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

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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.

While the cluster population balance model requires observationally-constrained inputs for the nucleation rate pre-factor K and the growth-enhancement factor Γ, calculation of the new particle formation criterion $L\Gamma$ requires only inputs for the pre-existing aerosol surface area and the nucleation mode growth rate (which is equivalent to having both the peak value of [H2SO4] and the growth enhancement factor). At a particular location, while fairly reasonable estimates of the pre-existing aerosol surface area can be used as model inputs, there are, at this point, only empirical means of determining Γ, either from measurements of nucleation mode particle composition or by comparing measured and sulfuric acid-limited growth rates. There is evidence that NPF at a particular site is characterized by a relatively narrow range of growth enhancement factors, such as in Atlanta, Boulder, and Mexico City. For those types of NPF events, application of this NPF criterion would provide a reasonable estimate for the relative strength and frequency of NPF. The predictive power of this criterion will improve as better estimates are made regarding particle growth rates.

b) The growth enhancement factor Γ is introduced in order to take into account multiÂňcomponent processes in particle nucleation and growth. However, since it is simply a multiplier of the sulfuric acid concentration to tweak the condensational growth paÂňrameterization, 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 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?

Assuming that particles throughout the nucleation mode undergo the same enhancement to growth does lead to some uncertainty in the growth rate for particles smaller than 3 nm, since the measured growth rate scales directly with Γ. The extrapolation of growth rates from aerosol measurements larger than 3 nm to growth just after nucleation, however, is not reasonable, given that growth rates obtained from time-shifts between [H2SO4] and N3-6 (growth rates below 3 nm) are comparable to modal diameter growth rates (growth rates above 3 nm).

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.

Figures 1 and 3 presented results whose primary purpose was to validate the cluster population balance model. Inclusion of the EUCAARI data set would not have added anything new to those particular conclusions and would have required a substantial amount of analysis. Since the main purpose of this study was to demonstrate the validity of the NPF criterion, results from EUCAARI were presented in Figure 4, showing the histogram of measured $L\Gamma$ values. Figure 3 presents model results from each campaign.

d) The core of the NPF criterion development resides in Eq. 17 which defines $L\Gamma$ as the ratio of the scavenging loss rate to the growth rate. $L\Gamma$ 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 beÂňtween 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.

The nucleation rate pre-factor K and the monomer-monomer coagulation coefficient β11 are conceptually similar in the case of a kinetic nucleation mechanism where they both define the probability that a monomer-monomer collision results in the formation of a stable dimer. However, in our study, setting the nucleation pre-factor K = β11 results in the gross over-estimation of the nucleation rate by several orders of magnitude, which would overestimate, among other things, the effects of cluster-cluster coagulation. It has been stated in the manuscript (pg. 16 lines 20 – 25) that the derivation of the NPF criterion $L\Gamma$ is independent of the form of the nucleation rate, so, in principle, any value of the nucleation rate pre-factor K or the nucleation rate exponent P can be used. What is important, however, is that the magnitude of the resulting nucleation rate is constrained to measured values so that the resulting competition between cluster-cluster coagulation, cluster scavenging, and cluster growth are representative of ambient processes.

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, CIMS, others?).

References to the appropriate gas-phase measurements of H2SO4 by the CIMS and to the measurements of aerosol size distribution by DMPS are included in the citation of [Kuang et al., 2008].

f) The histogram classes of $L\Gamma$ are hard to interpret from Fig. 4. Please indicate if the

LΓ bins are on a logarithmic scale or something similar.

The histogram bins in Figure 4 are logarithmically spaced to more clearly show the two orders of magnitude spread in the values of LΓ for the analyzed NPF events. This clarification has been added to the caption of Figure 4.

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