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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models" *by* J. A. Curry and V. I. Khvorostyanov

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General Comment

I cannot recommend this manuscript for publication.

The authors have invested much effort during the last decade into the development of theoretical descriptions of ice nucleation and other processes. Theory is appealing and useful toward conceptualizing various aspects of ice nucleation, and potentially, in quantifying it. The nature of homogeneous freezing appears to allow for quantitative application of theory to a population of particles acting first as CCN. Such predictions are testable versus collected data on freezing in the laboratory. Extension then to use

in cloud models would seem reasonable. Heterogeneous ice nucleation introduces a variety of dependencies on particle surface (and chemical) properties that complicate conceptualization and quantification using theoretical calculations. The synergy needed between experimental and theoretical studies in order to achieve quantitative predictions is much stronger. It remains to be seen if a theoretical model can quantitatively represent atmospheric populations of ice nuclei and their variability. Some seek useful approximations to directly represent observations via parameterizations. Parameterizations can also have issues and require careful validation and/or statement of limitations.

The present paper demonstrates a lack of effort in consideration of the consistency or inconsistency of the Khvorostyanov and Curry (hereafter KC) modified classical theory with existing heterogeneous ice nucleation data or ice crystal data that clearly identifies ice nucleation. In attempting to meet criticisms of previous applications of the theory, my opinion is that they succeed only in highlighting the very issues that led others to their conclusions regarding potential pitfalls of applying the theory in cloud model simulations. Namely, through application of an ice nucleation model that has not been validated in the proper manner versus the types of observations that matter, comfort continues to be taken in the apparent correct simulation of certain clouds, even though this occurs largely due to an artifact of application. A path toward meaningful validation and application that includes consideration of ice nucleation observations is ignored and only mentioned in the Conclusion section of the paper. Thus, the key flaw of this paper in my opinion is the failure to acknowledge the following: the acid test for a theoretical model is by comparison to the specific observations it intends to duplicate. A nucleation parameterization is not validated by simulating cloud systems or comparing to bulk cloud phase distributions in the atmosphere that reflect the net effects of cloud dynamics, microphysical feedbacks, redistribution of the ice phase, secondary ice generation and so forth. These are exercises that can be more confidently done after parameterization validation to gauge the impact of a scheme in the context of all the other the processes at play. Only then can the true state of knowledge regarding cloud

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and precipitation processes be evaluated and inconsistencies addressed via further targeted study. This was a basic point made by both Phillips et al. (2008), hereafter PDA08, and Eidhammer et al. (2009), hereafter EDK09.

An apparent tenet put forth here that I had not previously imagined is that use of classical theory, with some selection of parameters (not known a priori), is to be trusted as ground truth for atmospheric ice nucleation. While important challenges remain for ice nuclei measurements, this presumption about the utility of theory as presently formulated faces overwhelming contrary evidence from laboratory cloud chamber studies and existing ice nuclei instrument measurements in both the laboratory and field indicating that ice nuclei represent a strongly limited population, such that it is not correct to apply the gross assumption that ice nucleation activity can be predicted using simple assumptions applied to the entire aerosol size distribution as the population of relevance. While this may be a potentially elegant approach if enough differentiation on the appropriate distribution of relevant surface properties amongst particles is known and specified, the calculations presently appear wrong in their assumptions based on any experimental assessment. To follow through with these assumptions as a manner of critiquing the measurements and parameterizations thereof is fruitless. Judging validity based on cloud model simulations alone is an egregious abuse of the theory in my opinion.

If the authors wish to move forward with a form of this paper toward a useful point, such as how to further develop the theoretical model for realism and stringently test it, the paper should be shortened by at least half. Removal of comparisons that are irrelevant as validations and removal of the MPACE simulations are a few means for doing that. The MPACE simulations give a false notion about the ability/inability to simulate these clouds. If the authors apply more realistic descriptions of particle properties responsible for ice nucleation via the mechanism targeted, they will likely find that their model has the same problem that all others do in simulating long-lived Arctic clouds. Why would it not? The ice nuclei measurements should be sensitive to the

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mechanism that the theoretical model seeks to describe. Useful additions to the paper would be comparisons versus laboratory ice nucleation data and simulations of recent studies targeting cloud types that offer the opportunity to isolate the ice nucleation signal. My specific comments mostly repeat and expound on a number of these points below. While I regret the length of this comment, this paper bears a strong response because previous papers including cloud model simulations have not been met with the appropriate level of constructive criticism.

Specific Comments

To facilitate author response, I order these comments by section, and italicize these headers.

Abstract

a. Line 9-13: Using theory to provide restrictions on empirical schemes is an interesting concept, but a strange one if the schemes are actually based on data collected in the "forbidden" regime. It is fine if the point is that the schemes should not be used outside of their valid range, but the abstract is not clear on the fact that the thermodynamic restrictions are for a specific ice formation mechanism as quantified by theory not yet validated using specific ice nucleation data.

b. Lines 14-16: Clouds sometimes remaining mostly liquid to as low as -35°C are facts based on documented cloud observations (see references later). The parameterizations reflect actual ice nucleation data. It is flippant to ignore these facts. The comparison of the KC scheme to field campaign data is not to ice nucleation data, but via simulation of all cloud processes. This is not a valid approach for evaluating the scheme itself.

c. Lines 17-18: Not applying the KC parameterization to the entire aerosol distribution is an approach that corrects for invalid assumptions in the simplified model, not a misapplication. A parameterization of ice initiation should only act on the relevant

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source nuclei population, by way of explicitly (e.g., some particles have no active sites) or implicitly limiting this population.

d. Lines 20-23: This statement should be clear on which previous study is being referred to. It misrepresents what was done by EDK09, who restricted ice nucleation to the size range and compositions from which natural ice nuclei are known to come. The statement also treads from a level of lack of proper consideration of experimental data in Khvorostyanov and Curry (2004, or KC04) to near disdain in the comment regarding ice crystals exiting an experimental device.

1. Introduction

a. Page 2671, lines 11-15: KC have created a framework for describing ice formation (potentially in all of its dependencies) in clouds from a theoretical basis. The improvements mentioned here are needed before any further application is made to simulate cloud systems (see EDK09).

b. Page 2673, lines 3-8: It was not appropriate for this parameterization to appear published in an open access journal prior to my ability to first publish and describe it. The paper is in review. The coefficients listed here are incorrect in their final form. Had the nature and philosophy of the parameterization been known and discussed here, it would have been clear that the "DM10" parameterization is already hardwired for failure in the assessment it is used for in the present manuscript, as it is to be applied for global models only at the far right axis of Fig. 1 on the basis that under many circumstances the left side is mostly irrelevant. Justification for this is outside of the subject of this review. I recommend removing DM10, and if the figure is retained, comparing instead to PDA08, which does seek to resolve all ice nucleation in the phase space shown.

c. Page 2675, equation 8: The problem with this equation is that not all CCN and not all aerosols are potential ice nuclei. Use of such an equation necessitates differentiation of (and summation over) the variety of specific physical and chemical properties of particles at each size and over the distribution. It should not be applied absent such

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knowledge or without qualification.

d. Page 2675, lines 16-19: To continue with the previous thought, the statements here describe what the approach is capable of in theory. In practice, observations (e.g., Marcolli et al. 2007) indicate that even similar sized and chemically-similar particles possess a spectrum of ice nucleation abilities. The problem as I see it is that without such specific information, the application of the theory can lead to predictions that are grossly in error in terms of when a certain fraction of the aerosol population freezes.

2. Thermodynamic constraints on heterogeneous ice nucleation schemes

a. As noted above, this section could be fine as a hypothesis for the existence of thermodynamically-restricted regimes, but this should serve as the basis/motivation for experimental evaluation and refinement of theory. At present it can only show differences between the theoretical and empirical predictions, not determine which is correct.

3. Evaluation of phase state simulations

The following comments lead me to suggest that this entire section is invalid and needs to be removed.

a. Page 2679, lines 18-26: It is not clear why it is important to note the similarity between the DW04 and KC schemes, a point made by EDK09 to show that both have some undesirable features due to idealization of the ice nucleation behavior of a population of particles.

b. Page 2680, discussion of parcel model simulations: This section repeats what we already know from EDK09, although the authors misinterpret the meaning. The simulation performed by EDK09 using the PDA08 ice nucleation scheme was an idealized parcel simulation simply intended to cover a wide temperature range. There was no intent to mimic a specific cloud case, just specification of a steady updraft for an unrealistically long period. The simplest comparison was sought to make the simplest

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point – showing what the different nucleation models do in any circumstances that do or do not necessarily involve strong feedbacks from ice crystal growth on overall cloud phase. The authors have done much to elucidate the latter topic, but it is irrelevant as regards testing the ice nucleation scheme. Do the authors know circumstances where a 50 cm/s updraft persists for 4 hours for which they should be judging the phase transitions that occur in the clouds without any other mixing, secondary ice formation, and so forth? Yet, the simple issue and metric should be ice nucleation, period, not what we think we need to reproduce based on cloud case studies.

c. Page 2681, more discussion of the parcel model simulations: "...the "constraints" imposed in the PDA08 scheme lead to a substantial underestimation of heterogeneous ice nucleation." What is the basis for this conclusion? The PDA08 scheme attempts to represent present measurements of ice nuclei. Fig. 3 thus reiterates that the theoretical parameterization as constrained by the authors overestimates ice nucleation by two orders of magnitude. Number concentrations of 1000 L^{-1} nucleated do not represent a realistic cloud phase state due to known heterogeneous ice nucleation mechanism in all but rare circumstances in the atmosphere (e.g., high dust concentrations). Such values are not corroborated by observations in cases that can solely be attributed to heterogeneous ice nucleation, and it is grossly misleading to continue to perpetuate this notion.

d. If the authors believe that present measurements greatly underestimate ice nuclei number concentrations active by the mechanisms they purport to describe theoretically, they need to provide hard evidence as to why measurements are in error. Further, they might like to explain:

1) Low ice crystal number concentrations measured in laminar flow orographic wave cloud scenarios for vertical motions well above 1 m s^{-1} and to temperatures below -30°C (Heymsfield and Milosevich, 1993; Eidhammer et al. 2010). I will happily provide an advance press copy of the latter paper.

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2) The general agreement of average ice nuclei number concentrations as measured (EDK09) with ice crystal concentrations found in clouds when ice initiation is presumably isolated (Cooper, 1986), a reference the authors oddly twist to corroborate their model in Section 4.

3) The presence of liquid water to low temperatures in local regions of cumulus clouds (e.g., Rosenfeld and Woodley, 2000; Fridlind et al. 2004).

4) Ice nucleation results from laboratory studies, such as shown in Figures 1 and 2. Some other examples are present in the published literature, but I would like to offer a concrete example for the authors use that includes recent data from two types of measurements and is also open about issues the measurement community has to grapple with in interpreting ice nucleation data. First, some discussion of KC model predictions should preface discussion of these observations. One may note in Figures 5 and 6 of KC04, that given an unlimited vapor supply, deliquescence-freezing ice nucleation rates would result in complete ice phase transition for all CCN over a few % RH transition and no liquid water formation would be possible under any circumstance below about -20°C. These calculations are for nuclei of radii 0.46 μm, but as Fig. 9 of KC04 shows, very little size dependence is predicted. I have not attempted to update these calculations for the temperature dependent active site parameter used in some simulations in the present manuscript. This only changes the RH range over which the entire population of particles would be predicted to freeze heterogeneously, not the final number. Figure 3 and EDK09 should make it clear, already, that such predictions do not reflect present ice nucleation observations.

In Figure 1 are shown data from an ice nucleation experiment in the Aerosol Interactions and Dynamics in the Atmosphere (AIDA) cloud chamber using a polydisperse size distribution of a surface-collected sample of dust from the Saharan desert. These data were collected during an international ice nucleation workshop in 2007 (Möhler et al. 2008), which utilized several mineral dust and bacterial ice nuclei sources. Figure 1 shows a timeline of temperature, saturation ratios from a tunable diode laser hygrom-

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eter, total aerosol (condensation nuclei or CN) concentration, and liquid droplet, and ice crystal number concentrations inferred from optical particle counter measurements (Möhler et al. 2006). Note that the ice concentration measurements are on a scale that differs by two orders of magnitude compared to the aerosol and cloud droplet scale. Also note that CN concentrations during the liquid cloud stages are not interstitial values but reflect inefficient sampling and evaporation of cloud particles by the aerosol collection system. There were two cloud experiments performed by evacuating the chamber after particles had been introduced to AIDA. Total dust number concentrations were 200 cm^{-3} initially, for a distribution described with lognormal parameters (dg = 0.18 μm, sg = 1.8). The first cloud formation had a warmer base temperature around -20°C, activating nearly all of the mineral dust particles into cloud droplets (no other CCN were provided) and producing very little resolvable ice (>100 per liter for the OPC method). Note that the peak humidity at the cloud formation point is not resolved and may easily have exceeded a few % for the CCN concentrations and expansion rates used. As the humidity decreased toward ice saturation following suspension of the expansion, a new expansion was initiated triggering cloud formation around -22°C and continued cooling to -25°C. Note that all particles present prior to this second expansion were also activated into droplets. Higher concentrations of ice crystals formed in the second expansion (up to 1 cm^{-3}), but notably, the mixed phase cloud persisted for more than a few hundred seconds. The maximum sustained cooling rate during the expansions was about 2°C min^{-1}, equating to a vertical motion of around 5 m s^{-1}. Also notable is the fact that less than about 0.5% of all particles activated ice formation for this equivalent vertical motion. Experiments like this at a variety of temperatures are highly suited for validating nucleation models. I encourage the authors of this paper to consider if results such as these are consistent with predictions of the present assumptions of their nucleation scheme. I think they are not and now offer a simple analysis that also indicates how some portable ice nuclei instruments compared to the cloud expansions during the workshop.

Figure 2 shows how the ice nucleating fraction of all particles was measured by the

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Colorado State University aircraft CFDC as the relative humidity was slowly scanned (1% RH per minute) from low to high values during sampling from the large AIDA chamber volume prior to the start of the cloud expansion shown in Fig. 1. This requires some explanation, in advance of a more considered publication, of a relatively consistent feature of CFDC-type measurements of certain ice nuclei that has practical implications for everyday IN measurements and requires further research. Similar to the second AIDA expansion result, water saturation is required in the CFDC prior to the onset of significant ice nucleation. Near equivalence of the RH conditions for activating a fraction 0.001 of all particles was seen between the two measurements over about a 20°C range of temperatures and for RH as low as 70% (DeMott et al. 2008). Nevertheless, higher supersaturation is required in the CFDC to activate the maximum fractions of dust particles as detected via immersion freezing in AIDA. It should be understood that to give a result for comparison to the AIDA cloud conditions, supersaturation must rise to the level that all particles should be immersed in drops on adjustment to CFDC temperature and humidity conditions. This is likely the case by about 101% RH. Yet, ice nuclei concentration increases strongly to 104%, where a weaker increase is then noted toward higher RH. While first noting that RH uncertainty in CFDC measurements is 3% (DeMott et al. 2009), we presently hypothesize that this behavior may additionally reflect a surface chemistry impact on ice nucleation – faster dilution of surface impurities caused by larger droplet growth at higher supersaturation facilitates ice nucleation on all sizes of dust up to 1.5 μm. This feature was observed for all CFDC instruments measuring mineral dusts during the workshop and is also noted in early CFDC field measurements of natural ice nuclei (DeMott et al. 1998). While not shown here, I can report that such a feature is not present for sampling bacterial ice nuclei, which possess more unique active site features and likely do not have surface chemical factors to impact CCN activation or ice nucleation. The behavior is not indicative of any classical time dependence of nucleation, as CFDC growth times were approximately constant. Aside from such details, the results in Fig. 2 show that the maximum ice nuclei concentration measured in the regime favoring condensa-

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tion/immersion freezing in the CFDC is the same as measured in the AIDA expansions. This was a typical experiment, neither the best nor worst comparison between results from AIDA expansions and ice nuclei measuring instruments at the workshop. If higher concentrations/fractions of particles could activate as ice nuclei at this temperature, a CFDC, by design of providing replenished vapor to maintain RH during strong ice growth (Rogers, 1988), would measure concentrations up to 100's or more per cm^{-3} (DeMott et al. 2009; Richardson et al. 2010). Thus, the results from AIDA and the CFDC support that ice nuclei contain a strongly heterogeneous set of surface features responsible for ice nucleation with the consequence that only a small fraction of dust particles are active depending on temperature.

As a quick check on what the KC parameterization might predict in this case, I pulled approximate values of heterogeneous nucleation rates off of the figures in KC04 (active site parameter α = 0) and applied these at specific RH values for a CFDC steady-RH regime residence time of about 5 seconds in this case. Predicted ice nucleation occurs rapidly well below water saturation and all particles activate to ice crystals within a narrow range of RH, as shown by the dashed curve in the figure. Again, no consideration is given to the Bergeron-Findeisen process in this simple calculation, but little consideration should be given on the basis of AIDA cloud phase results. The focus is on the fact that ice nucleation is incorrectly predicted. The phase transition is too strong and is predicted at too low RH (and at too warm temperatures).

e. Page 2681, paragraph starting line 18 and continuing through the next page: The comparison of idealized parcel model ice mass fraction versus climatological values measured in clouds or prescribed in global models has potentially nothing at all to do with nucleation on measurable heterogeneous ice nuclei. This is an "apples versus oranges" comparison that is both misleading and dangerous. It lets the entire community of cloud scientists off the hook by falsely promoting that we have a complete understanding of all ice physical processes in clouds based on the parcel model behavior of a theoretical ice nucleation scheme. And it encourages continued application

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of a static cloud phase schemes in global models, something that many scientists are working hard to replace. For example, strangely, the authors later in the paper highlight the "Morrison microphysics scheme" for CAM3, a purposeful attempt to replace the temperature-dependent ice mass fraction they promote as validation here.

f. Page 2682, lines 21-23: I believe that the prediction of homogeneous freezing influence beginning at -34.5°C in EDK09 is fully expected for the onset of action of homogeneous freezing in up to 40 micron drops grown in the adiabatic parcel over the long times associated with the slow cooling rates in this highly idealized simulation.

g. Page 2682, line 25, on through to the end of section 3: This is another instance of turning an EDK09 argument around inappropriately. The high values of the DW parameterization are no more realistic than the high predicted values of KC04. None of the comparisons shown here give reason to reconsider those conclusions. See above.

4. Assessment of parameterized ice particle concentrations

Having made my key comments and attempting now to be briefer, this section has a similar issue to the previous section and should be removed or made the topic of a future paper once the ice nucleation parameterization has been reassessed properly. The comparisons made are not appropriate for validating ice nucleation parameterizations. The simulations include the impacts of microphysical feedbacks, which should not enter the direct evaluation of quantification of ice nucleation (EDK09). What has vertical motion to do with an ice nucleation measurement made at a set temperature and relative humidity? What vertical motion values are most applicable to Cooper's parameterization based on ice crystal concentrations measured in clouds? I have a suggestion – not the synoptic scale values plotted in the figures. Equation 13 encapsulates the effects of nucleation and negative feedbacks and so is not recommended for application in a global model until the nucleation scheme is modified properly. This would otherwise be an interesting suggestion for another paper. To compare it to ice

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nuclei data in Fig. 5 is another fairly preposterous exercise, especially in suggesting there is some range of vertical motion that equates to the IN measurements (cf., comment earlier in this paragraph). Curve KC in Fig. 5 as translated from PDA08 does represent one published form of the KC theory, as intended by those authors to show the ice nucleation prediction absent any simulated microphysical feedbacks on supersaturation that limits ice nucleation rate. Ice nucleation rate is not fundamentally limited in many experimental devices, so their data shows that the rates in the theoretical parameterization are incorrect. This section mainly explains why it is that the KC theory implemented in cloud models can artificially give an apparently correct simulation in some cases. There is no true prediction of ice nuclei, just a large reservoir for use as needed until the Bergeron-Findeisen process shuts down further ice nucleation. It was exactly a point of EDK09. Figure 6 is likewise of little meaning.

5. Simulations of Mixed-Phase Arctic Cloud Experiment (MPACE)

Following from comments to this point, I consider the exercise done here to be premature and, intending no disrespect, to be yet another instance of publishing cloud model simulations that give the false notion that the authors have solved the topic of ice formation in the atmosphere. Arctic clouds, surprisingly, continue to represent a strong challenge for correctly simulating cloud phase and ice distribution. This paper presents no new insights in this regard compared to referenced work already published; it only shows that if the ice nucleation model has ready ice nucleation at its disposal, without regard to the numbers of ice nuclei available via the mechanism prescribed (it is absolutely not the total CCN population), then it seems possible to simulate the clouds. It solves no mysteries and cannot be trusted until the nucleation scheme is validated. I have far too many comments on this section to list, so limit it to a few regarding issues mostly not touched upon to this point.

a."IN particles in the tail of the 2^{nd} aerosol mode with maximum surface area and potentially highest ice nucleability were excluded from CFDC measurements, while the concentration of large particles only 0.01 cm^{-3} = 10 L^{-1} would produce a significant

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effect. An additional explanation could be that the time of IN processing in the CFD chamber, 7–15 s (Rogers, 1982, 1988; PDA08), is much smaller than the timescale of heterogeneous ice nucleation of 15–240 min determined from models (e.g., Lin et al., 2002; KC05; EDK09)." Now these are issues worth discussing, and issues the measurement community is finely focused on at the moment. The first point, one also made by Santachiara et al. (2009), is a valid concern if all of the 10 L^{-1} are active as ice nuclei. This seems unlikely in this case based upon the best measurements presently published regarding the similarly limited fractions of large aerosols active as ice nuclei (Berezinsky and Stepanov, 1986). It is a more general concern to be examined for other aerosol distributions. Strong time dependence of heterogeneous ice nucleation remains to be explored experimentally, but is not consistent with present evidence. Vali (2008) provides an excellent discussion of this subject and indicates that, to a first approximation, the specific character of ice nuclei immediately active at any temperature dominates over the duration of supercooling in heterogeneous freezing nucleation by immersed impurities.

b. ". . .concentration Nc,ch at the exit of the chamber, but not as the concentration Na of original aerosol particles at the entrance of the chamber that may potentially become ice crystals." I will state this once again: based on the preponderance of evidence, the potential number that may become ice crystals at any temperature is not the total aerosol number.

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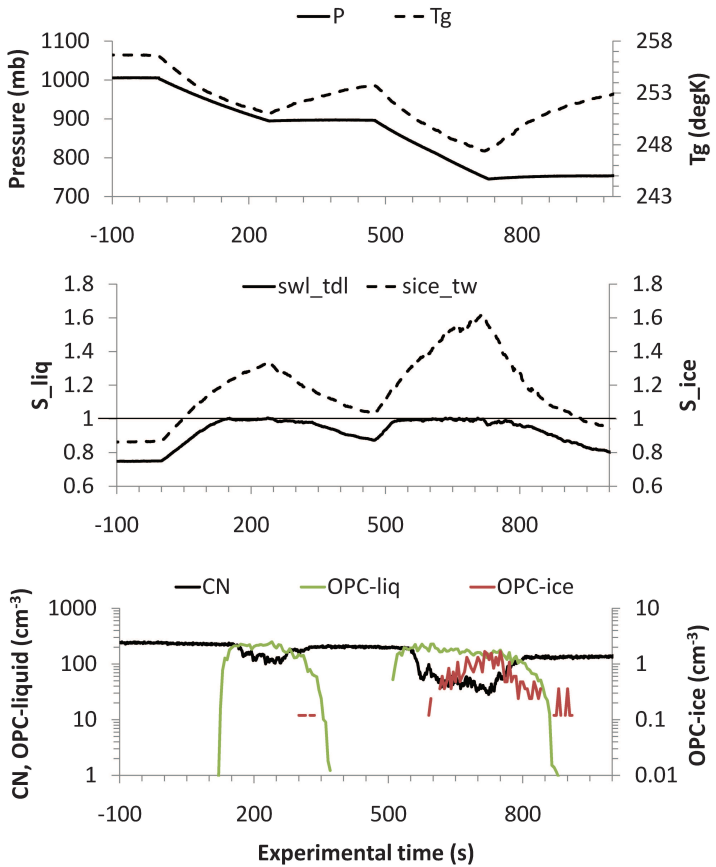

Fig. 1. AIDA experiment for Saharan dust particles. P, T, ice and liquid saturation ratios, and aerosol and cloud liquid (OPC-liq) and ice (OPC-ice) number concentration data courtesy of O. Möhler.

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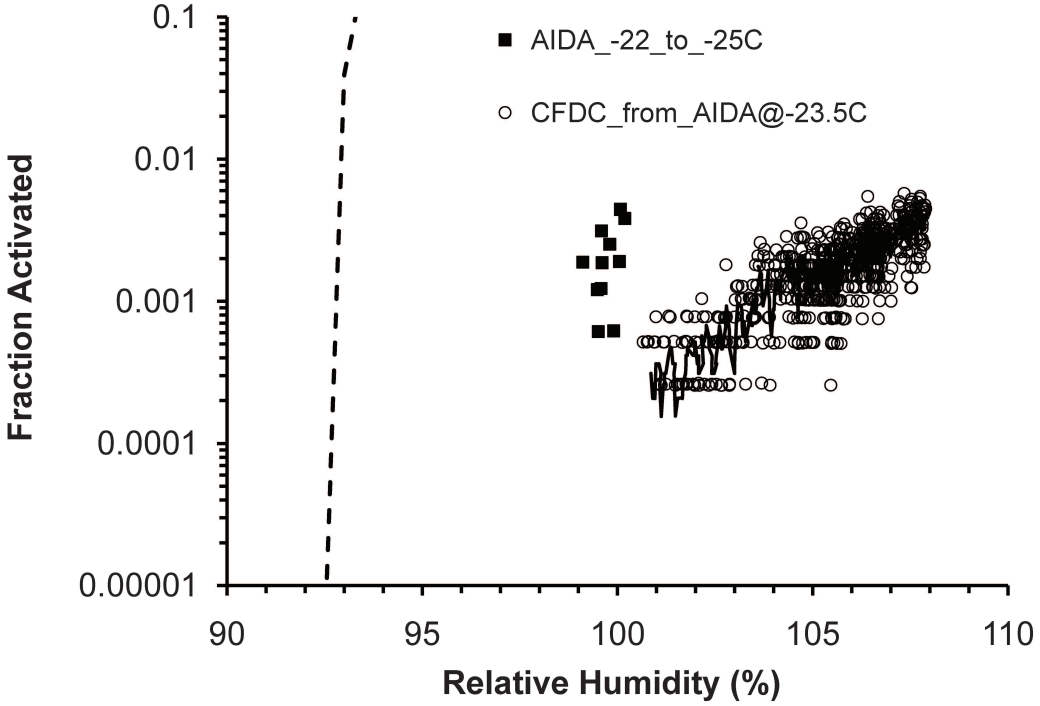

Fig. 2. Ice active fractions from AIDA (0.2 Hz) and the CFDC (1 Hz and 5 s running mean) for the second cloud experiment shown in Fig. 1. The dashed curve is a KC model prediction (see text).

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