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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The size-dependent charge fraction of sub-3-nm particles as a key diagnostic of competitive nucleation mechanisms under atmospheric conditions" *by* F. Yu and R. Turco

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Review of "The size-dependent charge fraction of sub-3-nm particles as a key diagnostic of competitive nucleation mechanisms under atmospheric conditions" by F. Yu and R. Turco

This paper revisits the standing debate on the role of ions in aerosol nucleation in Hyytiälä, Finland. The authors use a box model of ion-mediated nucleation (IMN) and aerosol microphysics, which uses chemical and meteorological measurements as in-

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Interactive Discussion

Discussion Paper

puts, to explore the evolution of aerosol charge as particles grow. The main conclusion of the paper is that although measurements show that the contribution of charged processes to the formation rate of 2 nm particles (J2) is low (order of 10%), the contribution of charged processes to the formation rate of 1.5 nm particles is virtually 100% at Hyytiälä.

The paper is well written and the figures are clear and interesting. The paper is certainly of interest to the ACP readership. However, I found two issues in the analysis/discussion that add uncertainties to the conclusions of the paper. I feel that additional discussion, or even analyses, would make this paper stronger and more informative. Once these issues have been addressed, the paper should be published in ACP.

General comments

1. The authors state on Page 11286 lines 3-7 that Kulmala and colleagues underestimate the importance of IMN by treating all neutral clusters with Dp < 2 nm (including those formed through ion-ion recombination) as formed through neutral processes. However, Manninen et al. (2009) did account for ion-ion recombination in their estimate of the IMN contribution at Hyytiälä ($\sim$10%).

As I read the current manuscript I was left wanting a thorough discussion/analysis about why these the Manninen and the current manuscript's estimates differ by such large amounts. In Manninen they work backwards from the measurements, in the current manuscript the model works forward from the Yu IMN model. Are there poor assumptions in the Manninen formulas? Are there uncertainties in the current scheme? Why don't these agree?

Please add a thorough discussion/comparison.

2. The box model used in this study only has H2SO4 as a condensible species. Riipinen et al. (2011) showed that H2SO4 accounted for <20% of the measured growth

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rate of 1.5-3 nm particles at Hyytiälä, and the balance must be by low-volatility organics. Yu (2011) showed how important organic condensation processes are to ultrafine particle growth in the global atmosphere.

Ignoring organic condensation in the box model of Hyytiälä implies that predicted growth rates may be biased low. If growth rates were faster in the box model (due to organic condensation), there would be less time for recombination to occur between 1.5 nm and 2 nm, and the predicted charge fractions at 2 nm would increase. Its not obvious if the model would agree as well with observations when this is taken into account.

Has a sensitivity analysis been done on this? How well do the modelled growth rates compare to the measurements? Is this a reason for the discrepancy between the current analysis and the Manninen et al. (2009) paper? It seems like it could be very important to the conclusions. Please discuss.

Riipinen, I., Pierce, J. R., Yli-Juuti, T., Nieminen, T., Häkkinen, S., Ehn, M., Junninen, H., Lehtipalo, K., Petäjä, T., Slowik, J., Chang, R., Shantz, N. C., Abbatt, J., Leaitch, W. R., Kerminen, V.-M., Worsnop, D. R., Pandis, S. N., Donahue, N. M., and Kulmala, M.: Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations, Atmos. Chem. Phys., 11, 3865-3878, doi:10.5194/acp-11-3865-2011, 2011.

Yu, F.: A secondary organic aerosol formation model considering successive oxidation aging and kinetic condensation of organic compounds: global scale implications, Atmos. Chem. Phys., 11, 1083-1099, doi:10.5194/acp-11-1083-2011, 2011.

Other comments:

Please be consistent for the 0 vs. neutral superscripts throughout.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 11281, 2011.