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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of nucleation on global CCN" *by* J. Merikanto et al.

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We thank Referee 1 for the constructive comments on our manuscript. The referee asks us to justify some of methods used in our calculations and further discuss some of our results. Below we will answer each of the comments and describe the changes we will make to the manuscript.

In experiment 8 we model the atmospheric CN and CCN concentrations without any primary emissions. The referee asks how we treat the 2.5% fraction of sulfur dioxide normally emitted as primary sulfate. We will revise the description of experiment 8 (page 13009, line 23) to:

"8. UTN+BLN: Runs with UTN represented with Kulmala et al. (1998) parameterization and BLN using $A = 2{\times}10^{-6}$. The run contains no primary emissions and the 2.5% fraction of sulfur, normally emitted as primary sulphate, is emitted as SO_2. This run

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examines the effect of primary emissions on nucleation."

Referee 1 notes that according to Figures 1a and 1b CN and CCN concentration are slightly reduced above $\sim$ 5 km in runs with BLN. This reduction is now discussed in the revised manuscript. However, this reduction is fairly minor since higher UT concentrations are obtained using Vehkamäki et al. (2003) $H_2SO_4 - H_2O$ nucleation parameterization and smaller concentrations with Kulmala et al. (1998) parameterization. In runs with BLN we only used the Kulmala parameterization for $H_2SO_4 - H_2O$ nucleation. To make this clear, we modified the text starting from line 5, page 13010:

"The red areas in Fig. 1 show the resulting profile with primary emissions and upper tropospheric nucleation with the range showing the effect of using two different $H_2SO_4 - H_2O$ nucleation formulations (runs 2 and 4). The smaller concentrations produced by $H_2SO_4 - H_2O$ nucleation are obtained with the Kulmala et al. (1998) parameterization and higher concentrations with the Vehkamäki et al. (2003) parameterization. Finally, the green areas show the profile with standard primary particle emissions, upper tropospheric nucleation using the Kulmala et al. (1998) parameterization, and boundary layer nucleation represented with varying rates of activation nucleation (runs 3 and 6)."

We will add to page 13011, line 2: "BLN causes a minor reduction of CN and CCN above $\sim$ 5 km. The resulting reduction is due to enhanced condensation of sulfuric acid and secondary organics on boundary layer particles. Hence less sulfuric acid and secondary organics are transported to UT where they would contribute to nucleation (sulfuric acid) and subsequent particle growth (sulfuric acid and secondary organics)."

Referee 1 asks why we use still use Kulmala (1998) $H_2SO_4 - H_2O$ nucleation parameterization instead of the newer Vehkamäki (2003) parameterization as the standard scheme. In hindsight the Vehkamäki parameterization would indeed be preferable to the Kulmala parameterization since the Vehkamäki parameterization is based on a more rigorous theoretical analysis and is in better comparison with laboratory experi-

ACPD
9, C6464–C6466, 2009

ments. We use the Kulmala parameterization as the standard scheme in the current paper for two reasons: First, in Spracklen et al. (2005) we showed that the Kulmala parameterization produces observed CN concentrations in the marine boundary layer and, according to our new (yet unpublished) results, the Kulmala parameterization can also explain observed CN in many UT measurement stations. The Kulmala parameterization seems to describe UT nucleation reasonably, and it is not clear if the Vehkamäki parameterization does a better job when compared to observational data. Also, using the Kulmala parameterization here means the results presented in this paper will be consistent with our other work. Secondly, while the Vehkamäki parameterization yields higher nucleation rates in upper troposphere, the concentrations (particularly in the boundary layer) are only mildly affected. However, we do agree that Vehkamäki rates are more trustworthy and should be utilized in future studies.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12999, 2009.