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Interactive comment on “Simulation of particle size distribution with a global aerosol model: contribution of nucleation to aerosol and CCN number concentrations” by F. Yu and G. Luo

F. Yu

yfq@asrc.cestm.albany.edu

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The authors thank the referee for the helpful comments. Our responses to the comments are given below.

Major comments

One of the significant conclusions of this work is that ion-induced nucleation can explain a large fraction of observed particle number. This conclusion is contrary to a number of existing studies (e.g., Kulmala et al., 2007; Boy et al., 2008) which suggest that ion induced nucleation is not a dominate particle formation mechanism. However, there is no reference to these previous studies or any explanation for this significant difference

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in conclusions. At the very least I think a discussion of these previous studies should be included, if it is not possible to account for the difference in conclusions.

The controversy about the importance of IMN based on the analysis of nucleation events observed in boreal forest has been discussed in Yu et al. (2008) and Yu and Turco (2008) and associated ACPD online open discussions. We think that Kulmala and colleagues (Laakso et al., 2007; Kulmala et al., 2007; Boy et al., 2008) might have underestimated the importance of IMN.

Laakso et al. (2007) extrapolated measured charging states of nucleation mode particles (3 - 7 nm) down to smaller sizes (1-2 nm) and concluded that the contribution of IMN to total nucleation rate was either negligible or relatively small in a large fraction of days considered. However, Yu et al. (2007a) demonstrated that the results of Laakso et al. (2007) may actually support the significance of IMN, contradicting their stated conclusion but not their data. The work of Kulmala et al. (2007) has been discussed in Yu et al. (2007b) and it has been emphasized that a substantial contribution from ion-mediated nucleation could not be ruled out based on the data reported in Kulmala et al. (2007).

Boy et al. (2008) simulated 4 days of nucleation events using atmospheric input data from the SMEAR II station. They calculated the IMN rate based on the model of Kazil and Lovejoy (2007), and used the empirical activation and kinetic nucleation formulas to represent neutral nucleation. By comparing their calculated ion and neutral nucleation rates, Boy et al. (2008) concluded that IMN contributes between <0.5 to 12% to the total number of particles nucleated inside the mixed layer in the boreal forests. The conclusion of Boy et al. (2008) is not surprising because they used the ion nucleation model similar to that of Lovejoy et al. (2004), which under-predicts ion nucleation rate by several orders of magnitude (Yu and Turco, 2008). In addition, Boy et al. (2008) used the empirical activation and kinetic nucleation formulas to represent neutral nucleation. It remains to be established whether the empirical formula indicates a new nucleation mechanism or if it simply represents an empirical fitting of exiting nucleation process

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such as IMN (Yu and Turco, 2008).

The focus on the present paper is on global aerosol modeling. To address the referee's concern, the existence of the controversy has been briefly discussed in the revised paper.

Minor comments

P10600, L20: Change "Pierce and Adam" to "Pierce and Adams"

Corrected.

P10601, L13: Change "w" to "W"

Corrected.

P10601, L16: Spracklen et al., 2006 was the first study to include this empirical mechanism in a global model.

The reference has been added.

P10601, L20: This implies that Hyytiala is the only location where this parameterization has been shown to match observed nucleation events. However, this parameterization has now been shown to work at a number of locations (e.g., Riipinen et al., 2007; Kuang et al., 2008).

To address the referee's concern, we have changed "in Hyytiala, Finland" to "in a number of locations".

P10604, L27 and L28: Should this be GEOS?

Corrected.

P10614, L1 Change to "Existing"

Corrected.

Table 1. Are some of these observations not CN3 (> 3 nm dry diameter)?

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All the data given in Table 1 are CN10.

Additional References:

Lovejoy, E. R., J. Curtius, and K. D. Froyd (2004), Atmospheric ion-induced nucleation of sulfuric acid and water, *J. Geophys. Res.*, 109, D08204, doi:10.1029/2003JD004460.

Kazil, J. and Lovejoy, E. R. (2007) A semi-analytical method for calculating rates of new sulfate aerosol formation from the gas phase, *Atmos. Chem. Phys.*, 7, 3447-3459.

Yu, F., et al. (2007a), Interactive comment on “Ion-mediated nucleation as an important global source of tropospheric aerosols”, *Atmos. Chem. Phys. Discuss.*, 7, S6602–S6608. www.atmos-chem-phys-discuss.net/7/S6602/2007/

Yu, F., et al. (2007b), Interactive comment on “Ion-mediated nucleation as an important global source of tropospheric aerosols”, *Atmos. Chem. Phys. Discuss.*, 7, S6754–S6766. www.atmos-chem-phys-discuss.net/7/S6754/2007/

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 10597, 2009.

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