

Ice nucleation efficiency of mineral dust surrogates in the immersion mode

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Atmospheric background

37% of the total aerosols (5700 Tg / yr; IPCC, 2001) injected in the atmosphere is desert dust. Desert dust is injected in the atmosphere every year, these aerosols can reach high altitudes (7 km) and can be transported over long distances. Desert aerosols can also act as ice nuclei (IN) modifying the optical properties of clouds and their lifetime (indirect effect). Nucleation process in the atmosphere can occur either by homogeneous or heterogeneous freezing; while the first process occurs at temperatures below ~236 K, when an insoluble particle is present in a droplet, it can freeze at warmer temperatures (De Mott et al., 1997).

Introduction

Mixed phase clouds present either ice crystals then water droplets, they are founded at middle altidudes between 3 and 6 km and their structure favors precipitations. Our study is based on understanding which mineral component of desert dust aerosol is mostly responsable for the heterogeneous ice nucleation and why.

Illite, kaolinite and montmorillonite are some of the most abundant components of desert dust aerosols (Glaccum and Prospero, 1979; Fig. 1). The aim of this study was to investigate these minerals and their behaviour as IN in immersion mode with a Differential Scanning Calorimetry (DSC).

As Marcolli et al. 2007 investigated Arizona Test Dust (ATD), here three different kinds of mineral were studied following the same procedure: illite (NX and SE) provided by Arginotec; kaolinite, the first one provided by Fluka and the second/third one (KGa-1b/Kga-2) provided by Clay Minerals Society; two kinds of montmorillonite provided by Fluka (Montmorillonite K10, Montmorillonite KSF



Saharan dust collected at Sal Island, Barbados and Miam = mica/illite; K = Kaolinite; C = chlorite; M = montmorillonite (upper limit 5%); Q = quartz Mc = microcline: P = plagioclase: Cc = calcit

Figure 1: Glaccum and Prospero et al., 1980

Illite

Montmorillonite

Kaolinite

Illite is a phyllosilicate or layered alumino-silicate. Its structure is constituted by the repetition of Tetrahedron – Octahedron – Tetrahedron (TOT) layer (Fig 2).

<u>Chemical formula: (K,H₃O)(AI,Mg,Fe)₂(Si,AI)₄O₁₀[(OH)₂,(H₂O)]</u>



Figure 2: illite structure

Illite NX

Mineral composition: (wt%)

Illite 86 Kaolinite 10 Calcite traces Quartz Feldspar traces

Size distribution: median diameter: 0.313±0.002 μm standard deviation: 0.39±0.05 μm

Droplets size distribution: median diameter: 1.73 ±0.05 µm Illite SE

Mineral composition: (wt%) Illite 77 Kaolinite 10

12 Calcite Quartz traces Feldspar traces

Size distribution: median diameter: 0.327±0.001 μm standard deviation: 0.33±0.03 µm

Droplets size distribution: median diameter: 1.60±0.04 μm 2 tetrahedral sheets sandwiching a central octahedral sheet (TOT) (Fig 3.).



Montmorillonite KSF

chemically treated: completely activated due to the high sulfuric acid content.

completely de-lamellated.

Size distribution: median diameter: 0.428±0.009 μm standard deviation: 0.42±0.03 µm

Droplets size distribution: median diameter: 1.56 ±0.05 μm standard deviation: 0.34±0.04 µm

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Montmorillonite K-10

Chemically treated with strong acids: the aluminosilicate sheets are partially disrupted

Size distribution: median diameter: 0.406±0.002 μm standard deviation: 0.441±0.003 µm

Droplets size distribution: median diameter: 1.42 ±0.04 μm standard deviation: 0.32±0.04 µm

ATD

Arizona test dust was investigated by Marcolli et al. 2007, one sample with a nominal 0-3 μ m particle diameters (fine ATD) and the other one with 0-7 μ m (coarse ATD).

It is a layered silicate mineral, with one tetrahedral sheet linked through oxygen atoms to one octahedral sheet of alumina octahedra (Fig 4).

Chemical formula: $Al_2Si_2O_5(OH)_4$



Figure 4: kaolinite structure

Kaolinite (Fluka)

Grade: purum Quality: natural

Size distribution: median diameter: 0.472±0.003 µm standard deviation: 0.34±0.05 µm

Droplets size distribution: median diameter: 1.45 ±0.05 μm standard deviation: 0.34±0.05 µm KGa-2

Infrared spectroscopy shows a high defected kaolinite, collected in Georgia (USA).

KGa-1b

Infrared spectroscopy shows a well crystallized kaolinite, collected in Georgia (USA).

standard deviation: 0.39±0.05 µm

Onset points



Instruments

DSC, TA instrument: emulsified and bulk samples have been investigated with a differential scanning calorimeter.

<u>SMPS</u>: a scanning mobility particle sizer was used to estimate particles size distribution.

Electron microscope: pictures of the emulsion were taken with an electron microscope with a magnification of 50x (Fig 6).



Figure 6: example of an emulsion

DSC experiments

The emulsions (Fig. 6) consist of 80 wt% of a mixture of lanoline (Fluka) and mineral oil (Aldrich) and 20 wt% of aqueous suspensions of mineral dust (made with distilled and deionized water, 18.2 M Ω).

Three freezing/melting cycles were run, the first one and the last one with a cooling rate of 10 K min⁻¹, while the second cycle, used for the evaluation, was run with a cooling rate of 1 K/min.

Fig 7 shows a tipical cooling cycle of 1 K/min for the Kga-1b sample, the first peak corresponds to the heterogeneous ice nucleation while the second one to the homogeneous one.



Universal V4.3A TA Instrument Exo Up Temperature (K)

Figure 6: Tipical DSC thermogram

Figure 5: onset points of the different studied samples. The ATD samples were investigated by Marcolli et al. 2007.

Summary and comments

Three kinds of desert dust surrogates were investigated. The different behaviour of the ice nuclei not only depends on which mineral is used, but it also depends on the crystal structure of the sample (Fig 5).

Previous studies (Marcolli et al., 2007, and Möhler et al,2006) show that the ATD particles act as very efficient ice nuclei. Marcolli et al. found onset points in the range 247-252 K.

The montmorillonite K10 is often used to study the behaviour of mineral dust like an atmospheric component, but it is not rappresentative of real dust

Future work

• Three different samples of real desert dust coming from three different deserts (Sahara, Israel and Taklamakan) will be investigated.

• A parameterization of nucleation rate developed by Marcolli et al., 2007, will be updated with new results of real desert dust samples.

•Desert dust surrogate surfaces will be chemically treated in order to understand how these modifications can affect the ice nuclei activity.

Conclusions

• Different crystal structure does not let shift the onset points to higher temperatures (Kga-1b, Kga-2)

• Completely destroying the crystal structure the montmorillonite presents the heterogeneous freezing onstet at lower temperatures (Mont K10, Mont KSF).

· Different mineral compositions of dust can affect the efficiency of ice nucleation, thus every real desert dust might react in different way.

• Increasing the concentration, the probability to have more active surfaces in the droplets increases.

References

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