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Interactive comment on "Impacts of coagulation on the appearance time method for sub-3 nm particle growth rate evaluation and their corrections" *by* Runlong Cai et al.

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Review of "Impacts of coagulation on the appearance time method for sub-3âĂĽnm particle growth rate evaluation and their corrections" by Cai et al.

The manuscript presents an attempt to correct apparent nanoparticle growth rates, obtained by the so-called appearance time method, to remove errors arising from coagulation. It is true that the appearance time method may cause significant artefacts in the experimentally deduced growth rate and consequently in its interpretation and comparisons with models. The purpose of the work, i.e. to test the derived corrections against a particle dynamics model (rather than to compare the used models to mea-

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Discussion paper

surements), is clarified in the revised manuscript. However, my main concerns are that (a) it seems that the suggested corrections do not generally work, and (b) the modeled test cases are quite limited, and for example assume that sub-3 nm particles do not evaporate which is quite a restrictive assumption. Below are the detailed comments that I would ask the authors to address:

1. Figure 6 does not look very convincing in terms of the performance of the suggested correction approaches – do other test cases exhibit similar behavior in the size-dependent errors? Rather than listing the average discrepancies in the corrected growth rates e.g. in the Abstract and Conclusions, the maximum errors should be stated, since they are up to 150 % (!) for the present test cases. In general, complex particle population dynamics cannot normally be described by simplifications, so it is not so surprising that the corrections do not work very well, or cannot be reliably applied on realistic data. This is one of the main results of the work and should be highlighted – even a negative result is a result. Instead of stating "the feasibility of the corrected method was verified" it can be said that the method was "tested", since based on the presented results it doesn't look like the method was verified. Also the title is a bit misleading since it talks about sub-3 nm particles, while later in the test it's concluded that neither the conventional nor the corrected method work reliably at sub-2 nm. I would therefore reformulate the essential parts of the text to discuss "suggestions" or "attempts" to correct, instead of "corrections".

2. The omission of evaporation in the simulations limits the applicability of the results, since atmospheric small clusters and particles generally evaporate significantly. (A single test with a size-independent evaporation rate doesn't help very much, since for the smallest particles the rate may vary even by orders of magnitude with the size and composition, and the used rate is also quite low.) Therefore, I'd ask to include test cases where the smallest clusters have strongly size-dependent evaporation rates of at least around 10ˆ-2 sˆ-1, or even higher. For example the highest values in Schobesberger et al., Atmos. Chem. Phys. 15, 55-78, 2015 could be used for upper-limit

estimates for the evaporation effects. Specifically, since often the evaporation rates of the smallest clusters exceed the vapor condensation rate, it's essential to include cases where β*N1-E (Sect. 4.1, page 9) is negative. Stating that "the bias caused by this size dependency of evaporation is similar to that of coagulation" is odd, since evaporation and coagulation processes are very different: the former moves particles along the size axis within the studied size range, while the latter removes particles from the size range. Can you show how their effects are similar (for arbitrary evaporation and coagulation rate constants)? In Figure 2, how would a test case with both non-zero sink and non-zero evaporation look like? The effects of evaporation may be different at sinks of different magnitude.

3. Sect. 4.2: it is concluded that CoagSrc does not have a major impact on the apparent growth rate. This statement could be softened, since the test cases are so limited, and also the vapor concentrations here are not extremely high. Maybe CoagSrc may still have a larger role at the higher end of atmospherically relevant vapor concentrations, e.g. at ca. 10ˆ8 cmˆ-3?

4. The particle size is mainly expressed through the diameter. The results should thus be dependent on the size of the vapor molecules: the diameters of very small nanoparticles containing equal numbers of molecules are different for e.g. large organic molecules and sulfuric acid. Also, the behavior of apparent GR with respect to diameter may be different for multi-component particle formation where the sizes of the molecular species differ significantly. Which molecular size was used in the simulations? In Sect. 4.3 it is stated that "one should be cautious about the sub-2 nm size-resolved growth rate" – could the 2-nm-limit be something else for different molecules, for example could the diameter threshold be larger for highly oxidized organic monomers or dimers? I recommend to bring up the fact that the diameters may not be very meaningful for such small clusters.

5. Derivation of the corrections in the Appendix: Can you elaborate how the first very first equality (GRconv = . . .) in Eq. (A9) is deduced; it is not obvious. Also, in the

derivation of Eq. (A6, it is assumed that the concentration at the appearance time is 50 % of the final value. It should be noted that in the "conventional" appearance time method it is actually defined that the appearance time concentration is 50% of the maximum value (Lehtipalo et al., 2014), which is not the same as the final value in a strong clustering event. This can affect even "ideal" cases with a constant vapor concentration. In general, the correction seems to be derived for a situation where the particle distribution relaxes into a steady state (equations in the Appendix with N_i,inf). Thus I don't see a reason to believe that the appearance time method, or any "corrected" version of it, would work for realistic, dynamic atmospheric environments with varying vapor and particle concentrations, so the failure in Fig. 6 is not surprising. Why could e.g. Eq. (A8) be applied to an atmospheric non-steady-state situation?

Minor comments: 1. After Eq. (1): "Note that Eq. 1 is expressed in the discrete form, i.e., it does not assume a continuum particle size": one can note that it does however assume a well-defined GR typical for the continuum space, i.e. no spreading of the size distribution, and no negative average net condensation flux in the Lagrangian presentation (see comment 2). 2. Sect. 4.1: Has the appearance time method actually been derived in the original paper (Lehtipalo et al., 2014), or is this something done only in the present manuscript? Is e.g. the original choice of 50 % concentration increase arbitrary? 3. Table A1 and Figure 6: Would a varying coagulation sink affect the results? The growth of the boundary layer in daytime typically causes a time-dependent sink.

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