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Atmospheric
Chemistry
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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.

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Answer to Referee 3 :

The authors thank the referee for their helpful remarks which certainly led to an improvement of the paper.

1) Dedicated paragraph about measurement uncertainty in section 3

Following the referee's suggestion, the following text was added to section 3 to include measurement uncertainties in the measurement section. "Systematic quality checks of the measurements were performed during and after the campaign, including compari-

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son between sampling methods (high and low volume samplers) and in particular the temperature dependence of the EC, OC determination with the VDI method (John and Kuhlbusch, 2004). The comparability of the different methods was in general satisfying, with differences below 10 to 15%. Nevertheless, systematic uncertainties remain, which could not directly be tested. In particular, evaporation of nitrate (and ammonium) above about 20°C outside temperature (e.g. Schaap et al., 2004a) can lead to a substantial (but difficult to quantify) underestimation of these ions especially in summer. During an intercomparison campaign at a Dutch rural sites, total carbon measurements obtained with high volume samplers agreed within 10% (ten Brink et al., 2004). However, EC measurements performed after the VDI 2564/2 method as in our study showed larger values than the average (about 50%), while OC measurements showed lower values (about 25%) than the average. Rather than indicating a systematic bias in one of the methods used in this study, these results indicate the range of measurement uncertainty which can be reasonably expected for this type of measurements."

2) is there a way to check the correctness of NOx emissions ? No, there is no way to check the correctness of NOx emissions in the frame of this study. Differences in the annual NOx means for the urban station MP42 are a bit more than 20% (observations 29.6 μg/m3 , nested simulations 23.1 μg/m3). However, they are due to different error sources impossible to discern, among which are : errors in emissions, in dispersion, problems of representativity of a site for the model grid (here 4 * 4 km). A way to check the validity of average emissions would be to perform flights at different height levels all around isolated urban areas, and measuring either NOx or NOy. In doing so, only the average wind speed over an area has to be correctly estimated (and can be measured on the same air craft), and not complex dispersion patterns over complex terrain. This has been done during the EVA campaign around the Augsburg town (Mannschreck et al., 2002) and around the Paris area during the ESQUIF campaign (Vautard et al., 2003). In both cases, an uncertainty in NOx emissions of about ś 30% was found.

3) reference for time dependence of emissions

The best reference in the open literature is the paper Cuvelier et al. 2006, where the preparation of emissions for the City Delta exercise is described. The same procedure is used in the present work.

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 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).

4) line 24 - 36 is broken in two references

5) The formula for Carters 1-product isoprene is the following (Carter, 1996):

ISOP + OH -> 0.088 RO2-N + 0.912 RO2-R + 0.629 HCHO + 0.912 ISOPROD + 0.079 R2O2 + 1.079 RO2 + 0.283 -C

where ISOPROD represents all unique compounds produced by the isoprene + OH reaction (= which are not produced by the oxidation of other VOC’s), in particular methacrolein, methyl, vinyl ketone, hydroxymethylacrolein, two unsaturated aldehydes. The scheme is calibrated against chamber measurements of ozone formation from isoprene oxidation. The meaning of the other acronyms can be found in Carter, 1996. The formula was not added in the text, because explanation of all acronyms would be awkward and not provide much relevant information for this study.

6) Terpenes are lumped into -pinene and limonene, because these two species are the lumped species representing terpenes in the SORGAM module which is used for calculation of SOA formation (Schell et al., 2001).

7) Explanation for 11 - 39 % underestimation of sulphate for nested simulations.

First, values for nested and regional simulations of sulphate are similar, because sulphate formation is slow compared to the transport time through grid cells in the regional model (30 km resolution, see detailed discussion in our reply to Referee 2, point 1).

Second, the underestimation in the simulations is not severe taking into account the various error sources (SO2 emissions, chemical transformation, deposition) affecting sulphate formation. These error sources are mentioned at several places in the manuscript. For example in page 7301, line 17, we write : “Still, uncertainty in simulated SO42- is large, as it crucially depends on the cloud liquid water content, which is not available from the observed meteorological fields and which had to be estimated for typical low level clouds.” It may be frustrating, but the fact is that it is hardly possible to discern these different error sources (as also discussed in other parts of our reply to referee remarks).

8) better correlation with observations for large scale than for nested simulations.

Smaller correlation for refined spatial resolution is not unexpected. Refined spatial correlation adds variability to the simulations, but, inevitably, also noise. Clearly, the nested simulations better show the urban-regional contrast in primary species than does the regional one, but for the price of some additional noise. To make this clearer in the paper, the following text was added in section 4.3 (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).”

As far as specific error sources for the small scale simulations are concerned (emissions, chemistry), we discuss this point in section 5.2 for EC. Indeed, the EC scaling with NOx suggests that EC emissions might be overestimated in the Berlin area. Chemistry, as far as secondary inorganic ions are concerned, is not expected to be very different in the nested and regional simulations, especially the correlation coefficients are nearly identical for sulphate and ammonium, a bit lower in the nested version for nitrate.

9) the indicated text was rephrased.

10) physical meaning of “annual average over correlation coefficients Ě.

In fact, it was meant: “the average (i.e over all HOVERT sites) of the annual correlation coefficients”. This is corrected in the text.

11) distinguish between potential error sources : effect of emissions or transport ?

As argued under point 2, it is impossible to discern between errors due to emissions and due to transport from the HOVERT measurements. However, for EC, a special scaling procedure with NOx was applied, which allows to normalise out dispersion and representativity errors (section 5.2). This was possible, because both EC and NOx observations were available and because both are strongly correlated. This procedure suggests an overestimation in the urban EC emission, under the assumption of correct NOx emissions (or in other words an overestimation of the urban EC/NOx emission ratio).

12) no clear support found for the following sentence “As an overall conclusion, this evaluation study has helped to gain increasing confidence in simulations of chemical constituents of particulate matter with the RCG model. “

To make the subjective nature of this sentence more evident (but implicitly, it was already), we added “in our opinion”. Still, we are very convinced of what we are saying: the HOVERT campaign offered an unpreceeded opportunity to test aerosol simulations with the RCG model. Biases for most of the aerosol species are lower or comparable compared to previous studies. Model / observation comparisons for EC and OC have not done before in this detail. Thus, clearly confidence in simulations is increased through this study. Anyway, throughout the paper, we also highlight observed differences and try to give explanations, whenever possible.

13) Quality of figure 2 is now improved. Titles over the panels are improved in the sense that the species name will be indicated.

14) Figure 3 caption is now more detailed. Black columns stand for observations, the red ones for nested simulations, the green ones for continental simulations.

15) A list of acronym definitions is added.

References :

Carter, W.P.L.., 1996, Condensed Atmospheric Photooxidation mechanism for isoprene, Atmos. Env. 30 , 4275 - 4290.

Mannschreck, K., D. Klemp, D. Kley, R. Friedrich, J. Kühlwein, B. Wickert and F.Slemr, 2002, Evaluation of an emission inventory by comparisons of modelled and experimentally derived concentration ratios of individual NMHC, CO and NOX, Atmos. Environ. , 36, 81-94.

Vautard, R. Martin, D, Beekmann, M. Drobinski, P., Friedrich, R., Jaubertie, A., Kley, D., Lattuati, M., Moral, P., Neininger, B., Theloke, J., 2003, Paris emission inventory diagnostics from ESQUIF airborne measurements and a chemistry transport model, J. Geophys. Res., 108, D17, 8564.

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

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