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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of individual ice nuclei by the single droplet freezing method: a case study in the Asian dust outflow region" *by* Ayumi Iwata and Atsushi Matsuki

Anonymous Referee #3

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The authors attempted to characterize the physico-chemical properties of atmospherically relevant ice-nucleating particles with the new off-line approach (i.e., a combination of several off-the-shelf techniques), which could potentially complement the in situ approach of ice residual studies. The topic itself is an important addition to ACP and the atmospheric science community. However, the experimental and analytical methods are unfortunately poorly explained and partly erroneous. Further, the manuscript contains a number of ambiguous statements and over-interpreted results without proper quantitative analyses as well as conclusive performance verifications. I have numerous suggestions for critical revisions. Additional tests of IDFM are necessary, and I do believe that the revision of the manuscript could be time consuming and result in a

significantly different paper.

Major comments and suggestions for additional tests are listed below:

The title is misleading. "Characterization of individual ice nuclei…" should read "…ice residual…" unless the authors could provide the evidence of particular individual particles repeatedly form ice over several cooling-thawing cycles. The properties of the particle may alter during/after ice activation, and the particle should be considered as a residual after thawing.

In the introduction section, the authors need to logically address why it is particularly important to study the aerosol mixing state to improve our understanding of atmospheric ice nucleation in mixed-phase clouds. What is special about the mixing state on ice nucleation as compared to other general properties, such as size and bulk composition? The authors may want to do a careful and rigorous literature review, digest the contents in a diplomatic manner and describe your thoughts to the reader along with your own story line.

Why did the authors arbitrarily pick -30 dC as the end cooling T (no explanation given)? Why didn't the authors carry out the T-binned analyses (e.g., up to -20 dC cooling vs. up to -30 dC cooling)? Such capability (seemingly feasible) and approach can resolve the issue, which the authors point out in the manuscript (e.g., P15L15-23).

Because differences in particle composition may be correlated with particle size, it is difïňĄcult to determine which characteristic fundamentally drives cloud-nucleating ability. Why didn't the authors conduct the size dependent analysis (i.e., coarse vs. fine) to examine if the size can be a nucleation-triggering factor? It is not appropriate to generalize the results based on all-size population.

The cooling rate of 0.5 K/s seems aggressive and not atmospherically relevant (i.e., ∼1 K/m may simulate a typical convective cloud updraft). Why not 1 K/min just like other numerous cold stage techniques do?

Show the profiles of dew point and T throughout the experiment rather than show snapshot pictures (i.e., Fig. 1).

Quantitatively evaluate the capability of immersion/condensation freezing method relative to other cold stages using the ice nucleation parameters, such as frozen fraction or ice nucleation active surface site density. The onset discussion (P711-18) seems qualitative, speculative and over-simplified. Description of the background contribution (contamination/impurity) should be included. The authors can assess it by putting soluble salts (e.g., NaCl) on the silicon wafer and cooling the cold stage down to the homogeneous freezing T.

Why the homogeneous freezing occurs over 3.5 K (P7L6-8)? It should be abruptly spontaneous in a narrow range of T, if the water saturation condition is well-controlled. The size of droplets might not be a substantial factor for the observed deviation. Technical validations seem necessary.

Fig 1: What particles are they? Regardless, the immersion freezing active fraction at -30 dC is 1/16. Is it comparable to other cold stage techniques? The authors may want to test reference samples (K-feldspar, quartz etc.) and estimate their n_s for to quantitatively compare to Atkinson et al. (2013, Nature).

Fig. 6: Two more pie charts should be added. Show the composition of total aerosols measured before cooling as well as that of after thawing to eliminate the artifact of cooling-thawing.

Sea salts are not ice nucleation inhibitors (P1L23), it is just not as active as other known INPs (e.g., aluminosilicate).

P2L9: Koop, 2000 (Nature) missing

How were reference sample powders dispersed/aerosolized onto silicon wafers? Some details of single particle techniques (laser intensity of Raman, and its influence on composition detection) are also missing.

Describing the experimental uncertainties in temperature measurement, particle size detection/limit etc. would be beneficial to the reader.

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