Response to Reviewer 2 (Reviewer comments in bold text)

The paper presents results from an intercomparison of ice nucleation measurement techniques, especially those from the University of Toronto continuous flow diffusion chamber (UT-CFDC). For that purpose, heterogeneous ice nucleation of biological aerosol particles, various mineral dust samples as well as soot was investigated at the AIDA cloud chamber during the International Workshop on Comparing Ice Nucleation Measuring Systems (ICIS 2007).

In the paper the importance of reliable measurements of heterogeneously freezing Ice Nuclei (IN) is described as well as the difficulty to intercompare the various IN counters, and a state of the art of laboratory measurements is given. The experiments at the AIDA chamber and the results of the UT-CFDC and other instruments are efficiently summarized. But, the paper presents not only an instrument intercomaprison but also contributes a perspective of the freezing properties of the most important ice nuclei in the atmosphere. Overall the paper is well structured and fluently to read. I suggest the paper for publication in ACP after addressing some points listed in the Specific Comments.

We thank the reviewer for the useful comments. We indicate the changes made and their location in the revised manuscript.

Specific comments:
• P 20862, line 4: Please indicate if the size is diameter or radius.

The size bins are based on diameters. This has been clarified on *page 7, para.1, line 14*.

• P 20863, title and first sentence of Section 2.2:
I suggest as title 'Experimental overview of AIDA expansions and CSU experiments at ICIS-2007'

We have added the word 'AIDA' into the sub-title for *section 2.2*

and for the first sentence
'The AIDA cloud expansion experiments ...'

We have added the word 'AIDA' into the first sentence of *section 2.2*

• P 20863, line 12: What is the ice threshold size of the Welas OPC and how it is determined?

The particle, therefore the ice threshold size can be as low as 0.8μm (*already indicated on page 9, para. 2, line 9*). The threshold for each experiment was determined based on the largest particle sizes from the size distribution of the aerosol, and a size significantly higher than the largest aerosol size or any liquid drops forming was chosen. This varied from one experiment to another since the size distribution for the various dusts and soot had different maximum sizes. We have explained this further and provided a reference beginning *page 9, para. 2, line 9*.

• P 20863, line 18: Please give the special issue references for the CSU and the AIDA.
The section referred to above discusses the experimental set up/operation of the AIDA expansion chamber and CSU-CFDC. The references for both these methods are given in various location of this section, for example *page 9, para. 2, line 3 and page 10, para. 1, line 12,* respectively. Where available, special issue references have been added (*page 10, para. 1, last line*).

• P 20863-64, CSU: As far as I understood, the CSU can also discriminate between water and ice at temperatures > 250K, yes? Can you please mention this.

The CSU-CFDC discriminates between ice and water droplets based on their differing optical sizes (ice crystals remain much larger), following a water evaporation region, however there exists a water evaporation region (maintained at ice saturation) which only allows ice crystals to survive to their optical particle counter. Nevertheless there is no phase discrimination device. The evaporation region has already been described in *page 10, para 1, line 11*.

• P 20866, line 8: '.. the RH's for 0.1% of particles activating.' To be clear, I would suggest here and throughout the text to always say '0.1% of particles activating ice'.

This change has been made where applicable throughout the text.

• P 20866, lines 10-11: 'To infer modes of ice formation we assume that activation below water saturation proceeded by deposition nucleation and activation at or above water saturation proceeded by condensation freezing.'

This classification of freezing modes should be explained. And, why you think that the drop freezing process is condensation and not immersion freezing? This needs to be discussed.

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 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 temperatures and within the time scale of the particles in the chamber (7-10 seconds). We have now added an explanation for this on *page 13, para. 2, line 2*.

• P 20866, line 24: '.. , it is possible that large particles were lost while sampling from AIDA.' Only a comment: a pity that you didn't connect a size resolving particle counter (for example an APS system) to the sampling lines from AIDA and APC. Even a rough estimate of the size dependence of heterogeneous freezing would have further improved the study.

We agree with the reviewer. We have referred to other studies (*page 14, line 20)* and presented data (in Figure 3) where size resolved data have been presented. We have also pointed to the special need for size resolved studies for the future in the conclusions section. However one of the goals of this study was to assess the ice nucleation activity of typical size distributions of dust particles found in the atmosphere.

• P 20867, line 1: 'The bacteria samples mostly activated *ice* around water saturation, ...'

We have not included the change suggested above to include the word 'ice' but instead have added more information to the text to be explicit about the discussion of ice and water activation for Snomax® and live bacteria samples (*page 15, para. 1).*

• P 20867, line 14: 'to deduce 0.1% activated *ice* fractions, ...'

We have added the word 'ice' here.

• P 20867, line 25 - P 20868, line 6: I read this paragraph several time but didn't get the message ... could you rephrase it?

In this paragraph we are just noting the method of AIDA expansions for ice activation is different than for CFDC techniques. For CFDCs particles just spend a few seconds in the chamber and relative humidities are calculated from steady state equations/assumptions. For AIDA once a cloud is formed due to CCN activation at RH_w>100% the measured relative humidity, which is actively measured, then relaxes to 100% since a liquid cloud forms prior to the observed freezing, which is then recorded to be at RH_w = 100%. We have added a sentence here to clarify this point. (*page 16, line 10*).

• P 20871, line 8-10: '... suggest a transition from condensation to deposition freezing for GSG at about 234 K which is lower than the transition temperature for dusts ...' Doesn't that mean that GSG does not nucleate ice at all, because the temperature for homogeneous drop freezing is 235K? Please explain.

We have now added a line for homogeneous freezing in Figure 7. This makes the above point more clear. From the Figure we now see that some data are at or above the RH required for homogeneous freezing, while for others GSG nucleates a little below the transition border of homogeneous freezing. That being said, for the data points below 100% RH_w and with solid particles such as GSG, we do believe that the soot particles have nucleated ice, and we do not think that homogeneous freezing has occurred. We have now clarified this point on *page 20, para. 1, line 6*.

• P 20871, title section 4: I suggest
'Inter-comparison of UT-CFDC, CSU-CFDC and AIDA expansion techniques'

We have added the word 'AIDA' to the title.

• P 20871, line 18-21: 'UT-CFDC gave higher onset RHs of ice formation for 0.1% of particles freezing than did the *AIDA* expansion chamber. This could partly be an issue of differences in the "residence time" of particles in each of the chambers. For

the CFDC techniques, particles pass through the chamber within seconds whereas in the expansion chamber, particles remain in the chamber throughout the experimental run that can last for a few minutes, albeit with continued cooling.'

I have a comment and a question to this discussion:
1) Comment: If the particles in the AIDA chamber stay longer until ice formation while continuous cooling, they will grow further, yes? Could this explain the higher onset RHs of ice formation? Please discuss.

Presumably the reviewer means 'lower onset RHs' and not higher onset RHs? We think the 'increased residence time' could indeed play a role in lower onset RH for ice activation if there is a time dependence to the nucleation process. However, we don't think the onset value of RH reported would be influenced by 'further growth' of an ice crystal beyond its detection size threshold (~ 2 – 5 μm in this study). A difference in onset RH reported could arise from the difference in time that it takes for a crystal to grow to the detection limit of an optical particle counter in use for a system. If the detection limit for what constitutes a crystal is a diameter of 5 μm as it was for the UT-CFDC, then one can expect at lower temperatures where the growth rates are slower, that there maybe be a time delay between detecting a 2 μm and 5 μm crystal.

2) Question: for the same experiment, the ice activation temperatures of AIDA, UT-CFDC and CSU-CFDC are different, yes? How the activation temperatures for the IN counters were choosen? What are the differences? Is it possible in the figures to see which experiments were the same ? This question popped up several times when reading the article so I think it would be good to discuss this in the beginning of the paper, maybe in section 2.

Most experiments for a given aerosol type in an overlapping temperature range were conducted on the same day. There could have been slight variations for ATD (see below). The experiments were organised based on sampling of a given aerosol type on a certain day. Each day was allocated to an aerosol type and a temperature range. The aerosol chosen to be sampled was then filled into the APC and AIDA chambers and the target range for the IN counters were chosen to be close to the operating range of the AIDA chamber for a given day. Since it was not possible to determine exactly the AIDA temperature of ice formation until after an expansion, non-AIDA investigators prepared their instruments to operate within ± 5K of the AIDA target. It was then up to each investigator to determine at what point during the day they would sample at the lower and/or upper end of the temperature range since each instrument had different cooling efficiencies/requirements. For ATD, three days were allocated with each day operating the AIDA chamber at progressively colder temperatures starting from about 250 K and going down to roughly 230 K. Therefore one could look at Figure 3 and conclude that data points from different instruments falling in over lapping temperature regimes were collected on the same day. However, it is possible for ATD that if in a given day an investigator did not have a chance to sample at a desired temperature, then they were free to conduct experiments so as to fulfil also their own goals of the desired temperature range rather than restrict themselves to the range of the AIDA chamber.

Comments on the Figures:
Though the Figures are quite nice, I have several suggestions which I think can further improve them to be more easy to understand:

• Figure 2-7 general: Some more information could be given in the Figures:

1) it would be nice to have: the water-saturation line, the ice-saturation line and the line of homogeneous freezing of solution particles in each Figure. Also, a vertical line could be drawn at 235K, the point of homogeneous drop freezing.

The water-saturation line is already a part of all figures where applicable, however we have added the ice-saturation and homogeneous freezing line for freezing of 0.1% of 300 nm solution drops (Ammonium sulphate). Since relative humidity (RH) is plotted on the vertical axis, we consider it sufficient to leave out a vertical line at 235 K indicating homogeneous drop freezing, since it will be clear from the intersection of the of the water and homogeneous freezing lines.

2) Further, it should be noted in the Figure captions that for UT-CFDC above 250K it is not possible to distinguish between water and ice.

We have now added in the figure captions that for T>250 K and RHw>100% data points could represent ice or water activation (see Figure 4, for example).

3) '... onset RHw for 0.1% activated *ice* fraction ...'

The word 'ice' has been inserted into all figure captions

• Figure 2: You could add a legend to the figure showing a black triangel (AIDA), square (APC) and star (AIDA 2nd cycle). This would help the reader.

We have indicated the above legend in form of text in the figure caption. Because the legend is already quite busy, we thing it is appropriate not to add more detail to the already existing legend, especially because the differences between AIDA and APC sampling are discussed further in Figures 3-7 and this is not the focus of Figure 2.

• Figure 3:
1) Print 'ATD' above the legend.

In addition the figure caption, we have added the title 'Arizona Test Dust' for Figure 3.

2) Please adjust the blue CSU symbols to those of AIDA (filled triangles –> sqaures, open triangels –> dots).

We would like to distinguish CSU from UT-CFDC results with colour and shape because of the number of data points in this figure, especially in light of open data points also being used for UT-CFDC size resolved data. Also, we would like to keep the CSU and AIDA in different colors, to make sure they are easily distinguished.

3) Indicate that UT/CSU(APC) and UT/CSU(AIDA) means smaller and larger particles.

This is not true for the CSU instrument since the instrument was placed closer to both APC and AIDA chambers and it sampled through a particle impactor and therefore should have sampled similar sized particles from both the AIDA and APC chambers (*page 16, para. 1, line 3*). The size only affects the UT-CFDC since the sampling line from AIDA was longer due to space limitations. Also, UT-CFDC didn't use an impactor for sampling. In addition, we present post-expansion results for the UT-CFDC chamber only which sampling smaller particles after losing the larger more efficient ones to ice nucleation and subsequent settling of crystals during the first AIDA expansion. These factors have already been discussed in detail in the manuscript (*see page 14, line 16, and page 17, para. 1, line 4*). Simply indicating APC and AIDA means larger and smaller particles respectively, may not be an accurate representation of the sampling conditions.

• Figures 3-7: indicate condesation/immersion freezing above the water saturation line and deposition freezing below.

This has now been addressed more explicitly in the text in *page 13, para. 2, line 2*.

• Figures 4-7:
1) Print the aerosol type above the legend.

This has been done for figures 3-7.

2) Use the same scaling for x- and y-axis for the Figures.

We have changed the scaling to be the same for all the y-axes. The x-axes however have been selected to optimise graph area usage, since the same temperature range was not covered for all sampled aerosols.

3) Figure captions 5-7: don't repeat the complete caption for each Figure, just say 'Same as Figure 4, but ....'

Figure captions have been adjusted accordingly.