Atmos. Chem. Phys. Discuss., 6, S203–S209, 2006
www.atmos-chem-phys.org/acpd/6/S203/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Efficiency of the deposition mode ice nucleation on mineral dust particles" *by* O. Möhler et al.

P. DeMott (Referee)

pdemott@lamar.ColoState.EDU

Received and published: 9 March 2006

General comments

This is a worthwhile contribution to ACP regarding ice nucleation by mineral dust particles in the regime of deposition nucleation at lower temperatures relevant primarily to cirrus clouds. Results are provided for a manufactured mineral dust that is being widely used as a surrogate for mineral dust ice nucleation studies and calibrations as well as two natural mineral dust samples. I would liked to have seen a comprehensive presentation of data on these particles across the full tropospheric temperature regime in a single paper rather than this one, the paper by Field et al. (2006) and one yet to appear. As the paper stands, the results span a narrow definition of cirrus, those below about

-50 °C, and perhaps this should be stated somewhere at the front of the paper. Nevertheless, this compilation of data provides an interesting contrast between the natural and manufactured dusts. Second, a number of excellent new analyses methods are introduced for experimental reports from the AIDA aerosol chamber. This highlights the growing capabilities of this unique facility and provides important information for evaluating all studies conducted in the chamber. Finally, the presentation of a suggestion that deposition nucleation data may not be treated in the same stochastic sense as homogeneous freezing is a worthwhile hypothesis for future exploration by those involved in such research. This point is important not only for model parameterization of ice nucleation processes, but for the appropriateness (or inappropriateness) of applying classical methods of nucleation in more explicit cloud models. My major critical comment concerns the strength of this assertion, which is primarily supported at present by the steadiness of the ice concentration signal during continued expansion of air following the achievement of peak ice saturation ratio in chamber simulations. More explicit testing of this observation by variations of cooling rate for all aerosol types as well as further process modeling is desirable. I have only a few more specific comments, suggestions and questions for clarification purposes.

Specific comments

1) Abstract: I find a few key points missing from the statements in the abstract.

a. The potential significance of the observation that repeated ice nucleation cycles did not lead to an increase in ice nucleation efficiency or alternately, to the lowering of the ice relative humidity for significant activation is not obvious and should be stated. It suggests to me that the phenomenon of preactivation noted by Roberts and Hallett (1968) does not appear valid for Arizona Test Dust. This is not really discussed in the paper. It may not be such a significant point, since the nature of the manufacture of this dust would imply the possibility of important differences (lack of aging or weathering) compared to natural dust particles. Thus, it might have been more interesting to have seen results for repeated cycles using the natural desert particles. Nevertheless, I

ACPD
6, S203–S209, 2006

assume there is a reason to highlight this result and it should be stated in summary form here and in detail within the paper, along with a careful description of the conditions to which particles were exposed during the cycling.

b. I suggest adding some words to note that the singular relation of ice activated fraction to ice supersaturation is valid "if it is assumed that time is only a secondary effect for deposition nucleation by these particles, which the data also support." I find this critically important as a point to be made to theoreticians and the numerical modeling community. I believe there may be a host of other recent data from various laboratories that may support this notion.

2) Introduction, page 1541:

a. I submit that Ström et al. (2003) did not measure ice nucleation, nor did that study measure ice nuclei, but rather they observed the result of nucleation interacting with cloud dynamics. Therefore, they did not show that cirrus clouds "are" nucleated by heterogeneous processes, but their data strongly suggest that this is the case. This may be a matter of opinion or semantics.

b. In a similar vein, I believe that Jensen et al. (1998) and Field et al. (2001) constrained the potential role of ice nuclei in cold wave cloud ice formation only through model sensitivity studies. DeMott et al. (1998), referenced elsewhere in the paper, did use measurements to constrain the role of heterogeneous ice nucleation in simulating the clouds considered by Jensen et al. (1998). As a second point on this issue, one should be careful about blanket statements regarding wave clouds as a class of clouds, since these span a range of temperature conditions in the atmosphere that sometimes include those where only heterogeneous ice nucleation is thought to be possible for ice initiation. That is, wave clouds may be present that are entirely warmer than -35 °C.

c. In regard to the reference to DeMott et al. (1998) later in this section, I must correct that we did not use the effective freezing temperature concept in the expression of the classical nucleation rate of pure water to calculate heterogeneous freezing rates. We

ACPD
6, S203–S209, 2006

EGU

did so for homogeneous freezing rate calculations in the parcel model simulations. For heterogeneous freezing at temperatures below the homogeneous freezing threshold, we assumed instantaneous nucleation of an observed freezing nuclei spectrum as given in Equation (1) in that paper, but substituted for temperature the effective freezing temperature of haze droplets. This was to account for the effects of associated solute composition (same effects accounted for in a water activity based parameterization) on heterogeneous freezing of mixed particles. Nevertheless, it is distinct from Kärcher and Lohmann (2003) because it assumes instant freezing as promoted in the present paper, not a freezing rate function.

3) Page 1546:

a. Line 10: The procedure used to convert the APS diameters to geometric diameters should be stated. For example, was bulk density of the mineral particle samples estimated and used?

b. Lines 14-16: It is not clear if a multiple charging correction for particles in the 200 nm size range has actually been made. Has it? Do you think it is a good correction or that some uncertainty remains unresolved?

c. Lines 23-25: Is it necessarily true that the structural morphology of particles implies that one will always overestimate surface area when assuming spheres? I rather think that the use of a combination of mobility and aerodynamic methods for measuring such particles creates a nebulous definition of spherical diameter that may lead to an underprediction or over-prediction of actual surface area.

4) Page 1548: Lines 1-3: Is it worth pointing out that the statement that transmission efficiency is equal to 1 for particles below 2 microns is only valid since your aerosol distributions do not include particles at the range of smaller sizes that would more effectively diffuse to the walls of the sample tube?

5) Page 1549:

ACPD

6, S203–S209, 2006

a. Lines 4-7: I do not see the conclusion of this statement evident in Figure 5. It looks like there are significant numbers of particles above 500 nm in the aerosol distributions, especially if small fractions of ice activated particles are measured and may come from this part of the aerosol size distribution.

b. Equation (2): Have you checked the water uptake properties of the dust particles to know if the simplification evident in this equation is valid? That is, have water uptake measurements been made or what is the basis of the statement that such dusts are not expected to take up water?

c. Lines 20-23: Is the growth of crystals to visible size the only delay to consider in the appearance of ice particles in the OPC spectra? There is another delay associated with the sedimentation time of crystals to the detector location, which may also depend on the constancy of conditions throughout the large chamber volume. I assume that such consideration of sedimentation is not necessary in the AIDA studies because the volume is mixed? This would be an important thing to emphasize to readers as regards ice crystal detection. Could the mixing impact diffusional growth times?

6) Page 1550, lines 23-25: The statements here led me to wonder if any "pulse" nucleation experiments, akin to those used by DeMott and Rogers (1990) to deconvolve the ice nucleation signal in their cloud chamber studies, have ever been done in the AIDA chamber. Are ice concentrations and conditions instantly sensed the same throughout the chamber or do some things like ice concentrations vary in time at one location, particularly at the bottom measurement position where sedimentation could play a role? This is similar to point 5c above. Have RH variations in space been examined as well?

7) Page 1553, line 1: Can you explain the meaning of the term "resonant vibrations"?

8) Page 1554: This comment concerns one of the major conclusions of the paper. The paragraph that transitions into this page ends by stating the conclusion that ice formation by deposition on dust particles depends only on ice saturation ratio and not on cooling rate. Cooling rate does not appear explicitly in Table 2 (rather dS_i/dt is

ACPD

6, S203–S209, 2006

listed) and it was not immediately apparent to me that this conclusion (independence of ice formation on cooling rate) was only explicitly tested with the manufactured ATD particles and then only in two sets of three subsequent expansions on the same initial aerosol. This conclusion deserves more direct study in the future, in duplicate experiments for fresh samples of natural dust particles and with varied cooling rates. The other supporting point made for dependence of ice formation only on supersaturation is that the concentration of ice crystals remains constant once S_{max} is achieved. It is not obvious to me that a constant ice concentration means that no additional ice crystals are forming during continued expansion in a large chamber. I think it may be necessary to repeat the fact that the chamber is continually mixed and that other factors related to the partitioning of water to ice crystals may be at play (e.g., ice crystal growth rates are slow). Otherwise, one might think that crystals should be falling out of the volume, in which case a constant concentration would imply continuing ice formation. The case for the absence of continued nucleation may ultimately be stronger once process modeling studies are conducted. This might also solve the issue of crystal growth time delays mentioned later for experiments at the lowest temperatures.

9) Pages 1557-1558: The separation of data between this paper and that of Field et al. makes it a little difficult to follow this discussion. The nature of the parameterization of deposition nucleation data (only valid below $S_i = 1.35$) is somewhat discouraging as regards using these data in models. Can the authors comment on the need for additional studies and the feasibility of providing modelers a seamless set of parameterizations to describe ice formation by mineral dust particles? For example, the parameterization encapsulated in equation (5) requires capping ice formation fraction at a value of 1 and also limiting it to $S_i < 1.35$. Other parameterizations will be required to extend these data to other valid ranges of S_i, even just for cirrus clouds.

10) Page 1559: The difficulty in resolving aerosol particle size effects suggests to me that future studies could utilize either the ability to examine finer size cuts of particles or examination of the residual particles from nucleated crystals, two capabilities that

ACPD

6, S203–S209, 2006

seem feasible in the AIDA chamber.

Technical corrections

The word "were" is spelled as "where" on lines 21-22 of page 1550 and line 29 on page 1554.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1539, 2006.