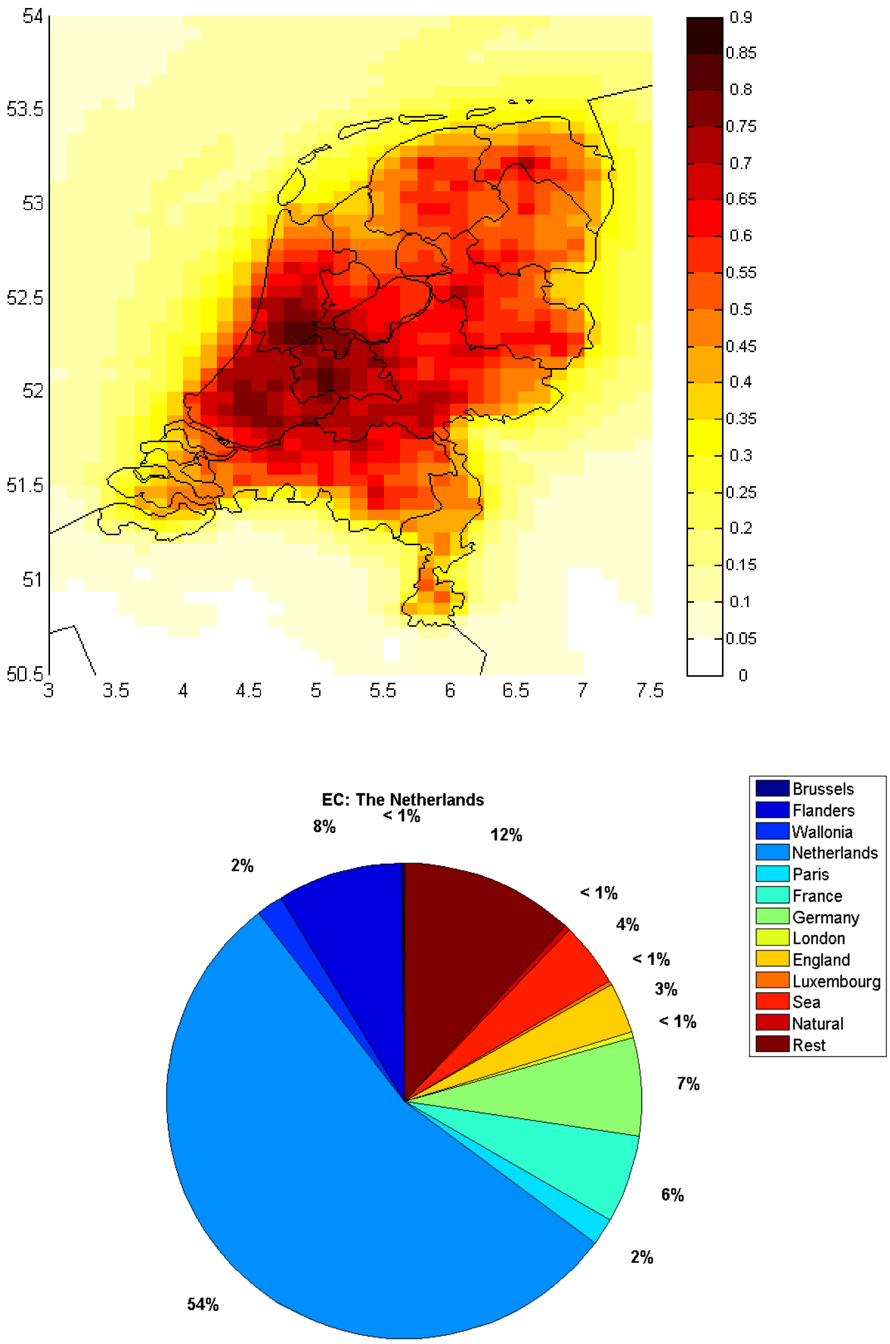

Figure 3 The fraction of EC concentrations over the Netherlands, due to Dutch emissions of EC (top panel) and the blame matrix for the Netherlands (bottom panel).

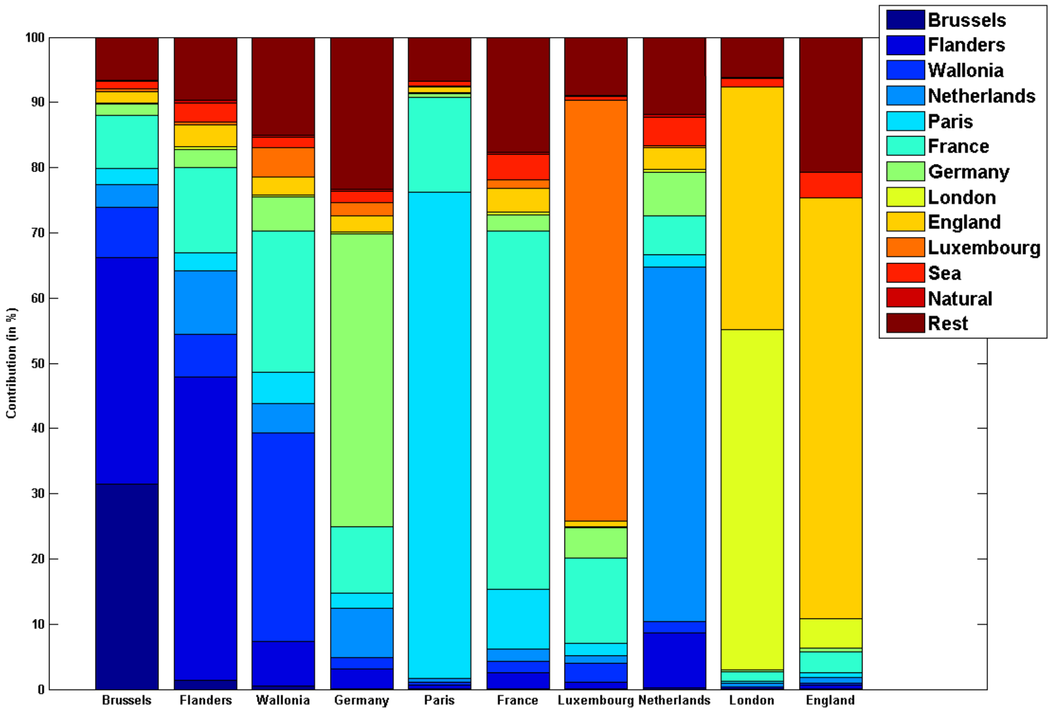

Figure 4 The blame matrix for the component EC as calculated with the LOTOS-EUROS model.

4 Results of the blame matrix for $PM_{2.5}$

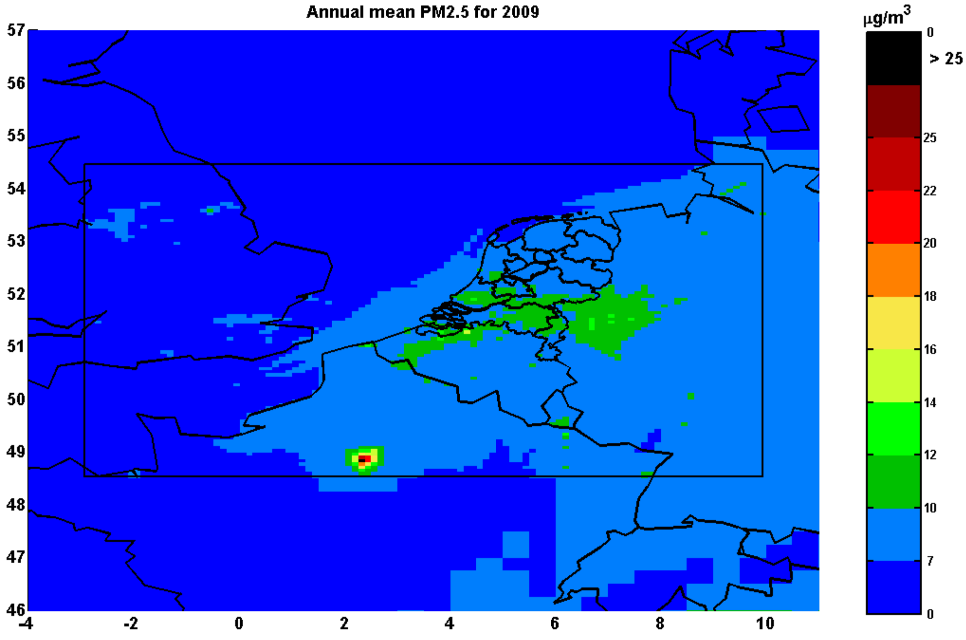

Figure 5 The concentration of $PM_{2.5}$ over the JOAQUIN domain for the year 2009.

Figure 5 shows the annual mean value for concentrations of $PM_{2.5}$ for the year 2009 as calculated with the LOTOS-EUROS model. One can see the areas with cities slightly enhanced in this map, with Paris being the hotspot of the $PM_{2.5}$ map. The latter is due to the emission which goes into the model for this particular city (see Adriaenssens et al., 2015). The resolution of 7x7 km^2 of the model, seems a good scale to represent this large-scale component pollutant. Therefore, contrary to EC, $PM_{2.5}$ is a more large-scale pollutant, which can also clearly be observed in the source apportionment analysis: compared to EC, each labelled country is contributing a smaller percentage of $PM_{2.5}$ to its own area. This can be seen from the source apportionment of the Netherlands for respectively EC and $PM_{2.5}$ (see Figures 3 and 6), and for the whole JOAQUIN domain (Figures 4 and 7).

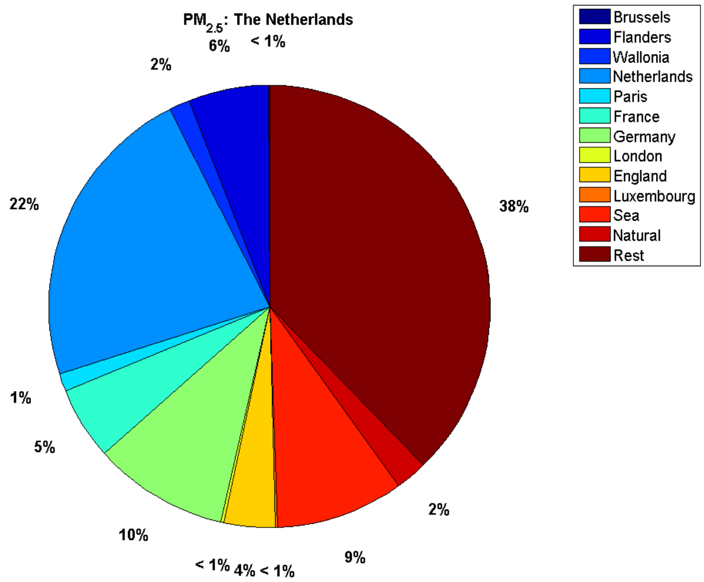

Figure 6 Blame matrix for $PM_{2.5}$ for the Netherlands.

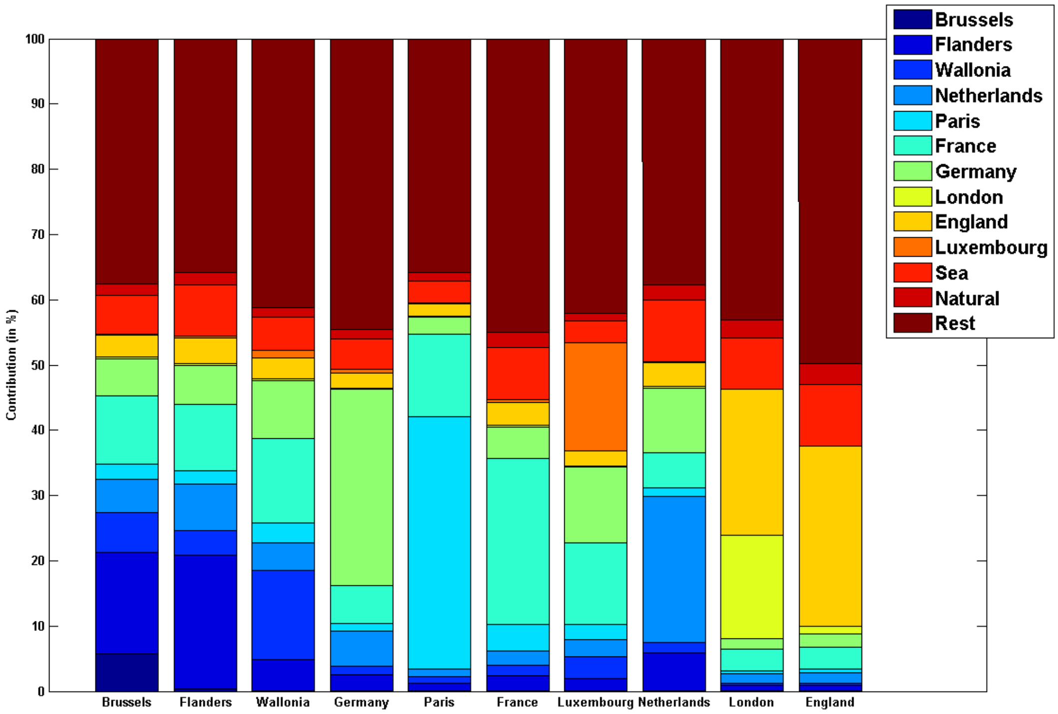

Figure 7 The blame matrix for the component $PM_{2.5}$ as calculated with the LOTOS-EUROS model.

Figure 7 thus shows the blame matrix for $PM_{2.5}$ for the JOAQUIN domain. If one compares this matrix with the one of EC, one can see the contribution of each country to its own concentration is smaller in the case of $PM_{2.5}$, reflecting its large-scale distribution over the surface area. Also, one can note that for a city like Paris, its contribution to $PM_{2.5}$ is still high as compared to all the other areas. This is because Paris is a relative large area, which also contains most of the emissions in the (larger) areas around the Paris region itself. Finally, the rest term for $PM_{2.5}$ is in the order of 40% for most countries. This rather large percentage is due to the fact that $PM_{2.5}$ concentrations are determined by large scale transport pollutants like the secondary inorganic aerosols nitrate, sulphate and ammonium.

5 Results of the blame matrix for PM_{10}

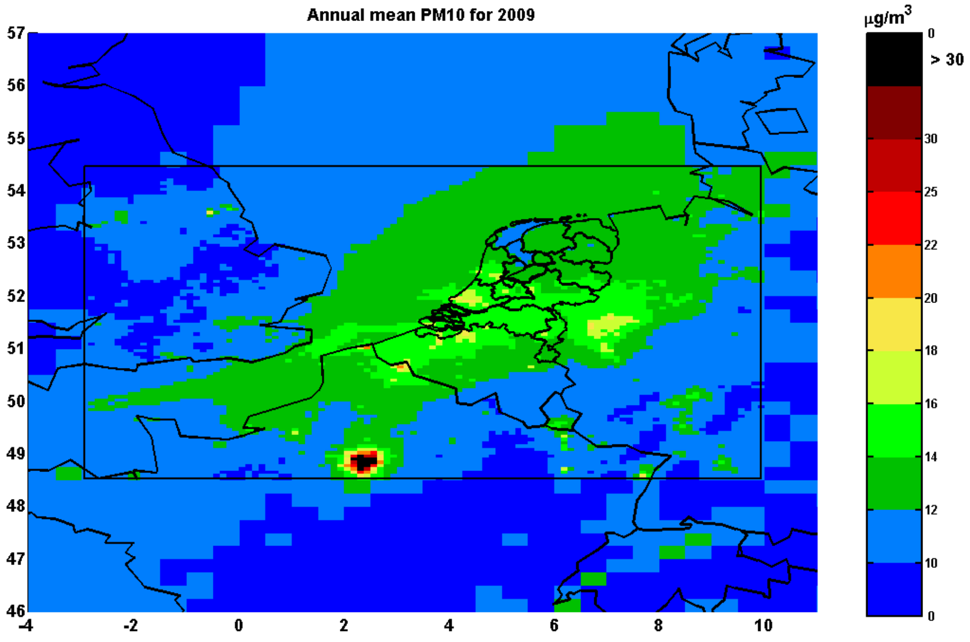

Figure 8 The concentration of PM_{10} over the JOAQUIN domain for the year 2009.

Figure 8 shows the annual mean value for concentrations of PM_{10} for the year 2009 as calculated with the LOTOS-EUROS model. One can see the areas with cities enhanced in this map, with Paris being the hotspot of the PM_{10} map. This is due to the emission which goes into the model for this particular city (see Adriaenssens et al., 2015). The resolution of 7x7 km^2 of the model, like $PM_{2.5}$, also seems a good scale to represent this large-scale pollutant.

Like $PM_{2.5}$, PM_{10} is a large-scale pollutant, which is also clearly visible in the source apportionment analysis (Figures 9 and 10): compared to EC in PM_{10} each labelled country is contributing a smaller fraction to its own area. One can also see that natural sources are contributing significantly, which is mainly the contribution of sea salt, an important ingredient of PM_{10} in for example the Netherlands.

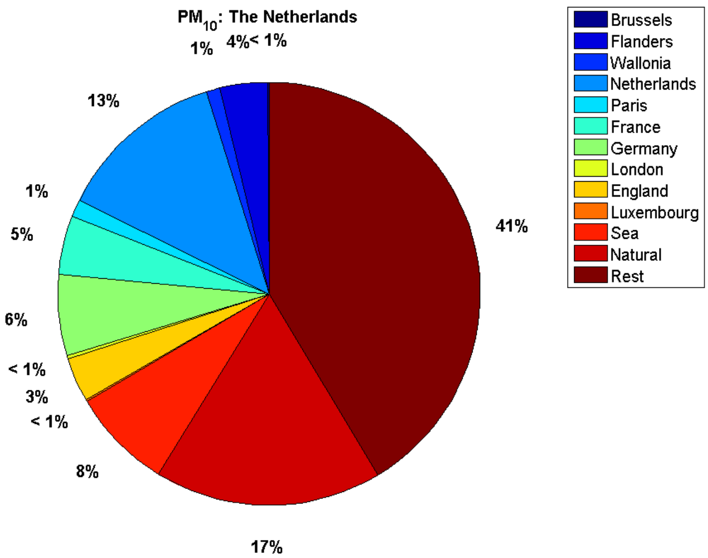

Figure 9 Blame matrix for PM_{10} for the Netherlands.

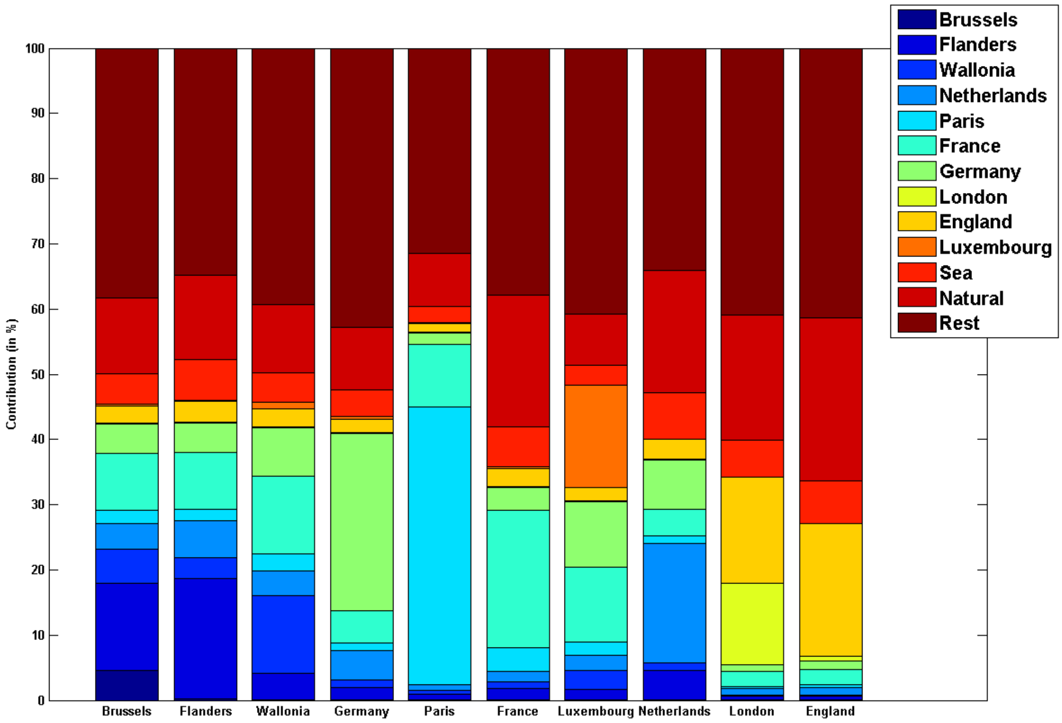

Figure 10 The blame matrix for the component PM_{10} as calculated with the LOTOS-EUROS model.

Figure 10 shows the blame matrix for PM_{10} for the JOAQUIN domain. If one compares this matrix with the one of EC, one can see that the contribution of each country to its own concentration is smaller in the case of PM_{10}, reflecting its large-scale distribution over the domain.

Contrary to $PM_{2.5}$, one can see that natural sources contribute significantly in the blame matrix for PM_{10}. This is due to the relative large contribution of sea salt to the PM_{10} concentrations. It is, as expected, relatively smaller in regions like Luxembourg and Paris which are further away from coastal areas than the other regions.

Finally, roughly 30-40% of the PM_{10} is coming from the rest term, which is due to the fact that PM_{10} is strongly influenced by large-scale transport from outside of the JOAQUIN domain as calculated here.

6 A comparison between BEL-EUROS and LOTOS-EUROS

In the JOAQUIN project, a source apportionment study has also been performed with the BEL-EUROS model. Here, a traditional methods of source apportionment was used, in which a source is reduced by a small percentage (Deutsch et al., 2014). As stated in section 2, LOTOS-EUROS explicitly calculates the contribution from labelled emission categories in order to track the origin of air pollutants.

In this section, we discuss the comparison of the results from LOTOS-EUROS and BEL-EUROS for this type of calculations. We will focus on the source apportionment of two regions within the JOAQUIN domain, i.e. the Netherlands and Flanders for the components EC and PM_{10}.

6.1 Elemental Carbon (EC) in Flanders and the Netherlands

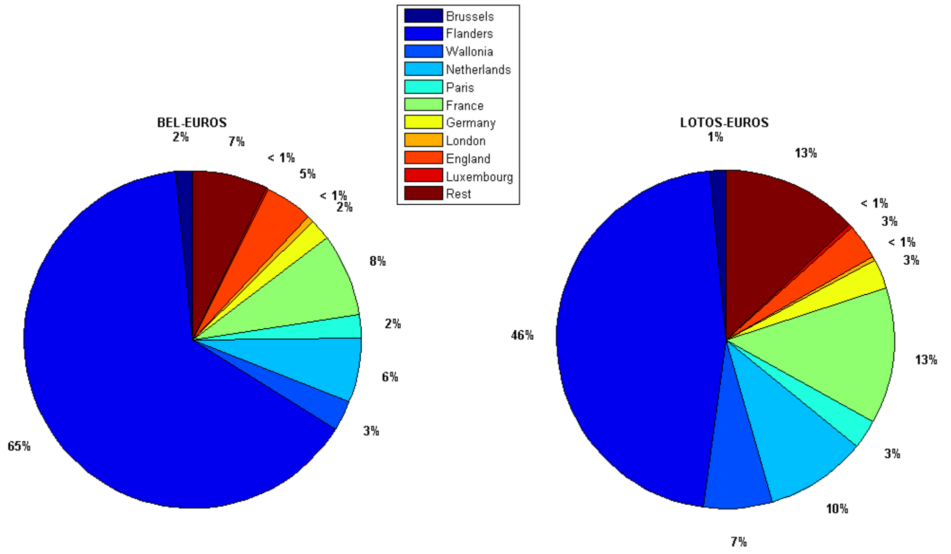

Figure 11 The blame matrix for the component EC in Flanders

Figure 11 shows the blame matrix for EC for Flanders as calculated with BEL-EUROS and LOTOS-EUROS. There is a slight difference in these two simulations between the contributions of EC from Flanders itself: 65% (BEL-EUROS) vs. 46% (LOTOS-EUROS). Another difference is the contribution of the rest term: 7% (BEL-EUROS) vs 13% (LOTOS-EUROS).

Figure 12 shows the blame matrix for EC for the Netherlands as calculated with respectively BEL-EUROS and LOTOS-EUROS. For the Netherlands, there is only a small difference between both models for the Dutch contribution to its own EC concentration: BEL-EUROS calculates 59%, and LOTOS-EUROS calculates 54%.

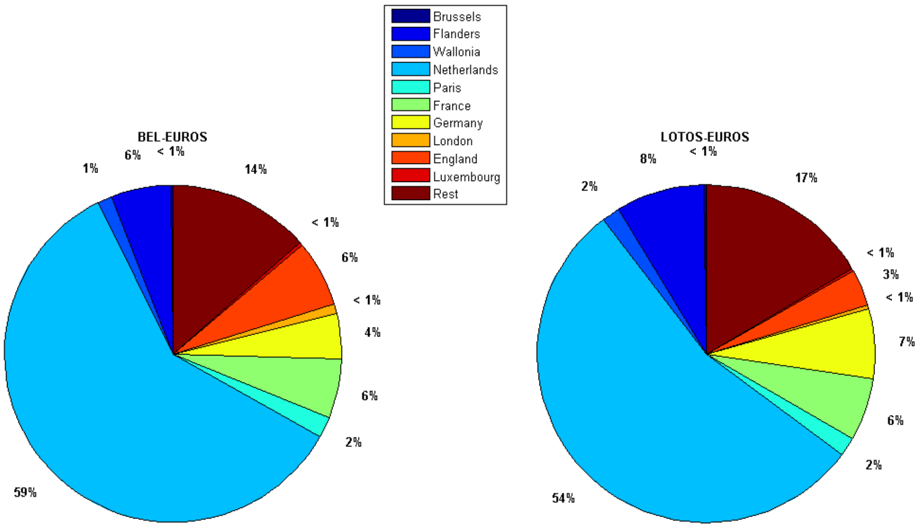

Figure 12 The blame matrix for the component EC in the Netherlands

In the Netherlands, high resolution EC maps (1×1 km^2) have been produced for the last few years in the context of the large-scale concentration maps for the Netherlands (GCN-project, see e.g. Velders et al., 2014). The calculations for GCN are performed using the OPS-model, which combines a Gaussian plume model approach for local applications with a Lagrangian trajectory model for long range transport. OPS has a good performance on local scales, in particular in areas close to sources In these studies, the contribution from Dutch emissions to EC concentrations is also roughly 50%, i.e. in the same range as calculated with BEL-EUROS (59%) and LOTOS-EUROS (54%).

For Flanders a larger difference is observed between BEL-EUROS and LOTOS-EUROS for the calculated EC contribution from its own emission sources to the mean EC concentration over its area (65% vs 46%). Here it is suggested that this difference of the model outputs is due to the difference between the EC concentrations themselves as calculated with respectively BEL-EUROS and LOTOS-EUROS. Figure 13 shows the EC maps of both models for the year 2009. One can see the difference in the magnitude of the EC concentrations in urban areas. The fact that EC concentrations are much more enhanced in the BEL-EUROS simulations in urban areas and its surroundings, indicate that the mass contribution due to (local) emissions in these areas might also be larger, as is indeed seen in the source apportionment calculations.

A small test using the CHIMERE model with reduced EC concentrations for Flanders, yielded a 41% contribution of Flemish emissions to their EC concentrations, very close to the LOTOS-EUROS source apportionment calculations. CHIMERE in general also has a very similar distribution of EC annual mean concentrations as LOTOS-EUROS (Adriaenssens et al., 2015). This confirms the hypothesis above that the difference between the BEL-EUROS and LOTOS-EUROS result for source apportionment is due to the difference in the distribution of the calculated EC concentrations between both models.

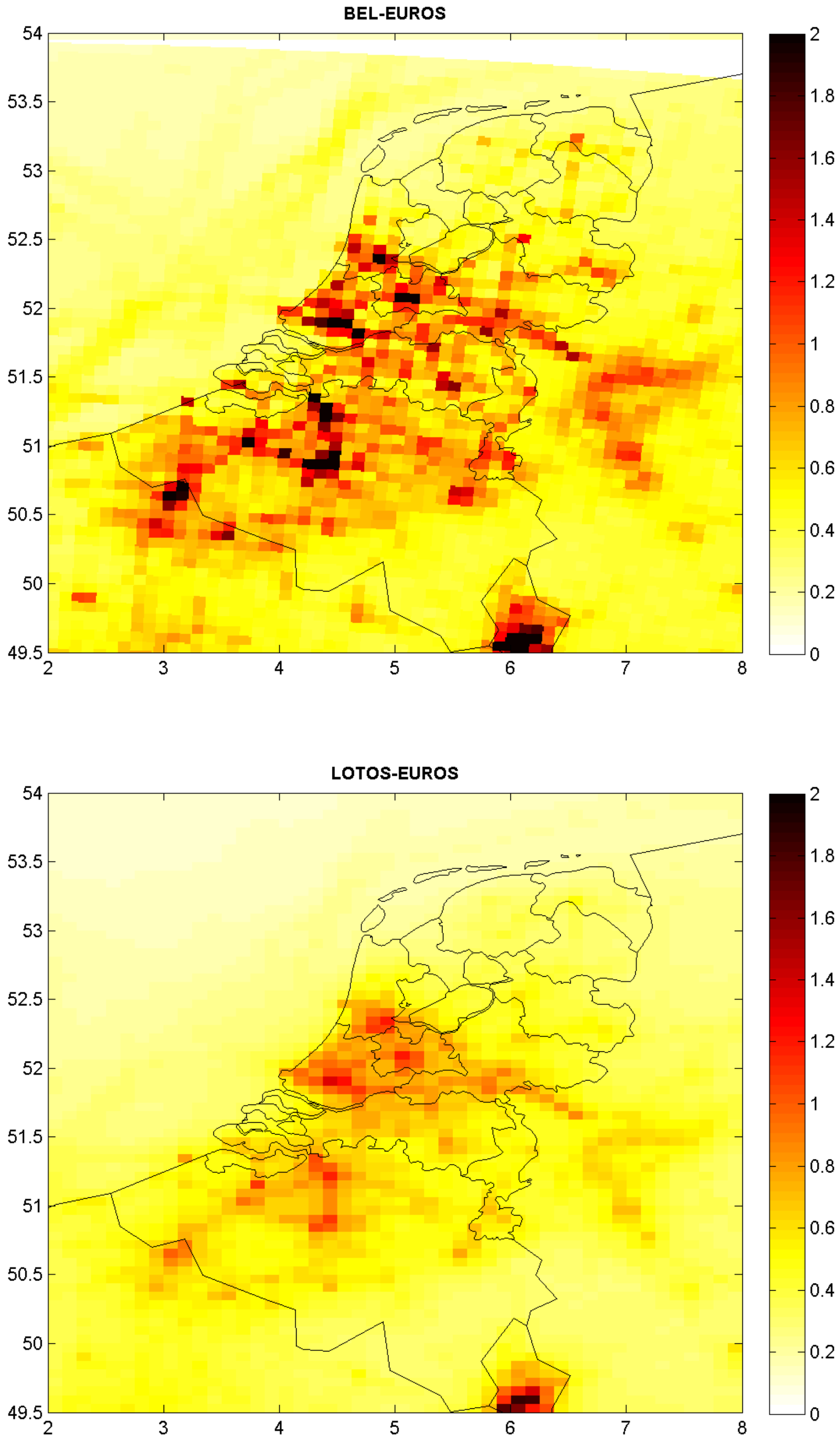

Figure 13 The annual averaged EC concentration (in µg/m^3) for 2009 as calculated with the BEL-EUROS model (top panel) and with LOTOS-EUROS (bottom panel).

6.2 PM_{10} in Flanders and the Netherlands

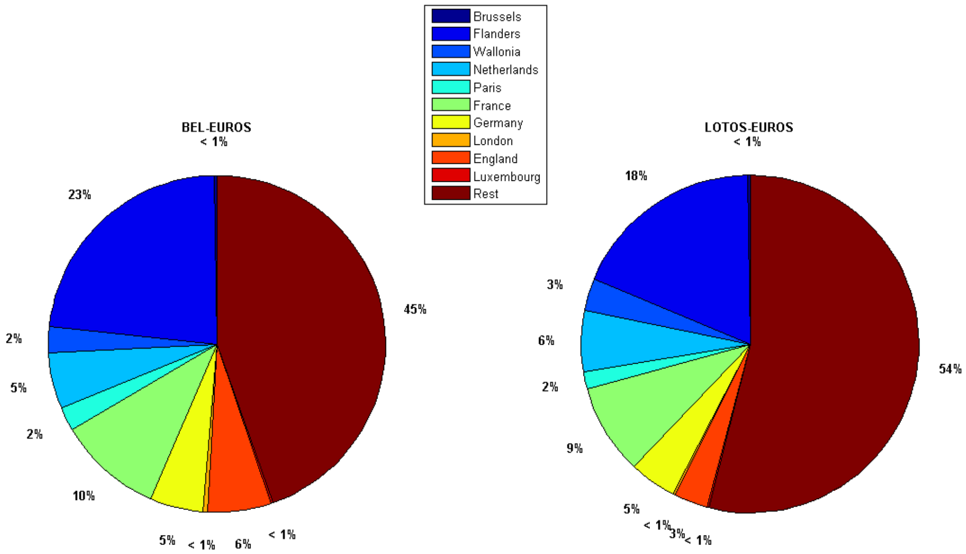

Figure 14 The blame matrix for the component PM_{10} in Flanders.

Figure 14 shows the blame matrix for PM_{10} for Flanders as calculated with BEL-EUROS and LOTOS-EUROS. The calculated percentages for the contributions of PM_{10} from Flanders itself are 23% (BEL-EUROS) and 18% (LOTOS-EUROS). Furthermore, the *Rest* term in both calculations: BEL-EUROS attributes 45% of the PM_{10} emissions coming from outside the JOAQUIN domain, whereas LOTOS-EUROS calculates this as 54%.

Figure 15 shows the blame matrix for PM_{10} for the Netherlands as calculated with respectively BEL-EUROS and LOTOS-EUROS. Also for the Netherlands, there is a small difference between both models for the Dutch contribution to its own PM_{10} concentrations: BEL-EUROS calculates 23%, whereas LOTOS-EUROS calculates 18%. The *Rest* term is again smaller in the BEL-EUROS calculations (49%) than in the LOTOS-EUROS calculations (60%).

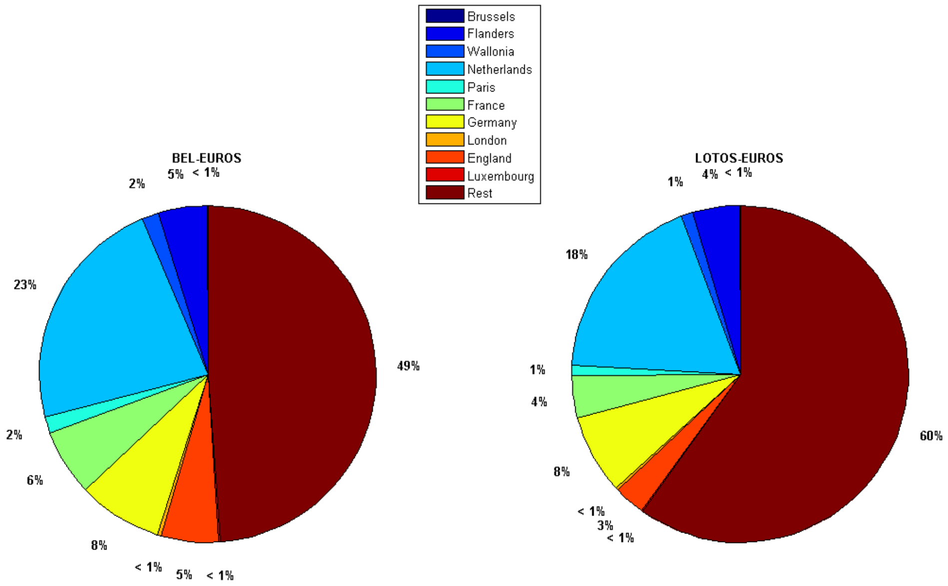

Figure 15 The blame matrix for the component PM_{10} in the Netherlands

Figure 16 shows the maps of PM_{10} as calculated by LOTOS-EUROS and BEL-EUROS. The two maps show quite some differences: the regional background areas over the Netherlands and Flanders are higher in BEL-EUROS than in LOTOS-EUROS. Scaling BEL-EUROS to the same background values as in LOTOS-EUROS (by subtracting 4.5 $\mu g/m^3$ (not shown)), the urbanized areas in the BEL-EUROS map are again enhanced with respect to the LOTOS-EUROS map, although less enhanced than for EC. This might again explain the difference between the BEL-EUROS and LOTOS-EUROS blame matrices for PM_{10}.

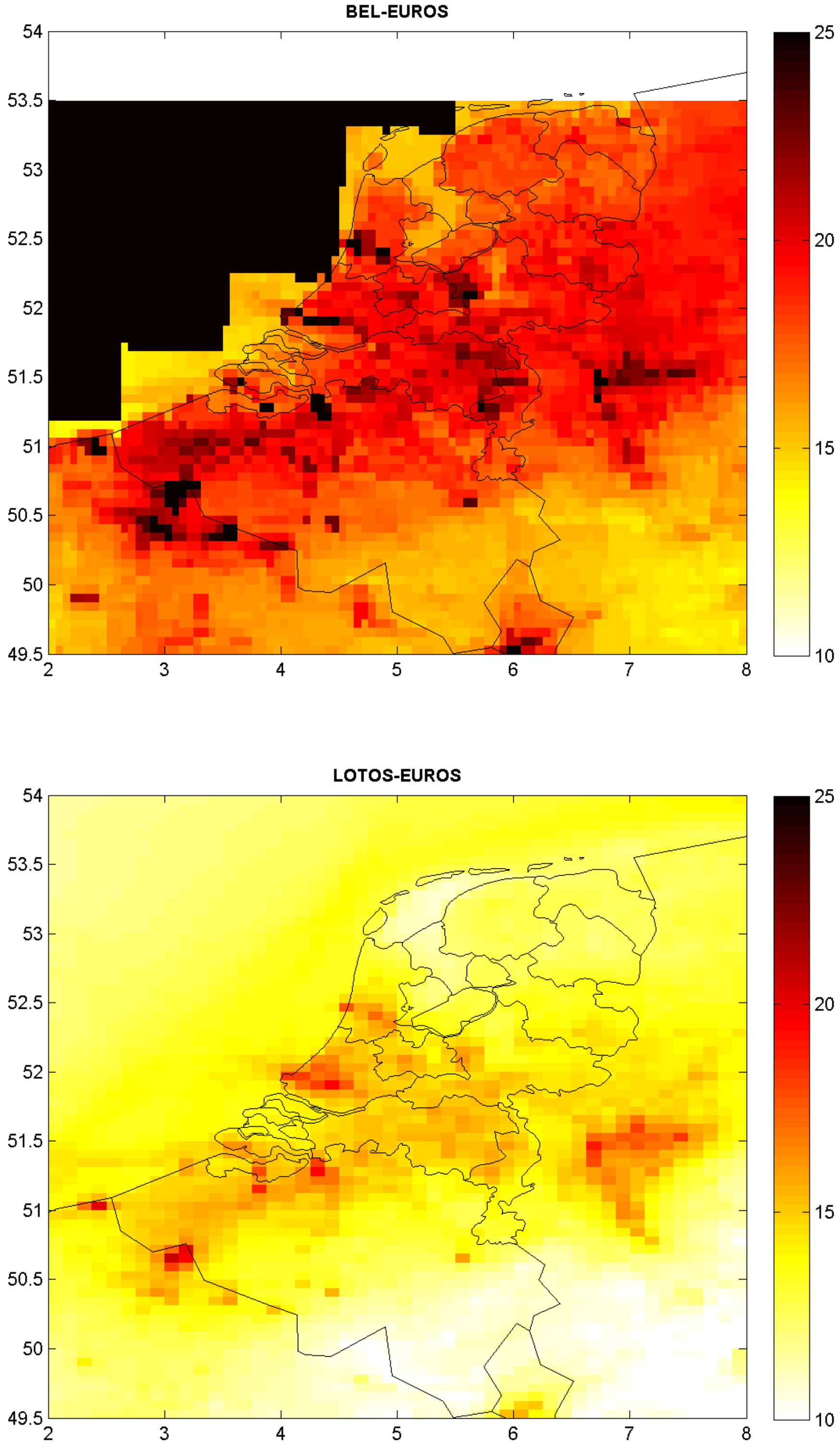

Figure 16 The annual averaged PM_{10} concentration (in µg/m^3) for 2009 as calculated with the BEL-EUROS model (top panel) and with LOTOS-EUROS (bottom panel).

7 Conclusions

Source apportionment calculations were performed with the LOTOS-EUROS model for the pollutants PM_{10}, $PM_{2.5}$ and EC over the JOAQUIN domain, i.e. Northwest Europe. It was shown that for EC, the local emissions from the ten considered areas (Brussels; Flanders; Wallonia; The Netherlands; Paris; France; Germany; London; England; Luxembourg) contribute significantly to its concentrations. The effect is most strongly seen for cities like Paris and London (50%), which are **the** examples of extended urbanized areas in the JOAQUIN domain with associated high EC emissions, which are directly visible in the EC concentration maps. In this context it is elucidating to emphasize that, for example in the Netherlands, we also find high contributions (up to 80%) to EC concentrations from Dutch emissions in areas like “de Randstad” (see Figure 3). Due to the spatial averaging (including less densely populated areas) for the blame matrix for EC, this fraction goes significantly down for the Netherlands as a whole (50%). Contrary to EC, for a large-scale component like $PM_{2.5}$ and PM_{10}, emissions in the regions themselves contribute only slightly (10-20%) to $PM_{2.5}$ and PM_{10} concentration in its own area. For regions close to the coastal area, natural sea salt emissions contribute significantly to the PM_{10} concentrations.

The above findings confirm that local policy measures have the potential of a much higher efficiency in reducing local EC concentrations than PM_{10} concentrations. As emphasized above this is especially applicable to densely populated areas like Paris or London. This study, however, also shows that for areas like de Randstad in the Netherlands, the same holds true.

Finally, it was found that there are slight differences between the source apportionment studies performed with the LOTOS-EUROS model (this study) and the BEL-EUROS model (Deutsch et al., 2014) for the JOAQUIN project, which is rather encouraging for the applicability of these type of source apportionment studies. It was shown that for both Flanders and the Netherlands, BEL-EUROS typically calculates slightly higher values for the region’s own contribution to EC and PM_{10}. We attribute the difference between both models to the calculated spatial distribution of the components and not to the source apportionment method used itself. Indeed, the maps of EC and PM_{10} as calculated with BEL-EUROS and LOTOS-EUROS show some differences, where the BEL-EUROS model shows enhanced concentrations of EC and PM_{10} in especially urbanized areas. This is in line with the higher contribution found by the BEL-EUROS model of a region’s own emission to its concentrations.

8 References

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