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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Refreeze experiments of water droplets containing different types of ice nuclei interpreted by classical nucleation theory" *by* Lukas Kaufmann et al.

Anonymous Referee #2

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Kaufmann et al. present new heterogeneous ice nucleation data for water droplets that contain one of four substances: Hoggar Mountain dust, Arizona Test Dust, nonadeconol, and birch pollen washing water. Experiments are performed using differential scanning calorimetry. The authors use repeated freezing and thawing cycles while tracking the change in the onset freezing temperature with run number. These refreeze experiments are interpreted through the prism of classical heterogeneous nucleation theory to infer if freezing occurs quasi-deterministic on special active sites or stochastic on truly random locations on the surface.

The experimental setup has been used previously by the same authors and the experimental protocols are sound. Overall the manuscript is well written. The analysis

is interesting, informative, and relevant to the ACP readers. At times the theoretical modeling seems to push against the limits on what can be in inferred in principle using the adopted methodology. If properly caveated, I recommend publication.

Comments

The concept of onset freezing isn't very clear. The manuscript states: "For the refreeze experiments, the onset of the freezing peak was evaluated. The evaluation was done using the implemented software "TA Universal Analysis" of the instrument." First, this part needs to be expanded to explain how the procedure works, what threshold is used to detect onset. Perhaps this could be illustrated with the help of Figure 1? For the Hoggar dust, the first freeze events were dominated by large droplets. I presume they are only sometimes present and the configuration may be more unstable? Do these runs coincide with the ones that are flagged by the rank correlation? Second, would it be possible to estimate how many drops or how much water volume needs to freeze to detect onset? Since this is used to repeatably probe the same active site, i.e. the most efficient nuclei in the sample, it would be important to estimate many different sites are able to compete for the "onset" detection. If it is not possible to estimate this, the authors should comment how competition from different active sites would affect the run cycle statistics and derived nucleation rates.

Introducing the beta factor to improve fits of jhet seemed like a promising approach to gain additional insight into the mechanism of freezing. However, the results show variation of beta from 10ˆ-9 to 10ˆ43, or 52 orders of magnitude. The observable range of the universe is 10ˆ-15 to 10ˆ26 m, only 41 orders of magnitude. A physical interpretation of beta seems to stretch credulity.

As an optional suggestion. The nucleation rates for the different samples, e.g. Figure 4, could be normalized by 'active site' strength, assuming ones buys into that concept. One way to normalize the rates is to anchor rates at T0, where T0 corresponds to a nucleation rate of 10ˆ-3 cm-2 s-1 (middle panel of Figure 4). All other temperatures would be relative to T0. The actual freezing temperature could be color coded onto the symbels. Such a plot might help to highlight similarities and differences in for the different active sites probed. If the graphs collapse, then a simple parameterization for a population of particles could be reported.

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