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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An improved criterion for new particle formation in diverse atmospheric environments" *by* C. Kuang et al.

Anonymous Referee #2

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Kuang et al. present an extended dimensionless theory for new particle formation building on previous theoretical approaches (McMurry, 1983; Lehtinen and Kulmala, 2003). Two key modifications are introduced based on recent experimental observations of atmospheric particle nucleation in different environments: (1) A parameterization of the nucleation rate as a power-law function of the gas-phase sulfuric acid concentration, and (2) a growth enhancement factor Γ to take into account multi-component condensation of other vapor-phase species. Appropriate scaling of the key variables leads to a set of dimensionless population balance equations that are applied to a diverse set of atmospheric new particle formation events. A dimensionless parameter $L\Gamma$ is shown to serve as a unique criterion for the occurrence of new particle formation in diverse environments.

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Interactive Discussion

Discussion Paper

The model development is presented in a clear and structured way. Each step is put in context with previous theoretical approaches, and the implications of introducing recent developments are discussed in detail. Observational data from six intensive measurement campaigns are used to evaluate the model performance, and to test the criterion for new particle formation.

I recommend publication of this manuscript in ACP after consideration of the following comments:

a) It is not clear if the authors see predictive capabilities in their criterion for new particle formation. There are statements in the manuscript that a new criterion "would form an important component of predictive models for aerosol formation" (p. 493, l. 20/21) and that it "can be used to predict the frequency and relative strength of NPF events" (p. 508, l. 10/11). However, given the high demand on observational data required as input for the model, the potential of the presented theory to predict new particle formation remains unclear. For example, the pre-factor K introduced in Eq. 3 is characterized as campaign-specific, indicating that it may not be considered universal even for a specific site. It must be determined individually for each measurement campaign. Also, the growth enhancement factor Γ is derived from an analysis of measured growth rates and condensation of sulfuric acid of individual NPF events. Thus, the potential of the new criterion for the prediction of NPF should be clarified in the manuscript.

b) The growth enhancement factor Γ is introduced in order to take into account multi-component processes in particle nucleation and growth. However, since it is simply a multiplier of the sulfuric acid concentration to tweak the condensational growth parameterization, the model is still based on single-component condensation of sulfuric acid. This is stated by the authors, together with two remarks that (1) the condensing species taking part in nucleation and growth are likely different (p. 494, l. 19/20), and (2) growth rates might depend on particle size (p.499, l. 9-10). A brief discussion of the uncertainties associated with the introduction of Γ would help the reader to appreciate the current limitations in studying aerosol nucleation processes. For example, what

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is a reasonable estimate of the uncertainty of growth rate extrapolations from aerosol measurements larger than 3 nm in diameter to growth just after nucleation?

c) Results of the EUCAARI campaign are not presented in Figs. 1-3, even though it is the largest individual data set in Tables 1 and 2. The authors should include the EUCAARI data in Figs. 1 and 3, or give a reasonable explanation for the different treatment of this data set.

d) The core of the NPF criterion development resides in Eq. 17 which defines LΓ as the ratio of the scavenging loss rate to the growth rate. LΓ is closely related to a similar expression (L) proposed by McMurry (1983) but it is derived from a more general form of the population balance equations. When comparing L and the new dimensionless L1 as defined in Eq. 11, it is striking that β11, the monomer-monomer coagulation coefficient, and K, the campaign-specific pre-factor used in the power-law parameterization of the nucleation rate, are utilized in a similar fashion. If K = β11, the two formulations collapse to the same expression. This interchangeable use of K and β11 motivates a different point of view of the nucleation rate parameterization in Eq. 3. In the manuscript, the authors keep the exponent constant P=2 based on the work of Kuang et al. (2008). This is consistent with a kinetic nucleation mechanism of sulfuric acid. However, the authors acknowledge that P has been shown to vary between 1 and 2 representing a mixture of activation of pre-existing clusters (P=1), kinetic nucleation (P=2), possibly classical ternary nucleation theory (P>2), and assuming a multi-component nucleation process. One could also keep the pre-factor constant K = β11, and allow 1 < P < 2. It may be worthwhile discussing this aspect in the present manuscript.

Minor comments:

e) Section 2.1 briefly summarizes the measurements utilized in this study by giving the relevant references. I would like to suggest adding one or two sentences mentioning the relevant aerosol and gas-phase instruments used in these field campaigns (DMPS,

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CIMS, others?).

f) The histogram classes of LΓ are hard to interpret from Fig. 4. Please indicate if the LΓ bins are on a logarithmic scale or something similar.

References:

Kuang, C., McMurry, P. H., McCormick, A. V., and Eisele, F. L. (2008) Dependence of nucleation rates on sulfuric acid vapor concentration in diverse atmospheric locations. J. Geophys. Res., 110, D10209, doi:10.1029/2007JD009253.

Lehtinen, K. E. J. and Kulmala, M. (2003) A model for particle formation and growth in the atmosphere with molecular resolution in size. Atmos. Chem. Phys., 3, 251–257.

McMurry, P. H. (1983) New particle formation in the presence of an aerosol: rates, time scales, and sub-0.01 μm size distributions. J. Colloid Interface Sci., 95, 72–80.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 491, 2010.