Atmos. Chem. Phys. Discuss., 6, S4363–S4371, 2006
www.atmos-chem-phys-discuss.net/6/S4363/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "PM measurement campaign HOVERT in the Greater Berlin area: model evaluation with chemically specified particulate matter observations for a one year period" *by* M. Beekmann et al.

M. Beekmann et al.

Received and published: 3 November 2006

Answer to Referee 2 :

The authors thank the referee for his thoughtful comments on the paper which certainly lead to a more profound discussion of several results presented in this paper.

Specific comments:

1)influence of spatial model resolution on results, in particular as far as emissions are concerned

This is an interesting point to which more attention will be give in the revised version. Throughout the paper, both results for continental scale and nested simulations are given, with respectively an about 30 km and a 4 km horizontal resolution. Differences between these simulations both depend on the nature of the site and on the nature of the compound.

For primary compounds at urban and traffic sites (MP42, MP174, BS), the nested simulations show as expected larger values then the continental ones and which moreover are closer to observations. This is evident for EC which is of primary origin only, but also for OC and PM10, which are partly primary, partly secondary. This behaviour is due to the fact, that urban area emissions on the nested grids are larger than on the continental grid, where they tend to be artificially diluted due to lower grid resolution. Please see also our answer to your minor comment 6.

On the other hand, for secondary species, and in particular for secondary inorganic ions (SIA), differences between continental and nested simulations are small, both for urban and rural sites. Even if urban SO2 and NOx levels (the SO42-and NO3-precursors) are larger for the nested run, the time scale related to the SO2 to SO42- and NOx to NO3- transformation (several hours to several days) is large enough to make differences in the grid resolution unimportant. Taking as an example a 10m/s average wind speed, the 30 km continental grid corresponds to a 1h transport time, which is small compared to the SIA formation time scale. These arguments are also supported by the fact that while differences in SIA between urban and rural sites are small, differences in EC, OC and PM10 are much larger.

For SIA a 30 km grid resolution appears then indeed as sufficient for air quality simulations. However, SIA are only one component of particulate matter, and the above statement is not valid for other compounds (EC, OC) or for PM10 as discussed above.

To strengthen this discussion in the text, the following sentence is added in section 4.1, at page 7296, line 25: “On the other hand, in agreement with the observations, the

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nested run exhibits a distinct urban-rural concentration gradient for primary particles, SO2 and NOx levels (not shown for SO2 and NOx) because of the better representation of the urban-rural emissions contrast. The regional scale run is not able to capture this urban signal because the urban-rural concentration gradient of primary pollutants is diffused due to the high spatial aggregation of the emissions. However, the time scale related to the SO2 to SO42- and NOx to NO3- transformation (several hours to several days) is large enough to make differences in the grid resolution unimportant for SIA formation."

Differences in the day to day variability between the nested and the regional version are also discussed in page 7300, line 15 : "This means that the spatial refinement in the model resolution also leads to the introduction of some additional noise in the model, as for instance possible errors in the urban emission inventory, which are "diffused" in the large scale data set with a higher spatial aggregation. It also has to be mentioned that the number of urban meteorological sites included in the optimum interpolation procedure is very small. Thus, the urban heat island effect might be not well resolved in the meteorological data base leading to too stable conditions, particularly in the night and in winter time. This discussion is relevant for all locally emitted primary species (EC, primary OC and PM10)."

2)Seasonal variation of particulate matter components

a)sulphate

The sulphate seasonal variation is influenced by a complex interplay between several processes : SO2 emissions (with a winter maximum in Europe), SO2 to SO42- transformation, with a summer maximum for the gas phase pathway, and a complex behaviour for the liquid phase pathway (more liquid water content during winter, but lower oxidant levels, transport including vertical mixing). Wet scavenging (a sink process) is stronger in winter, due to larger precipitation rates. From these individual processes, it is not clear whether a summer or winter sulphate maximum should be expected. However

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the ensemble of these processes does not lead to a summer maximum, neither in observations, nor in simulations. Also a comparison of the European scale CHIMERE CTM with EMEP observational data for the year 2000 does not show any seasonal bias in sulphate [Bessagnet et al., 2004].

In contrast, an earlier global model study by Kasibhatla [1997] shows a strong summer SO42-maximum over North America, but also over Central Europe. Many features are different in this latter model than in RCG, which makes the comparison of the simulated seasonal variations difficult. As an example, oxidant fields are prescribed in the Kasibhatla [1997] study and not explicitly calculated as in RCG. As the authors state, the SO2 to SO42- transformation could be overestimated due to this fact, because H2O2 is relaxed to monthly average after 1 h time, once it is reduced by SO2 within cloud droplets, while in reality it takes more than an hour to recover. This overestimation could change with season.

b)Biomass burning influence on EC and OC

Biomass burning (except residential wood burning) is not included in the RCG model, because emissions are thought to be small over Western and Middle Europe compared to anthropogenic emissions (very few fires occur in this region). Fires frequently occur over the Mediterranean region, but transport from this region to North-Eastern Germany is not frequent. However, during the simulation period (October 2001 - September 2002), the potentially most important biomass burning source were the large forest fires over European Russia in the July to September period (outside of the model domain). The question is then: did these events influence PM levels in North-Eastern Germany ? High levels of EC and OC appeared during the last 10 days of August, leading also to the enhanced monthly averages in August. In fact, these high levels are already explained by accumulation of emissions from the Berlin area, for instance they appear together with enhanced urban NOx levels. Moreover, analysis of weather maps during this period shows that conditions in lower layers were mostly cyclonic, not allowing for long range transport from Russia to the Berlin region.

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c)Sea salt influence on Sulphate

The sea salt emissions for Na+ and Cl- are included in the model, but those of sulphate are neglected. However, the seas-salt contribution to SO42- in the Berlin area should be very small on the average for two reasons. First, average Na+ levels measured during HOVERT are only about 0.30 μg/m3. Assuming that all observed Na+ comes from sea salt this gives an upper limit for the sea salt contribution to SO42- of about 0.30 * 0.26 = 0.08 μg/m3 (taking into account the mean sea water composition and the molar mass ratios). This means that the contribution of sea salt to sulphate levels over the Berlin area is marginal on the average.

d)Color bars on figure 3

An explanation of colour bars in figure 3 has now been added.

e)Why is PM10 summer minimum well represented in the simulations?

PM10 is the sum of different chemical species. Among them, nitrate has the most pronounced seasonal variation with a marked summer minimum, because of a higher vapour pressure in summer due to higher temperatures. The sulphate seasonal variation has been discussed before. EC is more of local origin, its emissions are lower during summer. So for these species, a summer minimum in concentrations is expected. More generally, the PM10 seasonal variation results from a complex interplay between different processes affecting the different species. Primary PM emissions, secondary PM formation, gas to particle phase equilibria, dispersion of local emissions, transport from outside the Berlin-Brandenburg region, wet and dry deposition, all influence this seasonal variation. A detailed budget study, addressing individually these source and sink terms and their seasonal variation is beyond the scope of this paper. This topic is addressed in a companion paper in preparation by Kerschbaumer et al. (2006).

The following sentence was added at the end of section 4.2 : “The fact that the seasonal variation (or its absence) is well represented by the model is encouraging. Apparently,

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the overall seasonality of a series of processes is well represented in RCG: Dispersion of primary PM emissions, secondary PM formation, advection to the Berlin area, wet and dry deposition."

3) Day to day variability

a)interpretation of correlation coefficient in relation to transport

The referee argues that (high) correlations are sensitive to the highest values and he doubts that those values were caused by transport regimes. In fact, in the paper interpretation of correlation coefficients is done separately for different species. For sulphate, sector analysis of the air mass origin is very instructive. Qualitatively speaking, when air masses originate from the southern and eastern sector, observed and simulated sulphate values are clearly enhanced as compared to the average of all sectors. This is due to the fact that for southern and eastern directions, strong SO2 sources are present in Saxony and Poland, which can be transformed into SO42- during transport to the Berlin area. Ammonium shows a similar behaviour. These results are based on back-trajectory analysis (E.Reimer, personal communication, 2006). So for SO42- and NH4+, variability both in observations and simulations , and thus correlation between both, seems indeed to be linked to different transport regimes. In the original version of the paper, the following discussion is already included to state this fact: "Probably, the good correlation in these time series is driven by different transport regimes made evident by trajectory analysis and which seem to be well taken into account in the model (E. Reimer, personal communication): larger observed and simulated SO42- and NH4+ values in the Berlin region are related to transport from the east and south east sector where strong SO2 sources are present (in Saxony, Southern Poland , while lower values are more related to transport from the western sector under low pressure conditions. Nearly identical correlation coefficients for the continental and the nested simulations are in line with this interpretation."

In the original version the following conclusion had been made at the end of section

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4.3:

“Good correlation seems to be driven by large scale transport patterns, as suggested by the consistently larger correlations obtained with the large scale simulations than with the nested ones.”

In the revised version, it was replaced by the following more balanced statement: “As discussed before, for SO42- and NH4+, correlation seems to be driven by transport patterns (transport from source regions located in the south-east sector). For EC and OC, smaller correlation coefficients and the fact that the refinement in model resolution does not lead to better correlation, indicate that the day-to-day variability in urban scale processes (emissions, horizontal and vertical dispersion) is difficult to represent in the input data bases.”

b)EC, OC underestimation at rural sites related to wet scavenging Yes, wet scavenging is one of the potential reasons for EC and OC underestimation at rural sites, but not the only one. Error sources for rural EC are discussed in section 5.2, page 7304 line 15 - 26. (“Uncertainties in measurements ĚĚĚ possible.”)

4)availability of CO measurements

No, unfortunately, CO measurements were not available and the well known EC/CO correlation could not be exploited. However, the EC / NOx correlation present at urban and traffic sites could be exploited.

Minor comments

Technical corrections 1 ,2 and 9 have been addressed in the paper (concerning wording).

3) SAPRC93 or 99

Indeed, the SAPRC93 version (an not SAPRC99) is available in the RCG model. Anyway, in this study CBM-4 is used.

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4) Mixing height and model top

During the one year simulations, the mixing height only very rarely is larger than the model height (2.5 km). In this case, the model mixed layer is bounded to an upper limit value, in order to keep room for the residual layers.

5) Meteorological data

Meteorological data are produced from surface and upper air observations employing a diagnostic meteoro-logical analysis system based on an optimum interpolation procedure. This has been made clear in section 2.3.

6) section 2.4 Emissions

As said in section 2.4 : “to ensure consistency between the urban/regional-scale and the continental-scale emissions, the Berlin/Brandenburg data were scaled sector-by-sector and species by species to the level of the EMEP data.” This procedure contributes to the consistent simulations between the nested and the continental scale model version for inorganic ions. The procedure of emission scaling has also been used for the City Delta exercise and is described in more detail in the paper Cuvelier et al. 2006. Surely, there are differences in the spatial distribution of emissions: in the nested grid (4 km resolution) Berlin urban background emissions are resolved whereas they are not in the 30 km grid. This leads to important differences in simulated urban EC, OC and PM10 in both model versions, as already pointed out above. We thought that giving an extra table numbers for emissions would not be very meaningful.

C. Cuveliera,*, P. Thunisa, R. Vautardb, M. Amannc, B. Bessagnetd, M. Bedognie, R. Berkowiczf, J. Brandtf, F. Brochetong, P. Builtjesh, C. Carnavalei, A. Coppallej, B. Denbyk, J. Dourosl, A. Grafm, O. Hellmuthn, C. Honoréd, A. Hodzicb, J. Jonsono, A. Kerschbaumerp, F. de Leeuwq, E. Minguzzir, N. Moussiopoulosl, C. Pertots, V.H. Peuchg, G. Pirovanos, L. Rouild, F. Sautert, M. Schaaph, R. Sternp, L. Tarrasono, E. Vignatia, M. Voltai, L. Whiteu, P. Windo, A. Zuberv “CityDelta: A model intercomparison

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study to explore the impact of emission reductions in European cities in 2010", which has been accepted for publication in Atmospheric Environment (ref. ATMENV-D-06-00252R1).

7) section 2.6 SOA formation

Both SOA formation from anthropogenic and biogenic VOC is included in the SORGAM module. This has been emphasized in the text. The referee may have been misled by the fact that only terpenes have mentioned in the text explicitly; this is because terpense had to be added to the CBM-4 chemical mechanism, because they only had been included before as a surrogate species.

8) section 2.7 EC wet scavenging

EC and OC wet scave nging coefficients have been chosen as equal to sulphate (1.0 10-4s-1), This value neglects the initially hydrophobic character of EC and OC. This potentially leads to an overestimation of EC, OC wet deposition, which in turn might explain the too low rural EC, OC concentrations in rural areas. The time scale needed to render EC and OC hydrophilic is still an open question in literature (e.g. see Kanakidou et al., 2005) .The following paragraph was added in section 2.7 to explain the treatment in EC and OC wet scavenging:

“Wet deposition of particles is treated in RCG using a simple scavenging coefficient approach. For EC and OC the same value as for sulphate is chosen, 1. 10-4 s-1. This factor implies that the particles immediately become hydrophilic.”

We hope that we could give satisfying answers to all questions raised.

Matthias Beekmann on behalf of the coauthors Créteil, France 03/11/06

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 7285, 2006.

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