

We thank the referee once more for commenting on our paper. The reviewers' 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.

Comments to the Author:

Please, make the corrections demanded by referee #1. Subsequently, the manuscript shall immediately be published.

Non-public comments to the Author:

final report of referee #1:

The authors have done a very good job in addressing almost all of the comments and the revised manuscript is much better than the original one.

I have only a couple of concerns that still need some thought (or more explanation, in case I have misunderstood or not understood what has been written):

1. section 2.4 (the justification of equation 21):

I still have problems accepting that $GR \times n = J$ when self-coagulation is important, and don't really understand all the details of the justification in the section. First of all, where does the 'initial equation' 16 come from? If I understand the equation correctly, it is 'designed' in such a way that each collision, between classes m and i , adds to the diameter a certain amount - and these amounts are then summed? Shouldn't this summation be done (first) for the volumes, and then transformed to diameters? Second, is the Taylor expansion (and the approximation of figure 3) valid with strong self-coagulation, e.g. in the case of sharp nucleation bursts causing a growing narrow mode? Of course I understand that by 'defining' $GR = J/n$ will force the equation to hold, but what does GR mean then? Will it represent anymore the growth rate of an average single particle? And, how is it estimated reliably from experimental data - especially for very small particles for which data can be very noisy?

1) The original equation (16) was derived for monomer collisions by Olenius et al. (2014). They also defined the growth rate (GR) of particles by dd_p/dt because the GR is usually expressed in units of nm/h (or m/s). The fact that the volumes need to be added if a particle of diameter $d_{p,i}$ collides with a particle of diameter $d_{p,m}$ is taken into account by adding the two diameters to the third power and subsequently raising the sum to $1/3$.

With respect to the manuscript we made a minor adjustment. Although Olenius et al. (2014) was mentioned in the beginning of section 2.4 it is mentioned again, to make clear that equation (16) was derived in a similar form for monomer collisions before. In addition, the definition of $GR = dd_p/dt$ was added to the equation.

Following a similar approach as Olenius et al. (2014) but taking into account cluster collisions and neglecting the effect of evaporation the particle growth rate for particles in the size bin m can be defined as

$$GR_m = \frac{dd_{p,m}}{dt} = \sum_{i=1}^m \delta_{i,j} \cdot \left((d_{p,m}^3 + d_{p,i}^3)^{1/3} - d_{p,m} \right) \cdot K_{i,m} \cdot N_i. \quad (16)$$

2) We will begin by answering the question what the GR means in case of strong self-coagulation (i.e. when cluster-cluster collisions contribute substantially to the growth of particles). In this case the definition according to equation (16) still describes the average growth rate of a single particle. Similar to the definition when monomers dominate the particle growth, clusters can contribute to the growth. This, however, means that an individual particle can take a "larger step" towards bigger

sizes compared to the monomer collision-only case where the particle can only move to the next larger size bin. Because the particle can now also skip the next larger size bin (e.g. due to a collision with a dimer), the definition of the formation rate J needs to be adjusted to this situation by taking into account all particles formed at a certain size or larger than this size (“ \geq ”-sign in equation (20)). When using the new method we recommend using the GR evaluated according to equation (24), which includes the measured particle number concentrations in the smallest size bins. It is true that the fluctuations of these concentrations can be significant in some cases. In a future study, it is planned to test our method on real data obtained in the CLOUD experiment. Since the GR is not expected to vary strongly with diameter over a rather narrow range of the smallest measured size bins it is probably the easiest to use the GR from an average over a certain size range in practice.

Regarding the question to what extent the Taylor expansion (equation (18)) is valid for conditions when self-coagulation is strong we want to highlight that Fig. 7 shows results of the method for the most extreme case of significant self-coagulation, i.e. the kinetic limit. The method gives quite accurate results for sulfuric acid concentrations $> 2 \times 10^6 \text{ cm}^{-3}$ when self-coagulation starts to become a dominant particle removal process (comparison between dashed black line and green and red solid lines). The stronger deviation between the accurate and the approximate solution for $J(d_{p,1})$ (comparison between green and red lines) is due to the slow growth rate in conjunction with the size-dependency of the GR . Therefore, the method seems to work well for situations where self-coagulation is strong.

In addition, equation (16) is not used anywhere in the method because calculating the GR according to this equation would require knowledge of the size distribution starting with the monomer; this knowledge is by definition not available. The purpose of section 2.4 is to justify that $J = n \cdot GR$ is valid within some limitations also for the situation where cluster-cluster collisions are important. By using the same relationship for evaluating the GR , the method is – in the words of the reviewer – “forced to work”. Despite this forcing, section 2.4 explains why this relationship is nevertheless a reasonable assumption. Future studies, should focus on examining the exact conditions under which the assumption holds and what the errors are.

2. Page 5, line 144 (and figure 1): The “measured” wall loss rate is amazingly smooth?

The dashed black curve in Fig. 1 (lower panel) does not represent individual measured wall loss rates over the entire range of diameters shown. Instead, the value of the empirical factor C' in equation (8) is derived from the wall loss rates measured for sulfuric acid monomers and particles of different sizes using a Scanning Mobility Particle Sizer (SMPS). The value of C' is determined from a least-square fit from equation (8) and the result is shown in Fig. 1. In order to clarify this point we have modified the sentences around equation (8) accordingly.

At small particle sizes the Cunningham correction factor is approximately proportional to d_p^{-1} , and so the wall loss rate can be approximated by

$$k_w(d_p) = \frac{C'}{d_p}, \quad (8)$$

where C' is an empirical constant determined from a least square fit by taking into account measured wall loss rates of sulfuric acid monomers and particles in different size bins. Fig. 1 shows the wall loss rate for the CLOUD chamber as a function of d_p (dashed curve, lower panel), where the value of C' is approximately 0.001 nm s^{-1} .

3. Page 7, lines 211-212 and equation 14: The reference to Lehtinen et al (2007) is probably not a good choice. This relationship ($J = GR \cdot n$) for condensational growth is much older, and is e.g. in

Friedlander's book 'Smoke, dust and haze' and also Seinfeld&Pandis, when the continuous condensation equation is derived.

We have searched for older publications reporting the relationship $J = GR \cdot n$ and added one to the reference list. The sentence before equation (14) has been reformulated accordingly:

The formation rate and the growth rate (GR) are connected by the following equation (Heisler and Friedlander, 1977; Lehtinen et al., 2007):

$$J_{m+1} = GR_m \cdot \frac{N_m}{d_{p,m+1} - d_{p,m}}. \quad (14)$$

New reference added:

Heisler, S. L., and Friedlander, S. K.: Gas-to-particle conversion in photochemical smog: aerosol growth laws and mechanisms for organics, *Atmos. Env.*, 11, 157–168, 1977.

4. Hanna Vehkamäki's comments on evaporation are interesting. The presence of evaporation also influences the validity of $GR \times n = J$, which is, I believe, strictly true only for a propagating (size distribution) wave. Evaporation deforms the shape of the distribution, by acting as a cause of diffusion in size space. This is another reason why the presence of strong evaporation will likely make the analysis in the manuscript less accurate (?)

It is true that evaporation will influence the accuracy of the derived method to some extent. Based on Hanna Vehkamäki's comment we have therefore already added a short discussion on this issue at the end of section 2.3. To our knowledge our study is among the first ones which has looked at the effect of self-coagulation on nucleation and growth regarding the derivation of the nucleation rate. We have discussed many aspects of this method and Fig. 7 gives an idea on the accuracy of the method for chemical systems, which we believe, are of relevance for the atmosphere. In any case, further studies are needed in order to explore, e.g., the influence of slow evaporation beyond the critical size.