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

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

We appreciate the comments and suggestions from the editor. They were addressed below and revisions of the manuscript were made accordingly. Equation numbers have been updated and they are quoted in the following responses correspond to the revised manuscript.

After reading the revised paper and final responses to referee comments, I still have a few minor issues to be considered:

1) Response to the third comment by the referee: I do not think it is enough to say that the third term is overestimated. The authors should, at the very least, explain shortly why this term is overestimated. Also, It would be very good to know what "slightly" mean in this context. Are we taking about differences <1%, a few %, <10 % or what?

Response: Eq. (A5) was written in the discrete form. When applying it in measured particle size distributions, it needs to be converted into the continuous form. Approximations are needed during the conversion. We agree with the referee that the conversion used in the previous version of our manuscript is affected by the bin structure and the estimated coagulation source has some deviation. In the last response, we argued that its contribution to the estimated $J_{1.5}$ is negligible since the coagulation source only accounts for a minor proportion to the estimated formation rate. For instance, the resultant uncertainty in the estimated daily maximum $J_{1.5}$ was less than 3.7%.

To address this question, we did further analysis and made additional revisions to the manuscript. A new conversion from the discrete form to the continuous form was derived and reported in the revised manuscript. This new conversion is not affected by the bin structure. Though this new conversion does not change the main findings in this manuscript due to the reason mentioned above, it serves as a method to properly estimate the coagulation effect from measured particle size distributions.

The revisions are given below (lines 324-332 in the revised manuscript):

"For the third term in the RHS of Eq. (A5), i.e., the coagulation source term, its summation sequence can be rearranged as:

$$\begin{aligned}
&\frac{1}{2}\sum_{g=k}^{u-1}\sum_{\substack{i+j=g\\ i,j\geq 2}}\beta_{(i,j)}N_iN_j \\
&=\frac{1}{2}\beta_{(2,\mathrm{k}-2)}N_2N_{k-2}+\ldots+\frac{1}{2}\beta_{(\mathrm{k}-2,2)}N_{k-2}N_2 \\
&\quad+\ldots \\
&\quad+\frac{1}{2}\beta_{(2,u-3)}N_2N_{u-3}+\ldots+\frac{1}{2}\beta_{(\mathrm{k}-2,\mathrm{u}-\mathrm{k}-1)}N_{k-2}N_{u-k-1}+\ldots+\frac{1}{2}\beta_{(\mathrm{u}-3,2)}N_{u-3}N_2 \\
&=\frac{1}{2}\sum_{g=2}^{u-3}\sum_{i=\max(2,k-g)}^{i+g\leq u-1}\beta_{(i,g)}N_iN_g
\end{aligned} \qquad \textbf{(A7)}$$

The formulae in both the far LHS and the far RHS of Eq. (A7) are equally accurate to estimate the coagulation source term. However, simply substituting the continuous particle diameter (e.g., d_g) for the discrete size (e.g., g) in the far LHS of Eq. (A7) will result in uncertainties when the size bins do not increase linearly in the particle volume space. As indicated in Fig. A1, substituting the continuous particle diameter for the discrete size in the far RHS of Eq. (A7) is independent of the bin structure for d_g and d_i.

Thus, Eq. (A5) can be rewritten as,

$$I = \frac{\mathrm{d}N_{[d_k,d_u)}}{\mathrm{d}t} + \sum_{d_g=d_k}^{d_{u-1}} \sum_{d_i=d_{\min}}^{+\infty} \beta_{(i,g)} N_{[d_i,d_{i+1})} N_{\left[d_g,d_{g+1}\right)} - \frac{1}{2} \sum_{d_g=d_{\min}}^{d_{u-1}} \sum_{d_i^3=\max(d_{\min}^3, d_k^3-d_{\min}^3)}^{d_{i+1}^3+d_{g+1}^3 \le d_u^3} \beta_{(i,g)} N_{[d_i,d_{i+1})} N_{\left[d_g,d_{g+1}\right)} + J_u$$

(A8).

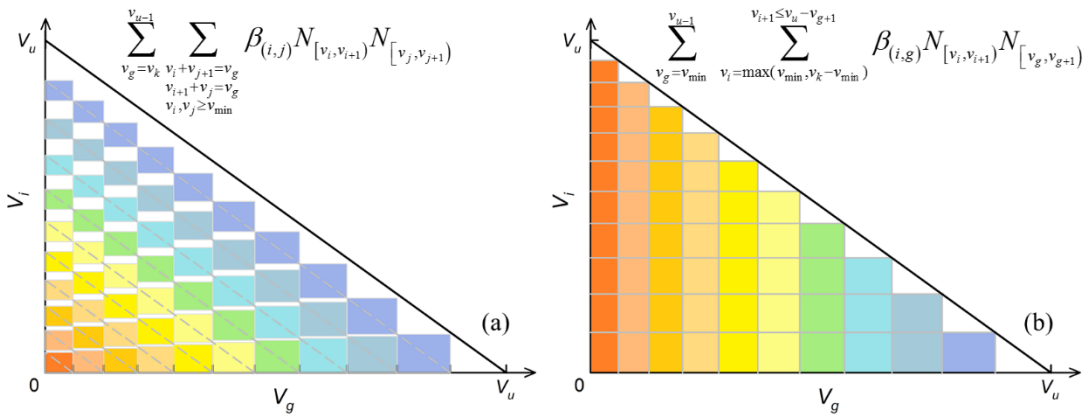

Figure A1: Schematic for two different summation sequences to estimate the coagulation source term. Equations in panels (a) and (b) correspond to the continuous forms of the far LHS and the far RHS formulae in Eq. (A7), respectively. The coagulation source term is denoted by half the area of the triangle (since the particles at the same diameter are accounted for twice). The colored areas are the estimated area using the two equations, respectively.

2) Lines 158-159: I am not sure you can call accommodation coefficient and coagulation efficiency the same because the former refers to condensation and the latter to coagulation.

Response: Thanks for the suggestion. We revised it as "**Mass accommodation coefficient**" in line 158.

3) Line 245: Can you measure the nucleation intensity simply by looking at particle number concentration? What is primary particle number emissions are very large, as they can be in polluted cities? Would change rates in number concentrations be a better measure? Please open this a bit.

Response: During typical NPF events, the sharp increase in total particle number concentration is mainly due to nucleation and the contribution of primary emissions is negligible. In cases that primary emissions

are significant, a high abundance of particles in various sizes is often identifiable in the 3-D contour plots of particle size distribution data. We agree that the change rate in particle number concentration, d*N*/d*t*, is good measure of nucleation intensity. The quantity of particles produced by nucleation can also serves as an indicator. The aim of this paragraph is to explain the reason for the dominant role of *CoagSnk* in urban Beijing. The expression of *CoagSnk* is determined by particle number concentration rather than its change rates. To better illustrate this, we revised it as "**As shown in Equation (1), CoagSnk is approximately proportional to the square of particle number concentration. Nucleation intensity in urban Beijing, characterized by number concentration of particles larger than 3 nm during typical NPF event periods, is found to be higher than those in Hyytiälä and Atlanta (as shown in Fig. 6(b)).**" (lines 246-268)

4) Grammatical comments:

The article (the) is missing from several places where it should be (lines 9, 10, 14, 16, 32, 41, 44, 70, 124, 157, 167, 175, 223, 236, 238, 254, 255, 270, 273, 274

criterions -- > criteria (lines 9, 92, 193, 263)

Line 14: … and formulae used widely in the literature.

line 37: has -- > had

line 40: …before they grow into larger sizes.

line 53: concentrations

line 66: …narrower size ranges, such as …

line 147: … there are no tall buildings nearby.

line 177: This indicates that the influence…

line 180: beyond what?

line 184: The reason for

line 192: what is meant by "proper large"? properly large?

line 209: bad wording (is because that the)

Response: We appreciated detailed editing from the editor that helps to improve this manuscript. We have made the suggested changes. In addition, we read the manuscript again to correct grammar errors.