

The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds

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The amount of ice present in mixed-phase clouds, which contain both supercooled liquid water droplets and ice particles, affects cloud extent, lifetime, particle size and radiative properties^{1,2}. The freezing of cloud droplets can be catalysed by the presence of aerosol particles known as ice nuclei². One of the most important ice nuclei is thought to be mineral dust aerosol from arid regions^{2,3}. It is generally assumed that clay minerals, which contribute approximately two-thirds of the dust mass, dominate ice nucleation by mineral dust, and many experimental studies have therefore focused on these materials^{1,2,4–6}. Here we use an established droplet-freezing technique^{4,7} to show that feldspar minerals dominate ice nucleation by mineral dusts under mixed-phase cloud conditions, despite feldspar being a minor component of dust emitted from arid regions. We also find that clay minerals are relatively unimportant ice nuclei. Our results from a global aerosol model study suggest that feldspar ice nuclei are globally distributed and that feldspar particles may account for a large proportion of the ice nuclei in Earth's atmosphere that contribute to freezing at temperatures below about -15°C .

Pure cloud droplets remain liquid until cooled to the homogeneous freezing threshold at around 237 K (ref. 2). At warmer temperatures, freezing can be catalysed by heterogeneous ice nuclei. Heterogeneous nucleation can occur in several ways depending upon temperature and humidity^{1,2}. Field observations and modelling studies of mixed-phase cloud formation have shown that supercooled liquid water is usually required before significant in-cloud ice formation occurs^{2,8}. Therefore, in this study we focus on heterogeneous ice nucleation by mineral dust particles immersed in supercooled water droplets.

Atmospheric mineral dusts are inorganic particles of rock and soil that have been lifted into the atmosphere, predominantly from arid regions such as the Sahara⁹. Mineral dusts from these regions are considered an important source of ice nuclei in mixed-phase clouds, owing to their nucleation efficiency^{7,10,11} and abundance in the atmosphere². The importance of mineral dusts as ice nuclei is also supported by their number concentrations within atmospheric ice crystals, which are enhanced relative to the background aerosol¹². At present the components in mineral dust responsible for ice nucleation are very poorly constrained. Although atmospheric dust concentrations and mineralogy vary spatially and temporally^{9,13} (Supplementary Fig. 1), a large fraction of observed atmospheric dust mass around the world is made up of just a few minerals. Individual minerals are classified by their crystal structure and chemistry and can be identified with diffraction techniques². The mineralogical composition of dust sampled from the atmosphere is shown in Supplementary Table 1. The clay minerals contribute approximately two-thirds of dust mass (kaolinites, 13%; montmorillonites, 2%; chlorites, 3%; micaceous minerals, such as the illites, 44%), with quartz (16%), feldspars (sodium/calcium feldspars (Na/Ca-feldspar), 8%; potassium feldspar (K-feldspar), 3%) and calcite (3%) responsible for much of the remainder.

Previous studies have investigated the ice-nucleating behaviour of dusts sampled from arid source regions or dusts selected as proxies for

natural dust^{1,2}. Studies of ice nucleation by individual minerals of varying purity immersed in water have focused on the clay minerals^{1,2,4–6}. However, minerals are rarely available in a pure state, and quantification of secondary minerals associated with a particular sample is often neglected. Such characterization is necessary because a minor component may dominate ice nucleation. In this study, we present measurements of ice nucleation by samples of individual minerals in which the impurities were quantified using X-ray diffraction.

To determine the ice-nucleating behaviour of each mineral, we used an established droplet freezing technique^{4,7}. Hundreds of micrometre-sized droplets containing a known amount of solid material were cooled at 1 K min^{-1} , and freezing was monitored by optical microscopy. The freezing temperatures of individual 14–16- μm -diameter droplets containing a range of different minerals are shown in Fig. 1a. In experiments with similar dust surface areas, the temperature at which

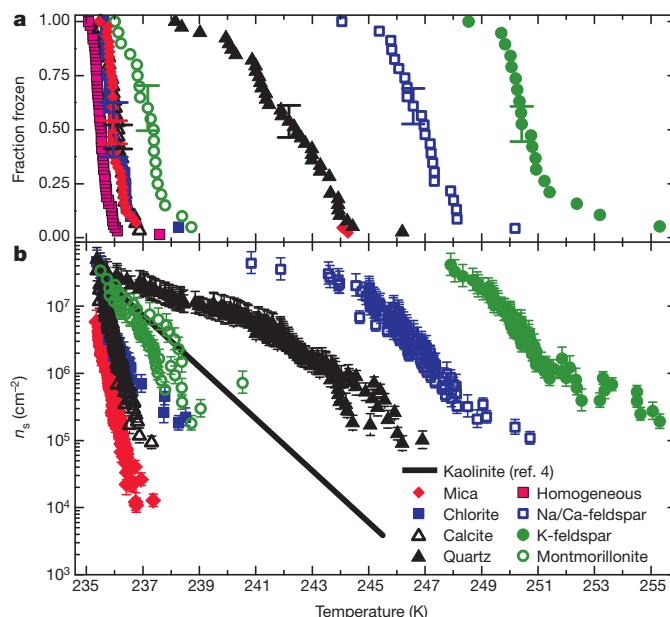


Figure 1 | Experimental freezing results for the individual minerals.

a, Fraction of droplets, 14–16 μm in diameter and containing a range of mineral dusts, frozen as a function of temperature during cooling. An experiment in which droplets contained no solid inclusion and froze homogeneously is shown for comparison. Temperature uncertainty (not shown) is estimated at $\pm 0.2\text{ K}$. The indicated uncertainty in the measured fraction frozen is the root mean squared error (68% confidence limit) determined from the pairwise differences between the data and the fraction frozen calculated from the log-linear best fits to the n_s values. **b**, Nucleation site densities for droplets between 9 and 19 μm in diameter collected into four 2.5- μm -wide bins. The uncertainty in n_s is primarily due to droplet size measurements, and temperature uncertainty is as in **a**. The kaolinite parameterization is from ref. 4.

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50% of droplets were frozen was 250.5 K for K-feldspar, 247 K for Na/Ca-feldspar, 242.5 K for quartz and less than 237.5 K for the clay minerals and calcite. These results suggest that it is the minerals of the feldspar group, in particular K-feldspar, that make mineral dust particles effective immersion-mode ice nuclei in the atmosphere. This contrasts with the prevailing view^{1,2}, which is that clay minerals are the most important component of atmospheric mineral dust for ice nucleation.

Droplet freezing temperatures are dependent on experimental parameters such as droplet volume and mineral surface area, and are therefore of limited value². To normalize the efficiency with which a material nucleates ice, we determine the number of nucleation sites per unit surface area^{2,11,14}, n_s (Fig. 1b and Supplementary Information). This method of quantifying ice-nucleation efficiency neglects the role of time dependence in nucleation on the basis that the particle-to-particle variability of ice nuclei is more important than the time dependence of nucleation^{2,11,14,15}. Our derived n_s values for 9–19- μm -diameter droplets are shown in Fig. 1b. These data show that the feldspar minerals, in particular K-feldspar, are the most efficient mineral dust ice nuclei per unit surface area.

In airborne dusts, the abundance of clay minerals tends to be greater than the abundance of the feldspars, and it is therefore not immediately clear which minerals dominate ice nucleation in the atmosphere. The n_s values presented in Fig. 1b were combined with the average mineralogical composition of atmospheric dust to estimate the temperature-dependent ice nuclei concentration (Fig. 2). We have assumed that all particles are spherical to estimate their surface area and have made two limiting calculations, one assuming that dust particles are internally mixed (that is, each particle contains all eight minerals) and the other assuming they are externally mixed (each particle is composed of an individual mineral). The mixing state of atmospheric dust is poorly constrained but falls between these two limiting cases¹⁶. Despite accounting for only 3% of atmospheric dust by mass, K-feldspar dominates the number of ice nuclei above 248 K in both the internally mixed and externally mixed cases. One potential caveat to this conclusion is that clay mineral particles may have a smaller particle size than feldspar or quartz¹³, and therefore may have a greater surface area per unit mass, which would increase the concentration of clay ice nuclei. However, even if the surface area of the clays were 100 times higher (probably an

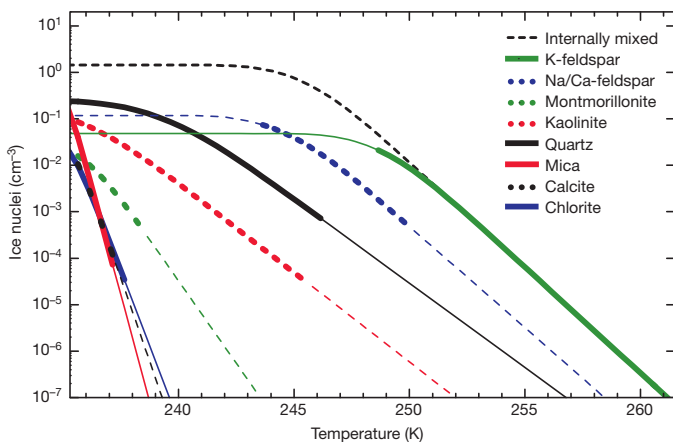


Figure 2 | Concentration of ice nuclei due to various minerals, for externally and internally mixed cases. Ice nuclei concentrations were estimated using the abundance of various minerals from Supplementary Table 1, taking the annually and globally averaged dust concentration (1.4 cm^{-3}) and size distribution from the GLOMAP model together with the n_s values in Fig. 1b (the K-feldspar line uses the n_s parameterization in Fig. 3). Lines with specific mineral names refer to individual minerals in an externally mixed case. Thick lines denote the range of experimental data and thin lines denote extrapolations outside this range.

overestimate⁷), the feldspars would remain the dominant ice-nucleating minerals (Supplementary Fig. 4).

Because clouds glaciate over a wider range of temperatures than is achievable in the experiments presented above, it is important to quantify the nucleating efficiency of atmospheric ice nuclei over a broader temperature range. For example, an ice nuclei concentration of as few as 10^{-5} cm^{-3} in a cloud at $\sim 266 \text{ K}$ may be able to trigger substantial glaciation through the Hallett–Mossop ice multiplication process¹⁷, and so measurements are needed at these temperatures. To extend the data for K-feldspar to this temperature regime, we performed a series of experiments with larger droplets, allowing for much larger particle surface areas per droplet and correspondingly lower values of n_s to be quantified. Results for K-feldspar using droplets $1 \mu\text{l}$ in volume are shown in Fig. 3. These data extend the range of experimental data up to 268 K. Combining the two techniques allowed us to determine n_s values over a range of eight orders of magnitude. The data show that the ice-nucleation efficiency of feldspar is measurable at temperatures relevant for the Hallett–Mossop process (about 265–270 K).

In Fig. 3, we compare our K-feldspar results with cloud-chamber-derived n_s values for a range of natural mineral dusts sampled from arid source regions¹⁰. Because feldspar is a major component of Earth's crust², it is ubiquitous in soils around the globe. K-feldspar makes up as much as $\sim 24\%$ by mass of soils throughout the Asian and African dust belt¹⁸ and is also present in airborne dust in concentrations ranging from a few per cent¹³ to 25% (ref. 19). We have estimated n_s values for natural dust in Fig. 3 by assuming that between 1% and 25% of the surface area of the dusts tested in ref. 10 was feldspar. In the temperature range where the data sets overlap, the agreement is very good.

Results presented here may explain discrepancies in existing experimental data for ice nucleation by mineral dusts. For example, a kaolinite sample from the Clay Mineral Society⁴ (CMS) had much lower n_s values

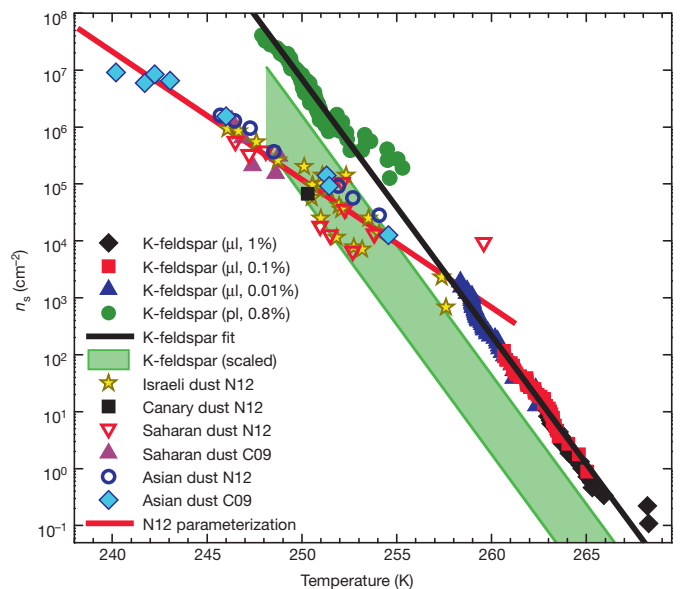


Figure 3 | Nucleation site density for K-feldspar and natural dusts. Data from Fig. 1b (picolitre experiments) extended to higher temperatures by use of microlitre-sized droplets, with a fit provided ($\ln(n_s) = -1.038T + 275.26$, valid between 248 and 268 K). Experimental K-feldspar concentrations in weight percent are provided in the key. Temperature uncertainties for microlitre experiments (not shown) were estimated at $\pm 0.4 \text{ K}$ and uncertainty in n_s (not shown) is estimated at $\pm 25\%$. We also compare with data for several natural dust samples from ref. 10 (N12) and ref. 11 (C09). The mineralogical compositions of the dusts used in those references are unknown, but feldspar mass content in natural soils typically varies between 1% and $\sim 25\%$ (see text). Hence, we have scaled our n_s values assuming K-feldspar is present at between 1% and 25% of the natural dust particles' surface.

than a sample obtained from Sigma-Aldrich^{2,5}. In this study, we have determined the mineralogical composition of these dusts and shown that the Sigma-Aldrich kaolinite contained 4.5% K-feldspar whereas the CMS kaolinite contained no detectable feldspar (Supplementary Table 2). It is likely that many individual particles of Sigma-Aldrich kaolinite contained feldspar, as is the case in atmospheric dust particles¹⁶. We have also quantified the mineralogy of the four montmorillonite samples used in a previous study of ice nucleation⁶ (see Supplementary Table 2 for sample mineralogy). Three of the samples (M SWy-2, M KSF and M K-10) contain K-feldspar and had higher freezing temperatures than the MSTx-1b sample, which did not contain measurable quantities of feldspar. Arizona test dust had the highest freezing temperature of any mixed-mineral dust proxy⁶ and also contained the most K-feldspar (~20 wt%). In general, the more feldspar a sample contains, the higher the freezing temperature. We propose that the feldspar component controlled the nucleation of ice in these experiments, highlighting the need to characterize sample mineralogy in such work.

The mineralogical composition of soils in arid regions around the world varies substantially. Therefore, to quantify the global contribution of feldspars to ice nuclei concentrations it is necessary to use a global aerosol model. Global maps of dust number concentration, feldspar mass fraction and ice nuclei concentration from an aerosol and chemical transport model, GLOMAP, are shown in Fig. 4a–d. To calculate ice nuclei concentration, the mineral-resolved size distribution of dust was combined with our parameterization for the ice-nucleation efficiency of feldspar at 253 K (ice nuclei concentrations at other temperatures are shown in Supplementary Fig. 7). In addition, we also make the assumption that the minerals in mineral dust aerosol are externally mixed; this assumption produces a better match to the observational

ice nuclei data at lower temperatures than does the opposing internally mixed assumption (Supplementary Fig. 8), although in reality the mixing state of atmospheric dust will lie somewhere between the internally mixed and externally mixed states.

The model data show larger ice nuclei concentrations close to the major dust sources in North Africa and Asia, which results in Northern Hemisphere dust ice nuclei concentrations around one to two orders of magnitude larger than in the Southern Hemisphere (comparing similar latitudes; Fig. 4c, d). Stratiform clouds in the Southern Hemisphere typically glaciate at lower temperatures^{20–22}, consistent with a lower concentration of ice nuclei than in the Northern Hemisphere.

To investigate the importance of dust mineralogy for modelling ice nuclei concentrations, we compared ice nuclei concentration in Fig. 4c with that predicted by a parameterization for natural dusts sampled from arid source regions¹⁰. Figure 4e shows that the two parameterizations are in agreement close to dust sources. However, the natural dust parameterization predicts ice nuclei concentrations up to 70% higher in regions remote from sources. This higher prediction arises because feldspar is more common in the larger-particle-size fractions (>2 μm) and therefore sediments out more rapidly than the minerals in the small-size fractions¹³ (Fig. 4b). Hence, atmospheric mineral dust becomes less efficient at nucleating ice during transport through a non-chemical ageing process. Our results may also help to explain the chemical ageing process of dust ice nuclei, which is known to reduce the ice-nucleating efficiency of dust²³. Feldspars are susceptible to emissions of acid gases such as SO_2 , which can convert the surface of feldspar grains to clay minerals²⁴. This may block ice-nucleation sites and reduce the efficiency of feldspar as an ice nucleus, which provides an explanation for the observed sensitivity of mineral dust ice nuclei to acid processing²³.

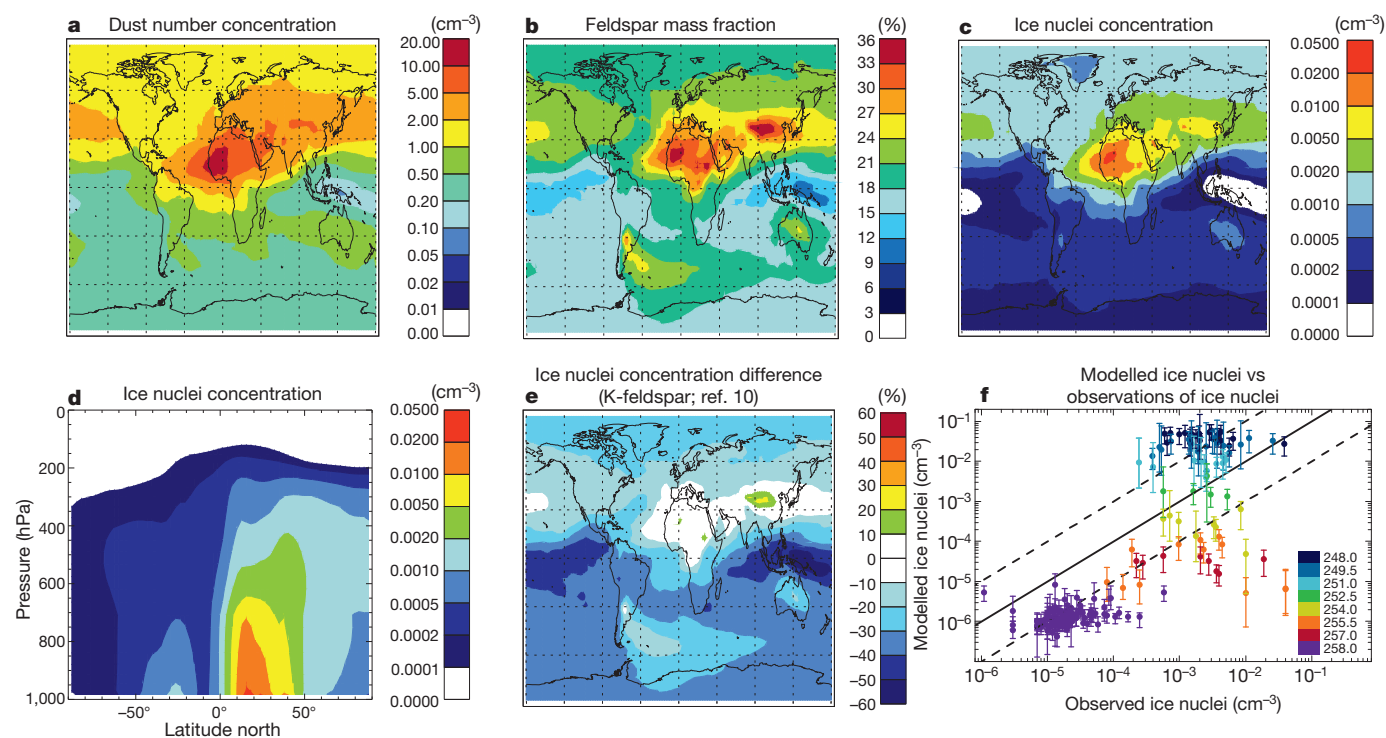


Figure 4 | Dust aerosol modelling study results. **a**, Modelled dust number concentrations. **b**, Total feldspar mass fraction of dust. **c**, Ice nuclei concentration due to K-feldspar at 253 K, calculated using our n_s parameterization and modelled particle surface areas. On the basis of observations in Supplementary Table 1, we assume 35% of feldspar mass is K-feldspar. **d**, Latitudinal zonal mean values of ice nuclei from **c**. **e**, Comparison of **c** versus ice nuclei concentrations calculated using a mineralogy-independent parameterization based on desert dust samples at 253 K (ref. 10). **f**, Comparison of model ice nuclei concentrations from K-feldspar mineral dust

with field measurements of total ice nuclei. Annual mean modelled ice nuclei concentrations are taken at the same pressure level as the field observation, with the observation temperature used to calculate n_s . Only observations between 248 and 258.15 K are shown; a comparison at higher temperatures is shown in Supplementary Fig. 9. Vertical error bars represent the maximum and minimum modelled monthly mean values. See Supplementary Table 4 for field campaign details. Parts **a**–**c** and **e** use concentrations at an altitude where the pressure is 600 hPa.

We also compare GLOMAP mineral dust ice nuclei concentrations with field measurements of ice nuclei concentrations (where the aerosol-processing temperature was ≤ 258 K) from around the world in Fig. 4f. The data are scattered around the 1:1 line, indicating that feldspar is one of the most important ice nuclei in Earth's atmosphere. The model tends to over-predict ice nuclei concentrations at temperatures below ~ 249 K. It is important to note that we do not include nucleation scavenging, where dust is removed when it serves as ice nuclei or cloud condensation nuclei, and we may therefore over-predict dust concentrations in regions remote from the source. Also, the cluster of data at 258 K, from a ship-borne study around southern Australasia (Supplementary Table 4), is consistently below the 1:1 line. This may indicate that in addition to mineral dust, other ice nuclei sources were also important in this region. At temperatures higher than 258 K, feldspar mineral dust is much less important as ice nuclei and cannot account for the observed ice nuclei concentrations (Supplementary Fig. 9). At these warmer temperatures, other types of ice nuclei, possibly of biogenic origin²⁵, may become increasingly important.

Finally, recent work suggests that human activity has led to a substantial increase in atmospheric dust concentrations and that the sources of this dust have changed^{9,26}. Because potential dust sources around the world have very different feldspar contents¹⁸, changes in the location of dust sources may have consequences for the concentration of ice nuclei in the atmosphere and the associated aerosol indirect effect.

METHODS SUMMARY

Experimental method. Picolitre experiments (Fig. 1) were performed using a freezing assay of micrometre-sized droplets in a manner similar to previously described⁴⁷. The main variation between this and the previous method is the development of a new cold stage configuration to improve the thermal stability and control of the system, with improvements to temperature measurements reducing uncertainty to ± 0.2 K. The microlitre experiments were performed with a separate system in which cooling, temperature measurement and control were provided by a Stirling engine-powered flat-plate chiller (Grant-Asymptote EF600). Droplets were then deposited onto a large, siliconized glass slide using a pipette, and freezing was monitored using a digital camera without magnification. The analysis of the data from the picolitre and microlitre experiments was identical. Mineral dust characterization methods have been described previously⁷.

Modelling with GLOMAP. GLOMAP is a size- and composition-resolving two-moment microphysical aerosol scheme²⁷, run within the TOMCAT chemical transport model²⁸. GLOMAP has previously been used to study atmospheric processing of mineral dust²⁹. The model is driven by reanalysis meteorology for the year 2000. GLOMAP was extended to represent eight mineral types, as specified by the surface mineralogy map of ref. 18. Dust is represented in 12 size bins, ranging in diameter from 0.1 to >20.0 μm . Dust emissions are prescribed from AEROCOM recommendations for the year 2000³⁰. The experimentally derived n_s values were combined with modelled atmospheric particle sizes, composition and concentrations (annual global averages at the altitude corresponding to a pressure of 600 hPa) to estimate ice nuclei concentrations (Fig. 2). Two particle mixing states were considered. In the internally mixed case, all particles contain the considered minerals in the same ratios, whereas in the externally mixed case individual particles are composed of single minerals with the overall population composition controlled by the dust mineralogy.

Received 30 January; accepted 7 May 2013.

Published online 12 June 2013.

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Supplementary Information is available in the online version of the paper.

Acknowledgements We thank J. Cuadros, L. Neve and R. Finch for help sourcing mineral samples, P. DeMott for providing ice nuclei observational data, and T. W. Wilson for discussions. We acknowledge the European Research Council (FP7, 240449 ICE) and the Natural Environment Research Council (NE/I013466/1, NE/I020059/1, NE/I019057/1) for funding. K.S.C. is a Royal Society Wolfson Research Merit Award holder.

Author Contributions J.D.A. conducted the picolitre experiments, analysed the data and wrote the paper, and T.F.W. performed and analysed the microlitre experiments. K.J.B. and D.O. contributed to the experimental study, and S.D. helped draft the manuscript. M.T.W. led the global modelling study in collaboration with K.S.C. T.L.M. did the X-ray analysis of the mineral samples. B.J.M. oversaw the project and helped to write the manuscript.

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