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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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The major shortcoming of this work is an inaccurate modelling of atmospheric aerosols, due to the assumption that they are a simple combination of ammonium-sulfate-nitrate, and the effect that this has on results of aerosol phase and optical properties. Specifically, single particle instruments and the Aerodyne aerosol mass spectrometer have shown that organics form a substantial fraction of particle mass in many atmospheric locations, especially the upper troposphere [Murphy et al., Science, 1998]. Recent laboratory studies (some referenced in the manuscript) indicate that organic components can have a significant effect on particle properties, namely phase, which is at the heart of this work. This will be critically important in the upper troposphere, where the au-

thors conclude that significant crystallization will take place, contrary to what is now being shown of aerosols which may contain 50% organics by mass.

Likewise, questions of phase and optical properties will be strongly driven by heterogeneous inclusions which have been shown to appear in many, and in some regions of the atmosphere a majority, of particles. The effect on aerosol phase remains largely unknown and the optical effect of highly absorbing heterogenaities, such as soot or biomass burning residue, is not described. The treatment of these issues, in two paragraphs in the Conclusions, does not adequately address substantial limitations of, and in some cases contradictions to, this study. Results from single particle instruments, which have described the complex nature of atmospheric aerosols throughout the last decade, are noticeably absent.

While this work is a comprehensive treatment of the ammonium-sulfate-nitrate aerosol system, the Earth's aerosol burden is clearly not only ammonium-sulfate-nitrate. It is very likely that the remainder, not described in this work, renders many of the conclusions inaccurate or invalid. This work should be withheld until these issues can be resolved on the same scales for which conclusions are drawn (two paragraphs is obviously inadequate). As currently constituted, this manuscript presents an incomplete, and largely false, picture of the interaction of aerosols with global radiative forcing.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 5399, 2003.

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