Responses to Reviewers' Comments on Manuscript ID ACP-2022-262

(The effectiveness of coagulation sink of 3–10 nm atmospheric particles)

We thank the reviewer again for the efforts and comments that help to improve this manuscript. The reviewers' comments are addressed in the following paragraphs and the manuscript has been revised accordingly. We rewrote the method section on the theoretical coagulation coefficient and provided the details in the supporting information. We also added a table to the supplementary information to provide more details on the experiments.

The comments are shown as sans-serif blue texts and our responses are shown as serif black texts. Changes are highlighted in the revised manuscript and shown as "quoted underlined texts" in the responses. References are given at the end of the responses.

I believe the authors still need to consider and revise the manuscript to address prior comments. The revised submission contains minimal modification to the manuscript and as it stands the authors do not fully clarify the calculation methods they apply, and they do not fully analyze the results from their experiments.

Equation 5 is still misleading without more explanation. The enhancement factor, if expressed in this way, would not only be a function of just the Hamaker constant and particle diameters, but also the gas pressure, gas temperature, and composition $E(A, d1, d2, kT, P)$. It is actually combination of the free molecular enhancement factor and continuum enhancement factor, and depends on the Knudsen number (really it is $(A/kT,))$. I recommend in describing calculations, the authors start with theoretically more appropriate expressions (those of Sceats, as shown in Stolzenburg et al, 2020, the equations in Alam 1987, or in Ouyang et al 2012) and then show where $E(A/kT, Kn))$ that they apply comes from. With regards to enhancement factor calculation, the authors only state: "The value of E(A) was obtained using a model accounting for the van der Waals attractive force and the methods to compute size-dependent E(A) can be found in previous studies (Alam, 1987; Sceats, 1989; Chan and Mozurkewich, 2001)." What was the exact equation applied? These references take slightly different approaches from one another. Assuming they are using equation (24) from Alam, 1987 for $E(A/kT, Kn))$, then the authors could clearly show their $E(A/kT, Kn)$ function, as Alam 1987 uses separate enhancement factors for the continuum and free molecular regimes. If they were instead using Chan & Mozurkewich equation (27), which does not show a transition regime enhancement factor explicitly but instead a transition regime coagulation rate, then they would still derive a $E(A/kT, Kn)$ expression with separate continuum and free molecular enhancement factors within it. The authors also need to clarify their method of calculations for the free molecular enhancement factor, was Marlow's equation used, as in Alam 1987 or Sceats's equation as in Chan and Mozurkewich?

A final note on this topic- the authors state "We took the multiplicative factor from this model instead of the van der Waals coagulation coefficient to avoid the minimal differences among the coagulation coefficients given by different models (Lehtinen and Kulmala, 2003; Alam, 1987; Ouyang et al., 2012) at A = 0 J." This is tangential to the issue with equation (5). The issue with current presentation of equation (5) is that it hides that there are different enhancement factors for different regimes, and for van der Waals potentials, the enhancement factor in the free molecular regime is typically considerably larger than in the continuum regime. Particular care has to be taken to explain what the Knudsen numbers are for the processes examined, and how enhancement factors are calculated.

Response: We rewrote Section 4.2 on the theoretical coagulation coefficient to clarify how the theoretical enhancement factor was computed in this study and added computation details in the supplementary information. $E(A, d_1, d_2)$ has been revised as $E(A/kT, \mathrm{Kn})$.

In the revised manuscript, we give the coagulation coefficients for the continuum regime and the free molecular regime,

$$\beta_{\mathrm{C}} = 2\pi(d_1 + d_1)(D_1 + D_2) \cdot E\left(\frac{A}{kT}, 0\right) \tag{8}$$

$$\beta_{\mathrm{FM}} = \frac{\pi}{4}\alpha(d_1 + d_1)^2\sqrt{c_1^2 + c_2^2} \cdot E\left(\frac{A}{kT}, \infty\right) \tag{9}$$

We clarified that Eq. 5 can be reduced to Eqs. 8 and 9 as Kn approaches the continuum limit and the free molecular limit, respectively.

We first compute $E(A/kT, 0)$ and $E(A/kT, \infty)$ using the formulae reported in Chan and Mozurkewich (2001), which were fitted to numerical solutions to the formulae in Sceats (1989). We then use the interpolation formula in Alam (1987) to obtain $E(A/kT, \mathrm{Kn})$, as Kn for particles in the chamber experiments were in the transition regime (1.0-1.9). We also compared the interpolation formula in Alam (1987) to those in Sceats (1989) and Ouyang et al. (2012), finding that the difference (< 4 %) among these interpolation formulae was smaller than the experimental uncertainties.

The above procedure has been clarified in the manuscript as:

"For the coagulation between 3-10 nm and 100 nm particles, Kn ranges from 1.0-1.9. Consequently, coagulation occurred in the transition regime. To obtain the theoretical $E(A/kT, \mathrm{Kn})$, we first computed $E(A/kT, 0)$ and $E(A/kT, \infty)$ using the formulae reported in Chan and Mozurkewich (2001), which were fitted to the numerical solution to the integral from Sceats (1989). The results were then extended to the transition regime using the interpolation formula in Alam (1987). We also compared $E(A/kT, \mathrm{Kn})$ to the results interpolated using the methods in Sceats (1989) and Ouyang et al. (2012), finding good consistencies among those methods. More details on the theoretical β and $E(A/kT, \mathrm{Kn})$ can be found in the Supporting Information (SI)."

In the revised SI, we added Eqs. S1-S17 to clarify the exact equations for $E(A/kT, 0)$, $E(A/kT, \infty)$, and the interpolation formulae for $E(A/kT, \mathrm{Kn})$. A new figure was also added to show the consistency among these formulae and indicate the range of Kn for test particles in the chamber experiments.

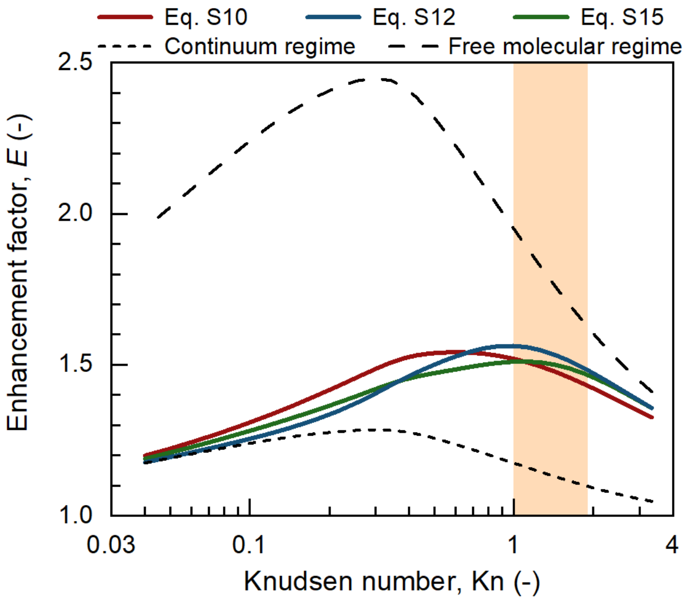

Figure S1. The van der Waals enhancement factor for the collision between a 1-1000 nm particle and a 100 nm particle. The shaded area indicates the Knudsen number of particles used in the chamber experiments.

2. The authors have dismissed comments (3) and (6) from prior review without completely modifying the manuscript. As written, the manuscript (and response to reviewer comments) come off as quite hasty. I recommend that the authors include, perhaps as a supplemental table, the number concentrations of 100 nm particles the initial number concentrations of 3-10 nm particles (ideally broken up into sub ranges, perhaps 1 nm increments) for each experiment performed. The Kn range or average Kn for each bin should also be shown, as shown a best fit (A/kT,) for that size range. There appear to be a number of potential valuable data in Figure 3, but they are not provided in a way that can be easily used by others.

Response: We followed the reviewer's suggestions and added a table to the SI, which summarizes the Kn and best fit coagulation coefficient β_{meas} for each particle size. The measured (A/kT,), which is determined as the ratio of β_{meas} to the theoretical hard-sphere collision coefficient, is also given, though their values are not used in the main text.

This table also includes the concentration of sub-10 nm particles in the chamber during each experiment in Figs. 3 and 5. The concentration of 100 nm particles can be found in the horizontal axis or the figure legend. The inlet concentrations are not given because they are not used to determine β_{meas} and the inlet concentration of 100 nm particles varied with time, yet we have clarified in the method section that "The concentrations of 100 nm and sub-10 nm particles were controlled to be 10^4-10^5 cm^{-3}". The data in Fig. 3 and other figures have been uploaded to an open-access server so that they could be used by other colleagues.

Unfortunately, we might not fully understand the comment "……the initial number concentrations of 3-10 nm particles (ideally broken up into sub ranges, perhaps 1 nm increments) for each experiment performed". The 3-10 nm particles were classified by a DMA and "the aerosol and sheath flow rates of the DMA were 3.5 and 10 L min^{-1}, respectively". For each experiment, only particles with sizes closes to the DMA centroid diameter were selected. As can be seen from Table

S1, the total number concentration of the sub-10 nm particles was relatively low. To minimize random uncertainties, we used the total concetration to compute the coagulation coefficient instead of the size-segrated concetration.

In the revised manuscript, we added:

"They were charged and then monodisperse particles with a certain size were classified using a Vienna-type differential mobility analyzer (DMA)."

"For each experiment, we evaluated the CoagS of particles with a certain size. The size-dependency of CoagS was obtained by repeating the experiment for different particle sizes."