We thank Veli-Matti Kerminen for carefully reading our manuscript and for providing constructive comments, which have improved the quality and clarity of our manuscript. His comments are repeated in full below, with our replies indicated after each comment in blue font. Text which has been added to the manuscript is shown in red font.

V.-M. Kerminen

This manuscript presents an analysis on how nucleation rates should be derived from experimental particle formation rates, including those obtained from chamber experiments. Noting the increasingly important role of chamber experiments in studies of new-particle formation and growth, the topic of this paper is definitely a very important one.

The manuscript brings up an essential, yet not surprising, result: aerosol dynamics in an environmental chamber can be very different from that in the ambient atmosphere. Related to this, the authors 1) first demonstrate that the original method by KK2002 in deriving nucleation rates from measured particle formation rates is not applicable to chamber experiments, and 2) then introduce a revised method which is generally applicable to such experiments. I very much welcome the point 2. I also think that it is valuable to discuss the limitations of earlier approaches (point 1), but such a discussion should be done in a correct way. I mainly agree with comments by the first anonymous reviewer. In addition to that, I have a few issues of my own for the authors to consider before recommending acceptance of this paper for publication.

The authors (K2014) spend two subsections (2.1 and 2.2 in K2014) to demonstrate why the KK2002 method does not perform well with typical chamber experiments. At the end of page 27236 (lines19-21) and also on page 2740 (lines 2-4), K2014 give readers the impression that the failure of KK2002 method in chamber experiments is due to some other reason than violation against the assumptions 1-3 (see line 10 on page 27236) which were stated very clearly in the original manuscript by KK2002. This is not correct. K2014 shows that in chamber experiments, the main sink for small nucleated clusters is either chamber walls or small (<10 nm) nuclei formed earlier during the experiment. A large chamber wall sink clearly violates the assumption 1, while a large sink by small nuclei violates either the assumption 1 or assumption 3 depending on how “pre-existing larger particles” are being defined (admittedly, this definition was a bit loose in KK2002). Most atmospheric aerosol scientists would not call 3-10 nm particles as “larger pre-existing particles”, in which case the large sink by such particles would violate the assumption 1. K2014 tends to categorize these 3-10 nm particles as part of the sub-population of “larger pre-existing particles”. In this interpretation, the large sink caused by such particles violates the assumptions 3, since this subpopulation undergoes major changes during the experiment. In summary: KK2002 fails in chamber experiments because the conditions in such experiments violate the very clearly-stated assumptions in the original paper by KK2002. This is definitely a valuable point to bring up in K2014, even though essentially the same information is already available in KK2002. At least some revisions of the in subsection 2.1 and 2.2 are necessary based on the arguments above.

(1) We have modified the first two sub-sections of section 2 substantially based on the criticism made by the reviewer. Since the first anonymous reviewer had very similar comments regarding the first part of the manuscript the following text is a copy of the reply given to reviewer #1:

In our revised manuscript section 2.1 now includes a broader overview over the series of the four papers (Kerminen and Kulmala, 2002; Lehtinen et al., 2007; Anttila et al., 2010; Korhonen et al., 2014) without deriving any of the equations. Section 2.1 only mentions the most relevant equations from Kerminen and Kulmala (2002) and Lehtinen et al. (2007), which are necessary to calculate $J(d_{p2})$ from $J(d_{p1})$, where $d_{p2} > d_{p1}$.

The revised section 2.2 includes a relationship between $J(d_{p2})$ from $J(d_{p1})$ by taking into account two relevant loss process in chamber nucleation experiments, i.e. wall loss and dilution. We acknowledge that this relationship can also be derived from the equations presented by Lehtinen et al. (2007). However, we feel that it is important to highlight what considerations and requirements go into the previously published method and how it can be applied to chamber experiments. To our knowledge this has not been described explicitly before. While this is not a main finding of our paper, it is an important aspect that is worth explicit mentioning and brief discussion.

In summary, the first two sections of the second paragraph have been substantially revised according to Veli-Matti Kerminens' suggestions. They will now serve as an overview for readers who are not experts regarding the Kerminen and Kulmala equation and its follow-up publications (section 2.1). The second section (2.2) formulates an easy-to-use formula for nucleation rates from chamber nucleation studies.

K2014 introduces a new method to derive nucleation rates from measured particle formation rates which works for chamber experiments. Very good, in principle! However, I am not confident that the new method can be called universal. The method takes into account the influence of self-coagulation on the nuclei number concentration (the term j=m in the r.h.s of Eq. 25), but does it take into account the influence of self-coagulation on the nuclei growth rate (Eq. 4)? The times scales over which nuclei self coagulation affects their number concentration and growth are so similar that these two processes become important under roughly the same conditions.

(2) Based on the comment by Hanna Vehkamäki we have removed the word "universal" in the context of the new method. However, Hanna Vehkamäki had different reasons for requesting to remove this term.

In the context of the comment above, the question is addressed whether self-coagulation is taken into account not only in terms of the nuclei concentration but also regarding the nuclei growth rate.

While it was maybe not that obvious from our originally submitted manuscript that both effects are taken into account the revised version now sheds more light on this question (see also reply to comment (2) by reviewer #1). The central point is how the particle formation rate and the growth rate are defined. If they are defined in an appropriate way the relationship $J = GR*N/\mathrm{d}d_p$ also holds when cluster-cluster collisions become important. Section 2.4 in the revised manuscript deals with the question if the relationship between *J* and *GR* is valid in such cases. Based on our arguments in this section (visualized in the new Fig. 3) we can conclude that the effect of self-coagulation on the *GR* is adequately taken into account by our method. The growth rate used in the reconstruction method is based on the definition in equation (24). The formation rate $J_{\geq m+2}$ in that equation is taking into account not only the contribution from monomer additions to the clusters in the next smaller size bin (*m*+1). Since all particles equal or larger than *m*+2 are considered also all cluster collisions contributing to the *GR* on the LHS of the equation are considered. Therefore, a method is presented, which takes into account self-coagulation both in terms of the nuclei number concentration as well as the growth rate.

Furthermore, is it possible that J_m in Eq. 4 is independent of the nuclei number concentration in the previous size bin (*m*-1)?

(3) The formation rate J_m is definitely affected by the number concentration J_{m-1} in the next smaller size bin. However, the formation rate can be calculated in two different ways (see also reply to comment g) by reviewer #1):

(I) From the production by looking at all possibilities how two smaller clusters (index *i* and *j*) can form a cluster, which is equal or larger than a selected size (here index *m*+1):

$$J_{\geq m+1} = 0.5 \cdot \sum_{i+j\geq m+1} \delta_{ij} \cdot K_{i,j} \cdot N_i \cdot N_j,$$

where both *i* and *j* have to be smaller than *m*+1.

(II) From the losses by using equation (11, referring to the revised manuscript):

$$J_{\geq m+1} = \frac{dN_{\geq m+1}}{dt} + \sum_{i=m+1}^{n+m} \left(k_{w,i} \cdot N_i\right) + k_{dil} \cdot N_t + \sum_{i=m+1}^{n+m} \left(\sum_{j=i}^{n+m} \delta_{i,j} \cdot K_{i,j} \cdot N_j \cdot N_i \right).$$

We have verified with our numerical model that both ways of calculating *J* yield exactly the same result. Using only experimental data method (I) does not work because it would require knowledge on the concentrations of all clusters smaller than d_{p2}. Therefore, method (II) needs to be used, which obtains the formation rate from the losses of clusters/particles, which all have sizes equal or larger than d_{p2} (index *m*+1). Together with equation (14, referring to the revised manuscript) the method can be used to calculate N_m as a function of the larger clusters/particles. From the method it follows directly that N_{m-1} is not required to obtain N_m. If this would be the case the method would not work since equation (15) would need to be solved for two unknowns.

One more related issue: the approach used in the new method is not entirely original either. Already K2004 (Aerosol Sci. Technol. 38, p. 1001-1008, equations 7 and 8 and the text) introduces the idea of adding extra bins below the size m shown in Fig. 3 by K2012, and then corrects the apparent particle formation rate when calculating toward the initial nucleation diameter. Of course, K2014 goes beyond K2004 in that K2004 does not take into account the effect of self-coagulation.

(4) We have added a reference to K2004 in section 2.3 of the revised manuscript:

"Also Kerminen et al. (2004) introduced extra size bins in order to increase the accuracy of their analytical formula, which connects the nucleation rate and a formation rate at a larger size similar to equation (2)."

Other issues:

Since there are several pieces of work on the issue addressed in this work between K2014 and the original paper by K2002, and since most of the readers probably will not go through all the details in sections 2 and 3, I would recommend adding a brief review of the conducted work made after K2002 into the last paragraph of section 1.

We have added a sentence in the introduction, which briefly describes the effects that have been considered following KK2002:

"Several publications followed Kerminen and Kulmala (2002) to include additional effects, like a better description of the coagulation sink from particle size distribution measurements (Lehtinen et al., 2007), self-coagulation (Anttila et al., 2010), and a size-dependent growth rate (Korhonen et al., 2014)."

In section 4, the authors should more explicitly bring up the limitations of the new method in determining the original nucleation rate. First, none of the methods developed so far, including the one introduced here, cannot reproduce a correct nucleation rate unless the size-dependent nuclei growth rate is known down to the size where the nucleation occurs (d_{nuc}). This is a major problem because this information is almost never available and when it is, even small uncertainties in it cause large errors (see Figure 7 by K2014 as well as in earlier works on this topic). Second, we do not really know which value of d_{nuc} to apply when estimating the nucleation rate. This definitely affects the result as well. Third, does the new method really work when self-coagulation becomes important?

This is a very good suggestion and we have included a short discussion to section 4 addressing these issues (see added text below). (1) We agree that the size-dependency of the growth rate can be a crucial point and that it can introduce large errors especially when the GR is small and the losses are high or when there is a pronounced size-dependency. However, this is a problem all other methods have to deal with as well. Nevertheless, we will highlight in section 4 that this can be a crucial point. (2) It is correct that the critical size (d_{nuc}) is generally not known and that it is therefore not easy to choose the right d_{p1} in any method reporting nucleation rates. For this reason the CLOUD experiment has chosen a reference size of 1.7 nm in mobility diameter, which should be at or close to the critical size for most chemical systems. Regarding our manuscript we will briefly mention this challenge in section 4. (3) We believe that self-coagulation is taken into account adequately in our method both in terms of the nuclei number concentration and growth rate (see previous reply (2)).

"One general issue with all methods, which are extrapolating formation rates towards smaller sizes, arises from the uncertainty in the growth rate. In most cases no measurement of the *GR* will be available down to the very small size since also the particle number concentrations are not available (otherwise no extrapolation of the formation rate would be necessary). A small size-dependency of the *GR* that is not taken into account can therefore lead to a big uncertainty. In addition, the critical size of the nucleating particles is generally not known. Ideally, one would choose d_{p1} to correspond with the critical size. However, since this is not possible a reasonable solution to this issue is to choose a size for d_{p1}, which is safely at or above the critical size to avoid extending the size distribution into the subcritical size regime. For this reason the CLOUD experiment has reported particle formation rates at a size of 1.7 nm in mobility diameter rather than nucleation rates (Kirkby et al., 2011)."

Additional changes

In addition to the points discussed above a few more modifications have been made to the manuscript, which are listed in the following:

(i) In the originally submitted manuscript, equation (24) was not showing the correct indices. The index on the LHS of the equation needs to be larger by one compared to the ones on the RHS since the formation rate is described from the flux of particles from the previous size bin. This has been considered in the revised version (equation (14) in section 2.3) and the subsequent equations have also been updated (equation (15)).

(ii) As part of rewriting section 2.1 and 2.2 we have removed the old Fig. 2. We feel that it is not necessary anymore to show this figure since it should be clear from the discussion that the different loss mechanisms lead to different correction factors between $J(d_{p1})$ and $J(d_{p2})$. Instead, we have added the dilution loss rate to the lower panel of Fig. 1 since this loss mechanism is now described in a bit more detail in the revised section 2.2 (see also equation (10)).

(iii) We have extended the range of the linear size bins in the model (section 2.5 and see also Fig. 4) to 100. With the reported density and molecular weight the maximum upper diameter covered by the linear size bins is now ~3.1 nm in geometric size and therefore covers both d_{p1} and d_{p2}. In the originally submitted manuscript the linear size bins ranged only up to ~2.1 nm in geometric diameter, which lead to a transition between geometric and linear bins during the reconstruction going from d_{p2} to d_{p1}. Using linear size bins throughout the relevant size range makes some formulations easier, e.g. equation (19), which includes the growth rate due to monomer and cluster collisions. The results from the reconstruction are not affected in any significant way due to this modification, however, the formulation of the equations become easier. In the same context, the index corresponding to d_{p1} has been changed from “1” to “x”. Due to this change in the notation the index “1” now belongs to the bin containing only one molecule (or better a single building block for all larger clusters and particles). Based on this new definition Fig. 2 (previously Fig. 3) has also been updated.

(iv) As mentioned before the new Fig. 3 illustrates why equation (21, revised manuscript) is a good approximation also for the cases when cluster-cluster collisions are important. We feel that this is an important figure as it visualizes the complex equation (21) in an easy way.

(v) Section 3.3 (and Fig. 7) have been extended in a way that not just kinetic nucleation is used to demonstrate the capabilities of the new method. We have also included three more scenarios where we have allowed for non-zero dimer and trimer evaporation rates. In order to achieve this, the model described in section 2.5 was adjusted to include the relevant cluster evaporation rates. The results now shown in addition to the kinetic simulation in Fig. 7 demonstrate quite interesting aspects, which are relevant for chamber nucleation studies such as CLOUD where different chemical systems are tested, which can behave significantly different than kinetic nucleation.

(vi) Based on the comment by Hanna Vehkamäki we have not termed our new method “universal” anymore. However, based on the new results described above (point (v)) we have evidence that the method can be used for many different conditions. The effects of particle evaporation in the size range between d_{p1} and d_{p2} remains to be studied. Nevertheless, we want to mention that the effect of evaporation on the relatively large clusters/particles at 1.7 nm is probably small for most chemical systems relevant to the atmosphere. In addition, the study by Nieminen et al. (2010) has also neglected the effect of evaporation on the growth rates and the theory is widely used in describing aerosol growth.

(vii) Based on the implemented changes a couple of additional references have been added, which are listed in the following

New references

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