

## ***Interactive comment on* “The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales” 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.

Detailed comments

1. *A significant amount of discussion is spent on the boundary layer; however this is never defined, nor is it explained why it's important for evaluation of the nucleation mechanism. How many model layers describe the boundary layer?*

We include a more detailed description of the advection and transport schemes used in the model. We give a description of the boundary layer scheme and

model resolution in the lowest model layers. We add the following text (p7330, l6):

‘Tracer advection is performed using the scheme of Prather et al. (1986). Sub-grid transport is calculated using the convection scheme of Tiedtke et al. (1989). Turbulent mixing in the BL and BL height are calculated using the parameterization of Holtslag et al. (1993).’

We describe vertical model resolution near the surface by adding the following text (p7330, l5):

‘with the lowest layers centered at about 30 m, 140 m, 340 m, 600 m, 930 m, 1300 m, 1710 m above the surface.’

2. *It is not clear to me whether the modeled values concern the average of the layers that describe the boundary layer or refer only to the surface layer.*

Model values refer to the surface layer. Where this is not clearly stated we add additional clarification.

3. *Model predictions concern the average of a grid cell while measurements are taken in a specific point". How does this affect the comparison? A discussion would be rather useful.*

Model values are interpolated in 2-D to the latitude and longitude of the observations. However, as the model has a global domain, horizontal model resolution is relatively coarse ( $2.8^\circ \times 2.8^\circ$ ). Recent work has shown that particle formation events are often regional phenomena (Tunved et al., 2003) and so a global model should be capable of modelling them.

4. *How well is the boundary layer captured by the model? Especially for Hyytiälä a discussion on how the model predictions of the boundary layer are compared with measurements and their implication for the overall model performance would be necessary.*

We add information on a general evaluation of the BL scheme in our model and also a comparison of model BL height at Hyytiälä with observations. We add the following text (p7330, l6):

‘A comparison of the BL scheme with observations is given by Wang et al. (1999). They show that model BL height is in good agreement with Holtslag et al. (1993). Including the scheme results in an improved seasonal cycle of model tracers.’

and (p7338, l27):

‘Maximum mixed BL height at Hyytiälä during August 2001 was calculated as between 1000 and 2200 m (Spirig et al., 2004). Radio soundings launched at Hyytiälä on 4 April 1999 recorded maximum BL height of 2180 m (Nilsson et al., 2001). At Hyytiälä model mean BL height is 550 m and maximum BL height is about 2000 m.’

5. *Page 7330, lines 10-11 - The activation of particles is a strong function of supersaturation; thus not all particles above 50 nm activate to form cloud droplets. Please revise appropriately.*

In GLOMAP all particles larger than a critical size are assumed to activate to form cloud droplets when clouds are present (Spracklen et al., 2005a). Activation in clouds is a complex process depending on the aerosol size distribution and cloud updraught velocity. However, the updraught velocity in clouds will be a very difficult parameter to include in global models. In Spracklen et al. (2005b) we evaluate the impact of varying a fixed aerosol activation diameter on model CN and CCN. Surface CCN concentrations are quite sensitive to the selection of activation diameter (global surface CCN is reduced by about 30% when the activation diameter is increased from 50 nm to 80 nm) whereas total particle number is relatively insensitive. In this work we focus on the prediction of CN concentrations so our use of a relatively simplistic aerosol activation scheme will not impact our results greatly. In Spracklen (2006) we include more detailed

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activation schemes and evaluate the impact on marine boundary layer aerosol size distributions.

6. *Page 7331, line 26. Could you please clarify what do you mean by ‘boundary layer formation mechanism’?*

We change ‘Secondary sulfate particles are formed through homogeneous nucleation as well as through new boundary layer formation mechanisms, while sea salt, BC and OC are added as primary particles.’ to ‘Secondary sulfate particles are formed through binary homogeneous nucleation above the boundary layer (Kulmala et al., 1998; Spracklen et al., 2005a) as well as through particle formation within the boundary layer. For a description of the particle formation mechanisms used in the model see section 2.2. Sea salt, BC and OC are emitted as primary particles.’

7. *Section 2.2 - Please, if possible, provide a more detailed description of the nucleation mechanism. How is the growth rate (GR) calculated and how is it affected by the presence of organics? What is the time step for these calculations? How is the overall mechanism implemented into the model? A plot of the spatial distribution of the nucleation rate would also be quite informative.*

The growth rate (GR) is calculated from the concentration of condensable vapours using the expression from Kerminen and Kulmala (2002). We add the following (P7333, L8):

‘Growth rate is calculated using the following approximation from Kerminen and Kulmala (2002):

$$GR = \frac{3.0 \times 10^{-9}}{\rho_{nuc}} \sum c_i M_i C_i, \quad (1)$$

where index  $i$  goes over condensable vapours and  $C_i$  is gas-phase concentration (molecules  $\text{cm}^{-3}$ ),  $c_i$  is molecular speed of condensing vapour ( $\text{m s}^{-1}$ ) and  $M_i$  is

its molecular weight and  $\rho_{nuc}$  is nuclei density ( $\text{kg m}^{-3}$ ):

We assume that only sulfuric acid contributes to the growth of nuclei between 1 nm and 3 nm diameter but that both condensible organics and sulfuric acid contribute to the growth of particles larger than 3 nm (see P7333, L9-12).

Page 7333, l14. We have added:

‘The timestep in the model for this process is 3 minutes.’

8. *Page 7334, section 2.2, last paragraph - It is not clear enough why the authors chose to apply different new particle formation mechanism for the boundary layer and above it. How different would the results be by adopting a single nucleation mechanism for the whole domain? It is also quite clear that ammonia plays a significant role in nucleation. How different would the results be if the key role of ammonia was considered?*

The particle formation mechanism that we include in the model has been determined empirically (Kulmala et al. 2006; Sihto et al., 2006) based on observations at a surface station at Hyytiälä. Applying the new particle formation mechanism throughout the depth of the atmosphere does not limit particle formation events to the BL and the observed Z-shaped profile of particle number (Schroder et al., 2002) is no longer produced by the model. This suggests that  $J^*$  depends on more than sulfuric acid and that equation 1 breaks down above the BL. See P7334, L11-20.

In Spracklen et al. (2005a) we showed that a BHN mechanism was able to reproduce observed particle formation in the FT and UT but that this scheme did not cause any particle formation in the BL. We add the following text (P7334, L12):

‘In Spracklen et al. (2005a) we showed that this nucleation scheme is capable of reproducing observed particle number in the FT and UT.’

We change the following (P7334, L13):

'Our limited measurements above the boundary layer at Hyytiälä suggest that this is a valid assumption. Aircraft observations showing CN enhancements in the European boundary layer and a minimum just above it (e.g., Schroder et al. 2002) are also consistent with this assumption, although primary emissions at the surface also play a role.'

to:

'Our limited measurements above the BL at Hyytiälä and aircraft observations showing CN enhancements in the European BL and a minimum just above it Scroder et al. (2002) suggest that new particle formation is confined to the BL. However, the reason for such behaviour is not understood. '

9. *There is no information on the additional computational effort required for the nucle-ation mechanism. This is an important issue that needs to be addressed.*

The additional computational effort of including the nucleation scheme is negligible compared to the overheads of calculating tracer advection etc.

10. *Please provide a definition for the condensation sink; How sensitive is it to the nucleation rate? Would the degree of agreement be the same if you replaced the nucleation mechanism with other expressions (e.g., binary or ternary nucle-ation)?*

We add a definition for condensation sink adding the following line:

'The reduced condensation sink (CS') is calculated by summing over the aerosol size bins j:

$$CS' = \sum_j \beta_j r_j N_j \quad (2)$$

where  $\beta_j$  is the transitional correction for the condensational mass flux (Fuchs et al., 1971),  $r_j$  is the particle radius,  $N_j$  is the particle number. The condensation

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sink (CS) has units of  $\text{s}^{-1}$  and is calculated from the reduced condensation sink by

$$CS = 4\pi DCS', \quad (3)$$

where D is the vapour diffusion coefficient.

Binary homogeneous nucleation schemes do not calculate nucleation in the BL. Nucleation is calculated in the upper troposphere and entrainment of these particles to the BL produces surface CN concentrations of  $100\text{--}200 \text{ cm}^{-3}$  which is 10-100 times lower than observed (see P7340, L11-25). We do not consider ternary nucleation in this work. Gaydos et al. (2005) used their model of ternary nucleation to simulate particle formation events in the Eastern U.S. In Hyytiälä ammonia is in excess and so will not experience the same behaviour as in the Eastern U.S. where ammonia is the limiting species. See P7332, L8-20.

11. *It has been reported in the literature that low (with top below 700 hPa) and middle level clouds (with top between 450-700mb) are not always captured correctly by ISCCP (both in amount and occurrence frequency). How this affects your results?*

Monthly mean cloud fields from ISCCP are used to specify when and where aerosol is processed ( $\text{SO}_2$  converted to particulate sulfate). Error in ISCCP fields will not directly impact gas-phase sulfuric acid concentrations or nucleation rates.

12. *The analysis considers monthly mean emissions of terpenes.  $\alpha$ -pinene is used as a surrogate for all terpenes and 13the secondary organic aerosols (SOA) formed are assumed to be condensable vapors. Moreover emission rates are not affected by temperature or light intensity. A discussion regarding the abundance of  $\alpha$ -pinene at Hyytiälä would be useful. Are the assumptions made justified by experimental evidences? How the presence of sesquiterpenes would affect the results? A paragraph discussing the implications of the simplifying assumptions made here would be important.*

Monoterpenes were measured at Hyytiälä and we make a comparison between observed and model monoterpenes (see P7339, I27). In April the model agrees with observed monoterpene concentrations but in March the model is about a factor of 10 too low. Further work is required to modify our emission estimates to include a daily cycle and to take into account meteorological variables such as light intensity and temperature.

Condensable organic concentrations are difficult to directly measure but have been inferred by various groups using a variety of different techniques. These techniques estimate that condensable vapour concentrations at Hyytiälä are between 2 and 10 times the value in our model. Particle growth in the model is slower than observed which also indicates that condensable organic concentrations are too low in the model. We add the following text (P7340, L9):

‘Condensable vapour concentrations at Hyytiälä have been determined by a variety of techniques to be between  $2.0 \times 10^7$ – $1.3 \times 10^8$   $\text{cm}^{-3}$  (Kulmala et al., 2001; Dal Maso et al., 2002; Lehtinen et al., 2004; Korhonen et al. 2005). Model condensable vapour concentrations range from  $0.5$ – $2.0 \times 10^7$   $\text{cm}^{-3}$ . This is a factor of 2–10 lower than observed.’

We carry out a sensitivity run with 10xGEIA monoterpenes that is intended to account for the uncertainty in the processes leading to SOA.

We add the following text (p7335, L5):

‘There are also uncertainties regarding the conversion to condensable organic species and the potential for organic species other than monoterpenes to produce SOA. We explore the effect of changing the monoterpene emission rate by a factor of 10 to take account of uncertainties in secondary organic aerosol yields and precursor emissions.’

### 13. *How are SOA distributed among the different size bins?*



The condensation of condensible organics onto the aerosol size distribution is described using the modified Fuchs-Sutugin equation (Fuchs et al., 1971) and the implementation of this in GLOMAP is explained in Spracklen et al. (2005a).

14. *Although as a first attempt the good agreement between the model results and the dataset from Hyytiälä provides some confidence, there are also other datasets that could be considered. A discussion would be necessary.*

The particle formation mechanism that we include in the model has been determined empirically (Kulmala et al., 2006; Sihto et al., 2006) based on observations at Hyytiälä. We therefore feel that it is best to interpret our model results based on the observations made at this site. A full comparison of the model against other surface sites is a major task. This is currently being undertaken but is beyond the scope of this first study.

15. *Page 7345, line 6-8 - Explain why we expect to have nucleation events in Southern Ocean.*

Many global aerosol models underestimate aerosol number in the Southern Ocean (Adams and Seinfeld, 2002; Easter et al., 2004; Spracklen et al., 2005a). The recent work of Pierce and Adams (2006) suggests that model neglect of super-fine sea spray emissions may result in some underprediction of total particle number concentrations but with a relatively small contribution between 30 and 45°S. The BL particle formation mechanism causes enhancements of a factor of 3-10 to particle number concentrations in the Southern Ocean, resulting in good agreement with observations (P7345, L12-19). The model vertical profiles of particle number show that nucleation is occurring at the top of the BL and that these particles then mix to the surface. The timeseries of model CN number in the Southern Ocean does not show nucleation bursts but elevated total particle number due to particles mixing down from the top of the BL (P7345, L20 - P7346, L4).

Future work should include a detailed analysis of aerosol distributions in the Southern Ocean. We remove the following text from the abstract:

‘Particle concentrations are enhanced by a factor 3-10 over the remote Southern Ocean (30-70°S), resulting in total concentrations of 250-1000 cm<sup>-3</sup>, in good agreement with observations. Particle formation tends to peak towards the top of the marine boundary layer and there is a lack of obvious burst-like behaviour at the sea surface. This result suggests that new particle formation in the marine boundary layer could be confused with entrainment from the free troposphere.’

16. *In their conclusions, authors state that in urban areas CN are not affected by new particle formation. However, observations of nucleation events in urban areas (e.g., Po Valley, Delhi, Mexico City) do not fully support this statement. As a matter of fact, such observations should be used to improve our understanding regarding nucleation theory and attempts to capture such events should be made.*

The model does simulate nucleation events in polluted areas with a similar frequency to observations (see P7344, L14-L24). We show here that these events only lead to relatively small enhancements to total particle number concentrations (generally less than a factor of 2) whereas over remote continental areas enhancements up to a factor of 8 are modelled. Further work is required to fully test the model against observations in urban areas.

We change (P7344, L22):

‘There is little evidence that the formation events have a lasting impact on CN in regions with such high primary emissions’

to:

‘The model suggests that although particle formation does occur in polluted regions, particle concentrations in such regions are dominated by primary emissions.’

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1. *Page 7331, line 19 - Adams and Seinfeld (2002)" rather than Adams and Seinfeld (2003)".*

We have changed 'Adams and Seinfeld (2003)' to 'Adams and Seinfeld (2002)'.

2. *Page 7331, section 2.1, last paragraph - SO<sub>2</sub> emitted as particulates" implies that SO<sub>2</sub> can exist as particle which is not true. Please correct all phrases related to that.*

We have changed all references of SO<sub>2</sub> emitted as particulates to 'anthropogenic sulfur emitted as particulates'.

3. *Page 7339, line 27 - Figure 1d" rather than Figure 1c"*

We have changed 'figure 1(c)' to 'figure 1(d)'.

4. *Page 7348, line 10 - Increase in" rather than Increases"*

We have changed 'Increases' to 'Increase in'.

5. *Table 1 - Please provide the source strength of sea salt.*

We have added the source strength of sea salt as 13 500 Tg NaCl / yr<sup>-1</sup>.

6. *Figure 1c, Caption - blue" rather that "red".*

We have changed 'red' to 'blue'.

7. *There is an inconsistency in CS units between the text (in m<sup>2</sup> ) and in Figure 1e (s<sup>1</sup> ). Please revise appropriately.*

We have changed the symbol CS (condensation sink) in equations 2 and 3 to CS' (reduced condensation sink).

we include a definition of CS and CS' in the text.

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$$CS = 4\pi DCS', \quad (4)$$

where  $D$  is the vapour diffusion coefficient.  $CS$  has units of  $\text{s}^{-1}$  and  $CS'$  has units of  $\text{m}^{-2}$ .

8. *Figure 2 - The axis labels and the legend do not clearly read. Could you please revise the figure?*

We have altered axis labels and legend to make more readable.

9. *Figure 6 - Could you please explain why there is an exponential increase in CN concentrations when both primary emission and formation events are included in the calculations?*

The response of European CN concentrations to primary emissions and BL particle formation events is explained on p7346/7347. Including current day primary anthropogenic BC/OC emissions in the model leads to European mean CN concentration of about  $6600 \text{ cm}^{-3}$ . Including BL formation events as well as primary emissions leads to mean CN concentration of  $9300 \text{ cm}^{-3}$  (a 40 % increase). Reducing primary emissions without BL formation events leads to a reduction in model CN number. When primary anthropogenic emissions are zero model CN concentrations are about  $500 \text{ cm}^{-3}$ . Reducing primary emissions with BL formation events leads to a non-linear change in CN number. Reducing primary emissions reduces aerosol surface area which results in additional particle formation.

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