

***Interactive comment on “A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties” by D. V. Spracklen et al.***

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We appreciate the helpful comments of the referee. The issues raised are well taken and we have responded to every comment made by the referee.

General comments

1. The referee asks that additional comparisons with observations are included in this paper. We have added the following comparisons:
  - (a) A table comparing observed and model annual mean DMS and SO<sub>2</sub> (Table

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- 3).
  - (b) A figure showing annual zonal mean DMS and SO<sub>2</sub> (Figure 2).
  - (c) Figures showing observed and simulated monthly mean DMS and SO<sub>2</sub> (Figures 5 and 6).
  - (d) Figures showing observed and simulated vertical profiles of DMS and SO<sub>2</sub> (Figures 7 and 8).
  - (e) A figure showing comparison between global, annual average, meridional CN concentrations from the MBL with Heintzenberg (2000) (Figure 11).
  - (f) Comparison with observed CN12 number (particle larger than 12 nm) from the central Pacific Ocean during the MAGE92, RITS93, RITS94 field campaigns.
  - (g) Comparison with aerosol number from the North Atlantic during JGOFS.
  - (h) A figure showing comparison between simulated and observed MBL aerosol size distribution in the North Atlantic made during the JGOFS cruises (Figure 23).
2.
  - (a) The referee asks for a discussion of the reasons why the model underpredicts BL CN concentrations. We have added a discussion of the likely reasons for this in section 4. Two possible reasons are that the model lacks a mechanism for aerosol nucleation in the boundary layer (as do all global models) and that we neglect primary particles. We mention that this latter issue is dealt with in the second of our papers, currently on ACPD, and in Adams and Seinfeld (2003).
  - (b) The referee asks for a discussion of the likely impacts of not including carbonaceous aerosol. We have added a discussion of this in section 4.
3. The referee asks for additional citations and comparisons with other models.

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- (a) p181, I19. The referee asks for additional citations of carbonaceous aerosol studies. We have added citations for Kanakidou et al. (2000) and Chung and Seinfeld (2002).
  - (b) p181, I21. The referee asks for more examples of models using empirical relationship indirect effect. We have added citations of Boucher and Lohmann (1995) and Menon et al. (2002).
  - (c) p181 The referee asks for other aerosol indirect approaches to be described. We have added Abdul-Razzak and Ghan (2002), Chuang et al. (2002) and Nenes and Seinfeld (2003).
  - (d) p181, I27 The referee asks for a description of sectional, modal and moment microphysical approaches. We have added a short description to the introduction.
  - (e) The referee asks for additional comparison with other aerosol microphysical models. We have added detailed comparison with the TOMAS scheme (Adams and Seinfeld, 2002) at many points through the paper.
4. The referee has asked for budget tables to be included. We have added a sulfur budget table (Table 2) and compared with earlier global sulfur models.
  5. The referee asks that we show the amount of sulfate that deposits onto sea salt aerosol. Our model configuration, which treats sea salt and sulfate aerosol in the same distribution, means that we cannot investigate this. A version of the model (currently under development) which treats different aerosol constituents in different distributions will be used to investigate this in the future.
  6. The referee asks that we perform model runs with only natural sulfur and sea salt emissions. We present these runs as the fractional contribution of natural sulfur to surface and zonal mean aerosol concentrations. These results appear in figures 17, 18 and 19.

1. p180, l17. "For this sulfur sea-salt system" added.
2. p181, l6. 'particles' added after largest to clarify.
3. p183, l13. The referee asks why the concentration of HO<sub>2</sub> is needed. HO<sub>2</sub> is used to recalculate the regeneration of H<sub>2</sub>O<sub>2</sub>. See p187, l2.
4. p183, l14 The referee asks for clarification of a "6-hourly monthly mean". We use monthly-mean global 3-D oxidant concentrations specified at 6 hour intervals (4 times during a daily cycle) at 00:00, 06:00, 12:00 and 18:00.
5. The referee asks what emissions of CS<sub>2</sub>, COS and H<sub>2</sub>S are included in the model. We have added the following: "Anthropogenic COS and CS<sub>2</sub> are emitted as constant molar emission ratios of SO<sub>2</sub> where COS/SO<sub>2</sub>=8×10<sup>-4</sup> and CS<sub>2</sub>/SO<sub>2</sub>=3×10<sup>-3</sup> (Pham et al., 1995). Biogenic emissions of COS and CS<sub>2</sub> are taken as constant molar emission ratios of DMS where COS/DMS=1×10<sup>-2</sup> and CS<sub>2</sub>/DMS=1×10<sup>-2</sup> (Bates et al., 1992)." There are no emissions for H<sub>2</sub>S in the model.
6. p186, l19 The referee asks for the formulation for dry deposition to be described. We have added the following: "Deposition rates are dependent on particle size, land use category (e.g. forest, ocean etc) and wind speed."
7. p186, l20. The referee asks whether the model includes aqueous-phase oxidation in convective clouds. GLOMAP does not include aqueous-phase oxidation in convective clouds. We have added the following text to the paper: "Precipitating, deep convective and frontal clouds are assumed to only remove sulfur gases by wet deposition. Formation of sulfate in convective clouds is limited by availability of S(IV) and oxidants (Laj et al., 1997)."

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8. p186, l7. Condensation has been given its own paragraph.
9. p195, l10. The referee asks why we don't include settling of large particles from upper layers. Previous studies have found this process to be particularly important for the long-range transport of wind-blown dust, which can be carried high into the troposphere. We will consider including this process in future versions of the model.
10. Figure 12. The observations in figure 12 are from July. The July simulation from GLOMAP accurately simulates the number of small particles.

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