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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} and show that feldspar minerals dominate ice nucleation by mineral dusts under mixed-phase cloud conditions, despite 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 ice nuclei in the Earth's atmosphere below about -15°C.

Pure cloud droplets remain liquid until cooled to the homogeneous freezing threshold at around 237 K². At warmer temperatures freezing can be catalysed by the presence of aerosol particles known as ice nuclei (IN). This process, referred to as heterogeneous nucleation, can proceed via several modes 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 IN in mixed-phase clouds due to their nucleation efficiency^{7,10,11} and abundance in the atmosphere⁹. The importance of mineral dusts as IN is also supported by their enhanced number concentrations within atmospheric ice crystals relative to the background aerosol¹². At present the components in mineral dust responsible for ice nucleation are very poorly constrained. While atmospheric dust concentrations and mineralogy vary spatially and temporally^{9,13} (see Supplementary Figure 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% and micaceous minerals, such as the illites, 44%), with quartz (16%), feldspars (sodium/calcium feldspars 8%, potassium 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 characterisation 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.

In order to determine the ice nucleating behaviour of each mineral an established droplet freezing technique was used^{4,7}. Hundreds of micron-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 droplets containing a range of different minerals are shown in Figure 1a. In experiments with similar dust surface areas, the temperature at which 50% of droplets were frozen was 250.5 K for K-feldspar, followed by 247 K for Na/Ca-feldspar, 242.5 K for quartz, and below 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 an effective immersion mode IN in the atmosphere. This data contrasts with the prevailing view^{1,2} 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. In order to normalise the efficiency with which a material nucleates ice we determine the nucleation sites per unit surface area^{2,11,14} (n_s ; Figure 1b; see supplementary online material). This method of quantifying ice nucleation efficiency neglects the role of time dependence in nucleation, on

the basis that IN particle-to-particle variability is more important than the time dependence of nucleation^{2,11,14,15}. Our derived n_s values for 9 -19 μm size droplets are shown in Figure 1b. This data shows that the feldspar minerals, in particular K-feldspar, are the most efficient mineral dust IN per unit surface area.

In airborne dusts the abundance of clay minerals tends to be greater than the feldspars, hence it is not clear which minerals dominate ice nucleation in the atmosphere. The n_s values presented in Figure 1b were combined with the average mineralogical composition of atmospheric dust to estimate the temperature-dependent IN concentration (shown in Figure 2). We have assumed that all particles are spherical in order to estimate their surface area and have made two limiting calculations, one assuming that dust particles are internally mixed (i.e. 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 atmospheric dust falls between these two limiting cases¹⁶. Despite only accounting for 3% of atmospheric dust by mass, K-feldspar dominates the number of IN above 248 K in both the internally 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 IN. However, even if the surface area of the clays was 100 times higher (likely an overestimate⁷), the feldspars remain the dominant ice nucleating minerals (Supplementary Figure 4).

Since clouds glaciate over a wider range of temperatures than is achievable in the experiments presented above, it is important to further quantify the nucleating efficiency of atmospheric IN over a broader temperature range. For example, Crawford *et al.*¹⁷ suggest that an IN concentration of as few as 10^{-5} cm^{-3} in a cloud at $\sim 266 \text{ K}$ can trigger substantial

glaciation via the Hallett-Mossop ice multiplication process, hence measurements are needed at these temperatures. In order 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 of 1 μL volume are shown in Figure 3. This data extends the range of experimental data up to 268 K. Combining the two techniques allowed n_s values to be determined over a range of eight orders of magnitude. The data shows that feldspar has measurable ice nucleation efficiency at temperatures relevant for the Hallett-Mossop process (about 265-270 K).

In Figure 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 regions by Niemand *et al.*¹⁰. Since feldspar is a major component of the 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 percent¹³ to 25%¹⁹. We have estimated n_s values for natural dust in Figure 3 by assuming that between 1 and 25% of the surface area of the dusts tested by Niemand *et al.*¹⁰ was feldspar. In the temperature range where the datasets 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 Society (CMS)⁴ had much lower n_s values 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 contains 4.5% K-feldspar whereas the CMS kaolinite contained no detectable feldspar (Supplementary Table 2). It is likely that many individual Sigma Aldrich kaolinite particles contained feldspar, as is the case in atmospheric dust particles¹⁶. Pinti *et*

*al.*⁶ studied ice nucleation by several montmorillonite samples. Three of these samples (M SWy-2, M KSF and M K-10) contain K-feldspar and had higher freezing temperatures than the M STx-1b sample which did not contain measurable quantities of feldspar. Arizona test dust had the highest onset freezing temperature of any mixed-mineral dust proxy¹⁰ and also contains the most K-feldspar (20 wt%). In general, the more feldspar a sample contains the higher the freezing temperature. We hypothesise that that the feldspar component controlled the nucleation of ice in these experiments, highlighting the need to characterise 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 IN concentrations it is necessary to use a global aerosol model. Global maps of dust number concentration, feldspar mass fraction and IN concentration from an aerosol and chemical transport model, GLOMAP, are shown in Figures 4a-d. In order to calculate IN concentration, the mineral-resolved size distribution of dust was combined with our parameterisation for the ice nucleation efficiency of feldspar at 253 K (IN concentrations at other temperatures are shown in Supplementary Figure 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 IN data at lower temperatures than the opposing internally mixed assumption (Supplementary Figure 8), although in reality the mixing state of atmospheric dust will lie somewhere in between the internally and externally mixed states.

The model data clearly shows larger IN concentrations close to the major dust sources in North Africa and Asia, which results in northern hemispheric dust IN concentrations of around 1-2 orders of magnitude larger than in the southern hemisphere (comparing similar latitudes, Figures 4c-d). Stratiform clouds in the southern hemisphere typically glaciate at

lower temperatures²⁰⁻²², consistent with a lower concentration of IN than in the northern hemisphere.

To investigate the importance of dust mineralogy for modelling IN concentrations we compared IN concentration in Figure 4c with that predicted by a parameterisation for natural dusts sampled from arid source regions from Niemand *et al.*¹⁰. Figure 4e shows that the two parameterisations are in agreement close to dust sources. However, the natural dust parameterisation predicts IN 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 bins (Figure 4b)¹³. Hence atmospheric mineral dust becomes less efficient at nucleating ice during transport through a non-chemical ageing process. Our results may also help understand the chemical ageing process of dust IN, 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 IN to acid processing²³.

We also compare GLOMAP mineral dust IN concentrations with field measurements of IN concentrations (where the aerosol processing temperature was ≤ 258 K) from around the world in Figure 4f. The data are scattered around the 1:1 line, indicating that feldspar is one of the most important ice nuclei in the Earth's atmosphere. The model tends to over-predict IN 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 IN or CCN, and we may therefore over-predict dust concentrations in regions remote from source. Also, the cluster of data at 258 K, from a ship-borne study around southern Australasia (see Supplementary Table 4), is consistently below the 1:1 line. This may indicate that in addition

to mineral dust, other IN sources were also important in this region. At temperatures warmer than 258 K feldspar mineral dust is much less important as an IN and cannot account for the observed IN concentrations (Supplementary Figure 9). At these warmer temperatures it appears that other IN types, possibly of biogenic origin²⁵, may become increasingly important.

Finally, recent work suggests human activity has led to a substantial increase in atmospheric dust concentrations and that the sources of this dust have changed^{9,26}. Since 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 IN in the atmosphere and the associated aerosol indirect effect.

Methods summary

Experimental method

Picolitre experiments (Figure 1) were performed using a freezing assay of micron sized droplets in a manner similar to previously described^{4,7}. 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 siliconised glass slide using a pipette, before freezing was observed using a digital camera without magnification. The analysis of the data from the picolitre and microlitre experiments was identical. Mineral dust characterisation 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 Nickovic *et al.*¹⁸. Dust is represented in 12 size bins, ranging from 0.1 μm to $>20.0 \mu\text{m}$ diameter. 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 (600 hPa annual global average) to estimate IN concentrations (Figure 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.

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Supplementary information is linked to the online version of the paper at www.nature.com/nature.

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global modelling study in collaboration with KSC. TLM did the X-ray analysis of the mineral samples. BJM oversaw the project and helped to write the manuscript.

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Figure legends

Figure 1. Experimental freezing results for the individual minerals. **a**, Fraction of droplets, 14-16 μm diameter and containing a range of mineral dusts, frozen as a function of temperature during cooling. An experiment where droplets contained no solid inclusion and froze homogeneously is shown for comparison. Temperature uncertainty (not shown) is estimated at ± 0.2 K. The statistical uncertainty in fraction frozen is indicated. **b**, Nucleation site densities (n_s , cm^{-2}) for droplets between 9 and 19 μm diameter collected into four bins 2.5 μm wide. n_s uncertainty is primarily due to droplet size measurements, temperature uncertainty as in **a**. The kaolinite parameterisation is from Murray *et al.*⁴.

Figure 2. The 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 Figure 1b (the K-feldspar line uses the n_s parameterisation in Figure 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.

Figure 3. Nucleation site density (n_s , cm^{-2}) for K-feldspar and natural dusts. Data from Figure 1b (pL) extended to higher temperatures by use of μL 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 μL experiments (not shown) were estimated at ± 0.4 K, $n_s \pm 25\%$. We also compare to literature data for several natural dust samples from Niemand *et al.* (N12)¹⁰ and Connolly *et al.* (C09)¹¹. The mineralogical compositions of the dusts used by these authors 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.

Figure 4. Dust aerosol modelling study results. **a**, Modelled dust number concentrations. **b**, Total feldspar mass fraction of dust. **c**, IN concentration due to K-feldspar at 253 K, calculated using our n_s parameterisation and modelled particle surface areas. Based upon observations in Supplementary Table 1, we assume 35% of feldspar mass is K-feldspar. **d**, Latitudinal zonal mean values of IN from **c**. **e**, Comparison of **c** versus IN concentrations calculated using a mineralogy independent parameterisation based on desert dust samples at 253 K¹⁰. **f**, Comparison of model IN concentrations from K-feldspar mineral dust to field measurements of total IN. Annual mean modelled IN 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 K and 258.15 K (-15 °C) are shown; a comparison at higher temperatures is shown in Supplementary Figure 9. Vertical error bars represent the maximum and minimum modelled monthly mean values. See Supplementary Table 4 for field campaign details. Parts **a,b,c,e** use concentrations at 600 hPa.







