Authors' response to reviewers' comments for "Microfluidic Immersion Freezing of Binary Mineral Mixtures Containing Microcline, Montmorillonite, or Quartz" by Nadia Shardt, Florin N. Isenrich, Julia Nette, Christopher Dreimol, Ning Ma, Zamin A. Kanji, Andrew J. deMello, Claudia Marcolli

We thank the reviewers for their time and their suggestions that have significantly improved our manuscript. Below, we provide our responses and summarise the changes that we have made with page and line numbers referring to the uploaded document with tracked changes. Other minor revisions were also made to improve the manuscript, and we updated the TOC graphic.

Reviewer 1

The manuscript describes the heterogeneous freezing of water microdroplets containing pure microcline (mc), montmorillonite (mm), or quartz (qu) particles, as well as binary mixtures thereof, as observed and analyzed in a microfluidic droplet assay device.

As natural mineral dust is typically a mixture of various minerals, one key assumption underlying many experiments on the ice nucleation ability of mineral dust is that individual dust particles do not interact in their ice nucleation ability; i.e., their joint effect can be described as the surface-weighted sum of the constituents.

This manuscript presents a counterexample to this assumption by showing that the addition of montmorillonite to a microcline suspension can even decrease the ice nucleation ability of the microcline particles. The authors discuss this in the light of chemical interactions between the two minerals. This is an important observation that will probably stimulate additional work, and therefore, the manuscript should be published in ACP.

The manuscript is written very well, and the findings are presented clearly and concisely. I have several minor questions and suggestions, which are listed below.

In total, the manuscript contains four parts:

1. Description of the apparatus, especially the addition of a semi-automated freezing detection scheme using a neural network for image analysis (Figs 1, 2)
2. Presentation and discussion of the results obtained for the individual minerals mc, qu, mm. (Figs. 3, 4)
3. Presentation and discussion of the results obtained for mixtures of the minerals at different mixing rations (Figs. 5, 6).
4. Presentation of the results obtained for a sample of Arizona Test Dust (ATD)

General remarks:

Part 4 of the manuscript seems somewhat unrelated to the main part, could the authors comment in the introduction why it was included?

> **Authors' response**
>
> We agree that the link between the binary mixture results and the ATD suspensions was not explicitly stated, as also noted by Reviewer 2. To strengthen the link between these sections, we have now added fitting of the experimental n_s vs. T values for each pure mineral and predictions of frozen fraction for ATD suspensions assuming additive surface areas of the constituent minerals.
>
> **Changes to manuscript**
>
> - Abstract: added "Predictions are also made for Arizona Test Dust from the obtained pure mineral fits, and general agreement with experiments is observed."
> - Page 4 starting on line 31: added "A comparison between the ice nucleation activity of the controlled mixtures and that of ATD is drawn to investigate whether samples mixed

from the pure components can explain the ice nucleation activity of samples that were collected as mixtures from the air or ground."

- Page 23, Fig. 8: added predictions of frozen fraction for ATD at three concentrations using the pure minerals' n_s fits from Fig. 3. The caption has the following additional text: "The dash-dotted lines are predictions (not fits) of frozen fractions calculated using Eq. (4) with inputs of each pure mineral's nucleation site density vs. temperature shown in Fig. 3 assuming an ATD composition of 29 wt.% microcline, 23 wt.% quartz, and 25 wt.% montmorillonite."
- Page 23 starting on line 17: added a new paragraph stating: "The median freezing temperatures at each concentration are in the same range as those that would be expected for the fraction of microcline present. If we assume the composition of ATD to be that reported by Kaufmann et al. (2016) (i.e., 29 wt.% microcline, 23 wt.% quartz, and 25 wt.% montmorillonite, we obtain the predictions of frozen fraction shown by the dash-dotted lines for each concentration of ATD in Fig. 8 using Eq. (4) with the fits obtained from each pure mineral's nucleation site density (shown in Fig. 3). There is general agreement between the experimental frozen fractions for ATD and the predicted frozen fractions obtained from the activity of the pure components. This agreement suggests that samples mixed from the pure components can explain the ice nucleation activity of samples that were collected as mixtures from the air or ground, such as ATD. Such an observation can inform future work in: (*i*) the design of experimental campaigns with controlled bottom-up mineral mixtures acting as surrogates of real field samples without their variability and complexity, and (*ii*) the development of composition-aware parameterizations for ice nucleation in the atmosphere."

Remarks on Abstract:
Line 3: Please delete the word "Unfortunately". Previous lab experiments have focused on individual constituents for a good reason, and for most of the results shown in this manuscript, the above-stated assumption on the additivity of ice nucleation efficiency holds. C.f. also to my remark 3 on part 3.

Authors' response and change to manuscript
Upon the reviewer's suggestion, we have deleted the word "Unfortunately" in the abstract.

Remarks on part 1:

1. Please give some references to the CNN setup and/or software packages used. In the central panel of Figure 1, there are abbreviations in the legend, that are not explained.

   Authors' response
   We thank the reviewer for finding this omission. We have made the modifications below.

   Changes to manuscript
   - Page 8 on lines 8 and 10–12: added "implemented with the TensorFlow and Keras packages in Python" and "The last layer used rectified linear unit (ReLU) and softmax activation functions to determine the final classification."

2. In Fig. 2, it appears that the number of droplet freezing events detected varies strongly between the 5 mixtures. Please explain. What was the total no. of droplets within the field of view for each experiment in Fig. 2? Please give this number in the description or in the Figure caption.

   Authors' response
   The number of droplet freezing events changes because the number of droplets in the field of view varies between experiments. We have now clarified this in the text and added information to the caption of Fig. 2.

Changes to manuscript

- Page 6 starting on line 15: added "Based on these recommendations around 100–200 droplets and three repeats were used for experiments in the current study. There were inherent variations in the number of droplets observed in each experiment because of variability in the process of droplet generation between experiments. The primary goal of droplet generation was to obtain a stable, monodisperse population of droplets with sufficient spacing to avoid droplet coalescence. The spacing proved to be slightly variable depending on the exact fluid flow rates required to achieve a stable droplet population, which influenced the number of droplets in the field of view."
- Figure 2 caption: added "The total number of droplets present in the field of view is assumed to be equal to the total number of freezing events detected, with variations between experiments arising from differences in droplet spacing during droplet generation."

Remarks on part 2:

1. In all freezing curves (Figs 3, 5, 6) a frozen fraction curve for pure water is reproduced from Isenrich et al. 2022. Shouldn´t blank reference measurements with pure water be repeated regularly to exclude contaminations in the tubing? Please comment on this in the manuscript.

   Authors' response

   We report the pure water data from Isenrich et al. (2022) as our background data, because it was performed contemporaneously as part of the same experimental campaign, and it is representative of the freezing behavior for pure water in the setup. To prevent contamination in the tubing between runs, the same tubing was only reused for experiments with the same mineral, and the lower concentration experiments were completed first. The tubing was also flushed with isopropanol after each day of experiments to avoid contamination.

   Changes to manuscript

   - Page 5 starting on line 24: added "To remove the possibility for contamination in the inlet PTFE tubing to the microfluidic device, the same tubing was only reused for experiments with the same mineral. The tubing was also flushed with isopropanol after each day of experiments. If multiple mineral concentrations were investigated on the same day, the lower concentration of each mineral suspension was investigated first to avoid potential contamination of subsequent experiments."

2. In Fig. 3, the n_s values collapse to a single curve for quartz, as they do in Fig. 7 for ATD. For mc and mm however, they are disjunct between the concentrations used. Please comment on this surprising finding in the manuscript.

   Authors' response

   We agree that this discrepancy between different concentrations for microcline and montmorillonite should be investigated further. Potential causes of this may be a real change in the nucleation site density for the concentrations and particles sizes we investigated or simply variability in the actual suspension concentration compared to the nominal concentration. This variability may also play a role in the interpretation of binary mixture frozen fractions (i.e., the microcline–montmorillonite mixture), which we now discuss in more detail in the manuscript.

   Changes to manuscript

   - Page 13 starting on line 4, added "For microcline and montmorillonite, the derived n_s values do not agree between different mineral concentrations, which suggests that there are differences in the number of nucleation sites per unit surface area as a function of concentration. However, we cannot exclude the possibility that these apparent discrepancies may be due to deviations in the actual suspension concentrations from the nominal ones in the process of preparing the samples from the stock suspension. To obtain a representative model of the site density, we therefore consider all n_s data over all mineral concentrations of a single mineral. The dashed lines in the plots of n_s in Fig. 3 are fits obtained using Eq. (2) for microcline, quartz, and montmorillonite with standard deviations (SD) for $\log_{10} n_s$ of 0.47, 0.19, and 0.22, respectively. The highest standard deviation is obtained for microcline due to the spread of the experimentally-derived values of n_s between different nominal suspension concentrations. The obtained fitting parameters for each pure mineral were used to calculate frozen fractions for each concentration using Eq. (1), as illustrated by dashed lines in Fig. 3, showing close agreement with experimental observations for quartz and montmorillonite. Some of the predicted frozen fractions for microcline, however, deviate from those obtained experimentally, with a steeper slope in frozen fraction predicted for the highest concentration of 0.1 wt.% and a shallower slope for the concentration of 0.001 wt.%. As in our analysis of n_s for microcline, we attribute these discrepancies to potential variation in the actual concentration of the suspensions. The three replicates shown at each concentration were prepared from the same starting suspension and show high reproducibility, while different concentrations were prepared independently, and variability may be greater."

- Page 19 starting on line 20: added "Yet we cannot exclude the possibility that experimental variability in the determination of microcline's ice nucleation activity explains the observed tail of the microcline–montmorillonite mixture compared to the microcline–quartz mixture. As shown in Fig. 3, the trend of n_s vs. T for microcline derived from experimental frozen fractions does not collapse onto a single line over all studied concentrations, and in Fig. 6, the obtained fit for n_s to this data thus yields a frozen fraction at lower temperatures (dashed blue line) than those observed experimentally at a nominal microcline concentration of 0.001 wt.% (open diamonds). If the true activity of 0.001 wt.% microcline is represented by the dashed blue line, then the frozen fraction observed in Fig. 6a for microcline–quartz is explained by the inherent activity of a 0.1 wt.% quartz suspension, and the frozen fraction observed in Fig. 6b for microcline–montmorillonite is explained by the additive activity of each constituent, as depicted by the solid purple line calculated using Eq. (4)."

3. It would be helpful to discuss the expected error margins in the data as given e.g. in Fig. 3. If possible, give error bars for one exemplary sample.

   Authors' response
   We have added the uncertainty in temperature to the caption of each relevant figure (being 0.2 K). Some indication of the variability between runs is shown through the three replicates, but this variability may not be classified fully as experimental error, since nucleation is a stochastic process and observations of nucleation must inherently show variability. We designed our experiments to align with the results of Monte Carlo simulations performed by Alpert & Knopf (2016), where a minimum number of droplets and minimum number of experiments were recommended to ensure adequate accuracy and interpretability from the results obtained.

   Changes to manuscript
   - Captions of Figs. 3, 5, 6, and 8: added "(temperature uncertainty is 0.2 K (Isenrich et al., 2022))"

Remarks on part 3:

1. In all freezing curves, but particularly in Figs. 5 and 6, the individual symbol shapes are hardly discernible. I do not have an easy solution for this, but plotting symbols open instead of closed and/or at a smaller size might help the reader.

Authors' response and changes to manuscript

We thank the reviewer for the suggestions to improve the clarity of the figure. We have now used open symbols for the pure mineral frozen fractions and reduced the opacity of the filled symbols for the mixture data. We also note that the raw data that is shown in the figure will be made available online for interested readers to better see the trends of specific datasets.

2. It would be helpful if expected "fraction frozen" curves could be constructed for the various mixtures from the results of part 2 using the individual n_s values and surface areas (the expected additive outcome) to compare to the measurement results. This would probably also avoid plotting too many individual data points in Figs. 5 and 6.

Authors' response

We thank the reviewer for this suggestion, and we have now added these predicted frozen fraction curves based on the pure mineral site densities. In each relevant figure, we plot the predictions in solid lines.

Changes to manuscript

- Page 10: added "Section 2.4 Governing Equations for Fitting and Prediction" to describe the methods used for fitting of the pure mineral n_s vs. T data and for predicting the frozen fractions for both binary mixtures and ATD suspensions.
- Page 12, Fig. 3: added dashed lines to indicate the fits of n_s and the corresponding frozen fractions obtained from these fits
- Page 12, Fig. 3 caption: added "The dashed lines are model fits for n_s as a function of temperature, from which frozen fraction curves are calculated using Eq. (1). The equations are given by: $n_{s,mc} = 10^{2.3344}\,[-1.6598(T + 23.054)]^{3.2291}$, $n_{s,qu} = 10^{0.8924}\,[-1.2756(T + 21.865)]^{3.7223}$, and $n_{s,mm} = 10^{-0.2189}\,[-0.5956(T + 20.744)]^{4.8178}$."
- Figs. 5, 6, and 8: added dashed lines for pure component frozen fractions calculated using the n_s fits, solid lines for predicted frozen fractions based on additive surface areas of the pure minerals, and dash-dotted lines for frozen fraction predictions for ATD suspensions.

3. Discussion section: Even though the non-additive behavior in the ice nucleating efficiency of mc and mm is surprising and interesting, it should be noted that it is only apparent if mm is present in a 100x higher concentration than mc, a situation that is not very likely to be encountered in nature in an individual droplet.

Authors' response

Due to the possibility of variability in our measured pure microcline n_s values explaining the tail end of the microcline–montmorillonite mixture, we have tempered our discussion and conclusions relating to interactions.

Changes to manuscript

- Page 25 starting on line 32: deleted "Interestingly, the median freezing temperature of the microcline (0.001 wt.%)–montmorillonite (0.1 wt.%) mixture was lower than that of the microcline (0.001 wt.%)–quartz (0.1 wt.%) mixture, likely due to ion exchange at the microcline surface."
- Page 26, starting on line 8: deleted "It highlights the dominance of microcline as an ice-nucleating particle in the investigated bottom-up mixtures, and it identifies a potential for interactions in an internal mixture of montmorillonite and microcline (depending on their relative weight fraction)."

No specific remarks on part 4 or the appendices.

Reviewer 2
Schardt et al. performed a study of the ice-nucleating activity of mixtures of three different mineral types, in addition to the activity of the individual minerals within a droplet microfluidic device. The main findings showed that K-feldspar microcline dominated the ice-nucleating activity in the binary mixtures when present, even at much lower concentrations than the other mineral in the mixture; While this is often generally accepted as being the case, this is the first time to my knowledge that it has actually been shown rather than being simply expected. An interesting finding was that a mixture of montmorillonite-microcline showed a slight decrease in ice-nucleating activity at cooler temperatures compared to microcline only and the authors provide a theoretical explanation for this phenomenon.

The experiments are well designed and performed and the results confirm some pre-existing notions while offering new interesting findings about montmorillonite-microcline that warrant further exploration. However, I am not convinced that there is enough here for publication as a research article in for ACP as is and may be more suitable for publication as an ACP Measurement Report or perhaps as a paper in AMT. What would elevate the manuscript for publication in ACP would be a further small subset of experiments to test the hypothesis about the effect of Na+ ions in solution from the montmorillonite influencing the microcline. This could, for example, be through testing of the leaching of Na+ from montmorillonite at different concentrations via methods such as ICP or atomic absorption spectroscopy and testing of the influence of these concentrations of montmorillonite on microcline and the influence of Na+ concentrations on microcline. Given this, I have a few major comments and some minor comments.

Major comments:

1. When each experiment was performed three times, does this mean that a new suspension was prepared and tested each time, or three repeats were performed on the same suspension? I'd be intrigued as to whether a new suspension of "mc 0.001 wt% + mm 0.1 wt%" would act in the same manner. It is also interesting that the bulk of the slope for this binary mixture is very similar to one of the mc 0.001 wt% repeats; there is more of a "tail" as the authors mention, but overall the main part of the line does not look vastly different to this particular mc 0.001 wt% experiment.

   Authors' response
   The three repeats for each experimental condition were prepared from the same starting suspension. We agree with the reviewer that additional experiments from independent suspensions of microcline and montmorillonite at those concentrations would be warranted to test our hypothesis more rigorously. One example where we do have multiple independent suspensions at the same concentration is that of 0.01 wt% microcline (pure microcline, mixed with 0.1 wt% quartz, and mixed with 0.1 wt% mm). In each of these experiments, the frozen fractions were indistinguishable, with the same temperature for the onset and end of freezing. However, as the reviewer mentioned, we cannot exclude the possibility that experimental error or variability contributed to the observed "tail" of the mc+mm mixture, as shown by the predicted frozen fraction that assumes lower microcline nucleation temperatures.

   Changes to manuscript
   - Page 5 line 31: added "from the same starting suspension"
   - Page 19 starting on line 20: added "Yet we cannot exclude the possibility that experimental variability in the determination of microcline's ice nucleation activity explains the observed tail of the microcline–montmorillonite mixture compared to the microcline–quartz mixture. As shown in Fig. 3, the trend of n_s vs. T for microcline derived from experimental frozen fractions does not collapse onto a single line over all

studied concentrations, and in Fig. 6, the obtained fit for n_s to this data thus yields a frozen fraction at lower temperatures (dashed blue line) than those observed experimentally at a nominal microcline concentration of 0.001 wt.% (open diamonds). If the true activity of 0.001 wt.% microcline is represented by the dashed blue line, then the frozen fraction observed in Fig. 6a for microcline–quartz is explained by the inherent activity of a 0.1 wt.% quartz suspension, and the frozen fraction observed in Fig. 6b for microcline–montmorillonite is explained by the additive activity of each constituent, as depicted by the solid purple line calculated using Eq. (4)."

1. Page 15: This section is an interesting discussion and could explain some of the effects seen in the mm + mc curve. This is arguably the key finding of the paper,and would benefit greatly from exploration of this. For example, could the concentration of Na+ ions leached from the montmorillonite be estimated or measured and then experiments performed on microcline using aqueous solutions containing similar concentrations of Na+ or a range of concentrations to test the hypothesis?

   Authors' response
   We thank the reviewer for their suggestion for testing the hypothesis of ion exchange at the microcline surface. However, given the variability we now quantify in the nucleation site density of microcline and its importance for explaining the observed frozen fractions, further experiments would also need to be done to determine the variability in the observed "tail" of the microcline–montmorillonite mixture (as the reviewer alluded to in their first comment). Within the scope of the present paper, we do not aim to discriminate between the effect of ion exchange and the variability of the observed "tail".

1. Page 16, line 11: The ATD discussion should be in its own section. However, it is unclear why ATD is being discussed here. What is the context for its inclusion amongst a discussion about binary mixtures of minerals? I appreciate that ATD is in itself a mixture of minerals, but there is no apparent link or discussion between ATD and the other results shown here.

   Authors' response and changes to manuscript
   Upon the reviewer's suggestion, we have added a section heading for the results pertaining to ATD, and we now make predictions of frozen fraction for ATD based on the measurements performed on the pure minerals, with changes as outlined in our response to the first reviewer's first general remark.

Minor comments:

1. Page 2 line 7: Was XRD not performed on the quartz sample to ensure purity/composition?

   Authors' response
   In previous work from our group, the same quartz sample purchased from Sigma Aldrich was characterized by XRD to confirm the manufacturer's specification with 98.9% quartz (Kumar et al. Atmos. Chem. Phys., 19, 6035–6058, 2019).

1. Page 5, line 14: Some quartzes have been shown to exhibit changes in ice-nucleating activity even when suspended in water for a relatively short time (see Harrison et al. (2019; doi: 10.5194/acp-19-11343-2019)), while sonication is also a very energetic technique. Have the influences of these effects been assessed here to ensure there is no loss in quartz activity due to being in suspension for too long (particularly if also being heated by the sonication process)? Or the potential effect of sonication itself on activity – though I am unaware of any mention in the literature that sonication has an adverse effect.

Authors' response
All quartz samples, whether suspended alone or in a mixture, were treated in the same manner in terms of time spent in solution, so we assume that any potential impact of aging would be the same for all samples. We also note that the aging of quartz samples in water was reported by Kumar et al. (2019) to occur only in glass vials over a time period of days. In polypropylene Falcon tubes (as used in our study), the activity of quartz did not change after at least five days.

1. Page 5, line 20: What was the SEM analysis for? Determination of particle size distribution? EDS of the particles for composition analysis?

   Authors' response
   The SEM analysis was used in a qualitative manner to confirm that the particle sizes after filtration were indeed sufficiently small for improved uniformity in encapsulation between droplets. The quality and quantity of SEM images to determine particle size distribution was insufficient; rather, we used DVS to determine the BET surface area of the particles. The small sizes and morphologies of particles in the SEM images corroborates the high BET surface areas obtained.

1. Section 2.3: The CNN-Expert method seems to be the best way of analysing the data is a rapid but more accurate (than CNN only) manner, but not as good as "Expert only". Is the slight lack of accuracy compared to "Expert only" accounted for in the uncertainties in the data later on?

   Authors' response
   In our analysis, we assume the variability between replicate runs to be a greater source of variation in the results for frozen fraction than that arising from the slight difference between the CNN–Expert vs. the Expert only approaches for image analysis. While the Expert only is designated as the "true" classification, it may itself be subject to expert classification error (which is challenging to quantify in a rapid manner).

1. Page 10, line 8: Include reference to Harrison et al. (2019) when referring to variation in activity.

   Authors' response and change to manuscript
   We have added Harrison et al. (2019) to this sentence for improved completeness.

1. Page 10, line 25: As the authors discuss, it would make sense that the ns(T) values may be lower than expected since the larger particles have been removed compared to in the literature. Have the authors also considered that these very dense particles might also be settling in the syringe and tubing, thereby further reducing the size range of the particles that can actually enter the device for analysis?

   Authors' response
   We estimate that the sizes of particles where gravity separation would be relevant exceed those that are present after filtration. Given the time scale of the experiments performed, the actual size range of the particles entering the device would not be expected to change substantially.

1. Page 10, line 25: Also, it is very interesting to see the results for microcline compared to the literature, with the ns(T) values falling short of the "expected" activity. Both Peckhaus et al. (2016; doi: 10.5194/acp-16-11477-2016) and Tarn et al. (2018; doi: 10.1007/s10404-018-2069-x) saw similar behaviour when comparing microcline results from similar sized droplets

to the literature and this phenomena likely warrants some attention in future. Further, Harrison et al. (2016; doi: 10.5194/acp-16-10927-2016) demonstrated that different microclines can have varying activity. Many microcline studies tend to specifically use one of a handful of samples to ensure results can be compared across techniques/experiments.

Authors' response
We thank the reviewer for discussing these results from the literature to interpret our reported results. Because of this variability in activity we designed our experiments to systematically measure each pure component (especially microcline) over a wide range of concentrations before performing experiments on binary mixtures. We provide a brief discussion of the variability in the manuscript, including the effect that the assumed specific area has on the calculated n_s.

1. Page 10, line 27: As noted in an earlier comment, some quartzes can lose activity after being suspended in water for even a relatively short period of time and this is worth bearing in mind here.

   Authors' response
   As we indicated in our previous response, there is no loss of activity in quartz expected when polypropylene Falcon tubes are used for sample handling.

1. Figure 3: Please add y-error bars to the ns(T) values, these should at least be calculated from the uncertainty in droplet volume, uncertainties in the measurement of concentration when preparing the suspensions and the BET values.

   Authors' response
   We thank the reviewer for the suggestion. We have chosen to illustrate the experimental reproducibility and the corresponding variation in experimentally-derived n_s values by plotting each experiment as an independent data series. A large portion of the uncertainty in the derived n_s values can be attributed to the stochasticity of nucleation, as investigated in the literature through Monte Carlo simulations. We used the recommendations by Alpert & Knopf (2016) to design our experiments with around 100 droplets and three replicates to reduce the uncertainty in the obtained frozen fractions and derived quantities. A further sensitivity analysis of the most important contributions to the uncertainty of n_s values would be an interesting future direction.

1. Page 12, line 8 (and Figure 4): It is unclear what the SEM analysis is for, I had assumed particle size distribution as mentioned in the text but this does not seem to have been measured.

   Authors' response
   As mentioned in our previous response to a similar note earlier, we used SEM as a complementary technique to qualitatively confirm the particle size as support .the BET measurements.

1. Page 13, line 1: This part on binary materials should have its own section with heading.

   Authors' response and change to manuscript
   We agree with the reviewer's suggestion and have added section headings "3.1 Freezing Temperatures of Pure Mineral Suspensions" and "3.2 Freezing Temperatures of Binary Mineral Mixtures".

1. Was there no background (pure water) assay performed for these experiments? The baseline data is all from Isenrich et al. (2022), which is quite unusual given that some of the results in this study encroach into the homogeneous freezing area.

   Authors' response
   We report the pure water data from Isenrich et al. (2022) as our background data, because it was performed contemporaneously as part of the same experimental campaign, and it is representative of the freezing behavior for pure water in the setup. We also performed another series of experiments on the homogeneous freezing of water with two droplet sizes and cooling rates to confirm the robustness of the instrument and observed the same nucleation rates across all tested conditions (*Phys. Chem. Chem. Phys.*, 2022, **24**, 28213-28221 DOI: 10.1039/D2CP03896J). To prevent contamination in the tubing between runs, the same tubing was only reused for experiments with the same mineral, and the lower concentration experiments were completed first. The tubing was also flushed with isopropanol after each day of experiments to avoid contamination.

   Changes to manuscript
   - Page 5 starting on line 24: added "To remove the possibility for contamination in the inlet PTFE tubing to the microfluidic device, the same tubing was only reused for experiments with the same mineral. The tubing was also flushed with isopropanol after each day of experiments. If multiple mineral concentrations were investigated on the same day, the lower concentration of each mineral suspension was investigated first to avoid potential contamination of subsequent experiments."
   - Page 11 line 15: added "contemporaneously-measured pure water droplets ~~using the same experimental setup~~"

1. Page 15, line 7: "SWy-2 montmorillonite is a clay mineral that releases Na+ ions when suspended in water and these ions may exchange with the K+ ions at the surface of the microcline." – please provide a reference for this.

   Authors' response
   The cation exchange capacity of montmorillonite is indicated in the specification sheet of SWy-2 provided by the Clay Mineral Society, and the exchange with K^+ ions from the surface of the microcline is our hypothesized mechanism for the interaction between montmorillonite and microcline. Such an ion exchange mechanism has been hypothesized for other studied systems, such as microcline and dissociated species (Kumar et al. (2018)). Thus, we do not add a reference for our hypothesis directly but clarify it to be so based on the literature discussed in the sentences that follow it.

   Change to manuscript
   - Page 19, line 4: added "we hypothesize that"

1. Page 16, line 14: The ATD has also not been processed in the same manner as the other minerals apparently, since a specific surface area has now been assumed based on another study for a particle size range that is completely different to that used for the other minerals (which had been filtered to 0.45 micron). Was the ATD also filtered? This becomes more confusing as it is revealed that the particle size has a large effect on the specific surface area, but it is unclear what particle sizes were used in these experiments and in that case why a normalisation to 22 m2/g is used.

   Authors' response
   We confirm that the ATD has not been processed in the same manner. We attempted to filter the ATD, but there was insufficient material remaining to quantify the composition by XRD.

As a result, we performed experiments with un-filtered ATD as received by the provider (page 5 on lines 22–24). The ATD used was A1 Ultrafine, which has a documented size distribution with 95.5–97.5 % of particles being < 11 µm in size, and to represent this size distribution, we used 22 m^2/g as the specific surface area, according to the data reported by Ibrahim et al. (2018).

Changes to manuscript

- Page 22, moved the following sentences to line 7 instead of at the end of the paragraph: "The specific surface area has been shown to vary as a function of particle size; for example, Ibrahim et al. (2018) reported a value of 37.8 ± 1.7 m^2/g for the 0–3 µm nominal size range and a value of 2.8 ± 0.4 m^2/g for the 40–80 µm nominal size range. We assume a specific surface area of 22 m^2/g from Ibrahim et al. (2018) for the fraction of particle sizes between 5 and 10 µm to represent the specific surface area of the A1 Ultrafine ATD investigated herein, which is documented to have 95.5–97.5 % of particles being < 11 µm in size by the supplier."

1. Page 18, line 8: This should also be a new section.

   Authors' response and change to manuscript
   We thank the reviewer for their suggestion, and to improve the structure of the manuscript, we have moved this section to be placed before the presentation and discussion of ATD results and added a section heading "3.3 Freezing Temperatures of Arizona Test Dust (ATD) Suspensions".

1. Figure A1: Given that the feldspar sample contained 97 % microcline, should the literature parameterisations used for comparisons in the earlier figures not be scaled to this? I assume the literature parameterisation (e.g. Harrison et al. (2019)) is represented as 100 % microcline? Perhaps it would not make a substantial difference, but given the experimental values reported here are lower than the literature data this could be one of the reasons (in addition to those discussed elsewhere).

   Authors' response
   We agree with the reviewer that the 97% content of our microcline provides a negligible source of error compared to other contributions, i.e., variation in inherent sample activity from different sources and uncertainty in the specific surface area. It is also not fully clear whether the literature parameterisations normalise for the purity of the samples reported, such as in Atkinson et al. (2013), where the purity of K-feldspar was reported to be 80.4 wt.%.

1. Page 19, line 34: "the freezing behavior of Arizona Test Dust (ultrafine fraction) also followed its mineralogical composition" – I do not think that this was particularly discussed in this way in the Results section, it was very unclear what the point or outcome of these ATD experiments was, particularly since none of the other mineral data were plotted.

   Authors' response
   Also raised by Reviewer 1, we agree with the reviewer's observation, and we have now strengthened the link between the results of the pure mineral experiments and the results obtained with ATD. We have fit experimental n_s vs. T for each pure mineral and made predictions of frozen fraction for ATD suspensions.

   Changes to manuscript

   - See changes listed in response to the first comment by Reviewer 1.