

Response to Reviewer 1 (Reviewer comments in bold text)

This paper presents an instrument inter-comparison study of the investigation of heterogeneous ice nucleation for temperatures as low as 230 K. The following types of potential ice nuclei (IN) were employed for instrument evaluation: Arizona test dust (ATD), desert mineral dust (Saharan, Canary Island, Israeli), graphite soot particles, and live and dead bacterial cells. The ice nucleation experiments were performed at the AIDA (Aerosol Interactions and Dynamics in the Atmosphere) facility at Karlsruhe Institute of Technology (Institute for Meteorology and Climate Research). Ice nucleation onsets with respect to aerosol temperature and relative humidity (RH) were measured by the University of Toronto continuous flow diffusion chamber (UT-CFDC), CSU (Colorado State University) CFDC, and AIDA expansion chamber. At higher temperatures good agreement was found between the different instruments, at lower temperatures significant differences in ice nucleation onsets were obtained by the various instruments. Possible reasons for these findings are discussed.

Overall this is a well written paper and fits very nicely within the scope of Atmospheric Chemistry and Physics. Atmospheric ice nucleation is one of the least understood topics in atmospheric sciences and this instrument inter-comparison study is an important step to improve our understanding of how aerosol particles affect ice nucleation. The atmospheric science community will greatly appreciate the discussion of the various instrument performances.

I suggest this manuscript for publication after the authors have addressed the points given below.

We thank reviewer 1 for the helpful comments and suggestions. Below are detailed responses and references to locations in the manuscript where changes have been incorporated.

**The authors discuss their results in terms of deposition and condensation mode ice nucleation although none of the instruments can directly discriminate between these modes. Immersion mode freezing is left out entirely. The authors assume that activation below water saturation proceeded by deposition nucleation and activation at or above water saturation proceeded by condensation freezing. Is such a categorization valid?**

We acknowledge that we have referred to freezing above water saturation as condensation freezing and not immersion freezing. This depends on whether one views the freezing of a particle immersed in a water droplet formed by condensation to be different than the freezing of an immersed particle being cooled. We refer to immersion mode freezing when the liquid droplet activation of the particle has taken place at a different temperature (usually a warmer temperature) than the temperature of freezing. For condensation freezing the water activation followed by quick freezing are both taking place at the same temperature and within the time scale of the particles in the chamber (7-10 seconds). We explain this more explicitly on *page 13, para. 2, line 2*.

We also acknowledge that a categorisation such as that alluded to by the reviewer above is in fact an assumption and that there are no instruments that can define the activation mechanism below water-saturation. The instruments used in the current study therefore measure contributions from both deposition and condensation/immersion freezing modes and we define water saturation as the boundary of transition between the two based on equilibrium RH values with respect to water. However, one cannot eliminate the possibility of deposition mode proceeding concurrently with condensation freezing above water saturation. Similarly there could be some degree of water adsorption occurring at  $RH_w < 100\%$ . The onset of an additional ice nucleation mode (assumed to be condensation freezing) is usually inferred when a change in slope of an activation curve is observed such as that seen in Figure 1 below.

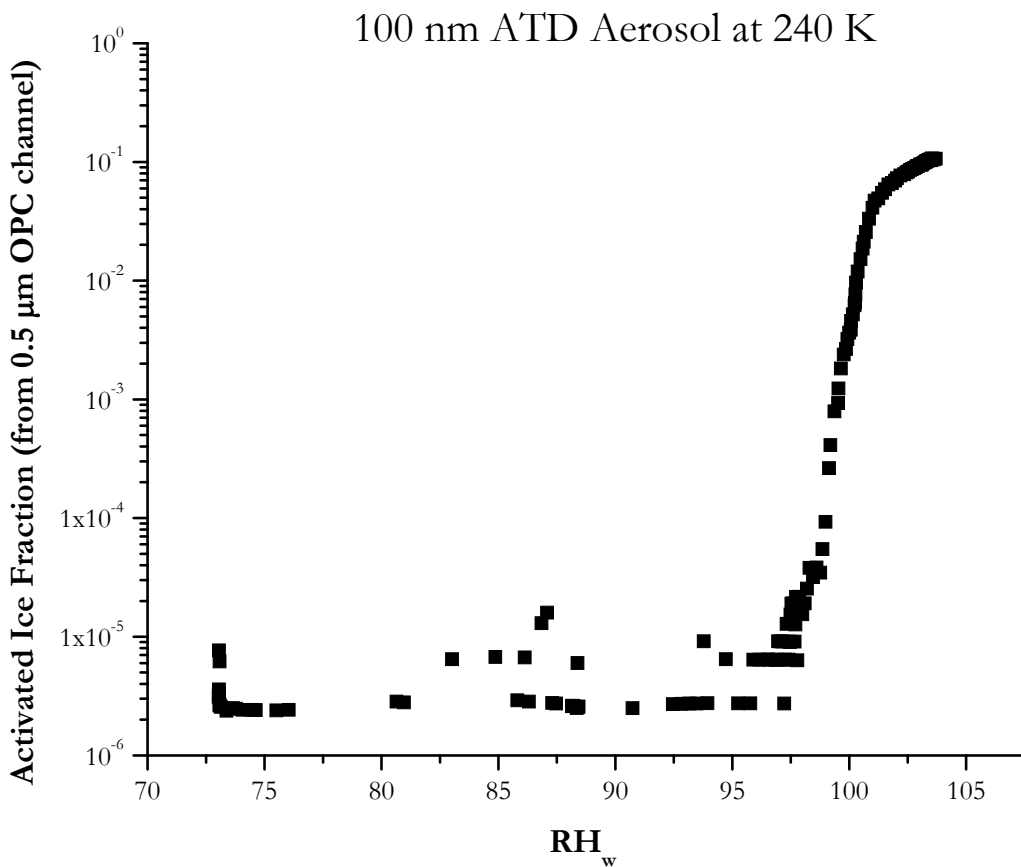


Figure 1. Ice activation curve showing a change of slope at  $RH \sim 100\%$  indicating the onset of an additional/different ice forming mechanism compared to that occurring at  $RH < 100\%$ .

**Discussion lines 1-8, page 20867: At temperatures above 263 K usually no deposition ice nucleation occurs. Looking at Fig. 2 and taking into account the experimental uncertainty it cannot be stated how Snomax and bacteria samples nucleated ice.**

We agree with the above comment. We have modified the paragraph in the revised manuscript on *page 15, para. 1, line 2* to be more explicit to state that at temperatures above 263 K due to uncertainty in RH we cannot state with certainty if the Snomax® and bacteria nucleated ice via deposition or condensation freezing and that deposition mode freezing is only relevant for the Snomax® data point at  $T = 247$  K.

**Snomax studied at 247 K may have nucleated ice via deposition mode. Also, Fig. 2 indicates that there is an ATD sample which nucleated ice at lower RH<sub>i</sub> than Snomax, so the statement “...which is more active than all of the dust particles sampled, forming ice at RH<sub>i</sub> that is 10% lower than that required for SD, ID and CID.” is in part not correct.**

The ATD sample which nucleates ice at lower RH<sub>i</sub> referred to in the above comment is at a different temperature than the Snomax® data point at 247 K. The efficiency (10% RH<sub>i</sub> lower than other dust samples) comparison is specific to  $T = 247$ K. We have now modified the sentence to be more specific on *page 15, para. 1, line 4* to read, “However, Snomax® activates ice well below water saturation, implying deposition freezing at  $T = 247$  K forming ice at RH<sub>i</sub> that is 10% lower than that required for SD, ID and CID at the *same temperature*.”

**Particle sampling seems to have a significant effect on the determination of ice nucleation onsets. All instrument supply lines except the one for the CSU-CFDC are at room temperature. At which temperature was the supply line for the CSU-CFDC?**

Only the supply line from the AIDA chamber to the CSU instrument was maintained at a temperature close to that of the AIDA chamber (from 263 – 230 K) depending on the experimental day. The sampling line from the APC to the CSU chamber was also at room temperature like the sampling line for UT-CFDC.

**How long was the residence time for particles in the warm supply tubing for the AIDA and UT-CFDC instruments? Was the residence time sufficiently long and the temperature warm enough to avoid pre-activation effects?**

The residence time of the particles in the sampling lines depended on sampling flow rates for each instrument as well as the exact length of tubing from each chamber to each instrument but was on the order of 5 seconds. This time would be enough for the particles to equilibrate to room temperature and therefore lose any memory effect. In addition, repeat expansions on same aerosol from AIDA shown in Figure 3 for example, do not suggest positive pre-activation (lower onset RH<sub>i</sub> for ice activation), effects. UT-CFDC data for post-expansion sampling also suggest no positive pre-activation effects. For the cooled AIDA-CSU line, pre-activation could only be a factor for post-expansion experiments, however no pre-activation effects were observed and we have not presented these data from CSU since the focus of the current study is the UT-CFDC instrument.

**The trend in ice nucleation onsets of ATD with regard to sampling from APC and AIDA is opposite for UT- and CSU-CFDC for ice nucleation at around 230 K. Could you elaborate on this?**

The data points for the CSU-CFDC sampled from AIDA and APC sampling are both within uncertainty of the other's RH measurements and therefore not significantly different (*see page 16, para. 1, line 3*). This is clearer in the revised manuscript where we have now added error bars for the data points in Figure 3. The same is true for UT-CFDC APC data points for the AIDA pre-expansion data point (*see Figure 3 in revised manuscript*). Details of the general spread in reported RH<sub>i</sub> for the 5 data points at ~ 230 K for the UT-CFDC have already been addressed on *page 14, line 12, and page 16, para. 1*.

**How much time has passed between the ending of an expansion experiment and sampling of the particles? In other words, how large can the ice crystals grow within the AIDA chamber to justify the argument that sedimentation could be an issue. How quick is the sedimentation rate and does this result in a bias of sampled particles. E.g. Does the CSU-CFDC at the bottom of the AIDA chamber receive more “active” particles?**

The time between the end of an expansion and the beginning of sampling by CSU and UT CFDCs was on the order of 15-25 minutes as this time was needed to re-pressurise the AIDA chamber to ambient pressure. We are sure that there are no settling effects on the distribution of aerosol particles throughout the chamber prior to an AIDA expansion. The settling velocities of the largest aerosol particle (3 μm) are about 2 cm per minute, which roughly matches the internal mixing time scale of the particles in the AIDA chamber. Vertical gradients can be expected for particles which settle more than about 1m/min, which is the case for droplets and ice crystals with diameter larger than about 50 μm. Therefore we can expect efficient removal of ice nuclei with large ice crystals. But this would affect only measurements after the first expansion for CSU-CFDC which is not a factor for the current work since we report only CSU data taken prior to an AIDA expansion.

**Looking at Fig. 3 at around 233 K, the range of ice nucleation onsets for the first expansion spans from 70% to about 84% RH<sub>w</sub> not accounting for pre- and post sampling and expansion experiments. This corresponds to about 102% to 126% RH<sub>i</sub>. If the instrument uncertainties are included the range becomes even larger. This is a significant range in RH when discussing ice nucleation onsets. When comparing this to Fig. 7, it could be argued that the differences in ice nucleation onsets for graphite spark generated soot particles at approx. 231 K is about 16% RH<sub>w</sub> (without instrument uncertainty), only a little more compared to Fig. 3. If the interpretation of Figs. 3 and 7 is correct, then it should be concluded that not only for graphite spark generated soot particles but also for ATD at the lowest sampled temperatures significant differences in ice nucleation onset values were observed by the instruments.**

This is a valid point as now made in the paper (*see for example abstract, line 10, page 17, para. 1, line 1 and para. 2, line 1, and page 23, para. 2 (conclusions section)*). For the ATD results, if we only consider the 1<sup>st</sup> AIDA expansion results and APC sampling, the RH<sub>w</sub> range spans 70-80%. This spread is 6% less than that observed for GSG (soot). If uncertainties are included the range does indeed become larger, however the significance of the spread becomes smaller. For the data from UT-CFDC AIDA sampling, the higher onset may be influenced by sampling smaller sized particles due to the long sampling line (hence loss of larger particles) as discussed on *page 14, line 20*.

**The authors state in chapter 4 and in the conclusions section, that there was generally good agreement between the instruments. What does “generally good agreement” indicate? When referring to the discussion above I am not sure if this statement is still valid for temperatures below about 240 K.**

We agree with the reviewer and therefore have modified the manuscript to highlight the spread in ATD onsets at lower temperatures (see comment above and below). In addition we have explained that residence time in section 4 (*page 21, para. 1, line 3*), aerosol size distribution (*page 25, para. 1, line 1-4*) and size thresholds for what constitutes a detected ice crystal (e.g. 2 $\mu$ m for CSU and 5 $\mu$ m for UT) incorporating kinetic effects (*page 22, para. 1, line 7*) all contribute to differences in the observed results.

**This may affect the abstract and conclusion section. For this inter-comparison study it would be very beneficial to state which maximum uncertainty in ice nucleation onsets is aimed for to advance this field and if this has been achieved in the presented campaign or not. Differences in RH<sub>i</sub> of over 25% might be too large to describe ice crystal formation in typically very dynamic atmospheric environments. A discussion of this point would benefit the reader and the broader community. This may also spark further instrument development. Clearly, more inter-comparison studies of these kinds are necessary.**

We agree with the reviewer. We had already indicated special needs for future work with regards to aerosol size (*page 25, para. 1, line 1-4*) but now have included a paragraph (*page 24, para. 1*) to highlight the special needs for more work at lower temperatures where kinetic effects become amplified and also what level spread in RH values would constitute good agreement.

**Technical comments:**

**Throughout the text and figure captions, when listing objects, a comma is missing before the final “and”.**

This has been modified wherever applicable (when there are three or more items in a series).

**Page 20868, line 5: Change “repeat” to “repeated”.**

The word has been changed

**Figures 2 and 3: Please give uncertainties for selected data points. Figures 3-7: It would be nice to have as second y-axis RH<sub>i</sub> given.**

Uncertainties have now been included as error bars in both Figures 2 and 3.