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Interactive comment on “Investigating the discrepancy between wet-suspension and dry-dispersion derived ice nucleation efficiency of mineral particles” by C. Emersic et al.

Anonymous Referee #2

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The authors present a number of interesting ideas and findings, including the potential role of aggregation and sedimentation of mineral particles to alter their ice-nucleating (IN) efficiency in suspension and the revised importance of atmospheric IN mineral particles especially at high temperatures, by combining experimental and modeling approaches. The authors are very knowledgeable on the subject matter, and the topic will be an important addition to ACP. However, some revisions seem necessary before this paper can be considered for publication. My major concerns are listed below, followed by some minor comments.

Major concerns: Potential effect of aggregation on IN: I agree with the potential of

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coagulation and settling (or flocculation) of mineral particles in “bulk suspension”, possibly altering the slope of the $n_s(T)$ spectrum at high temperatures. But, I do not think such processes in “droplets on a cold stage” will have substantial effect to inactivate any potential INPs since agglomerates/flocculation are made up of very loosely packed particles bound weakly together such that particle surface may be completely accessible to water. To support this, a number of previous cold stage studies suggest the negligible effect of aggregation on IN efficiency of mineral dusts. For example, the NC state cold stage experiments with 0.0001 and 1 wt% illite NX (with droplet populations of two different size ranges) collapse into a single $n_s(T)$ spectrum as reported in Hiranuma et al. This implies that aggregation plays a minor role. More recent study by Whale et al. (AMTD) reports similar results for microliter-sized droplets. In addition, several previous studies performed simulations to test for the role of statistical distribution of particles in drops and the role of random component due to drop placement, which is in principle stronger than that of aggregation within the drop. Briefly, Wright and Petters (JGR) and Hader et al. (ACP) demonstrated that the effect of random component could be small relative to the statistical variation due to nucleation rates. More specifically, Fig. 2 in Wright and Petters shows that the random placement in the drop is much less of an effect than time dependence, which seems minor as well. Concerning these points, the authors need to offer stronger counterarguments against these previous observations or direct evidence of the effect of aggregation in reducing IN efficiency to write some bold statements that appear in the current manuscript, i.e., P888 L10-12 “, revealed the...”, P888 L18-21 “revealed that...”, P899 L22-26, P902 L1-8, P905 L8-10. Otherwise, I would recommend carefully rephrasing and softening the tone.

Overall structure: A lot of things described in Sect. 4 (P 894-902) appear to be more relevant to the methodology. Consider reorganizing the sections with a narrative description of experiments & modeling methods independently (e.g. in Sect. 2) with subsections, e.g., “2.1. MICC experiment”, “2.2. n_s parameterization”, “2.3. wet-suspension vs. dry-dispersion”, “2.4. colloid experiment”, “2.5. ACPIM modeling”, “2.6.

coagulation modeling” etc. A similar structure for results followed by the discussion section may improve the clarity of the paper more.

Minor comments: P 888 L6 & L 13: I suggest maintaining consistency of terms with a previous publication. The correct notation should be either ice-active surface site density (IASSD; Connolly et al., 2009) or ice nucleation active surface-site density (INAS density; Niemand et al., 2012). The same applies elsewhere, e.g., P889 L 13 & L 16, P 894 L12.

P889 L5-6: Hiranuma et al. discusses the potential effect of agglomerates and multiple nucleation modes besides chemical aging effect.

P889 L11: Consider giving the description of Kaolinite (e.g., KGa-1b from Clay Mineral Society) here instead of Sect. 5.

P894 L3-4: So the surface area is scaled to the droplet number to calculate n_s ? If so, it is worth mentioning for clarity.

P896 L8: Awkward sentence. I suggest rephrasing.

P896 L15-16: The curves were manually fitted to the data.

P896 L19: Two lognormal modes according to Table 1?

P897 L1: 1-5% in mass? surface area?

P897 L26: This sentence seems incomplete - do not to well what?

P898 L2: wet-suspension according to its first appearance.

P899 L7: between to?

P900 L3 : I suggest using wt% to be consistent with what appears in figures. The same goes to other parts, e.g., P900 L 23.

P900 L4: $M\Omega$ cm

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P900 L6: 1 or 5 μm

P900 L19: H₂O

P901 L7-10: The authors give a justification to neglect the force of repulsion later in the Sect. 5.1.1. The variation in zeta-potential seems not responsible for the observed enhancement in aggregation either; i.e., why the point of zero charge, $\sim\text{pH}7$ (P904 L3), does not correspond to the smallest DLS size measurement? More clarification seems necessary here.

P901 L14: Hiranuma et al. offers several other potential reasons as mentioned above. In fact, the potential impact of agglomeration is discussed in their paper as a minor factor. So the word “alternate explanation” in P 901 L 21 seems misleading. Consider rephrasing.

P 901 L17-18: Please clarify the connection to the previous sentence. The “same result” seems to mean the discrepancy between wet-suspension method and dry-dispersion method.

P901 L18-21: In Fig. 9, K-feldspar does not exhibit a divergence between wet-suspension and dry-dispersion at high T as compared to other two minerals (i.e., the ratio is larger at high T for NX illite and Kaolinite). Any physical reasons? It may be worth discussing here.

P904 L16: MICC?

P904 L23-25: Consider stating what the authors observe is consistent with what is described in Hiranuma et al. for illite NX at least.

Table 1: Geometric surface area concentration? ($\text{m}^2 \text{m}^{-3}$)?

Figure 8: Can the authors present their experimental uncertainties in T and n_s in this figure? Or include them in the figure caption or text?

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