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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Attribution of Modeled Atmospheric Sulfate and SO_2 in the Northern Hemisphere for June–July 1997" *by* C. M. Benkovitz et al.

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As noted by the authors, this paper is a contribution to the growing body of knowledge indicative of the long range transport of submicrometer aerosols. Although this is a worthy goal, I think the authors need to strengthen the paper in a number of respects before this contribution should be published.

We thank the referee for his/her comments and respond briefly below.

Comment: My main reservations concern the very limited time period for which this study is conducted - with just 4 weeks of results. As noted by the authors, other papers have dealt with annual or multi-annual time-scales. Indeed, many of the authors were involved in the three-year simulation of Rasch et al. (2000). Six years later, I did not

see convincing arguments that the current study of a four week period extends upon the results of annual or multi-annual simulations. The authors may have good reason to suppose that this 4-week period is representative in some way, in this case they need to make this argument.

Response: We ran a six-week simulation (including two week spin up) because both the meteorological data used to drive the model and the measurements used in evaluation of model performance represent the actual conditions for the simulation period. Carrying out modeling runs and model evaluations in this way places a high demand on computational and personnel resources; for example carrying out roughly 5000 comparisons of modeled and observed sulfate and 12000 such comparisons for SO2 in the original study required assembling data from numerous sources, conversion of units and time bases, and the like. .The period of the simulation was chosen because the Aerosol Characterization Experiment II conducted during this period provided additional highly time-resolved data sets with which to evaluate the model, see Benkovitz et al. (2004) (B04). In the current manuscript we do not claim that the results are representative for any but the modeled time period; consequently the specific conclusions reached for this time period cannot be extended to other time periods or be taken as representative of, say, an annual average. Part of the motivation for modeling and comparing specific time periods is the high interannual variability of, for example, monthly averages that limits the utility of comparisons of, say, model calculations for one April and measurements from April of a different year or model calculations with GCM-generated meteorology.

Comment: Further, if focusing on such a limited period, I think the links between meteorology and concentration/burden need to strengthened, perhaps with the use of trajectory analysis.

Response: We have discussed several of the links between meteorology and concentration in the paper. We did not use trajectory analysis because a) our emissions and target areas are large and thus not suitable for this type of analysis, b) the accuracy of

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trajectory analyses decreases markedly after ~ 5 days, and c) trajectory analysis using different assumptions and methodologies can yield different results. The animations of the column burdens of SO2 and sulfate taken from the model output at 6-h intervals, available at http://www.ecd.bnl.gov/steve/model/junejuly97.html, are an effective way to follow the transport of air masses containing high amounts of these substances.

Comment: 1. p. 4026, line 28. Explain why the Graf study found similar fractional contributions of volcanic and anthropogenic emissions.

Response. Although Graf et al. (1997) do not explicitly address why their model yielded similar fractional contributions of sulfate from volcanic and anthropogenic emissions, several potential reasons can be identified. As noted by Graf et al., SO2 released from volcanoes into the free troposphere exhibits a longer atmospheric residence time than that emitted into the planetary boundary layer from anthropogenic sources, mainly because of a lower dry deposition rate, resulting in a greater sulfate yield (fraction of SO2 emissions that is converted to sulfate). Similarly, the sulfate formed from this SO2 might also be expected to exhibit a greater residence time, relative to wet deposition, than sulfate in the boundary layer. Graf et al. do not present the yield or the residence times of sulfate from the several source types that would permit quantitative attribution of the reasons for the greater amount of volcanic sulfate than anthropogenic sulfate, relative to emissions.

Comment: 2. A lot of the material here is taken directly (or almost) from Benkovitz et al. (2004) (B04). Figures 1 is identical, to B04's Fig.1, and Figure 2 almost identical to B04's Fig.4. Why give an Appendix with model description here when the model is described in B04? Table A1 here is modified from Table 1 of B04, but I actually found the B04 version clearer. The nomenclature used here with NA, Eu, etc., is explained in the text anyway. I would simply remove this material, unless it really is adding something new?

Response: The material in this paper that is similar to B04 was included to give an

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overview of the modeling that is the basis of this paper without having to refer to (B04).

Comment: 3. It would help very much if the symbols in Table A1 were replaced by proper numbers - giving the various budget terms. Although colour maps are illustrative, it also helps the reader to have Tables with numbers to refer to.

Response: The table is meant simply to identify the modeled species. Budgets (emissions, burdens, conversion rates by process, removal rates, etc.) of the several species were discussed in B04.

Comment: 4. p. 4028, lines 5-6. Indicate the length of the time-period over which the measurement comparison was done.

Response: As stated in the paper (page 4027 line 25) “A six-week simulation was performed; the first two weeks were considered model spinup time and results were not analyzed.”

Comment: 5. p. 4028, lines 10-11. The text here states that a substantial fraction of model observation differences was due to subgrid and/or measurement error. Was there no sign of substantial model error? No indication of problems with the model?

Response: Perhaps we should have explicitly indicated that the balance of the difference must be attributed to model error.

Comment: 6. The model seems to be using “labelled” S atoms to keep track of different source regions. Is this right? - I didn’t find it explicitly stated.

Response: As explained in Appendix A and summarized in Table A-1 the model tracks sulfur species according to source region, type of emission (biogenic, anthropogenic SO2, anthropogenic sulfate, volcanic SO2), and, for sulfate, (conversion process: primary, gas-phase oxidation, aqueous-phase oxidation).

Comment: 7. The 2-letter abbreviation for Europe (Eu) will be easily mistaken for the European Union (EU). I would suggest Eur instead. And why not Asia instead of As?

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Why save 2-letters?

Response: We trust that the abbreviations, used to make the paper less verbose, will lead to no confusion.

Comment: 8. p. 4030. The discussion of meteorology is limited to just one week which the authors state is representative. Still, the only figure of sulfate burden is presented for 29th June, while the meteorology is presented for 23-27th.

Response: In an effort to extend the applicability of our conclusions beyond the specific time modeled we noted that the circulation pattern toward the end of June 1997 is similar to the climatological mean from 1979-1995 with a slight shift eastward of the center of the main high pressure centers. The major features discussed in section 3 of the text are all related to the main high- and low-pressure centers present in the climatological map from the Climate Prediction Center found at (http://www.cpc.ncep.noaa.gov/products/precip/realtime/clim/annual/monthly/monthly.12.slp.html). The sulfate burden for the 29th of June was influenced by the meteorology of the previous days, not solely by the meteorology of the current date.

Comment: 9. 3-D trajectories would have been more instructive to understand the transport patterns, preferably in association with some of the sulfate burden figures shown later in the manuscript.

Response: See response to second unnumbered comment above.

Comment: 10. Summary and implications. This text does not mention the length of the simulation period.

Response: The reviewer is correct that that information is important and should be included in the summary.

Comment: 11. As well as a discussion of the limitations of a 1-month analysis, I miss here a discussion of the robustness of the results. How do these model results fit into those of other global S models, for example of the eleven models of the COSAM

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exercise (Roelofs et al., Tellus, 53B, 673-694, 2001)? This paper showed that current global models differ strongly in a number of features. What implications do such studies have for the conclusions presented in this paper?

Response: These questions are addressed in the Discussion section of B04; Table 10 of that paper summarizes quantities such as yields, conversion rates, deposition mechanisms, and residence times for the several species from our model and eight other models. One implication of the comparison is that modeled amounts of materials may differ substantially less than rates and fluxes, which can differ by more than a factor of 2. To some extent these differences may be due to different time periods modeled, but a concern is that the models may be getting the “right” results, that is results that agree with observations, for the wrong reasons. Confidence in the models requires detailed examination of processes and rates, not simply comparison of modeled and observed concentrations.

References Benkovitz, C. M., S. E. Schwartz, et al. (2004). "Modeling Atmospheric Sulfur over the Northern Hemisphere During the ACE-2 Experimental Period." J. Geophys. Res..109, 1029/2004DJ004939.

Graf, H.-F., J. Feichter, et al. (1997). "Volcanic Sulfur Emissions: Estimates of Source Strenght and Its Contribution to the Global Sulfate Distribution." J. Geophys. Res. 102(D9): 10727-10738.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 4023, 2006.