

Reply to Referee #1

We thank Anonymous Referee #1 for their helpful comments. We have answered to the comments below. The bold text is quoted from the referee's comments, and the text in italics has been added to the manuscript.

GENERAL COMMENTS

The paper explores the differences in estimated growth rates for sub-3nm particles by three different methods. The work seems solid and thorough, and it is of publishable quality. However, I'm unsure if the interest is broad enough for ACP as opposed to e.g. Aerosol Science & Technology. I'd recommend either GMD or AMT, though the paper is a modelling study that is of interest to observationalists, so it sits between those journals. I'll leave it up to the editor to make a judgement call. If the paper is to be published in ACP, I have several minor comments that should be addressed.

We believe that our paper fits into the scope of ACP (as also agreed by Referee #2) and is relevant for the readership of the journal, as the methods discussed in the paper are commonly used for analyzing experimental data on new particle formation both in the atmosphere and in chamber experiments. Experimentally determined growth rates are used to characterize particle formation and growth events, and, thus, they are routinely utilized in the analysis of particle distribution data (e.g. Kulmala et al., 2012). Moreover, growth rates extracted from measurements are often compared to those determined by theoretical or modeling approaches. Therefore, assessing the differences between different growth rate definitions is useful also for modelers, and, in general, for all researchers working with atmospheric particle data.

SPECIFIC COMMENTS

P2 L11: "CGR" doesn't correspond to the previous text. Can you define it? Is it "condensational growth rate"?

Yes, in this paper CGR is used as the abbreviation for the growth rate determined based on irreversible vapor condensation, and the letters stand for condensational growth rate. This has now been added to the text.

P3 L6-7: "AGR" and "FGR", same as above.

AGR is used as the abbreviation for the growth rate obtained with the appearance time method, and the letters stand for appearance time growth rate. FGR is used as the abbreviation for the growth rate corresponding to the molecular fluxes, i.e. flux-equivalent growth rate. These are now clarified in the text.

P4 L7-8 and throughout: I'm somewhat confused by the different sinks in this paper. Is the loss due to coagulation with larger particles included here? Is the loss due to coagulation between the model-resolved clusters included elsewhere when discussing the "loss coefficient"?

We agree that the processes included in the model are not explained very clearly. The model includes all the possible collisions between the clusters (up to the clusters consisting of 70 molecules), and also an additional, time-independent external sink. The external sink can be thought to represent the loss of the clusters due to the coagulation onto pre-existing larger particles in the atmosphere. The losses discussed in the paper always refer to the external sink, characterized by the loss coefficient of the vapor monomer (in 1/s). The loss frequencies of the clusters are somewhat

lower than that of the monomer (see Eq. (8)). The role of coagulation between the modeled clusters in the cluster growth dynamics is characterized by the non-monomer fraction of the flux.

To clarify the processes included in the model, we now added the following sentences in the beginning of the Sect. 2.2 (page 6, line 16):

We simulated the time-evolution of cluster concentrations in a one-component system using the Atmospheric Cluster Dynamics Code (ACDC; McGrath et al., 2012; Olenius et al., 2014). The model included the production of monomers, all the possible collision and evaporation processes between different clusters, and the losses of clusters due to an external sink.

Eqn 1: What about the source by a collision of two clusters that are not in the previous bin, but where the sum of their molecules would put the resultant particle in the current bin?

It is true that Eq. (1) considers only the flux originating from the previous bin $J_{i-1,i}$ although the collisions between clusters belonging to bins $< i-1$ may also contribute to the flux into bin i . This contribution is in practice notable only when the concentrations of large clusters are high, or the bin widths very narrow. In the simulations of this work, all possible collisions between clusters were considered but the studied conditions were selected so that the contribution of collisions from bins $< i-1$ to the flux into bin i was negligible for all size classes i . We added the following remark in Sect. 2.1 (page 4, line 8):

In situations with high concentrations of large clusters, the overall flux into bin i may contain the contributions of fluxes from smaller bins $< i-1$. This makes the analysis of the dynamics more complex and such situations are not addressed in this study.

Sect 2.2: Would it have made sense to do a simulation set where you simulation every cluster size from 1-70, so you don't have any numerical issues? This would be a useful comparison.

Actually, every cluster size from 1 to 70 was simulated. Only when calculating the growth rates and fluxes, the clusters were grouped into the size bins. This was done as the growth rate of a single cluster is ambiguous when clusters grow not only by monomer additions but also by cluster-cluster collisions. To make this clear, we modified a sentence in the Sect. 2.2 (page 7, line 4):

After simulating the time-evolution of the discrete cluster concentrations, the clusters were grouped into size bins containing an equal number of cluster (in most cases ten), for which fluxes and growth rates were determined.

Furthermore, we want to point out that the previous study by Olenius et al. (2014) discusses the case where clusters grow only by monomer attachments, and thus the growth rates can be unambiguously determined with the resolution of one molecule.

P7 L4: Why is the concentration in units $\text{cm}^{-3} \text{s}^{-1}$?

This was a typographical error that has now been corrected. The correct unit is cm^{-3} .

P7 L30: What values of monomer sources were used earlier (I think only steady state concentrations were given)?

The monomer source rates used in the different simulations are now presented in Table 1.

REFERENCES

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M., Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K. E. J., Laaksonen, A., and Kerminen, V.-M.: Measurement of the nucleation of atmospheric aerosol particles, Nat. Protoc., 7, 1651–1667, doi:10.1038/nprot.2012091, 2012.