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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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The article by Benkovitz et al. is an interesting paper that describes the relative importance of different source regions and different sulfate production mechanisms on the sulfate burden at various locations across the northern hemisphere. The tagging method to determine source regions and production mechanisms is a powerful way of analyzing the importance of these factors. I like this article because of the in-depth analysis that includes meteorological aspects. Plus it enlightens the reader as to possible strategies to address visibility problems at specific locations. The paper is well written and is nearly ready for publication. Some specific questions for discussion follow.

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

Comment 1: Page 4032, section 4: The authors choose 3 locations, Seattle, WA, USA, Sagres, Portugal, and Barbados, to study. While these 3 locations do exhibit influence of sulfate from different source regions, I am surprised that a location in Asia exhibiting influences from both Europe and Asia was not chosen. Examination of Figure 9 suggests that European sources are 10-20% of the sulfate column burden in eastern Asia. Do theauthors think there is substantial European influence in Asia?

Response. Yes we do. Figure 4, which summarizes for the period simulated the influences of the different source regions on the sulfate column burden, shows that the influence of Eu sources in the sulfate burden of northern and western Asia was ˜ 40 to 80%. The animations of the column burdens of sulfate and SO2 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 sulfate and SO2 from Eu to As. Further, in Section 3 the meteorological discussion describes the conditions which favor transport of sulfur from Eu sources over Asia.

Comment 2. Are the locations chosen for this study "extreme" examples, or are they representative for their region (i.e. is Seattle representative of the North American west coast, Sagres representative of the west coast of Europe, and Barbados representative of the Caribbean)?

Response. The locations were chosen based on the results shown in Figure 4 to discuss the mechanisms that allowed far source areas to be considerable contributors to the sulfate burden. Seattle is a representative location in northwestern North America where the fractional influence of As sources on sulfate column burden is large ($>$ 40%); however this influence decreases as one moves eastward, especially east of emission sources of SO2. Sagres was chosen as a representative costal European site where the fractional influence of North American sources on sulfate column burden was ˜ 40%; again, on the western edge of a continent This location is more likely to exhibit a

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higher fractional influence from NA sources than a source that is more subject to local emissions. Barbados was chosen as a trade winds location where the influence of Eu sources was ˜ 40%.

Comment 3. Figure 9. It would be helpful to zoom into the region of interest (continental scale instead of global scale). The printed, ACP formatted version of this article results in fairly small panels making it difficult to see a plume reaching Seattle.

Response. Actually one of the advantages of publishing in ACP is the high quality rendering of the figures in PDF format. By looking, for example, at figures 9 a-c at 1200% magnification one can clearly discern the swirls of higher or lower sulfate column burden at the several time periods. This detail is also readily discerned in the animations viewed at high magnification. We invite the referee (and others) to have a look, especially at the animations, and hope that this format of presentation might stimulate some further discussion here and also might encourage others to present their model results as animations, as we have done in this study and previously (Benkovitz et al., 2001). Benkovitz C. M., Miller M. A., Schwartz S. E. and Kwon O-U. Dynamical influences on the distribution and loading of SO2 and sulfate over North America, the North Atlantic and Europe in April 1987. Geochem. Geophys. Geosyst. 2, doi:10.1029/2000GC000129 (2001); http://146.201.254.53/publicationsfinal/articles/2000GC000129/fs2000GC000129.html . There is really a wealth of detail in these animations.

Comment 4. Page 4036, attribution to production mechanism, and Abstract, line 18. The authors conclude for regions with infrequent clouds, e.g. deserts, that gas-phase oxidation can be dominant. This seems perfectly logical and perhaps obvious. I'm not sure why it is one of the major conclusions of the paper (i.e. why it made the abstract). There are only 2 major production mechanisms for sulfate. If one pathway is suppressed, than the other pathway would dominate. What makes this conclusion interesting and unique?

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Response. One of the results presented in this paper was that there can be strong spatial (and temporal) variability in the fraction of the sulfate that is formed by aqueous-phase reaction vs gas-phase reaction. This was what we are trying to point out in the abstract. We will examine this sentence again and revise it to better define our conclusion.

Comment 5. Referee #2 is concerned that the 4 week simulation period during June-July 1997 presents limitations in generalizing the results. While s/he makes a good point, I would argue that detailed analysis of specific time periods should be carried out. It must be recognized that these authors have done similar studies for other time periods (e.g. October-November) and a collection of these detailed analyses benefits our understanding of sulfate concentrations in the atmosphere. What's missing in this paper is 1) justification of the model configuration and integration compare to global models, especially to Rasch et al. (2000) and Barth et al. (2000) who similarly discuss the attribution of sources to sulfate.

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. We would also stress that the time period of the earlier study noted by the referee (Benkovitz et al., 1994) is not a generic October-November but is a specific October-November, namely 1986; it would not necessarily be appropriate, and in fact we think inappropriate, to draw conclusions about an average or typical October-November from a run carried out for the meteorology of a specific October-November.

Technical Details: Abstract, line 17. It seems appropriate to round up to 62%.

Response. It will be done.

p. 4026, line 14, state that the same, or similar, model is used in the current study.

Response. The introduction is summarizing previous results, and thus we think it would

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not be an appropriate place for this comment. The link to our previous work was discussed in Benkovitz et al., 2004; we will add a statement on this to Appendix A.

p. 4027, line 5, lighting should be lightning.

Response. Touché. This is exactly the sort of excess precision that we try to avoid. We thank the referee for noting this. It will be changed.

p. 4037, lines 18-29. This is a summary paragraph for section 5 presumably, but is actually summarizing sections 4 and 5. It seems to be in an awkward place.

Response. We will include a change to indicate that this paragraph summarizes Sections 4 and 5.

Figure 5, the panels need to be marked a, b, c, d, e, f.

Response. It will be done.

References

Benkovitz C. M., Miller M. A., Schwartz S. E. and Kwon O-U. Dynamical influences on the distribution and loading of SO2 and sulfate over North America, the North Atlantic and Europe in April 1987. Geochem. Geophys. Geosyst. 2, doi:10.1029/2000GC000129 (2001); http://146.201.254.53/publicationsfinal/articles/2000GC000129/fs2000GC000129.html .

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

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