Cai et al focuses on evaluation of the coagulation rate coefficient between sub-10 nm particles and 100 nm particles in a chamber environment. In reading the manuscript, I have concerns on the manner the experiments were conducted (a large fraction of 100 nm particles remain charged after neutralization, and this is not considered in results) and and analysed (equation 5 is not a valid approach in the transition regime). I have the following comments for the authors to address:

1. Figure 1. It appears that after DMA selection, the authors sent particles through a bi-polar charger (labelled neutraliser, but the authors should note the type of neutraliser in terms of source type, and strength). For the 100 nm particles, roughly 47% of them are charged at steady-state (near 20% will be +1 net charge, near 20% will be -1 net charge, and 7% multiply charged). Therefore, it seems the experiment is not between neutral particles exclusively, but between a population of charged 100 nm particles and sub 10 nm particles, as well as uncharged 100 nm particles and sub 10 nm particles. It is not clear if the charged particle-uncharged particle potential is negligible in this instance, and in comparison to the often weaker effect of van der Waals potentials, it is likely not negligible. Without directly addressing the charge influence (or clarifying that charged particles were removed), it is not clear to me that the measurements and analysis reported are sufficiently done to merit publication, as they would not necessarily be representative of the atmosphere.
2. Section 2.1. The mixing within the chamber needs to be described in much greater detail. One concern in the experiments is that the inlet sub-10 nm particle concentration is orders of magnitude higher than the outlet concentration. The inlet flow is not diluted infinitely quickly upon entering the chamber, and there is certainly a region near the inlet where the sub 10 nm concentration is similar to the 100 nm particle concentration (their injected concentrations are similar)
3. Methods (General). The authors need to more explicitly state the number concentration ranges of the 100 nm particles used and the sub-10 nm particles.
4. Equation (1). The authors neglect the influence of sub-10 nm particle self-coagulation on the differential equation governing the sub-10 nm particle concentration. From the limited data in the main text, it appears that sub-10 nm particle concentrations in the chamber were much lower than the 100 nm particle concentrations, but this needs to be discussed more directly, and brief calculations to show it is negligible.
5. Alongside neglecting the potential influence of 100 nm particle charge on the coagulation rate, equation (5) in the manuscript is not an appropriate way to handle coagulation in the transition regime accounting for potential interactions. In the continuum limit (very small Kn), it is acceptable to write that the coagulation coefficient using the formula:

$$\beta_{Cont} = 2\pi(d_1 + d_2)(D_1 + D_2)E_{Cont} \quad \text{(R1)}$$

Similarly, in the free molecular limit (very large Kn), one can write:

$$\beta_{FM} = \left(\frac{\pi kT}{2m_{12}}\right)^{1/2}(d_1 + d_2)^2 E_{FM} \quad \text{(R2)}$$

where *k* is Boltzmann's constant and m_{12} is the reduced mass of the two particles. However, E_{Cont}, which can be calculated for spheres exactly using the approach of Fuchs for continuum collisions, and E_{FM}, which is calculated using an approach by Fuchs & Sutugin, Ouyang, or Sceats, are not one and the same, and for the van der Waals potential, usually $E_{FM} > E_{Cont}$. Because in the transition regime, the transport features controlling collision are evolving as particle diameter increases (Kn decreases), it is not correct to express equation (5) as a single enhancement factor applicable to all particles (the enhancement evolves as well). The authors should instead adopt an approach to fit E_{Cont} AND E_{FM} to results. The Ouyang (2012) reference of the manuscript attempts to do this, as does Fuch's limiting sphere approach when potentials are included and Sceats 1989 (I believe the Sceats approach is the one used in

Stolzenburg 2020; see equation 3 in their paper). When the authors state they used the methods from previous studies it is not clear if they mean E_{Cont} or E_{FM}, and only considering one is not correct in the transition regime. As inference of this parameter is the central purpose of this manuscript, this issue combined with neglecting charge effects (again, unless charged particles were removed) is concerning.

6. Equation (5), continued. The authors need to explicitly state how they are defining Kn (define lambda, the mean free path in term of other, better defined parameters, as this is not the gas mean free path). They should then discuss the Kn range examined in this study, to state whether their results apply to coagulation in the continuum limit, the free molecular limit, or a transition regime range. There is a chance Kn (which tends to be lower for particle-particle collisions) is sufficiently low that they are close to the continuum limit, such that they can simply use a continuum limit coagulation expression and E_{Cont}. However, as per comment 5 if the coagulation range studied is in the transition regime, then I am afraid the authors need to rethink their analysis.