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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of a three-dimensional chemical transport model (PMCAMx) in the European domain during the EUCAARI May 2008 campaign" *by* C. Fountoukis et al.

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1. *This manuscript presents a comparison of PMCAMx regional air quality model to EUCAARI AMS measurements in May 2008. Overall, the model is capable of reproducing the relative contributions of sulphate, organics, nitrate and ammonium to submicron aerosol mass, and predicts their absolute concentrations with a reasonable accuracy. Organics (especially oxygenated species) are found to be the dominant component in Northern and Central Europe. PMCAMx is applied to a European domain for the first time. The methodology used in this study is state-of-the-science, the results and*

assumptions are clearly presented and the manuscript is well-structured and mostly well-written. My main criticism is that the model is evaluated only against a very limited data set (one month and 4 ground stations + airborne measurements), which is hard to justify since much more extensive AMS data is available from the EUCAARI campaign. Since the model is compared only against May 2008 data, its performance in other seasons cannot be evaluated and AMS measurements from several other sites cannot be utilized. I therefore recommend that the authors run the model at least for one full year, if not for both 2008 and 2009, and use all available AMS data from this period. This should not be computationally too demanding to do. Another option is to combine this manuscript with the forthcoming one presenting a comparison between modeled and PMF-analysed AMS organic aerosol components – I cannot help thinking that this latter option might have been the authors' initial intension since the current introduction would fit an OA component paper much better.

We thank the reviewer for her good overview of the contents of the paper. We are indeed planning to run the model for the winter EUCAARI intensive measurement period (January/February) during which AMS measurements were taken in a number of European stations. There were no airborne measurements for this winter period. The two periods (summer and winter) are quite different as the first is dominated by photochemical processes while the second by primary emissions. We considered discussing both in the same paper, but the result is not satisfactory. The current paper focusing on just one period includes 26 individual graphs and 4 Tables, so it already tries to analyze a lot of information. Even more important there are some serious issues with the winter emission inventory (related to wood burning and ammonia emissions) that require lengthy discussion, sensitivity studies, etc. We are working in dealing with these issues and performing the required emission sensitivity tests. Trying to present the results of both periods would result in a very lengthy paper with approximately 50 individual graphs that would consist of two very different parts. We would prefer not to follow this path. The available EUCAARI AMS data do not cover a full year and of course not two. There are some additional AMS measurements collected during an

EMEP campaign (approximately a month long) in the fall of 2008.

For the scientific aspect of our evaluation, we are using around 1500 measurements for each PM component (for a total of approximately 8500 data points). This is the most extended evaluation, focusing on particulate matter, of a regional CTM in Europe at least to the best of our knowledge. While use of more measurements is always useful this is a large enough sample (statistically) for the model evaluation during photochemically active periods. Therefore our conclusions are scientifically valid.

The recommended detailed comparison with the results of the PMF analysis is obviously an important additional test of the model. Unfortunately this analysis has not been finalized and the results have not been published. Our intension is to perform a detailed comparison with all the PMF results (summer and winter) as soon as the complete final dataset becomes available.

Specific comments:

2. *If the authors decide to extent the current manuscript to cover a full year/two years, the introduction needs to be rewritten. Currently it focuses solely on organic aerosol (and to even specific OA components) while the focus of this manuscript is on general PM1 composition.*

In the second and third paragraph of the introduction the focus is indeed on organic aerosol and its components. This was mainly done to introduce (in the third paragraph) the VBS approach that the current version of PMCAMx includes for both POA and SOA as this is one of the main advantages of the model and is worth-mentioning. However, we understand that this might confuse the reader as to what follows in the manuscript and thus we have now revised the second paragraph of the introduction excluding references to techniques (e.g. the PMF method) that are not used in the following sections of the manuscript. We have also added some material about the rest of the aerosol components to balance this part of the paper.

3. *p. 14190, l. 15-16: Where are the boundary conditions obtained from?*

The boundary conditions are based on measured average background concentrations in sites close to the boundaries of the domain (e.g. Zhang et al., 2007; Seinfeld and Pandis 2006). This is now explained in the text.

4. *p. 14195, l. 11: Provide an explanation why the aerosol composition in the Mediterranean area behaves differently from the rest of the Europe.*

The major difference in the Mediterranean area is the higher photochemical activity during that period combined with higher aerosol sulfate concentrations (compared to the OA levels). These higher sulfate levels are due to the long-range transport of sulfur dioxide and the resulting sulfates originating from industrial areas of eastern Europe and the Balkans (Sciare et al., 2003; Lelieveld et al., 2002; Ganor et al., 2000; Mihalopoulos et al., 1997) plus additional contributions of sulfur dioxide from shipping and DMS from phytoplankton. The intense sunlight and photochemistry in this area convert rapidly the transported sulfur dioxide from less photochemically active areas to sulfate.

5. *Table 3: Several of the statistical metrics are never mentioned in the text (apart from their formulae) and thus this table can be substantially condensed. Why some metrics values are not shown for nitrate in Finokalia?*

We have now included in the discussion the FERROR and FBIAS metrics which were not previously discussed. Having a comprehensive set of metrics is useful in future comparisons of the performance of different models.

Measured nitrate concentrations at Finokalia were often below the detection limit and thus a calculation of NMB, NME, FBIAS, FERROR and the “percent within a factor of 2” for nitrate would be misleading. This is now explained in the revised manuscript.

6. *If the authors decide to extent the current manuscript to cover a full year/two years rather than combine it with organic component paper, Figure 5 should be extended to all four sites. Filter measurements shown in Figure 5 should be discussed in the text or omitted from the figure.*.

We have followed the reviewer's suggestion and included the filter measurements in the discussion of the revised paper. Presenting all the inter-comparison figures (at least 4 for each site) requires 16 graphs that add little new scientific information or new conclusions. Repeating this for multiple seasons will require tens of additional graphs.

7. *p. 14198, l. 11: "sulfate lay within the error". What error?*

Corrected to: ". . .lay within the 1:2 and 2:1 error lines."

8. *p. 14198, l. 14: While the OA description in PMCAMx is fairly sophisticated compared to many other models, it is still greatly simplified. Could this be a possible explanation to the bias?*

Indeed, possible sources of the bias include the volatility distribution of the primary organic aerosol emissions, the zeroth order parameterization of the chemical aging of the semivolatile OA, etc. Bias could also be introduced by emission errors in either the anthropogenic or biogenic emissions, errors in the meteorological input, etc. Some discussion of these issues has been added to the revised manuscript.

References

Ganor, E., Foner, H. A., Bingemer, H. G., Udisti, R., and Setter, I.: Biogenic sulphate generation in the Mediterranean Sea and its contribution to the sulphate anomaly in the aerosol over Israel and the Eastern Mediterranean, Atmos. Environ., 36 (6), 929-938, 2000.

Lelieveld, J., Berresheim, H., Borrmann, S., Crutzen, P. J., Dentener, F. J., Fischer, H., Feichter, J., Flatau, P. J., Heland, J., Holzinger, R., Korrmann, R., Lawrence, M. G., Levin, Z., Markowicz, K. M., Mihalopoulos, N., Minikin, A., Ramanathan, V., de Reus, M., Roelofs, G., J., Scheeren, H. A., Sciare, J., Schlager, H., Schultz, M., Siegmund, P., Steil, B., Stephanou, E. G., Stier, P., Traub, M., Warneke, C., Williams, J., and Ziereis, H.: Global Air Pollution Crossroads over the Mediterranean, Science, 298 (5594), 794-799, DOI:10.1126/science.1075457, 2002.

Mihalopoulos, N., Stephanou, E., Kanakidou, M., Pilitsidis, S., Bousquet, P.: Tropospheric aerosol ionic composition in the Eastern Mediterranean region, Tellus B, 49 (3), 314-326(13), 1997.

Sciare, J., Bardouki, H., Moulin, C., and Mihalopoulos, N.: Aerosol sources and their contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea during summertime, Atmos. Chem. Phys., 3, 291–302, www.atmos-chem-phys.org/acp/3/291, 2003.

Seinfeld, J. H., and Pandis S. N.: Atmospheric Chemistry and Physics, John Wiley, New York, 2006.

Zhang, Q. Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H. Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y. M., and Worsnop D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, Geophys. Res. Lett., 34, L13801, doi:10.1029/2007GL029979, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14183, 2011.