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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First comparison of a global microphysical aerosol model with size-resolved observational aerosol statistics" *by* D. V. Spracklen et al.

Anonymous Referee #1

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The paper presents a comparison of modeled aerosol size distributions calculated with a global chemical-microphysical model driven by analyzed meteorological data with averages of measurements selected to be representative of the model conditions, but not at the specific times and locations for which the model was run. The focus is on sulfate and sea salt. The model has previously been described and numerous comparisons with observations already presented; this paper focuses on size distributions. Several sensitivity studies are conducted, also focusing on size distributions. The paper is deserving of publication, but I raise a number of points that the authors may wish to address. These are of two categories, one focusing on the philosophy and utility of the sorts of statistical comparisons that are presented here. I also raise questions about

specific formulations of emissions and new particle formation employed in the model. I have looked at the published comments of Reviewer 2 and by and large concur in them, so in several instances have not repeated similar comments. The paper is suitable for publication after the authors have responded to comments and modified the manuscript as they see fit.

General comments

1. This paper, and the earlier papers in this series (Spracklen et al. (2005a, b) describing the model and the results of the calculations represent an important but hardly unique step in the right direction, namely driving an aerosol chemical transport model with aerosol microphysics by observationally derived meteorological data, specifically in this instance the European Centre for Medium-Range Weather Forecasts (ECMWF) analyses at 6-hourly intervals. In principle this approach permits detailed comparison with observations not just at the same location as in the model but importantly at the same time. To my thinking such an approach is absolutely essential. Variability in winds, clouds, and precipitation is responsible for much of the variability of concentrations and properties of atmospheric aerosol constituents; the balance is due to variability of emissions, of which a good fraction (seasalt, mineral dust) are again controlled to great extent by meteorological variability. For example here emissions employed for volcanic sources are 25 year averages, rather than emissions for specific dates.

2. In the past it has been all too common to compare, say, monthly average concentrations of measured and modeled aerosol constituents and assess the quality of the model by agreement within the standard deviations. Unfortunately the standard deviations are usually so large (often a factor of two or more) that you can drive a truck through them. In considerations of air quality or radiative forcing of climate change where a factor of two looms large in its implications, such an approach is hardly going to yield an assessment of model skill that is of much use in these contexts or that presents a challenge to the modelers (and measurers) to improve their skill. It was thus with some anticipation that I agreed to review this paper.

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3. That anticipation was reinforced by language in the paper such as the following: "The principal advantage of a global model over a box or column model is that it naturally takes account of the spatial and temporal changes in aerosol in the MBL and overlying FT driven by variations in meteorology." (page 8875, lines 7-10)

4. Unfortunately despite the title and the expectation that it engendered, the present paper reverts to a statistical comparison of observed and modeled quantities, rather than a point by point comparison at specific times. Thus "In this paper we use statistical analyses of observed remote MBL aerosol (Heintzenberg et al., 2000, 2004) to carry out a comprehensive evaluation of the factors controlling its properties." (page 8876, lines 6-8). For this reason the paper is somewhat of a disappointment, though I certainly do not feel that this should stand in the way of publication. However the authors might wish to speak to the consequences of their statistical approach rather than the comparison at specific times and places as others have done (e.g., Yu et al., 2003).

5. The paper makes it clear (page 8876, line 23) that the baseline calculations presented here are restricted to sea salt and sulfate. Results of several studies are presented that progressively introduce additional species or processes.

6. In that context the paper wavers occasionally from its stated mission to compare with observations, for example in Figure 10, which is more of a sensitivity study than a comparison with observations.

7. That said, with the recognition that the comparisons are statistical, the comparisons shown in Figure 2a should be taken as encouraging. I found it difficult to compare the contour diagrams in Figure 2b; perhaps this figure would be more effective if the (same) color scheme were used for modeled and observed, in two panels, rather than the superposition given. In panel a, my hunch is that the Heintzenberg climatology was for size without resolution of composition. But the model gives composition, so it would be interesting to see what fraction is sulfate, what fraction sea salt.

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8. I would concur with the authors in their explanation of the deep minimum in modeled number concentration (page 8884 line 10-15): Another difference between the model and observations is in the minimum between the Aitken and accumulation modes (apparent in Fig. 2). The modelled number concentration at the minimum is too low. The deep minimum is caused by the use of a fixed activation diameter (50 nm) during cloud formation in the model. In reality, this activation diameter varies according to variations in updraft velocity as well as the shape of the particle size distribution.

9. I find the comparisons in Figure 3 somewhat less encouraging especially in that in some instances the model exceeds observations by several fold, and in some instances vice versa; again, however, this could result from statistical rather than point to point comparisons. The reasons for this are not known and while the authors offer several speculations, none of them seems supported by any modeling results. Again the model might inform the comparisons by explicitly distinguishing the seasalt and sulfate in the "standard" results.

10. In contrast I find the comparison in Figure 7 a and b rather encouraging, again within the limitations of the measurements and model corresponding to different times and places.

11. Figure 9 is a novel and useful comparison of temporal autocorrelation of size dependent particle concentrations

12. The authors appropriately question the emissions size distributions from the various published formulations (Gong, Monahan). The recent book by Lewis and Schwartz sheds some perspective on the uncertainties in these emissions.

13. One final concern I would note in the paper is the use of the Kulmala et al. 1998 binary parameterization for sulfuric acid nucleation. The updated parameterization (from the same Helsinki group) is in Vehkamaki et al. 2002, who state that "The differences [between these two parameterizations] can be explained by several approximations in the old parameterization which are partly erroneous in the kinetic part". The authors

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might wish to comment on the consequences of the choice of parameterization.

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Lewis E. R. and Schwartz S. E. Sea Salt Aerosol Production: Mechanisms, Methods, Measurements, and Models – A Critical Review. (American Geophysical Union, Washington, 2004).

Vehkamaki, H., M. Kulmala, I. Napari, K. E. J. Lehtinen, C. Timmreck, M. Noppel, and A. Laaksonen, An improved parameterization for sulfuric acid -water nucleation rates for tropospheric and stratospheric conditions, J. Geophys. Res., 107(D22), 4622, doi:10.1029/2002JD002184, 2002.

Yu, S., Kasibhatla, P., Wright, D., Schwartz, S., McGraw, R., and Deng, A.: Moment-based simulation of microphysical properties of sulfate aerosols in the eastern United States: Model description, evaluation, and regional analysis, J. Geophys. Res.-Atmos., 108, 4353, doi:10.1029/2002JD00289, 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 8871, 2006.