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Atmospheric Chemistry and Physics Discussions

Interactive comment on "First comparison of a global microphysical aerosol model with size-resolved observational aerosol statistics" *by* D. V. Spracklen et al.

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. Title. We accept the comments of the referee. We omit the word 'First' in the title as suggested.
2. Abstract. The referee suggests that we modify the abstract to better account for uncertainties associated with assumptions made in our analysis. We change:

   'However, anthropogenic primary black carbon and organic carbon particles do

not explain the large discrepancies in Aitken mode number.'

to:

'However, anthropogenic primary black carbon and organic carbon particles (at the emission size and quantity assumed here) do not explain the large discrepancies in Aitken mode number.'

and we change:

'We show that a physically based cloud drop scheme is needed to explain the observed change in the accumulation mode geometric mean diameter with particle number.'

to:

'We show that a physically based cloud drop scheme better explains the observed change in the accumulation mode geometric mean diameter with particle number.'

3. Model description. The referee requests additional model description regarding our treatment of aerosol nucleation and aerosol components other than sea salt and sulfate.

   We add the following to P7, L5: 'This scheme is valid down to temperatures of 233 K. Below this temperature we use the rate at 233 K as described in Spracklen et al. (2005b).'

   As the referee points out the temperature dependance of the binary homogeneous nucleation mechanism results in very little nucleation in the boundary layer (BL) and lower free troposphere. BL nucleation events have been observed in many continental locations around the world (Kulmala et al., 2004b) but the contribution of these events to global and regional particle number concentrations is highly uncertain. We explore the contribution of BL nuceation events to total particle concentrations in Spracklen et al. (2006). We add the following text:

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'Other nucleation schemes also take place in the atmsophere resulting in observed BL particle formation events over many continental areas (Kulmala et al., 2004b). The impact of these events on BL total particle number is studied in Spracklen et al. (2006).'

The version of the model used here assumes that all sections of the particle size distribution have the same chemical composition (sulfate). This simplification is made to reduce the computational expense of treating multiple components. We clarify this in the model description section. As suggested we move the description of additional model components and a physically based aerosol activation scheme into the model description.

4. Observations. Throughout the paper the size of particles reported is the particle dry diameter. We add this to P8880, L27 and add the following text to P8881, L8 'with respect to particle dry diameter d_p.' In addition all relevant figure axes include 'Dry diameter'. The referee correctly states that the paper does not consider the large uncertainty in radiative forcing associated with water uptake by aerosols.

5. Focus. In this paper we focus on sub-micron aerosol. We add text in the introduction and abstract to ensure that this is clearly stated.

Specific comments.

1. P8874, L15. We remove the text suggesting that most previous global model studies have assumed aerosol from the free troposhere is only sulfate.

2. P8875, L12. We define di-methyl sulfide (DMS).

3. p8876, L9. We remove the line 'This is the first detailed comparison between a global sectional aerosol process model and remote MBL aerosol size distributions.'

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4. Sections 5.1 and 5.2, p8881-8882. The referee comments on the role of sub-micron sea salt emissions. We discuss this on P8882, L23 - P8883, L3. The referee also comments on the tri-modal distributions which were observed during ACE-Asia and ACE-2. Both these experiments showed ultra-fine modes in their median distributions (median ultrafine number of about 250 cm^{-3} and 30 cm^{-3} for ACE-2 and ACE-Asia respectively) whereas the other experiments only showed ultrafine modes at the 95th percentile (Heintzenberg et al., 2004). None of our model runs capture the observed ultrafine mode observed in ACE-2. We discuss this on P8889. The ultrafine mode in the ACE-Asia observations may be explained by primary anthropogenic emissions.

5. Section 5.3. We add the following comment to compare our calculated anthropogenic influenece with earlier studies.

   'In the SH oceans generally less than 20% of surface model sulfate is anthropogenic in origin. Earlier model studies have found that over NH oceans up to 80% of surface sulfate in anthropogenic in origin whereas this is generally less than 20% over most of the SH oceans (Chin et al, 1996; Koch et al., 1999; Chin et al., 2000; Rasch et al., 2000)'

6. Section 5.4.1, p8886. We clarify the assumptions we have made regarding emissions of primary sulfate particles.

   We modify the following sentence to clarify that here we are only referring to emissions of primary sulfate:

   'Including primary emissions has relatively little impact on the size of the Aitken mode and does not help to explain model underprediction of mode diameter.'

   to:

   'Including primary emissions of anthropogenic sulfate (with the assumptions on mode size and emission rate made here) has relatively little impact on the size

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of the Aitken mode and does not help to explain model underprediction of mode diameter.'

Globally in our model about 80% of total particle number comes from upper tropospheric nucleation and 20% from primary emissions of anthropogenic sulfate. However, primary anthropogenic sulfate particles are emitted at a larger size than nucleated particles and so have a greater CCN production efficiency (Pierce et al., 2006b).

7. Section 5.4.2, p8887. We add a reference for the injection height of large scale wildfire emissions.

   The referee points out the uncertainty relating to the best choice of size for primary particulate emissions. We add the following discussion:

   'There is some uncertainty as to the most appropriate size choice for primary emissions. As biomass burning aerosol plumes age the mode radius increases and the distribution width narrows (Dentener et al., 2006). Here we have chosen to emit particles at their freshly emitted size according to Dentener et al. (2006).'

   We clarify that BC/OC emissions are treated in the same distribution as sea-salt and sulfate and that all aerosol components are assumed to have the chemical composition of sulfate.

   In this work we only include primary emissions of organic aerosol. We do not include secondary organic aerosol (SOA). SOA will condense on existing aerosol and shift the distribution to larger sizes. Further work is required to establish the potential role of SOA in remote MBL aerosol distributions.

   In GLOMAP the particle sinks are coagulation (global average rate of 5×10^{-3} cm^{-3} s^{-1}), dry deposition (2×10^{-4} cm^{-3} s^{-1}) and wet deposition (1×10^{-4} cm^{-3} s^{-1}). The dominant sink for particle number is coagulation with dry and wet deposition being relatively unimportant sinks in terms of aerosol number. This

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result that has been found in other global modelling studies (e.g., Adams et al., 2002).

The referee suggests that we use diameter for emissions to be consistent with the rest of the manuscript. We use radii in description of emissions to be consistent with referenced work on emission size (Bond et al., 2003; Dentener et al., 2006).

8. p8889, l17. The referee suggests that an additional reason for our underprediction of aerosol number in the N. Atlantic could be our treatment of BC and OC aerosol as sulfate. We add the following line to p8889, l17:

   'In addition, our treatment of BC and OC aerosol as hydrophilic upon emission will likely increase the efficiency at which this aerosol is scavenged.'

9. p8890, l17. We replace the statement: 'In these model runs sulfur is the only condensable species' with 'The model runs do not include condensation of secondary organic aerosol.'

10. Section 6. The referee suggests that the difficulty of representing sub-grid scale growth processes in the global model may contribute to underestimated growth rates. This problem will be studied in an upcoming UK campaign ADIENT. We add the following line to p8890, l19.

    'An additional contribution to underestimated growth maybe be due to the problems of representing sub-grid scale growth in the global model.'

11. p8891, l10. We add: 'This will lead to a high bias in the observations.'

12. Section 7.2.

    The combination of relatively long transport times between UT and BL and the rapid coagulation sink for small particles means that large uncertainties in the upper tropospheric binary homogeneous nucleation rate only lead to small changes in model MBL particle number (Spracklen et al., 2005b).

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13. p8892, l4. We add 'in the number size distribution' as suggested.

14. p8893, l25. We change: 'The NSO3 scheme worsens the comparison'

    to:

    'The NS03 scheme combined with the updraft speeds chosen here makes the comparison of model 5th percentile with observations worse. This is especially true for the Aitken mode which is reduced both in size and number.'

15. Conclusions, p8895, l1. We remove the following sentence from the conclusions as requested:

    'Accurate average MBL aerosol number, 'closed' size distributions and a good comparison between model and observed persistence suggests that a binary homogeneous nucleation scheme correctly calculates the secondary source of particles to the MBL, at least as a global mean.'

16. Figure 1. Areas of campaigns are shaded and locations of observations is made more clear as suggested.

17. Figure 3,4,5 and 6. Captions are modified to explain BC/OC (Black carbon/organic carbon).

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 8871, 2006.