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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 #3

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Kuang et al. present a nice piece of work about new dimensionless parameter "L_Γ", characterizing the ratio of the particle loss rate to the measured particle growth rate, which is used to determine whether or not NPF would occur on a particular day. The topic of the manuscript sounds very interesting. The manuscript is definitely in the scope of ACP and I suggest publishing this article after considering the following suggestions and comments.

General comments: Referee 2 suggested many improvements to the manuscript about which I agree completely. For this reason I will not start all over again but concentrate on some issues.

(1) Γ is the ratio between $GR_{[Measured]}$ to $GR_{[H2SO4]}$

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I am very much concerned about how "practically" the authors have taken the growth due to other species, e.g. organics, into account? How the growth enhancement factor Γ is introduced in order to take into account multicomponent processes in particle nucleation and early growth. It seems that Γ is simply just a multiplier of the sulfuric acid concentration due to condensational growth? On p.499, just right after eq. 6, the authors state that "While the growth enhancement factor Γ incorporates the effects of multi-component condensation, Eqs. (5) and (6) are still based on a single-component formulation since Γ is calculated assuming a condensing molecular volume of hydrated sulfuric acid". Please explain this "main improvement point" in more detail in the revised version.

(2) I really cannot understand why nano particle growth rates are that high sometimes? Could you please explain? For example, in table 1, row EUCAARI 04/15/07, the values indicate a total growth rate of about 40 nm/h. At a H_2SO_4 concentration of 10^7 molecules cm^{-3}, the nanoparticle growth rate is about 1.5 nmh^{-1}. Surely, there are several condensing vapors obviously participating in the growth process as well, however total growth rates typically do not exceed 20 nm h^{-1}. There are exceptions such as coastal areas, where oxidation of iodine-containing vapors can rapidly produce large amounts of condensable matter and also highly polluted environments, but here the explanation should be different?

(3) Growth enhancement factor (Γ) spans the range from 1 (ANARChE) to over 100 (EUCAARI)? The authors claim that this might depend on the location. However, in some cases the growth enhancement factor varies in the same location by a factor of as much as 34 within the same month while the amount of measured sulfuric acid concentrations remains almost in the same range. How is this possible? I think the discussion of the variability of Gamma, as well as the related uncertainties and limitations should be discussed in more detail.

ACPD
10, C887–C891, 2010

(4) I would like to see a figure, instead of a table, showing number of event days and nonevent days as a function of Γ values for each measurement sites. It was hard to make any criteria based on Γ values to distinguish between events and nonevents while Γ here is presented as a key factor for the improved dimensionless parameter. This point should be well clarified in the manuscript. (Yes, I noticed that LΓ was presented as a function frequency of NPF and non nucleation in Fig. 4, but this is a different story and we will get back to it later.)

(5) As Gamma is presented in the tables, and used in the Equations following Eq. 6, please present an equation showing how it is calculated instead of just mentioning it inside the text.

I would also be interested to see the modeled formula for N_{3-4}nm.

(6) In the methodology:

How was Nm (peak [H2SO4] during NPF event) defined for Nonevent days?

To deal with the six intensive measurement campaigns similarly, the authors estimated K (pre-factor) values based on the power-law equation: J†=K[H2SO4]^P and by assuming nucleation exponent (P=2; i.e assuming Kinetic nucleation mechanism for all stations) and taking the time for event days to be "times when events take place i.e during NPF event" So my question is how you get K (Kinetic coefficient) values for nonevent days while J†is missing ?. For ambient data, in which K ranges from 10^{-14} to 10^{-11} cm^3 s^{-1}, have you used these ranges for K values for non-nucleation days??

Overall, a few sentences about the non-nucleation days would be very useful. (The reader necessary needs to know; how the non-nucleation data parameters were treated as well in order to be comparable with NPF days and therefore to be tested by the new criteria).

ACPD
10, C887–C891, 2010

Criterion was tested against measured 77 days of new particle formation and for 19 non-nucleation days BUT so far the classification of new particle formation days has usually been based on visual examination of the temporal development of size distributions ("banana plots") to distinguish between NPF and no nucleation days. The coauthors checked these classified days by using their criteria (calculating L_Γ factor for both data sets (NPF and no nucleation days)) and plotted against the Frequency (Figure 4), so isn't it so that the authors have made an interesting *observation* about event days versus some nonevent days, and I am afraid that the word "criterion" is not rightly used here.

P. 508 line 9-11: authors claimed that L_Γ is a robust parameter that can be used to **predict** the frequency and relative strength of NPF events. However to accurately predict NPF, it is necessarily to get from the observational data i) the measured growth rate, and ii) estimate the growth rate assuming only sulfuric acid condensation ; iii) estimate activation pre-factor K, to be able to present L_Γ factor correctly in the model. Please detailed discussion of this aspect in the revised version will be so much appreciated.

(7) The parameters L_1 and Γ_1 characterize the scavenging and growth processes, respectively.

Therefore these two parameters are the core of the developed "criterion" for new particle formation. Later on, these two parameters were combined as L_Γ ($L_{1/\ \ \Gamma_1}$) and were used to estimate the survival probability of a nucleated particle growing to the detection limit (3 nm) i.e. J_3, and as an expected result, J_3 depends only on the ratio L_1 and Γ_1. If the new modified L_Γ seems to be identical to the old L when the whole growth is caused by H_2SO_4 as in sulfur-rich environment, would it be more informative here for the authors to present the survival probability J_3 as a function of old L and the modified Lgama respectively in order to show how sensitive the modified growth enhancement factor improve the survival probability J_3 estimation?

As it was mentioned by the authors that L_Γ is independent of the nucleation rate pre-

ACPD
10, C887–C891, 2010

factor K and nucleation exponent P therefore the main KEY here is how the enhancement growth factor improves this NPF “criterion”? A figure would be useful to show how well the improved “modified L –factor theory” really worked?

(8) Figure 2: Identical behavior was also observed in the model results for the other measurement campaigns so was there any particular reason that authors picked MILAGRO station with 9 new particle formation days and zero non nucleation days?

Minor comments:

Page 497, line 11. k under the square root is Boltzmann constant (i.e K_B)not the cluster size k, Am I right? . Please correct.

Fig 3. in the figure captions $L_\Gamma$was defined as L_1 divided by Γ_1, while in page 506 ,equation 17 , L_Γ was defined as the ratio between L and Gamma ? Please unify your definitions.

Fig3. was it only for NPF days or all datasets? Please clarify.

In the reference list, the "a" is missing from Kulmala et al., 2004a

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