Atmos. Chem. Phys. Discuss., 3, S1902–S1906, 2003
www.atmos-chem-phys.org/acpd/3/S1902/

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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Received and published: 15 November 2003

Dan Cziczo's call for a treatment of individual particles—including their complex chemical compositions (e.g., soot, sulfates, nitrate, mineral dusts, and more), morphologies (e.g., coated versus mixed), optical properties, phase, and relative humidity histories—is a laudable but halcyon goal for a global model. Beyond limitations in computational power, there are limitations imposed by knowledge of the composition and structure of ambient particulate matter and of laboratory studies and parameterizations of the physical properties and chemical behaviors of mixed particles. One approach, called on by Dan Cziczo, is to do nothing in the face of such obstacles.

Another approach, which we followed, is to assess the state-of-the-art with regard to knowledge and understanding and to move forward in a significant and important increment. As such, we clearly elucidate at the outset the limitations of our treatment in the paper's title, "Effects of... tropospheric ammonium-sulfate-nitrate particles..." In this field, work has progressed from modeling atmospheric "sulfate", to the extent of its neutralization, to most recently the inclusion of nitrate, and finally to our treatment of aerosol phase in this manuscript. The recent antecedents in literature are discussed on pages 5404-5 of the manuscript. The list includes:

* Boucher, O.; Anderson, T. L. "General circulation model assessment of the sensitivity of direct climate forcing by anthropogenic sulfate aerosols to aerosol size and chemistry," J. Geophys. Res. 1995, 100, 26117.

* Haywood, J. M.; Roberts, D. L.; Slingo, A.; Edwards, J. M.; Shine, K. P. "General circulation model calculations of the direct radiative forcing by anthropogenic sulfate and fossil-fuel soot aerosol," J. Clim. 1997, 10, 1562.

* van Dorland, R.; Dentener, F. J.; Lelieveld, J. "Radiative forcing due to tropospheric ozone and sulfate aerosols," J. Geophys. Res. 1997, 102, 28079.

* Boucher, O.; Schwartz, S. E.; Ackerman, T. P.; Anderson, T. L.; Bergstrom, B.; Bonnel, B.; Chylek, P.; Dahlback, A.; Fouquart, Y.; Fu, Q.; Halthore, R. N.; Haywood, J. M.; Iversen, T.; Kato, S.; Kinne, S.; Kirkevag, A.; Knapp, K. R.; Lacis, A.; Laszlo, I.; Mishchenko, M. I.; Nemesure, S.; Ramaswamy, V.; Roberts, D. L.; Russell, P.; Schlesinger, M. E.; Stephens, G. L.; Wagener, R.; Wang, M.; J., W.; F., Y. "Intercomparison of models representing direct shortwave radiative forcing by sulfate aerosols," J. Geophys. Res. 1998, 103, 16979.

* Adams, P. J.; Seinfeld, J. H.; Koch, D. M. "Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model," J. Geophys. Res. 1999, 104, 13791.

* Haywood, J.; Boucher, O. "Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review," Rev. Geophys. 2000, 38, 513.

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* Adams, P. J.; Seinfeld, J. H.; Koch, D.; Mickley, L.; Jacob, D. "General circulation model assessment of direct radiative forcing by the sulfate-nitrate-ammonium-water inorganic aerosol system," J. Geophys. Res. 2001, 106, 1097.

* Jacobson, M. Z. "Global direct radiative forcing due to multicomponent anthropogenic and natural aerosols," J. Geophys. Res. 2001, 106, 1551.

* Metzger, S.; Dentener, F.; Krol, M.; Jeuken, A.; Lelieveld, J. "Gas/aerosol partitioning - 2. Global modeling results," J. Geophys. Res. 2002, 107, 4313.

* Colberg, C. A.; Luo, B. P.; Wernli, H.; Koop, T.; Peter, T. "A novel model to predict the physical state of atmospheric H2SO4/NH3/H2O aerosol particles," Atmos. Chem. Phys. 2003, 3, 909.

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While our manuscript clearly marks an important step forward in the treatment of atmospheric sulfate-nitrate-ammonium particles, we also recognize on pages 5434-5 that our treatment is not comprehensive of all factors influencing global aerosol direct radiative forcing and in particular particle phase, as follows:

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***** EXCERPT BEGIN ********* pages 5434 - 5435 **************

Some of the limitations of our study are readily identified, and in some cases we can predict whether delta-F-U,L will increase or decrease. For instance, aerosol phase is treated through contrasting assumptions of US and LS behavior, rather than by a prognostic treatment of RH history (Colberg et al., 2003), which suggests our delta-F-U,L values are upper limits. In contrast, our use of ISORROPIA including only four solids, when at least seven are known for the SNA system, increases the difference between FU and FL and suggests our delta-F-U,L values are lower limits.

In other cases, the change in delta-F-U,L is uncertain, even though the directions of change for F-U and F-L are individually clear. For example, we employ monthly RH averages with off-line radiative transfer calculations, rather than on-line RH simulation coupled to on-line radiative transfer. Inaccuracies introduced by the use of monthly RH are discussed by Haywood and Boucher (2000) (page 519) and Ramaswamy et al. (2001) (page 368). Although the use of monthly RH underestimates F-U and F-L, the effect on delta-F-U,L is unclear. F-U and F-L are principally underestimated because of underprediction of the occurrence of highly scattering aqueous particles a high RH, yet delta-F-U,L = 0 at high RH.

Our study limits aerosol chemical composition to sulfate-nitrate-ammonium-proton-water, yet authentic atmospheric aerosols have a more complex internally mixed chemistry. More detailed descriptions of aerosol direct radiative forcing include soot, organics, dust, and sea salt (Tegen et al., 1996; Haywood et al., 1997; Haywood and Shine, 1997; Tegen et al., 1997; Haywood and Ramaswamy, 1998; Jacobson, 2001b; Sokolik et al., 2001; Chin et al., 2002; Menon et al., 2002). Particularly affecting delta-F-U,L are dissolved low molecular weight organics, which are commonly internally mixed with anthropogenic sulfates and nitrates. Dissolved organics typically decrease hygroscopicity and decrease, increase, or leave unchanged CRH (Cruz and Pandis, 2000; Prenni et al., 2001; Brooks et al., 2002, 2003; Choi and Chan, 2002). Insoluble components such as mineral dust particles do not affect hygroscopicity directly but do serve as effective heterogeneous nuclei, thus increasing CRH and decreasing the RH hysteresis window over which delta-F-U,L = 0 (Han and Martin, 1999; Martin et al., 2001; Han et al., 2002).

***** EXCERPT END ********* pages 5434 - 5435 **************

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These processes and chemical constituents are important in quantifying the role atmospheric aerosols in radiative forcing and global chemistry. The sulfate, nitrate, and ammonium system, which we focus on, represents a baseline in that it is the largest an-

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thropogenic input to the accumulation mode on a mass basis and the most significant anthropogenic cooling contribution to light scattering on a global basis. This baseline has considerable uncertainty associated with it, and our modeling work on phase is important quantitative progress. In addition to its importance, this chemical system is tractable at high precision due to its simplicity; it lies within the nexus of global modeling capability, extant quantitative field measurements of chemical composition, and laboratory parameterizations of physical and chemical properties. Hence, the manuscript provides a baseline effect at high precision for a critical anthropogenic component of atmospheric particles. This baseline can be employed in future work as a metric to evaluate perturbations by the effects of organic molecules, mineral dusts, and soot on CRH and DRH and the related atmospheric effects.

To clarify the context of our work, we will insert the preceding paragraph on page 5435, line 13, of the revised manuscript.

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