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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 #3

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Overall comments:

The manuscript "The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales," by Spracklen and colleagues, describes a modeling study of the impact of nucleation on global aerosol number distributions. This study is important for a number of reasons. By isolating the individual contributions of nucleation and primary aerosol production, we can begin to assess its impact on atmospheric chemistry and climate. In addition, we can look at the response of the flux of new particles to environmental changes such as increases or decreases anthropogenic particulate emissions.

The authors have taken on a difficult challenge: new particle formation from nucleation is a highly non-linear, regional phenomenon whose mechanism is not fully understood. Thus it is necessary to make a number of simplifying assumptions if one is to model its global impacts. To the authors' credit, these assumptions are presented clearly and some discussion is given as to their implications. In my opinion, these simplifying assumptions form the basis from which the results and interpretations of the model results are to be judged. Thus I would like the authors, in the forum of this discussion or by direct modification of the text, spend some additional time in discussing the impacts of what I consider key simplifications. I will provide some topics for initial discussion:

1. While the basic interactions between sulfuric acid and pre-existing aerosol surfaces are accounted for, all condensable organic species are lumped together as a single compound whose emissions are associated with that of terpenes and are monthly averaged without regard to temperature or light. The reaction of this model compound is assumed to be that of a-pinene, and 13% of the secondary products are assumed to be condensable vapors. Clearly an experimental validation of this approach would be preferable. Were monoterpenes measured at Hyytiälä?

2. Sulfuric acid is primarily produced by cloud processing, thus the parameterization of cloud formation is key to accurately determining ambient levels of this compound. One assumption of cloud droplet formation relates to the criterion for droplet activation (a specific question relating to this issue is presented below). Another assumes that particle scavenging by clouds occurs only by nucleation, and not wet deposition. This latter assumption may result in a less accurate depiction of cloud droplet number.

3. The centerpiece of this study is a data set consisting of 22 days of measurements at Hyytiälä, Finland. Some additional insights are provided by average particle concentration data from Melpitz and Heidelberg. Certainly other datasets are also available with high quality measurements of sulfur containing gases and aerosol. Can the authors comment on why they considered only this data set interpreting their model results?

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4. I find that the days in which the model does not capture the observed nucleation events are the most interesting. A closer look at these days (e.g., days 78 and 82), might provide insights that may lead to model improvements.

5. It is not at all clear to me that CN are not affected by new particle formation in urban areas, although the authors state this as a main result of their model (ref page 7344, lines 20 - 23). Examples of recent observations in the Po Valley and Mexico City have shown that areas of high primary emissions can still regularly host new particle formation events. This could be merely an phenomenon related to vertical mixing, e.g., the breakup of the planetary boundary layer. These high sources of condensable material, which may or may not be adequately represented in the model, make these important areas to explore (as in point #4, worthy of more careful inspection).

Overall I commend the authors on tackling a difficult issue with a clearly presented approach. Certainly, as the authors state, these and other simplifications are only the first steps in a long process of modeling this complex phenomenon. However it is also true that results from studies such as these are often disseminated without regard to the conditions with which the conclusions are drawn. Thus I would encourage the authors to be more introspective of the implications of their approach. I would encourage some additional discussion before this paper is published in its final form.

Specific comments:

Page 7330, line 10: Please clarify your criterion for cloud droplet formation. As it currently reads, all particles larger than 50 nm activate to form droplets regardless of supersaturation.

Page 7330, line 19: Please clarify what is meant by “the new boundary layer formation mechanisms.”

Page 7332, paragraph starting at line 19: Please correct all statements in this paragraph that refer to “SO2 emitted as particulates,” as this implies that SO2 can exist in

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particles. From the abstract of the Adams and Seinfeld paper: “Both analyses show that the few percent of anthropogenic sulfur emitted as particulate sulfate results in an increase in CCN concentrations comparable to that resulting from much larger emissions of gas-phase sulfur dioxide.” Thus the correct statement should be “Ě industrial sulfur emitted as particulatesĚ”

Page 7334, paragraph starting at line 17: In this paragraph discussing the competing effects of condensate production and scavenging due to particulate surfaces it seems appropriate to mention the work of P. McMurry, who developed a simple parameter that predicts whether new particle formation will occur based on the ratio of the rate of formation of the thermodynamically stable clusters to the rate at which they are lost to pre-existing particles (McMurry et al., JGR, 2005).

Page 7336, line 12: Although this paper focuses primarily on the prediction of CN concentrations, their formulation for cloud production (i.e., any particle greater than 50 nm forms a cloud droplet) will be greatly influence by disparities between both the growth rates and CCN activities of biogenic SOA versus those of sulfate. For example, VanReken et al. (JGR, 2005) studied a variety of biogenic compounds in chamber studies and found that CCN activity differed from that of pure ammonium sulfate in all cases (lower), and that particles became significantly less hygroscopic as they aged. I suggest a sentence or two that convey the implications of your simplification regarding SOA formation.

Page 7339, line 27: correct the Figure reference. It should be 1d.

Page 7348, line 10: correct the word “Increases”

Table 1: should there be an entry for the source strength of sea salt?

Figure 1: caption for plot 1c: blue should be the color of the modeled data.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 7323, 2006.

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