

Responses to Reviewers' Comments on Manuscript acp-2017-199

(A new balance formula to estimate new particle formation rate:

reevaluating the effect of coagulation scavenging)

We thank the reviewer for checking the detailed derivation of the formula. We have addressed the comments in the following paragraphs and made corresponding changes in the revised manuscript.

1.) The authors have done a good job in answering most of my comments and those of the other reviewer. I still, however, think that additional explanations are needed to a couple of my initial concerns.

1. Derivation of A7 and A6 in the Appendix and double summation term in eq. A7:

Isn't the message in A7 self-evident? I.e. the rate of change of particle concentration in range $k \dots u$ is equal to condensational growth into range (I) minus condensation out of range (J_w) plus coagulation into range minus coagulation out of range ?

Response: We agree with the reviewer that Eq. A7 is conceptually self-evident providing that aerosol general dynamic equation (GDE) can be expressed in the continuous form. However, previous studies presented this relationship differently (e.g., Eq. 5-8) such that they underestimate new particle formation rates when analyzing intense NPF events in the polluted atmosphere.

2.) In A6, the rate of change of particle concentration above size k is equal to condensational growth into the range (I) minus collision rate in the range?

Response: Yes. Here k is the critical cluster size (line 303). Equation A6 is mathematically simplified from Eq. A5 given that u is large enough. The rate of change of particle concentration for the whole aerosol population is equals to the formation rate of new particles minus the coagulation loss rate.

3.) In my view, the only slightly difficult part to formulate is the coagulation into range term in A7, and this is not thoroughly explained here (and perhaps, slightly wrong?). Let's say that we have a linear bin structure in volume so that $v_1 = 1, v_2 = 2, \dots, v_k = k, \dots$ and we are looking at the range from 7 to 10, i.e. $dN(7,10)/dt$. According to the indexing in eq. A5, the following index-pairs contribute to the coagulation source-term into the range: 1 and 5, 2 and 4, 3 and 3, 4 and 2, 5 and 1, 1 and 6, 2 and 5, 3 and 4, 4 and 3, 5 and 2, 4 and 3, 1 and 7, 2 and 6, 3 and 5, 4 and 4, 5 and 3, 6 and 2, 7 and 1. The factor (1/2) takes care of the double counting and this is correct. But what about the 5 first pairs in the list? They also produce particles into the range 6 to 7 which is not in the range 7 to 10 ? Please explain! Also, what if you have a different bin structure, say logarithmically spaced? Then, the conditions under the summation term on the third term of the right hand side of equation A7, i.e. $v(i) + v(j+1) = v(g)$ cannot hold? If the equation in its current form is only applicable for a linearly spaced bin structure, it must be

clearly stated.

Response: The coagulation source term in Eq. 7 overestimates the formation rate due to coagulation. Since the continuous form GDE is expressed in the summation form (rather than the integral form), however, *CoagSnk* and *CoagSrc* have to be either overestimated or underestimated. To clarify this, we added **“The third term in the RHS of Eq. A7, i.e., the coagulation source term, is slightly overestimated. Since the coagulation source term is usually much less than the coagulation sink term when d_u is limited (e.g., $d_u < 50$ nm), however, this overestimation can be neglected.”** (lines 327-329). When d_u is infinite, the estimated *CoagSrc* is almost exactly the half of the estimated *CoagSnk* using Eq. A7.

4.) In my original review, I suggested removing most of the derivations in the Appendix. As they are located in the Appendix, they might as well stay there, if the authors wish so.

Response: We prefer to keeping the derivations in the Appendix. The above comments raised by the reviewer perfectly illustrates the necessity to include the derivations and key assumptions made.