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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

Anonymous Referee #2

Received and published: 13 March 2008

General comments:

The author presents an analysis of PM10 and AOD simulations from an innovative model application over Europe. To my knowledge, this is the first time the CMAQ model has been applied for a multi-year simulation over Europe. The design of the modeling study is sound, covering two full years thereby facilitating an evaluation of model performance on timescales ranging from daily to seasonal. The setup of all components of the model simulation (meteorological modeling, emissions processing and photochemical modeling) reflects best modeling practices and is described well in the manuscript.

The analysis presented in the manuscript goes beyond simply summarizing model performance through basic statistics such as bias and correlation coefficients as is often done in other studies. In particular, I find the comparison of various aspects of the observed and simulated distributions illustrative and hope it will be used more widely in model evaluation studies. I also commend the author for examining both surface level PM10 and column total AOD values simulated by CMAQ.

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.

Overall, I recommend publication of the manuscript after addressing my comments below.

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.

Page 1462, lines 6-9: What is the justification for this speciation of PM2.5 emissions?

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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 compounds appear to account for only 3% of PM2.5 emissions in this study. In urban areas, 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 60%/30% with only small contributions from 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.

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?

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?

Pages 1462-1465, Sections 2.2-2.4: These sections are well written and reflect a carefully designed modeling study.

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?

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

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

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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.

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.).

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 particle size classes?

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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.

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.

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.

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.

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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-Yuh Wu, 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, 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.)

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.

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.

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

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

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threshold. This would make it easier to quickly spot deviations from the assumed log-normal distribution.

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.

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