

Interactive comment on “Aerosol dynamics simulations on the connection of sulphuric acid and new particle formation” by S.-L. Sihto et al.

Anonymous Referee #1

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General comments

The authors investigate the correlation of the nucleation rate and of the formation rate and concentration of 3-6 nm particles with gas phase sulfuric acid, using a detailed aerosol model. The correlation of the concentration of 3-6 nm particles with gas phase sulfuric acid during nucleation events is available from field studies. Correlations produced by different nucleation mechanisms and their response to condensation of a non-nucleating compound are analyzed. This study is important as it shows how in situ measurements can be used to determine the mechanism responsible for a nucleation event.

In their simulations, the authors show that activation type nucleation is a good can-
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didate to explain the correlation often observed in the boundary layer, while ternary nucleation, implemented using a parameterization, is not. The authors therefore challenge the role of ternary nucleation in new particle formation from the gas phase in the boundary layer. With this conclusion the authors join the controversy on the role of ternary nucleation (of ammonia, sulfuric acid, and water) in atmospheric new particle formation. This controversy draws on numerous conflicting results, e.g. by Ball et al. (1999); Janson et al. (2001); Kulmala et al. (2002); Napari et al. (2002a); Anttila et al. (2005); Yu (2006); Kurtén et al. (2007).

In this context it should be noted that the majority of boundary layer nucleation observations has been conducted over continents, and many of these over forested areas, where organics seem important for nucleation. Over oceans and in polluted areas, however, organic molecules may play a secondary role; in a very recent analysis of nucleation events in the Pittsburgh area, e.g., Jung et al. (2008) find, using the same parameterization of ternary nucleation (Napari et al. , 2002b) as in the present work, that ternary nucleation works quite fine.

The manuscript is well written and the analysis of the results results thorough, providing new, relevant insights. Given the conflicting findings mentioned above and the uncertainties regarding the validity of the ternary nucleation parameterization used, the authors' quite general dismissal of the role of ternary nucleation in the boundary layer is courageous.

Specific comments

Page 11367, line 29:

The saturation vapour concentration of sulphuric acid is assumed to be negligible, i.e. it condenses with the maximum flux without Kelvin effect ($c_{sat}=0$).

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This is not a very good approximation for very small sulfuric acid/water aerosol particles, unless they contain sufficient amounts of ammonia. The growth rates of the particles will be overestimated, and consequently, Figures 5 and 6 show upper limits for the sulfuric acid vapor growth rate. More slowly growing particles are more affected by loss onto pre-existing aerosol and by self-coagulation. Hence there is a chance that if the actual condensation/evaporation rates of sulfuric acid were used, somewhat different correlations may have been obtained.

Page 11381, line 17:

Simulations with ternary H_2O - H_2SO_4 - NH_3 nucleation yielded too steep dependence on sulphuric acid, with regard to observed exponents of 1-2 in field measurements. The correlation exponents were always greater than 4 for both J_3 and N_{3-6} , suggesting that ternary nucleation is not the valid nucleation mechanism for particle formation in atmospheric boundary layer.

This is the passage I am referring to in the "General comments" section.

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