Atmos. Chem. Phys. Discuss., 8, S1998–S2007, 2008
www.atmos-chem-phys-discuss.net/8/S1998/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The aerosol distribution in Europe derived with the Community Multiscale Air Quality (CMAQ) model: comparison to near surface in situ and sunphotometer measurements" *by* V. Matthias

V. Matthias

Received and published: 25 April 2008

While the results presented in this study appear sound, I miss a discussion that puts them in the context of other CMAQ application and evaluation studies, particularly over North America. I realize that speciated measurements of particulate matter in North America tend to focus on PM2.5 while in Europe more emphasis is placed on PM10, but I would still urge the author to discuss his findings in the context of the numerous studies that have been published on evaluating CMAQ PM2.5 simulations over North America (examples are listed in my specific comments below). Specifically, I would like to see a discussion of the author's findings taking into account reported model results

for elemental and organic carbon as well as the CMAQ 'other unspeciated PM2.5' component over North America. In this respect, the author needs to strengthen the caveats on the impacts of the uncertainties in the speciation of primary PM emissions on the reported results.

A discussion in the context of the CMAQ results in the US has been added (section 3.3, line 629, line 660, sec. 4, line 886), altough it is sometimes difficult to compare the PM2.5 results to the PM10 values. A discussion on the speciation of the primary PM emissions has also been added (line 187). Certainly, if the fraction of EC and OC in the primary PM would be larger, the differences in the organic aerosol will be decreased. Nevertheless, the large discrepancies between observed and modelled PM10 will remain the same. Possible explanations will still point to the organic aerosol fraction that might be underestimated in the model.

Specific comments: Page 1460, line 1: Some of the sulfate and nitrate PM is emitted directly, at least according to the model. Therefore, the inorganic PM portion should not be referred to as 'secondary' here or in subsequent passages. Instead, I suggest using the term 'total inorganic aerosol' or simply 'inorganic aerosol' throughout the manuscript. The primary contribution to total inorganic aerosol may be small, but it is not zero.

It is true that the primary contribution to total inorganic aerosol is small in this modeling study but of course it is also true that it is not zero. I introduced an new term inorganic aerosol (IA) and secondary inorganic aerosol (SIA) is only used when no primary emitted aerosol is included.

Page 1462, lines 6-9: What is the justification for this speciation of PM2.5 emissions? Is it an average over all industrial sectors such as transportation, heating, power generation, etc.? Does it vary in space? In particular, PM2.5 emissions from diesel engines are known to be a significant source of EC and primary OC emissions but these two

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compounds appear to account for only 3areas, this number is certainly an underestimate. For example, a typical speciation profile for PM2.5 emissions from diesel trucks used in North American modeling applications assumes an EC/OC split of roughly 60from other species (sulfate, nitrate, 'other' unspecified fine particles). At a minimum, please provide more justification for the speciation approach used in this study and discuss the implications for the results presented in Section 3.3.

It has been taken from SMOKE as a typical PM profile connected with emissions from combustion processes. Because no information about the real split of the primary anthropogenic PM2.5 emissions was available no spatial variation was applied (see line 187).

Page 1462, lines 8-9: While coarse PM emissions are treated as unspecified species, doesn't CMAQ internally distribute primary coarse PM emissions into the model species ASOIL and ACRS using a 90% / 10% split?

Yes, that's true and this is now explained in the text (line 194).

Page 1462, line 26: Are the EC/OC emissions from the GFED dataset representative of the wildfire activity in the modeling domain for the years 2000 and 2001 simulated here, or do they represent long-term averages? If so, what are the implications for the results presented in Section 3.3?

The GFED data set for the year 2000 have been taken for both years 2000 and 2001. When the model runs were performed the data for 2001 was not available. The data represents monthly averages. Also the anthropogenic emissions were kept the same in 2000 and 2001, only the weekly cycles were adapted to the correct weekdays in 2001. The advantage of this approach is that changes in the concentrations can be more easily interpreted as driven by meteorology. It has been clarified in the text that all emissions were kept the same in both years (line 211).

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Page 1565, lines 11-25: Did you attempt to evaluate the model for monitors that are located in urban/suburban areas, at least for modeling results from the finer inner domain?

I restricted myself to the EMEP monitors that are of known quality and that better represent data from a larger area than other monitors (e.g. from AIRBASE) do. Monitors in urban or suburban areas often show quite high observations which is caused by a high impact of local emissions. Even on the 18 km scale the model is in most cases not able to reproduce such effects. If large differences are observed at one of the monitors it is diffcult to decide whether this is due to the poor model results or due to the special location of the monitor.

Page 1566, line 1: I suggest adding a column to Table 1 identifying the network to which each monitor belongs.

I introduced an additional column in Table 1.

Page 1567, line 2: please also provide the relative uncertainty of the AOD measurements in percent

The relative uncertainty has been added in the text (line 365).

Page 1467, line 25 / Table 3, also Tables 5-9: Given that a log-normal distribution best fits the data for both surface-level PM and AOD, it would be more appropriate to present differences in geometric means and standard deviations rather than arithmetic means and standard deviations since the latter are better suited to compare properties of regular normal distributions.

This is certainly true and I want to emphasize with the paper that it must be taken into account that statistics dealing with aerosol mass densities in the atmosphere should better be given with median (or geometric mean) than with mean values. However, most authors use mean values and for easier comparisons of these results with the

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literature, mean values were also provided here first. Geometric means and geometric standard deviations have been added in Tables 3, 5, 6, 7, 8 and 9.

Pages 1467-1472, Sections 3.2-3.3, also Section 4 (conclusions): Please discuss your findings in the context of the numerous studies that have been published on evaluating CMAQ PM2.5 simulations over North America (e.g. Tesche, T.W.; Morris, R.; Tonnesen, G.; McNally, D.; Boylan, J; Brewer, P. CMAQ/CAMx annual 2002 performance evaluation over the eastern US. Atmos. Environ., 2006, 40, 4906-4919; Boylan, J.W., and A.G. Russell. PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models. Atmospheric Environment 2006; 40, 4946-4959; Zhang Y, P. Liu, A. Queen, C. Misenis, B. Pun, C. Seigneur and S.-Y. Wu. A comprehensive performance evaluation of MM5-CMAQ for the summer 1999 Southern Oxidants Study episode-Part II: Gas and aerosol predictions. Atmospheric Environment 2006; 40: 4839-4855; Eder, BK, and S. Yu. A performance evaluation of the 2004 release of Models-3 CMAQ, Atmospheric Environment 2006; 40, 4811-4824; Mathur R., S. Yu, D. Kang, K. L. Schere (2008), Assessment of the wintertime performance of developmental particulate matter forecasts with the Eta-Community Multiscale Air Quality modeling system, J. Geophys. Res., 113, D02303, doi:10.1029/2007JD008580; McKeen S., et al. (2007), Evaluation of several PM 2.5 forecast models using data collected during the ICARTT/NEAQS 2004 field study, J. Geophys. Res., 112, D10S20, doi:10.1029/2006JD007608.).

I read these interesting publications carefully and included some comparisons to the CMAQ results in Europe in the text. This concerns particularly the sections 3.2 (line 500), 3.3 (line 630) and 4 (line 686) .

Page 1468, lines 18-21, also page 1475, lines 18-21: Please provide more details on the criteria used for deciding to use nine classes in this analysis. Were the same class definitions used for each station, or were the class boundaries adjusted for each site? Were the results sensitive to the choice of the number of classes and the width of the

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particle size classes?

There are some explanations in the text without going into the last details. The number of classes is a compromise between a high resolution with many classes and a minimum number of five elements per class. Therefore the number of classes also depends on the total number of elements. I kept the number of classes constant but I adapted the range in which the classes are found from case to case. That is written in the text (line 460). The results are somehow sensitive to the number of classes, but usually not in terms of passing the test or not passing it. For example the modelled PM10 values at CH02 and CH03 would not have passed the χ^2-test if a different number of classes would have been chosen. Additionally I chose two tests (not only one) to avoid an erroneous interpretation of one possibly misleading test result.

Page 1469, lines 19-29: I do not follow this argument. First, the author states that the differences in bias are relatively small in general and that there are stations with both increased and decreased bias. I do not think that this result justifies the conclusion that "this result confirms the assumption that a better model resolution gives more reliable results". I recommend to either remove this statement or expand the analysis of model results from both grids to investigate the effect of grid resolution further. The results presented now are inconclusive as to whether increased grid resolution improves performance or not. In line 25, I suggest replacing "improvements" with "theoretical improvements" because none of the results presented in this study actually show that systematic improvements occurred as a result of higher grid resolution.

This is now explained in more detail in the text. At two coastal stations a reduced bias is observed. This can be attributed to the fact that a lower influence of open sea grid cells in the 18 km grid enhances the modelled aerosol mass and by this reduces the bias. The opposite behaviour is observed at measurement stations in higher altitudes. In the 54 km grid the bias at these stations (particularly DE04) is low because the measured aerosol concentrations are relatively low. They represent higher altitude

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conditions that are not reflected in the model with the coarse grid resolution. In this case the bias is low, but for the wrong reason. The modelled values will decrease in the finer grid, because the orography effect is now better considered and therefore the bias will increase. The model shows the correct effect but it doesn‘t improve the model performance.
In line 25 “improvements” has been replaced by “the reduced bias at some stations” (line 527).

Page 1470, lines 16-19: This ranking should be contrasted to findings from North American studies where OC/EC were often found to also play a significant role in total PM2.5 concentrations. As discussed above, this different ranking may be related to the primary PM2.5 speciation profile used in this study, particularly the low fractions for EC and OC emissions.

Following PM measurements in Europe, OC also plays an important role and it is obviously too low in these model simulations. It is difficult to decide whether this is due to the low primary emissions or due to the low SOA formation. Secondary organic aerosols are again influenced by precursor emissions, that might be too low and by aerosol formation from precursors, that might be underestimated as well. A paragraph on the importance of organic aerosol was added at the beginning of section 3.3 (line 567).

Page 1471, line 25-Page 1472, line 1: Figure 6 is missing from the manuscript. Instead, Figure 5 is repeated and labeled erroneously as Figure 6. It is also not clear which observed quantity was compared to which model quantity. How was the observed “not specified” aerosol calculated-total PM10 minus (sulfate+nitrate+ammonium)? For the model data, doesn’t CMAQ internally distribute primary coarse PM emissions into the species ASOIL and ACRS? If so, shouldn’t they be included into the model definition of “remaining modeled aerosol”? The legend of Figure 6 mentions “dust”, but it is not clear if this refers to ASOIL.

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I am sorry that Figure 6 was not correctly given in the manuscript. I missed that error while reading the proofs.
The observed quantity is total PM10 - (sulfate + nitrate + ammonium + sodium + chloride). The same has been calculated from the model results. This has now been specified in the text. This means that OC, EC, soil and other coarse aerosol mass (as dust) is included in this "not specified" component. "Dust" means ASOIL, this has now been specified in the figure caption.

Page 1471, lines 27-28: Does the author refer to a missing biogenic source of primary organic aerosols (a pathway currently not included in CMAQ) or missing pathways of organic particle formation from gaseous biogenic precursor gases? The former would indeed be an emission inventory problem, while the latter is more related to deficiencies in the formulation of the CMAQ secondary organic aerosol module. Please clarify.

This refers to both. Primary biogenic aerosols are not considered in CMAQ and secondary biogenic aerosol formation might be underestimated in the current SOA module. Last not least biogenic precursors might also be underestimated in the emission inventory. This has been clarified in the text (line 623).

Page 1472, lines 11 -13: Again, I urge the author to refer to results reported in North American studies for comparison (e.g. Dazhong Yin, Weimin Jiang, Helmut Roth and Eric Giroux, Improvement of biogenic emissions estimation in the Canadian Lower Fraser Valley and its impact on particulate matter modeling results, Atmospheric EnvironmentVolume 38, Issue 4, , February 2004, Pages 507-521.; Betty K Pun, Shiang-YuhWu, Christian Seigneur, John H Seinfeld, Robert J Griffin, Spyros N Pandis, Uncertainties in modeling secondary organic aerosols: three-dimensional modeling studies in Nashville/western Tennessee., Environ Sci Technol. 2003 Aug 15;37 (16):3647-61; Tesche, T.W.; Morris, R.; Tonnesen, G.; McNally, D.; Boylan, J; Brewer, P. CMAQ/CAMx annual 2002 performance evaluation over the eastern US. Atmos. Environ., 2006, 40,

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4906-4919; J. Chen, R.J. Griffin, P. Tulet, and A. Grini, Modeling secondary organic aerosol formation through cloud processing of organic compounds, Atmos. Chem. Phys., 7, 5343-5355, 2007.)

Most of the work has been cited and discussed in the text (line 629).

Page 1476 line 20: When mentioning "aerosols of biogenic origin", does the author refer to a missing biogenic source of primary organic aerosols or missing pathways of organic particle formation from gaseous biogenic precursor gases? Please clarify.

Both. It has been clarified in the text (line 647).

Page 1477, line 7: Please elaborate-why do the correlation coefficients demonstrate that the dominant emission sources are correctly located? I agree with the statement that the main transport patterns appear to be captured, but I do not follow the argument for the location of emission sources, especially given that organic aerosols are a complex mix of primary and secondary particles and the model underestimates their total mass by a factor of three.

Measured aerosol concentrations are a result of emissions, chemical transformation and atmospheric transport. If the location of a dominant emission source would be wrong, the atmospheric transport, assuming it is correctly considered, would result in high or low concentrations at the wrong place or at the wrong time. A systematic shift in time would lead to low correlation coefficients, therefore both, the transport and the location of the emission source, must be correct to yield high correlation coefficients. The underestimation of the total mass means that the SOA formation rate is too low or the emission strength is underestimated.
It was not clear in the text that he sentence "The time series showed reasonable correlation coefficients between 0.47 and 0.81 ..." referred to the inorganic aerosol mass. The IA components show a relatively low bias and therefore I think that hte statement "that the dominant emission sources are correctly located" is true. I clarified in the text

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that this is not meant for the organic aerosol (line 865).

Page 1478, lines 25-26: Please include a discussion of findings from CMAQ applications in North America.

I added a paragraph in the conclusions (line 886).

Page 1488, Table 4 and Page 1496, Table 10: The header for the K-S test results should be "K-S", not "a". Furthermore, I suggest using bold font for those stations and observed/modeled distributions where their chi-square or K-S value exceeds the threshold. This would make it easier to quickly spot deviations from the assumed log-normal distribution.

The "a" is often used in the literature as the relevant threshold parameter for the K-S test. This has not been explained in the text and I modified the table as suggested.

Technical corrections: Page 1467, line 16: typo "aersol", please correct. Page 1478, line 21: typo "aersol", please correct. Page 1501, Figure 6: This is the same Figure as Figure 5. The actual Figure 6 is missing.

Corrected.

I thank the reviewer for his/her valuable comments that improved the manuscript considerably. Particularly, I hope that it can contribute to a fruitful discussion about CMAQ applications in the US and in Europe.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1457, 2008.