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Interactive comment on "A global off-line model of size-resolved aerosol microphysics: II. Identification of key uncertainties" *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. Additional model evaluation has been incorporated into the first paper of this series (Spracklen et al., 2005) as suggested in the reviews for that paper. The first paper now includes comparison of modelled and observed SO_2 and DMS, MBL and continental BL CN number, vertical CN profiles and number-size distributions. We now believe there is sufficient model evaluation in Spracklen et

al. (2005) to allow some exploratory sensitivity analyses to be carried out here. Papers covering the model description, sensitivity analysis and comparison with observations can be ordered in different ways. We felt it was important to demonstrate broad agreement with observations in paper 1, but to explore how robust the results were before proceeding to a detailed comparison with observations.

2. The referee asks for additional analysis of how the lack of aerosol constituents other than sea-salt and sulfate will affect our results. Spracklen et al. (2005) showed that GLOMAP simulates realistic MBL CN and CCN number but underpredicts CN and CCN over polluted continental regions. There are several possible reasons for this; including lack of carbonaceous aerosol, neglect of 'primary' sulfate aerosol (nucleation that occurs in power plant plumes at scales smaller than the model grid), and underprediction of BL nucleation events which have been widely observed (Kulmala et al., 2004). As the referee points out, over polluted continental regions carbonaceous emissions may be a dominant contributor to tropospheric CCN.

   The suggestion to focus on the marine regions where sulfate and sea salt particles are likely to dominate is a good one. We have therefore modified Figure 20 to show the change in CN and CCN as means over the oceans as well as global means as previously shown. We have also added clarification throughout the paper to warn the reader that our results are for sulfate and sea salt aerosol only and that over continental regions other aerosol constituents are likely to dominate the total aerosol.

3. We have changed the wording 'changes in the binary nucleation rate... cause a shift of the upper tropospheric CN layer by as much as 3 km, while changes in the absolute concentration are relatively small' to 'changes in the binary nucleation rate... cause a shift of the upper tropospheric CN layer by as much as 3 km, while the shape of the CN profile is essentially pre-served'. Although there are changes

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in the local and STP-corrected CN concentrations, the main point to note is that the altitude of the maximum has shifted.

Specific comments

1. p3422, l21. By aqueous-phase processing we mean liquid-phase oxidation of gas-phase SO_2 to sulfate. For clarity we replace 'aqueous-phase processing' by 'in-cloud oxidation' in the paper.

2. p3444, l3. Dry deposition only includes turbulent deposition and not sedimentation. We have added the word 'turbulent' for clarity. We have added the following text (p3444,l4): 'but with limited impact to accumulation mode particles demonstrating a good representation of size-resolved deposition.'

3. p3447, l9 and Figure 5. Coordinates were missing from the Figure captions. The following captions have been added:

   (a) and (c) Tropical Pacific (10°N-10°S, 210°-270°E)

   (b) and (d) Northern Europe (45°-60°N, 5°-25°E)

4. p3448, l28. The referee states that sensitivity to accommodation coefficient is likely to change when other aerosol components are added. The model is likely to underpredict aerosol surface area in polluted regions (due to lack of carbonaceous aerosol) and downwind of major dust sources (due to lack of dust aerosol). However, our argument here is that aerosol formation occurs primarily in the FT and UT so CN concentrations are unaffected by changes in aerosol surface area in the BL, which is where most primary aerosol resides. Basically, in a model in which particle formation occurs through binary homogeneous nucleation the CN source to the BL is decoupled from the surface area in the BL itself. This won't be the case in a model in which particle formation can occur also in the BL, which is something we are currently investigating, but goes beyond the current study.

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We have added the following text to the paper(p3449, l10):

'In addition, our simulations will underpredict continental aerosol surface area and so are likely to underestimate the impact of uncertainty in condensation rates on aerosol properties. However, in a model in which particle formation occurs through binary homogeneous nucleation the CN source to the BL is decoupled from the surface area in the BL itself.'

5. p3449, l28. Here we refer to the coagulation scavenging of nucleation clusters to the existing particle sink.

6. p3450, Section 5.2 and 5.3. The referee asks definition of activation,

   Activation in our model means that a fraction of the aerosol particles (above a fixed dry diameter, described in Spracklen et al., 2005) are assumed to grow into droplets in cloudy air and subsequently support oxidation of SO_2 to sulfate, which then remains on the particles as the air subsequently passes into clear air.

   We have added the following text to the paper (p3450, l13): 'Activated particles are assumed to grow into droplets in cloudy air and subsequently support oxidation of SO_2 to sulfate, which then remains on the particles as the air subsequently passes into clear air.'

   The referee asks how the activation diameter relates to scavenging and of how the effective radius for scavenging is chosen. We have added the following text to the paper:

   'An effective scavenging diameter of 0.206 μm is chosen above which nucleation scavenging may occur in the model. This value is intermediate between the values used by Adams and Seinfeld (2003) (0.03 and 0.082 μm for convective and stratiform clouds respectively) and that used by Capaldo et al. (1999) (0.250 μm).'

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Addition of in-cloud produced sulfate is the only growth mechanism in the liquid-phase. We do not include droplet collision and coallescence as a mechanism of increasing evaporated aerosol sizes.

7. p3453 and p3455, Section 5.4.

   We have added the following text to the paper(p3454 ,l16) to explain how we view the role of primary sulfate emissions:

   'Here we assume that a 1-5% emission of SO_2 as particulates is used as an estimate solely for sub-grid nucleation of sulfate in power plant plumes. It has been suggested that this method could also be used as a simple surrogate for primary carbonaceous emissions (Adams and Seinfeld, 2003). However, other models include carbonaceous aerosol in addition to emissions of primary sulfate. For example, the ECHAM5-HAM model includes 2.5% of SO_2 as primary sulfate in addition to emissions of primary carbonaceous emissions (Stier et al., 2005). These different approaches will result in different simulated CN and CCN concentrations. Our estimates of CCN sensitivity need to be repeated in a model with a more complete range of aerosol types.'

8. p3455, Section 5.4. The referee points out that we should not expect the model to accurately simulate polluted CN concentrations without all aerosol constituents. We are not saying that all primary particles come from sulfate, but only that the accepted range of 1-5% conversion leads to CN concentrations that span the observed range. So clearly there are very large uncertainties in emissions inventories and primary sulfate could make up a large fraction of total concentrations in some environments. If primary sulfate made a small contribution to CN then we could not confidently estimate its relative contribution without including other CN sources. The fact that primary sulfate more than explains observed CN on its own is a useful outcome of our sensitivity tests.

   We have added the following text (p2455, l2):

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'However, in polluted regions carbonaceous aerosol would be expected to contribute significantly to CN number. Including 3% of SO_2 as particulates (with the diameter and standard deviation assumed here) may cause an overestimate of the contribution of sulfate aerosol to total CN. Small changes in the assumptions about the size of primary sulfate emissions will cause large changes in simulated aerosol number. Including carbonaceous aerosol in the model will be required to test the contributions of primary sulfate and carbonaceous aerosol to total CN. It is likely that to accurately predict the contribution of carbonaceous aerosol to tropospheric CCN will require more extensive observations regarding the size distribution of primary carbonaceous emissions.'

9. p3456, Section 5.4. When we change the model sulfur emission rates we do not include any percentage of anthropogenic sulfur as particulates. The almost linear relationship between sulfur emissions and CCN concentration is not due to changing surface area caused by primary emissions.

10. p3460, l15. We have removed the word 'several' for clarity.

11. Section 7. We point out that the uncertainty in total CCN may well be lower in a model that includes a full description of all aerosol components. We have added the following text (p3463, l6):

    'In addition, estimation of total CCN number will require a model which contains the full range of aerosol constituents. Over polluted continental regions, carbonaceous aerosol may be the dominant contributor to total CCN.'

Technical corrections

1. Figures. Zonal means in figures 3a,b and 4a,b have been replotted with different contour levels and an additional colour bar to make them easier to read.

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2. Figure 1. Lines have been replotted to make easier to read.

3. Figure 5. Following caption added:

   (a) and (c) Tropical Pacific (10°N-10°S, 210°-270°E)

   (b) and (d) Northern Europe (45°-60°N, 5°-25°E)

4. Figure 5c,d and Figure 7. Standard values for a_e and for nucleating cluster size have been added.

5. 'Runhas' split to 'Run has'.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3437, 2005.