Interactive comment on “Growth of atmospheric clusters involving cluster-cluster collisions: comparison of different growth rate methods” by J. Kontkanen et al.

Anonymous Referee #2

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In their manuscript, Kontkanen et al. present a set of numerical simulations of a one-component system, with the purpose to study the effect of cluster-cluster collisions on the growth of particles, and also the impact of different definitions and methods of the growth rate. The study is interesting, and gives a good overview of the different ways of looking at fresh particle growth, and the author’s approach brings interesting new viewpoints to recent studies of nanoparticle formation in the atmosphere. The work is quite theoretical, and no comparison to actual experiments is made (as the studied one-component system would be quite difficult to produce). I think that the author’s work is useful and interesting, and also in the scope of ACP.

In my opinion the manuscript could be published in ACP, but I would appreciate it if some more information on the simulations and their analysis could be provided. I have listed my concerns below.

* I did not fully follow how the simulations were performed. Were all inputs kept constant over the whole time? The authors state that there was an initial cluster distribution corresponding to a low monomer concentration. I addition, it is stated that a ‘steady-state monomer concentration’ is reached at some point. This suggest that the aerosol formation and growth proceeds until some kind of a steady state is reached. It it is so, this has at least the following implications for the data analysis:

a) from which part of the process are the various size-dependent growth rates determined? For AGR, the GR is necessarily from the start, but for the other GRs some other times could be used. Was the steady-state situation used? What was the criterion for steady state? Were the different growth rates determined at different time points within one simulation? Were the growth rates time-dependent within one simulation?

b) A ‘steady state’ in which everything stays constant is rarely seen in nature, partly because the growing particles contribute to a growth in the sink and therefore the Q/S fraction also changes. This could, for example, introduce a coupling between the formation rate and the AGR of FGR, because high formation rate causes a fast-increasing sink quickly to the distribution. I understand that the sink in the simulations was not dynamic, and therefore this effect is omitted. Do the authors have an idea how this affects the results?

In any case, it would be useful to have a description of the time-dependent developing distribution, for example using a figure. The same figure could then be used to give a graphical explanation of the derivation of the different growth rates.

* A big question in particle formation research is the size dependence of growth in particle formation. From the results in this paper (Figs 1d, 2d, 3d, and 4d), it seems that for this one-component case, AGR and CGR tend to decrease with size while FGR increases with size. Is this a result that could be considered general? Is it possible that
this changes in a true, multi-component case?
* Finally, I think that the discussion in the conclusions could be improved. In the last paragraph, (p. 14, L 23) the authors state that "conventional methods used to determine particle formation rates from growth rates may give estimates far from their true values". Based on earlier discussion, it seems that the authors bring forward the idea that not a single ‘true’ value exists (for example, the growth rate can mean the AGR, FGR or CGR). In the study of particle formation, finding the values for the formation rate $J$ and the growth rate $GR$ has for some time been a key objective. It would be useful if the authors could discuss here which of the given growth rates is the true value meant.

Specific comments:

P7 L33: Regarding the monomer concentration ranges, I understand the limitation of the lower level. However, the upper level for the monomer concentration is less justified. This is a case of selecting a too long time step; reducing the time step in the differential equation solving would present usable results. In any case, the limits for the usability of the methods should be given.

P19, Table 1: For reproducibility, it is not enough to give the final monomer concentration, but also the monomer source rate is important information. I think that the monomer source rates used to obtain the final concentrations in Table 1 should be given also in Table 1.