

We thank both reviewers and Gabor Vali for their valuable comments on our manuscript. Since all three referees raised some similar and recurring criticisms, we would first like to address four general points, and thereby hopefully facilitate the readability of our responses to their individual points.

(1) Why we do not assume the presence of 'active sites' in our analysis. Our paper presents a modeling approach to explain experimental immersion freezing results from various laboratory studies. We emphasize that our aim is to describe these experimental data using only physical observables, i.e. parameters which are accessible from the experiments. We purposely have not invoked concepts or introduced parameters which are not theoretically supported or not available from the experimental results. The basis of most of the reviewers' comments is the assumption that particle surfaces possess 'active sites' or locations that exhibit substantially differing efficiencies of nucleating ice. However, as noted by some of the reviewers, there is currently no fundamental theory and/or physical observation to support an active site concept and any evidence claimed is thus far based only on the mathematical frameworks and fitting procedures that describe the experimental immersion freezing data. In other words, given the fact that in situ detection of ice active sites has yet not been achieved, it is lacking for the immersion freezing data sets being analyzed.

This manuscript therefore proposes another way to interpret the experimental observations. Application of classical nucleation theory (an accepted theory in many disciplines) in combination with experimental parameters allows us to test our scientific hypothesis: Can variability of ice nucleating particle surface area immersed in different droplets explain observations of immersion freezing without invoking unobservable parameters? Where so, the theory can be used to make statements and predictions about the nature of the experimental uncertainties and thus guide new experimental approaches. It is important to note that a good fit of any given model to experimental data is not sufficient to prove a concept or develop new axioms to derive a theory. In summary, here we take an intentionally conservative approach to existing theory and fitting parameters.

(2) Why we do not assume that each droplet contains the same ice nucleating particles (INPs) surface area, or ISA, and its implication for freezing analysis.

Droplets containing INPs for immersion freezing experiments are prepared or generated in a variety of ways and results in variable ISA per droplet. i) When droplets are subdivided from a bulk particle-in-water solution, the surface area per volume of solution is typically measured. When generated droplet diameters, D_{drop} , vary (Broadley et al., 2012; Wright and Petters, 2013), droplet volumes vary proportional to D_{drop}^3 . Finally, the ISA per droplet must also vary proportional to D_{drop}^3 as a result of scaling the droplet volume to the measured bulk surface area per volume. For example, if D_{drop} varies by a factor of 2 in an immersion freezing experiment, ISA per droplet will vary by a factor of 8. ii) INP immersed in droplets will have different sizes and surface features, such as cracks, pits, pores and edges. When subdividing droplets from a bulk solution, INPs are also subdivided from an INP population in the bulk

solution. Subdividing results in different particles and dissimilar ISA per droplet (e.g. Wright and Petters, 2013). This will result in additional variability beyond what can be expected from variability in droplet volume. iii) Coagulation or aggregation of particles in solution or in droplets may also lead to additional ISA variability (Hiranuma et al., 2015). iv) Other INP characteristics such as density, void fraction and shape factor may also contribute to variability in ISA (DeCarlo et al., 2004; Slowik et al., 2004; Zelenyuk et al., 2006; Schmid et al., 2007; Park et al., 2008). v) When size selecting particles in a differential mobility analyzer (DMA) for immersion freezing experiments, particles having multiple charges will be present. This will result in an ISA distribution that is biased toward larger particle sizes and thus, surface area. We also note that some effects listed above may also influence ISA variability when employing a DMA.

Equation (1) in our manuscript is the common starting equation applied in most studies to derive the expected change in the number of frozen droplets in an immersion freezing experiment. It does not assume that all droplets possess the same ISA, equal to some average value, A_{avg} , of the INPs. However, when identical ISA per droplet is assumed, the total available ISA is typically expressed as $A_{tot}=A_{avg}N_{ufz}$. Integration with respect to time yields the familiar logarithmic expression, $\ln(N_{ufz}(t)/N_{tot})=-J_{het}A_{avg}t$, and finally results in the well-known equation for the unfrozen fraction, f_{ufz} , in the final form of $f_{ufz} = \exp(-J_{het}A_{avg}t)$, where J_{het} is the heterogeneous ice nucleation rate coefficient and t is the nucleation time. The exponential form of f_{ufz} results directly from the assumption of same ISA in each droplet. This equation is of the same form even when considering multiple components or contact angle distributions for the same ISA per droplet. However, if droplets prepared in laboratory experiments exhibit variable ISA, this formula is no longer valid to describe the unfrozen fraction. Consequently, any other mathematical formulations or frameworks which stem from the f_{ufz} equation are also invalid when ISA varies. For these reasons, our approach to model the immersion freezing experiments differs from previous analyses that do assume that each droplet contains the same ISA.

(3) Why we do not assume uniform ice nucleating efficiency of ISA. We constructed our model to address the following question: to what degree can variable ISA account for previously published immersion freezing data? Our model and this question are independent of any assumptions of a uniform or variable ice nucleation efficiency of immersed particle surfaces. This is contrary to the statements of all referees claiming that we neglect the multicomponent nature of mineral dust particles, variety of ice 'active sites', or that we apply a single contact angle. We show that variable ISA can account for measured immersion freezing data. Conceptually, our results imply that a distribution of ice active sites is similar on a droplet to droplet basis. This can be referred to as an “internally mixed” case, in which an average J_{het} value can represent all sites on the ISA; in contrast to an “externally mixed” case, where rare but highly efficient locations on ISA in some droplets dominate immersion freezing (Broadley et al., 2012). For an internally mixed case, if ice active sites may be small in surface area (~ 10 nm², Marcolli et al., 2007) compared to the overall ISA (one active site representing a fraction

of 3×10^{-6} on a $1 \mu\text{m}$ spherical particle) and numerous, then our results again suggest that their distribution on a per droplet basis is similar. In summary, we do not assume a uniform INP surface, but we do show that a single function of J_{het} for a single particle type (e.g. NX illite, CMS kaolinite, K-feldspar, etc...) can substantially describe the experimental data, without invoking the presence of different (rare) and non-detectable ice nucleating sites or components present in some but not all droplets.

(4) Why our approach reduces the uncertainty in predicting immersion freezing. We believe that a shared overall goal in the ice nucleation community is to reduce the uncertainty in predicting ice formation, here immersion freezing. The current state of the art uncertainty in predicting immersion freezing rates ranges over roughly four orders of magnitude, based on experimentally derived ice active sites (Hiranuma et al., 2015). This uncertainty translates into a range of, e.g., 0.1-1000 ice crystals predicted per liter of air, too ambiguous to model atmospheric ice nucleation. Application of a theory and physical observables allows us to quantitatively assess and specify the uncertainty, allowing it to be minimized in future investigations for better predictive capability. This not only includes the contribution of uncertainties from temperature, ISA measurements, ISA variability and relative humidity to the overall predictive capability, but also stochastic effects (the random freezing process) and time. A stochastic uncertainty becomes important in experimental studies when operating close to detection limits, using too few droplets, or observing too few ice nucleation events. This can be especially important at the warmest freezing temperatures. The presented uncertainty analysis is able to explain the scatter in experimental immersion freezing data for two independently compiled and very large freezing data sets (Knopf and Alpert, 2013, Hiranuma et al., 2015), supporting the notion that a stochastic freezing process may have to be considered to further reduce the current state of immersion freezing uncertainty.

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