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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ion-mediated nucleation as an important global source oftropospheric aerosols" *by* F. Yu et al.

Anonymous Referee #2

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General comments

The authors present a global scale comparison of ion induced nucleation rates, calculated with a chemistry transport model, with observed particle formation rates. Despite the considerable difficulties posed by such an undertaking, the simulated spatial distribution of the nucleation rates resembles reasonably well that of observed particle formation rates and ultrafine particle concentrations. The authors indicate issues that may impede the comparison, which include:

- Measurements typically give formation rates of particles that are larger than 3 nm in diameter (at the instrument RH). Nucleation rates are, however, formation rates

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of supercritical particles that are larger than the critical cluster. The critical cluster contains, in conditions at which nucleation occurs only a few sulfuric acid molecules, and is hence much smaller than 3 nm. The nucleation rate and the formation rate of larger particles may therefore differ by orders of magnitude [4, 2].

- Global scale models do not resolve variability below a scale of $\sim$ 100 km, but measurements do. Due to non-linearity of the nucleation process subgrid scale variability may lead to nucleation in excess of what average conditions on the $\sim$ 100 km scale would give.

The comparison of the modeled annually and zonally averaged nucleation rate with the observed zonally averaged ultrafine particle concentrations in Figure 3 is meaningful, as well as the comparison of the nucleation rate with the emission rate of primary aerosol in Figure 4, which is discussed well. I concur with the points raised by Referee #3, except for point 8, which I explain below. I recommend the manuscript for publication subject to changes in response to the details below.

Specific comments

Page 13604, line 9:

At given values of [H_2SO_4], T, RH, Q, and S_0,J_{IMN} can be accurately decided using the look-up tables with an efficient multiple-variable interpolation scheme.

More details would help support the point on the precision of the lookup table interpolation. Can you give a measure of how well the interpolation of the lookup table reproduces nucleation rates calculated with the detailed model? What is the resolution of the lookup table in the different dimensions? What is the multiple-variable interpolation scheme exactly?

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Page 13602, line 15:

Due to the uncertainty of the emissions from sporadically erupting volcanoes, we only consider the continuously active volcanoes emission in this study. The emission from eruptive volcanoes is not considered in the study of Lucas and Akimoto (2006) as well.

The first rationale is much better than the latter.

Page 13606, line 7:

Observed nucleation events typically last for 3 h a day, ...

Why not giving a reference here, e.g. [1] or [3]?

Page 13606, line 18 and 25:

Most boundary layer nucleation events in the northern hemisphere (except over remote ocean areas, and Greenland) are associated with anthropogenic SO_2 emissions; in the southern hemisphere, nucleation is triggered both by oceanic DMS and anthropogenic SO_2.

The simulations also indicate that nucleation induced by anthropogenic SO_2 emission contributes to particle abundances in the southern hemisphere.

Could you say a few words how you came to these conclusions? While it seems safe to assume that SO_2 in the vicinity of strong anthropogenic sources is mainly responsible for nucleation, how do you identify the relative contributions of anthropogenic and natural SO_2 to nucleation farther away?

Page 13608, line 10-12: I disagree with Referee #3 on this point: Transport and mixing can make small particles ”pop” up quickly at a measurement location and thus mimick very fast nucleation in conditions that would support nucleation only at a much lower rate. This effect is more likely to occur the larger the smallest particles are that can be detected.

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Page 13609, line 27:

The general agreement between simulations and observations demonstrated above strongly supports the important role of IMN in generating new particles in global troposphere.

Given the considerable uncertainties involved in the comparison (see ”General comments” above) and without considering other nucleation pathways, such as neutral binary and ternary nucleation, and the possible role of organic molecules in aerosol nucleation, a slightly more cautious wording could be justified here.

Technical details

Page 13613, line 11: A. D. Clarke, not A. Clark.

Please check the English, such as:

Page 13599, line 4: ”dominant”, not ”dominate”

Page 13605, line 28: ”the increased production dominates”, not ”the increased production dominate”

Page 13605, line 29: ”thus [H_2SO_4] is generally higher”, not ”thus [H_2SO_4] are generally higher”

Page 13606, line 1: ”decreases with altitude due to the more rapid decrease of”, not ”decreases with altitude due to more rapidly decrease of”

Various occurrences: ”Continents”, not ”continentals”.

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References

[1] W. Birmili, H. Berresheim, C. Plass-Dülmer, T. Elste, S. Gilge, A. Wiedensohler, and U. Uhrner. The Hohenpeissenberg aerosol formation experiment (HAFEX): a long-term study including size-resolved aerosol, H_2SO_4, OH, and monoterpenes measurements. *Atm. Chem. Phys.*, 3:361–376, 2003.

[2] J. Kazil and E. R. Lovejoy. A semi-analytical method for calculating rates of new sulfate aerosol formation from the gas phase. *Atm. Chem. Phys.*, 7:3447–3459, 2007.

[3] L. Laakso, T. Anttila, K. E. J. Lehtinen, P. P. Aalto, M. Kulmala, U. Hõrrak, J. Paatero, M. Hanke, and F. Arnold. Kinetic nucleation and ions in boreal forest particle formation events. *Atm. Chem. Phys.*, 4:2353–2366, 2004.

[4] R. P. Turco, J.-X. Zhao, and F. Yu. A new source of tropospheric aerosols: Ion-ion recombination. *Geophys. Res. Lett.*, 25:635–638, 1998.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 13597, 2007.