

***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 #2

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This manuscript studies the effects of boundary layer nucleation on total particle concentrations on regional and global scales using a new particle formation mechanism in a global aerosol chemical transport model. Understanding the budget of particle number concentration is important because of the particles' potential to be cloud condensation nuclei (CCN) and change the lifetime and properties of clouds and because this particle-cloud interaction is a major source of uncertainty in quantifying human impact on climate. In this study, several model simulations were performed to investigate the contribution of anthropogenic particle emissions, non-boundary-layer binary homogeneous nucleation, and boundary layer nucleation events on total particle concentrations. In addition, several sensitivity simulations were performed to ex-

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amine some of the major uncertainties in the model results. The results point out the non-linear coupling between emitted and secondary particles such that reduction in primary emissions of particles could greatly enhance new particle formation. The manuscript presents clearly the modeling approach and also states clearly the simplifying assumptions made in the model. Due to the simplifying assumptions and significant uncertainties in nucleation rates in the ambient atmosphere, this study should be seen as a first attempt in predicting particle concentrations, as responsibly pointed out by the authors. Overall, the manuscript is well rewritten, presents interesting results, and warrant publication in ACP; however, the model needs better explanation and more detailed discussion should included.

Detailed Comments:

1. The manuscript never explain how boundary layer is defined in the model. What is the vertical resolution of the model near the surface? Does the boundary layer height extends above the first model layer? Are there measurements of boundary layer height at Hyytiälä; if so, how does model PBL heights compare to the measurements? Is it possible that PBL height contributes significantly to the discrepancies in modeled and observed concentrations?
2. The calculation of particle formation rate is not explained in enough detail in section 2.2. The parameterization for the apparent new particle formation rate J_m in Equation (2) assumes that the condensation sink term CS is constant, which requires a small time step. What time step is used for calculating J_m ? How is the growth rate GR determined? Does GR also include organics or just sulfate?
3. The model assumes that newly formed particles that grow to 3 nm consist of pure acidic sulfate. According to Kulmalaetal04, for cluster-activation nucleation represented by Eqn (1), the sulfate is likely ammonium bisulphate and organics are also involved. By not including organics in newly formed particles, the model is reducing gas-phase sulfuric acid faster than if organics were included, which could lead to decreased new

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particle formation rate.

4. There are several competing theories on nucleation mechanism in the literature. The manuscript should provide a brief discussion on why Eqn (1) is chosen for the boundary layer and why binary homogeneous nucleation rate of sulfuric acid/water particles is used above the boundary layer. What are the fundamental differences between the boundary layer and the rest of the troposphere such that a different particle formation scheme is required? While the actual nucleation mechanism is still undetermined, it is well known that ammonia plays a role. What is the implication by not considering ammonia in the nucleation mechanism?

5. In section 2.3, how is SOA distributed among to size bins?

6. In paragraph 3 of section 4.2, the statement “The modelled monoterpenes show the same diurnal cycle, with minima at night due to rapid reaction with NO_3 ” is misleading. The observed diurnal cycle is mostly like due to diurnal cycle of emissions, which is not modeled. Though not universal, monoterpene emissions tend to be high during day time and low during night time. This also means that oxidation by O_3 and OH is biased low during the day and NO_3 oxidation is biased high at night in the model.

7. The last paragraph of section 6.1 goes better with the discussion of the first paragraph of section 6.2. A plot like Figure 6 should be made for the sensitivity runs discussed in the first paragraph of section 6.2.

8. One of the major short-coming of this study is its modeling of aerosol-relevant gas-phase organics and secondary organic aerosol (SOA). The model uses α -pinene as a surrogate of all monoterpenes and assumes 13% aerosol yield under all conditions. In fact, both the gas-phase oxidation rate and aerosol yield can be very different for different monoterpenes. Furthermore, aerosol yield depends on temperature and availability of other organics. Sesquiterpenes, of which oxidation products can have high aerosol yield, is not included in the model. To some extent, the sensitivity simulation on emissions of monoterpenes incorporate all these uncertainties. The manuscript should be

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careful to note that the 10xGEIA emissions simulation is really only a proxy for all the uncertainties related to modeling SOA and SOA precursors.

9. In section 6.3, how does varying k affect CN over polluted regions and globally?

Minor Comments:

1. Last paragraph, page 5 : “Adams and Seinfeld (2003)” should be “Adams and Seinfeld (2002).”

2. Section 3 on page 9: All instances of “BHN of sulfuric acid aerosol” should be rephrased “BHN of sulfuric acid aerosol above the boundary layer” for clarity.

3. Caption for Figure 1(c): There is no red line in the figure; “red” should be “blue”?

4. In the caption for Figure 1(d), “(blue)” should be “(red)”.

5. There is an inconsistency in the units of CS: m^{-2} in the text and s^{-1} in Figure 1(e), which are off by a factor of diffusion coefficient.

6. Page 10, second line of second paragraph of section 4.2: “obvious” should be “obviously”.

7. In section 6.1 and Figure 6, what fractions of the emissions are considered anthropogenic?

8. Table 1: What is the source strength in $Tg\ yr^{-1}$ and $\#\ yr^{-1}$ of sea salt?

References

[Kulmala et al., 2004] Kulmala, M., Kerminen, V.-M., Anttila, T., Laaksonen, A., and O’Dowd, C. D. (2004). Organic aerosol formation via sulphate cluster activation. *J. Geophys. Res.*, 109.

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