The provided manuscript discusses a VBS-based model for pure organic neutral and ion-induced nucleation, including its formulation, assumptions, and comparison with CLOUD measurements. The authors claim that their model reproduces experimental nucleation rates across various conditions. The topic of the manuscript is important and well within the scopes of ACP. However, several critical issues need to be addressed before publication consideration in ACP.

Major Concerns:

1. The assumption that the critical cluster for both neutral and ion nucleation is always a dimer (see Equations BR5 and BR14) is likely invalid. This assumption raises fundamental questions about the model's physical accuracy and consistency. According to nucleation thermodynamics and kinetics, the critical cluster is defined as a cluster with equal forward and backward evaporation rates. As shown in Figure 9 and other figures, the evaporation rates of neutral dimers are 3-4 orders of magnitude higher than the forward rate, indicating that dimers are not yet critical clusters. Additionally, the size of the critical cluster should depend on monomer concentrations; a constant size regardless of concentration (as in Figure 9) is unphysical. Furthermore, charged clusters tend to be more stable, implying that the critical cluster size for ion-induced nucleation should be smaller than that for neutral nucleation. The authors should at minimum explicitly acknowledge the unphysical nature of this assumption and discuss its impact on the predicted nucleation rates.

2. Regarding Equations 16 and B4, clarification is needed on how the value of the kinetic (collision) rate coefficient, k^{kin}, was obtained as 10^{-10} cm^3 s^{-1}. According to Figure 13.5 in Seinfeld and Pandis (2016), the typical collision rate coefficient for organic molecules (~700 amu) should be approximately 7×10^{-10} cm^3 s^{-1}, about seven times higher than the value used. Since the neutral nucleation rate is proportional to k^{kin}, adopting the higher value would likely lead to deviations from CLOUD measurements. This discrepancy suggests that other parameters might need adjustment to reconcile the model with experimental data.

3. In Figure 2, the criteria for determining neutral and charged threshold values are unclear. Clarification on the methodology used for these determinations is necessary for proper interpretation.

4. In lines 300-301 and Figure 7, the manuscript omits comparison with CLOUD measurements at higher ionization rates, such as those achieved with π beams (see Fig. 4 in Kirkby et al., 2016). Including such comparisons would strengthen the validation of the model against experimental data.

5. At line 669 and Equation B9, the authors neglect 'other losses' or 'wall losses' in the final nucleation rate equation. It is important to justify this omission and clarify whether these losses are considered in the results presented in Figure 9, as they can significantly influence nucleation rates.

6. The rates used in lines 739-740 pertain to small molecules or charged clusters (~100 amu). For larger organic molecules (~700 amu), these rates are expected to differ substantially. The authors

should justify their choice of these rates and analyze the sensitivity of the predicted ion-induced nucleation (IIN) rates to these assumptions.

7. In line 741, the method for determining the wall loss rate should be explained. The sensitivity of model predictions to this parameter must be assessed. As I understand, wall loss for neutral and charged clusters are quite different. The authors should also discuss how different wall loss rates for neutral and charged clusters might affect the model performance.

Minor Concerns:

1. Lines 19-21 should include references to other groups' studies on organic nucleation, such as Zhang et al. (2004) and Yu et al. (2017), to provide a more comprehensive and balanced perspective.

2. Lines 27-29 contain a statement that may not fully reflect the current understanding. Several studies (e.g., Lee et al., 2003; Yu et al., 2010) have demonstrated the importance of ion-induced or ion-mediated nucleation in the global atmosphere. The introduction should acknowledge these differing viewpoints for a balanced discussion.

3. In Figure 9, including error or uncertainty bars for the measured data would improve the clarity and robustness of the comparison.

4. Lines 304-305. Similar general shape has been observed and predicted for H_2SO_4-NH_3-H_2O ternary nucleation (Dunne et al., 2016, Yu et al., 2018). The authors may want to point this out as well.

Overall, addressing these issues will enhance the physical consistency, clarity, and robustness of the manuscript, making it more suitable for publication in ACP.

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