Atmos. Chem. Phys. Discuss., 6, S3322–S3326, 2006
www.atmos-chem-phys-discuss.net/6/S3322/2006/

Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #1

Received and published: 25 September 2006

Overall comments:

The manuscript "The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales" by Spracklen et al., discusses a modeling study of the effects of boundary layer particle formation on the large scale particle number budget. This is an interesting and important issue since nucleation affects climate and visibility by changing the aerosol size distribution and the cloud condensation nuclei (CCN) concentrations. In their analysis, the authors perform simulations that investigate the individual contribution of primary aerosol production and nucleation to the boundary layer number concentrations. The latter is of particular importance since it

is a first step toward the improvement of our understanding for the contribution of new particle formation on atmospheric chemistry and the climate. Moreover, several sensitivity simulations were made to examine the impact of various uncertainties on their results.

The manuscript is well written, and both the modeling approach and the results are presented quite clearly. In addition, the simplifying assumptions that were made are presented clearly and some discussion on their implications for the results is also provided. Given the degree of simplifying assumptions, the reliability of the results provided by this study, at least on a global scale, needs to be evaluated with caution as also underlined by the authors. Overall, the manuscript is publishable in ACP if a more detailed discussion regarding the modeling framework and its configuration and the effects of the simplifying assumptions on the results will be included.

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?

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.

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.

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.

5. Page 7330, lines 10-11 - The activation of particles is a strong function of super-

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saturation; thus not all particles above 50 nm activate to form cloud droplets. Please revise appropriately.

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

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.

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?

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

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 nucleation)?

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?

12. The analysis considers monthly mean emissions of terpenes. α-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 α-pinene

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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.

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

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.

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

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.

Minor comments:

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

2. Page 7331, section 2.1, last paragraph - “SO_2 emitted as particulates" implies that SO_2 can exist as particle which is not true. Please correct all phrases related to that.

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

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

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

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

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7. There is an inconsistency in CS units between the text (in m^{-2}) and in Figure 1e (s^{-1}). Please revise appropriately.

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

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?

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