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Atmospheric Chemistry and Physics Discussions

Interactive comment on “Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on global aerosol direct radiative forcing” *by* S. T. Martin et al.

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We thank Mark Jacobson for his attention to our work and his considered call for attention to his earlier publication.

To understand the work of Jacobson (2001) in relation to our manuscript, the reader should focus on Tables 2a, 2b, 2c, and 3 as well as sections 2.1, 2.2, and 2.3 of Jacobson (2001). Jacobson (2001) combines (a) digitized model-study maps from other publications, (b) estimates of ion mass loadings over typical continental regions, the NH and SH oceans, and the polar and midtroposphere regions, (c) reported mean boundary layer sea salt concentrations, and (d) addition of ammonia to one-half the amount of sulfate. These source data are interpolated to provide a latitude-longitude-altitude-seasonal grid. The Jacobson (2001) approach brings together models having differing emissions and transport schemes and makes an assumption regarding ammonium neutralization. The approach is very different from that adopted by us, which is an emissions description followed by fate and transport modeling in the atmosphere.

The approach of Jacobson (2001) leads to 47 chemical species, which are entirely internally mixed. A thermodynamic model is applied to those chemical species. The resulting prediction is for a dominant sulfate solid of CaSO4.2H2O(s), for a dominant ammonium salt of NH4Cl(s), and for a dominant nitrate solid of KNO3(s). We believe these predictions arise from a crossover in the internal mixing, e.g., NH4+ associated with SO42- is internally mixed with sea salt containing Cl-, so NH4Cl(s) results. Similarly, CaCO3 from dust combines with SO42- from industrial activities. While CaSO4(s) formation is important in localized regions such as the Mediterranean due to mixing of CaCO3-rich dust out of Libya with European sulfate, in general this extent of crossover is not so great in atmospheric particles. In summary, we totally disagree with the conclusions of Jacobson (2001) regarding the major solids in the troposphere.

Jacobson (2001) calculates global aerosol direct radiative forcing for the contrasting cases of assuming the aerosol follows the upper versus lower side (US, LS) of the hysteresis loop. An effect of 0.05 W mˆ-2 of cooling is reported for upper versus lower side behavior. Comparison of this result to ours is difficult because this delta-US/LS-forcing depends strongly on aerosol chemical composition, particle diameter, and relative humidity and temperature fields. In our manuscript, we have dissected each of these effects in detail, but we do not have similar information available from the Jacobson (2001) report.

While carrying out our work, we closely examined Jacobson (2001). We made the decision not to include an extensive comparison to this work because the approach of developing global aerosol chemical fields differed so strongly from our own (points a, b, c, and d above) and because the predicted solid phases (CaSO4, KNO3, NH4Cl) do not appear reasonable.

The revisions below are more explicit about why we chose to compare our work in detail to a restricted set of previous publications.

We will revise point b of section 1.2:

ORGINAL calculation of the effects of US and LS of the hysteresis loop on aerosol direct radiative forcing assuming fixed aerosol chemical composition (usually ammonium sulfate)

REVISED calculation of the effects of US and LS of the hysteresis loop on aerosol direct radiative forcing (usually assuming fixed aerosol chemical composition of ammonium sulfate)

We will revise opening sentence of second paragraph of section 1.2:

ORGINAL Previous studies of category b (fixed chemical composition) find delta-F-U,L = 15 to 20%.

REVISED Previous studies of category b employing fixed chemical composition (usually ammonium sulfate) find delta-F-U,L = 15 to 20%.

We will add a third paragraph to section 1.2. NEW PARAGRAPH "Jacobson (2001) interpolates several sources of off-line data into a combined description of 47 internally mixed chemical species. The dominant predicted sulfate, nitrate, and ammonium solids are CaSO4.2H2O(s), KNO3(s), and NH4Cl(s). The degree of neutralization with for ammonium to sulfate is assigned as 50%. The optical calculations assume a black carbon core. Jacobson (2001) calculates global aerosol direct radiative forcing for the contrasting cases of assuming the aerosol follows the upper versus lower side (US, LS) of the hysteresis loop. An effect of 0.05 W mˆ-2 of cooling is reported for upper versus lower side behavior. Comparison of this result to ours is difficult because this delta-US/LS-forcing depends strongly on aerosol chemical composition, particle diameter, and relative humidity and temperature fields. In this paper, we dissect each of these effects in detail. However, similar information is not available in Jacobson (2001) so

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that we can make only limited comparisons between our results and those of Jacobson (2001)."

We will add a citation to Jacobson (2001a) in the first sentence of section 6.2.

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