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Citation: AIP Conference Proceedings **1527**, 914 (2013); doi: 10.1063/1.4803420
View online: http://dx.doi.org/10.1063/1.4803420
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/1527?ver=pdfcov
Published by the AIP Publishing

Immersion Freezing of Clay Minerals and Bacterial Ice Nuclei

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Abstract. The immersion mode ice nucleation efficiency of clay minerals and biological aerosols has been investigated using the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber. Both monodisperse and polydisperse populations of (1) various clay dust samples as well as (2) Snomax® (a proxy for bacterial ice nucleators) and (3) hematite are examined in the temperature range between -4 ˚C and -35 ˚C. The temperature dependence of ice formation inferred by the INAS (Ice Nucleation Active Surface-Site) density is investigated and discussed as a function of cooling rate and by comparing to predicted nucleation rates (i.e., classical nucleation theory with θ-probability density function nucleation scheme). To date, we observe that maintaining constant AIDA temperature does not trigger any new ice formation during the immersion freezing experiments with clay dust samples and Snomax®, implying strong temperature dependency (and weak time dependency) within our time scales and conditions of experiments. Ice residuals collected through a newly developed PCVI (Pumped Counter-flow Virtual Impactor) with the 50% cut size diameter of 10 to 20 μm have also been examined by electron microscope analyses to seek the chemical and physical identity of ice nuclei in clay minerals. In addition to the AIDA results, complementary measurements with mobile ice nucleation counters are also presented.

Keywords: heterogeneous ice nucleation, immersion, clay mineral, biological aerosol, PCVI
PACS: 42.68.Ge, 82.70.Rr, 64.60.Q-, 87.18.-h

CLAY DUST SAMPLES

The immersion freezing of various clay dust samples, including three illite (namely quartz rich illite from the Clay Mineral Society, feldspar rich German illite, and NX Nanopowder from Arginotec®) as well as two kaolinite standards (Fluka and Clay Mineral Society), has been studied in the temperature range -12 ˚C to -35 ˚C. The preliminary results are shown in **FIGURE 1**, where the INAS density, n_s, represents the number concentration of ice crystals (N_{ice}) normalized to the total surface area of

Nucleation and Atmospheric Aerosols
AIP Conf. Proc. 1527, 914-917 (2013); doi: 10.1063/1.4803420

particles (A). We observe the immersion mode ice nucleation activity, inferred by the INAS density, of clay minerals to be strongly dependent on the temperature. For instance, INAS density increases with decreasing temperature as previously described in the literature [e.g., 1, 2]. We also observe that the immersion mode nucleation of our dust samples is equally active as desert dust samples and even comparable to the deposition mode of Snomax®. Among our clay dust samples, kaolinite from Fluka is the most efficient immersion ice nuclei in the temperature range of -20 ˚C to -28 ˚C. An explanation of its ability to be more ice active compared to other clay minerals is given.

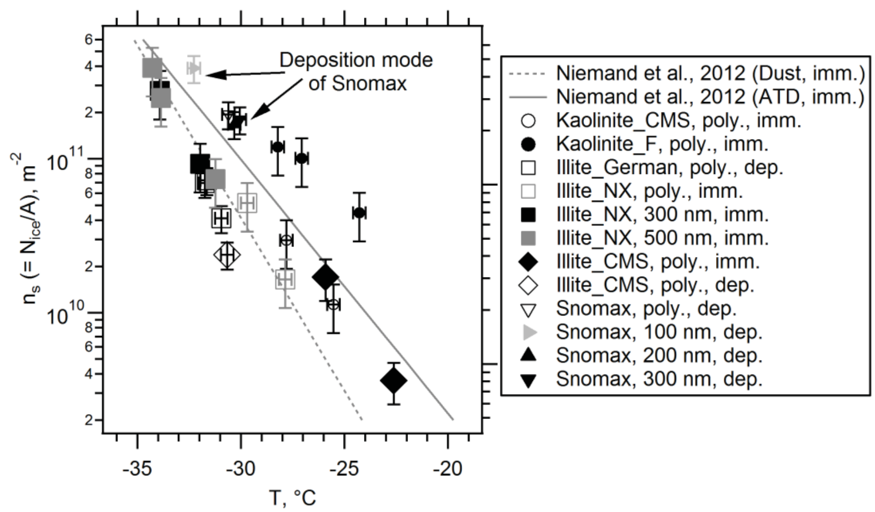

FIGURE 1. INAS densities, n_s, for deposition and immersion freezing of clay dust samples as a function of temperature, T. Lines represent fits to previously measured data of natural dusts (denoted as Dust) and Arizona Test Dust (ATD) taken from *Niemand et al*. [3], and references therein.

SNOMAX SUSPENSION

Snomax® (Johnson Controls Inc., Milwaukee, USA) is a commercially available product mainly composed of freeze-dried *P. syringae* bacteria and often used to induce artificial snow formation in ski resorts. As in previous studies, the particles generated from Snomax® solution/suspension (hereafter denoted as Snomax particles) trigger heterogeneous freezing in a narrow temperature range of -7 to -10 ˚C, and associated the immersion mode ice nucleation efficiency of Snomax particles is well characterized [e.g., 4, 5, 6]. Even less studied to date is the freezing behavior of size-segregated Snomax particles and its influence on overall freezing processes, including deposition, immersion, and contact mode. Given such a burden, for the first time, we conduct AIDA experiments with quasi-monodisperse Snomax particles for both deposition and immersion mode freezing. Results for the various expansions are plotted in **FIGURE 2**. The results indicate that while deposition mode ice nucleation of Snomax particles is nearly independent of size, immersion mode nucleation exhibits a strong size dependency (i.e., the larger surface carries more ice active protein complexes), suggesting a need for nucleation mode specific parameterization of size dependency.

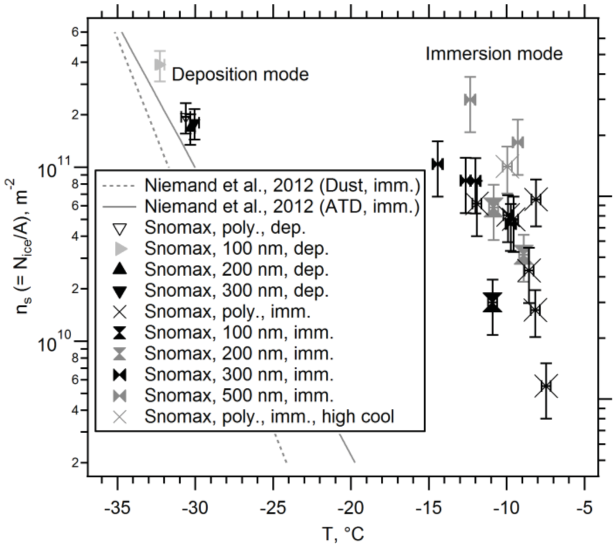

FIGURE 2. INAS densities, n_s, for deposition and immersion freezing of Snomax particles as a function of temperature, T.

HEMATITE AND THE EEFECT OF SURFACE MILLING

The influence of surface modification of hematite on the ice nucleation efficiency and freezing mode has been diagnosed as a function of degree of milling and AIDA temperature (<-30 ˚C). Milled hematite is generated by mechanically milling 800 nm diameter cubic hematite with 100 μm diameter bronze beads for more than 6 hours (**FIGURE 3**). An isometric experiment is conducted on un-milled cubic hematite subset. We observe two trends as expansion conditions are varied. First, as opposed to the speculation in *Möhler et al*. [7], the surface modification does not substantially enhance the overall ability of ice nucleation, inferred by the INAS density. Second, early deposition mode ice nucleation before the spontaneous formation of droplets is routinely observed for the milled subset. Thus, the important conclusion is that the surface milling may alter nucleation mode, presumably bolstering size independent freezing. Conclusions from AIDA experiments are still ambiguous, and complementary analysis with an Electro-Dynamic Balance is briefly discussed.

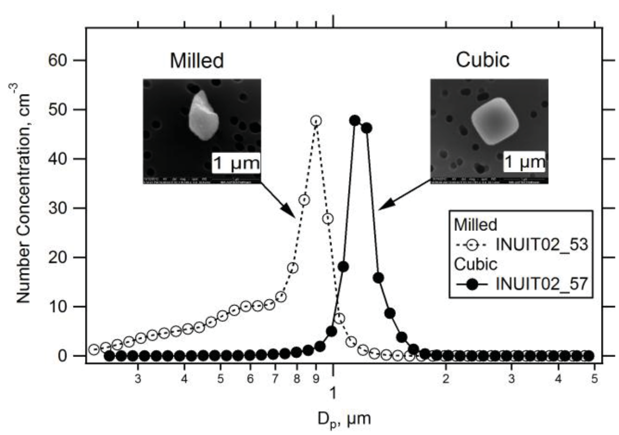

FIGURE 3. Size distributions of cubic and milled hematite.

ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support provided by the Deutsche Forschungsgemeinshaft under the DFG research unit 1525 INUIT. The work of D. J. Cziczo and O. Stetzer was supported by EUROCHAMP2 under the Transnational Access Activity E2-2012-05-14-0075. K. Höhler acknowledges the HALO priority program 1294 under the DFG contract number MOEH 668/1-2. The expertise of G. Kulkarni for the PCVI work is gratefully acknowledged.

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