Reviewer 2:

General comment:

This paper describes a new instrument to measure ice nuclei based on the design of the Colorado State University continuous flow diffusion chamber CFDC. The new instrument, the Manchester ice nucleation chamber MINC is described in detail and selected measurements from the ICIS2007 workshop are presented and compared to data taken with the CFDC itself during the same workshop. I suggest to accept this manuscript for publication in ACP after major changes which are discussed here, first in general and then in more detail with page and line references:

In general, I have the feeling the paper assumes the reader to be already familiar with similar instruments down to very detailed aspects such as the data collection of the OPC. Without this knowledge, some parts of the paper are hard to understand. I would recommend to revise the document such that someone without detailed knowledge on these instrument details can read and understand it.

The authors are presenting many important and in most cases valid facts. However, they leave it to the reader to make connections between them. The paper would be a much better and easier read, if these connections were made by the authors. I will give a few examples later but suggest that the authors critically revise the document with this in mind.

Another more general remark: At some points the authors argue based upon facts or data not presented in this paper. Please, wherever possible, add the data (e.g. calibration data) needed for arguing so the reader can follow the arguments and come to his own conclusion. I will give some examples later.

Please be consistent in supplying information for used materials and components, (Name, Manufacturer).

Response:

We thank the reviewer for all their comments. We agree that the paper can benefit from some additional introductory sentences, and these shall be added. In the attempt to keep the paper succinct, we omitted detailed explanation of the certain aspects, e.g. temperature calibration and inlet aerosol losses. As such, we now intend to include this information so that readers interested in the instrument design can access this information.

We have attempted to address the comment that states clearer connections be made within the text of the paper.

Each of the specific comments have been addressed separately below.

Specific comments:

1 - P19278, lines 25/26:

Please add a statement that it is clearer to the reader, that atmospheric IN mainly act as heterogeneous nuclei and the latter is the focus of the presented measurements. This connection is currently not made.

Response: We agree with the reviewer that a sentence to this affect would benefit the paper. After 'due to the presence of IN.', insert: 'Measurements of these atmospheric IN is the focus of the current study'

2 - P19279, lines 1 and 8,9:

Please make a direct connection between the mentioned mechanisms (line 1) and the conditions they are referring to (lines 7,8).

Response: We believe that this should be clearer now that extra information has been added to the sentence in line 1 - a brief description of each mechanism.

"There are currently four recognised fundamental heterogeneous ice nucleation mechanisms: deposition nucleation (vapour transferred directly to the IN surface), condensation-freezing (water condenses onto the surface of the IN to form a supercooled droplet, then subsequently freezes), contact-freezing (supercooled water droplet freezes upon contact with the IN) and immersion-freezing nucleation (IN becomes immersed inside a supercooled water droplet, then subsequently freezes), (Vali, 1985)."

3 - P19279: lines 9-13:

Runaway sentence is a bit hard to read because many facts are listed. Would be better to split this information over 2-3 sentences.

Response: agreed, we suggest the following changes:

Current version:

Typical atmospheric concentrations of IN are ~1 stdL⁻¹ at -20°C and ~10stdL⁻¹ at -30°C (DeMott et al., 2010), around 6 orders of magnitude less than typical urban total atmospheric aerosol concentrations, but can be subject to very large variations in time and space.

Replace with:

Typical atmospheric concentrations of IN are ~1 stdL⁻¹ at -20°C and ~10stdL⁻¹ at -30°C (DeMott et al., 2010). This is around 6 orders of magnitude less than typical urban total atmospheric aerosol concentrations. However, it is important to consider that IN concentrations can be subject to very large variations in time and space.

4 - P19279: lines 14... :

again a very long sentence and three instead of the mentioned two options are listed.

Response: I can see how the current wording is confusing, therefore we suggest that the following changes to the text are made:

Current version:

There are two ways in which to predict IN concentrations in models: use aerosol properties in conjunction with lab studies, or a constrained theoretical approach, on the effects of aerosol properties on ice nucleation, or, use IN climatologies of IN concentrations.

Reword to:

There are two ways in which to predict IN concentrations in models: use aerosol properties in conjunction with either lab studies or a constrained theoretical approach on the effects of aerosol properties on ice nucleation (e.g. Meyers et al., 1992), or to use IN climatologies of IN concentrations (e.g. Bigg, 1990).

5 - P19280, line 26: . . . in Rogers (1988). The main.....

Response: We agree that the suggestion above reads better.

6 - P19281, line 10:

It would be very helpful to have a drawing (to scale) of the cross section of the chamber head to assess the design and compare it to other instruments. The design of this region is crucial for a faultless operation of the instrument without artifacts in the measurements. How are the sample and sheath layers merged? How are the flows distributed equally over the angular layers (e.g. by series of orifices or gaps)?

Response:

We recognise that an additional figure would help here, but the design is essentially equivalent to Rogers (1988).

The sample enters the chamber via a knife edged ring, as with the CSU-CFDC design. This is to introduce the sample in-between the two sheath layers which enter the chamber slightly higher. The sheath layers are introduced to the chamber from two concentric rings of small holes (72 at 1mm diameter) on either side of the knife edge. These aspects of the design are now shown in a new version of figure 1, see below.

7 - P19281: line 27:

How do you control/measure that the air is slightly supersaturated with respect to ice in the evaporation section?

Response: Unfortunately with the current instrument described in this paper – this is not measured, but assumed. The assumption that the evaporation section is slightly supersaturated with respect to ice is based on calculations given in Rogers, 1988 paper.

8 - P19282, line 20:

Can you specify what material is used as hydrophobic plastic here (Type, Name). Next lines: Can you specify how the residence time is split between nucleation/growth section and evaporation section? Please make clear that growth of ice crystals only takes place in the first section.

Response: Hydrophobic plastic used here is polypropylene, this will be included in the text. The residence time issue is discussed in the discussion section, but we agree that extra wording here would help for understanding how the chamber works. However, it is possible that under certain conditions, namely, when above water supersaturation, that the ice crystals are still growing in the evaporation section as the ice crystals grow at the expense of any evaporating droplets. After '... is ~10s.' add in '(latter 4s in the evaporation section).'



9 - Referring to the fact mentioned on p19283, line 14

it would be good to know how long the section with stable established supersaturation really is.

Response: This is discussed in the discussion section as this depends on the issues of temperature variation along the walls, which is also addressed in the discussion section.

10 - P19283, lines 1:

Please replace (modern roof space insulation) with a brand name and supplier. Please specify thickness of the bubble wrap and the Armaflex layer).

Response:

Insulation: Aluminium thermal foil, B&Q – 4mm thick. Armaflex is 20mm thick. This has been added to the text.

11 - P19283, line 7:

What about the "funnel" section at the end of the evaporation section? Can you specify somewhere the cross section of this region (angle and width of gap)?

Response:

This in included on a new version of Figure 1, see comment 8. This part if the chamber is of the same design as Rogers, 1988.

12 - P19283, line 8:

Can you explain in more detail where and how a transient supersaturation may form. The following description of how you are trying to overcome this problem is then easier to understand.

Response:

Transient supersaturations may form in the top part of the chamber when relatively warm moist air enters the cold chamber (next to dry sheath air). We will add the following in after ' (e.g. Elliot, 1971).'

"Transient supersaturations can occur at the top of the chamber if the incoming sample air is saturated and colder than the sheath air."

13 - P19284, line 1:

Is the second expansion valve of the same type as the first one?

Response: We will include the model number of this valve in here. SEI-0.5-11-S

14 - P19284: line 25:

replace experiment conditions with supersaturated conditions (because the temperature in the evaporation section will also change with changing experimental conditions).

Response: This change will be made

15 - P19285, lines 4,5:

Can you explain this in more detail? As far as I know this detector normally operates at 30 LPM so this removal should not be necessary? It appears, that your detector has a standard flow rate below 10 lpm (p19287, lines 1-2). In this case: What is the effect of the removal of this orifice on the flow field inside the detector and therefore the detection efficiency? Turbulences within the detector due to the increased flow rate may influence the efficiency.

Response:

We have spoken with the manufacturer of this OPC to confirm what we state below. The Climet model used here (CI-3100-1158) has a normal operating flow rate of 28 LPM. The critical orifice was removed by Optical Sciences Ltd, who then recalibrated the instrument at 10LPM for the required size range that we intended to measure. Counting efficiency would not be affected by a change in flow rate, provided that you can see the smallest size of interest clear of the baseline noise (this is determined by the gain settings on the electronics). Increasing the flow rate makes it harder to see smaller particles as their signal is within the baseline noise. Smaller particles for this counter would be classified as 0.5 um (the smallest size this sensor would detect under normal operating conditions), however, we are using the sensor to detect particles larger than this (1um) that can easily be detected. The counting efficiency is given as 50% +/-10% for the smallest size detectable (in this case 1um).

Changes have been made to the text to reflect the above.

16 - P19285, Aerosol losses: I have some major concerns regarding the loss measurements and the corrections which are made based upon these: First, at these high number concentrations mentioned, coagulation may become an efficient enough mechanism to reduce the number and shift the mean diameter of a given aerosol concentration. However, I am even more concerned with the corrections which are made for measured data. Please explain in detail step by step (with formulas if necessary) how this correction is being applied. The reason why this may be or become critical is the fact that the IN efficiency of a given aerosol is known to be a function of particle size (you even mention this in the discussion somewhere). I don't see how it is easily possible to deconvolve these two effects

which are both size-dependent because for the ice crystals you detect you have no information about the initial particle size. This may have a significant impact on the quality of the derived data and may be the source of systematic errors. It gets even more complicated when measuring samples which may be externally mixed(which may be even true for some of the samples used in the ICIS workshop) where the IN efficiency may be significantly different for individual modes within the sample.

Response:

Both reviewers asked for clarification regarding the aerosol loss calibration, as such, this section has been expanded, please see the bottom of this document for this section.

Coagulation is certainly occurring in the APC during the time scale of experiments, but this is not a parameter we could control. It was the same for all investigators and so the best comparisons are made at the same times. It is something to consider in future workshops if the goal is to carefully characterize the ice nucleation behaviour of specific aerosols. However, this is not the focus of this paper.

It is agreed that size is an important factor regarding the IN efficiency, and the issue that the reviewer addresses here is an inherent issue with these types of instruments. A way to avoid this is to size select the sample prior to CFDC chamber entry to test different size ranges of the sample. Samples used during ICIS-2007 were mono-modal.

17 - P19286, line 14... :

It would help if you presented the data and the fit for the reader either in form of a figure or table. Please provide the data for the conclusion you make in the next sentence (. . ..were found to be much higher....) before and not after the conclusion. Again, a detailed table would be very helpful instead of the data listed in the text later on. Please specify e.g. by formulas with parameters how the corrections are made. It is also important to know how the sample position is determined because you mention that the conditions for the sample are corrected.

Response:

Both reviewers asked for this section to be expanded to include the data and more explanation. Please see section at the bottom of this document.

18 - P19287, whole section OPC:

The setup used here for detecting and counting particles by using an OPC as a source for analogue pulses which are then converted into binned digital counts (if I understand this correctly) is very hard to understand. I am not sure if the counting efficiency reported by CLIMET for the OPC for the two size classes (high gain, low gain) can be used for this setup as the detection efficiency depends on the setting of threshold values in the custom made electronics described later on line 11 (comparator, Schmidt trigger). For someone unfamiliar with the latter electronic terms, could you explain what they do to the signal? How many size bins do you have in your setup and what are the sizes of these bins? How do these correlate with the threshold of 3 micron you describe later? I recommend to re-arrange this section so that reader can easier follow the details of the setup (e.g. by first describing the whole setup by following the signals during processing). Please make a new paragraph where you only describe the calibration of the OPC with the APS, Grimm detector etc. If applicable, one could compare the PSL calibration then with the efficiency reported by the manufacturer but be careful here because the detection efficiency may be modified by the nonstandard flow rate anyhow. A figure depicting the signal and data flow and a figure/graph with the calibration system and data would help so that this section is easier to read and understand. In the end you are discussing calibration data and interpreting it which is not accessible to the reader. If you feel it is important for the reader, please present this data, otherwise don't discuss and comment on it.

Response:

The question regarding the counters efficiency in its modified state is addressed as part of comment 15 above.

Regarding the electronics and APS/GRimm section:

Current text:

The Optical Particle Counter (OPC) used was a CliMET 3100-1158. As the OPC was operated at a higher flow rate than standard, a peak voltage to size calibration was carried out by Optical Sciences Ltd using polystyrene latex (PSL) spheres and a standard oscilloscope. The OPC outputs two continuous analogue signals – high gain and low gain, from which concentrations of 1 -2 um particles and 3 – 8 um particles respectively were derived. The counting efficiency of the high gain channel is reported as 50% +/- 10% at 0.5um, whereas the low gain channel counting efficiency is reported as 100% +/-10% for 0.8um particles. The OPC sampled the total flow from the chamber. Ice numbers detected in the total flow are related to the sample flow to detect nucleated IN number concentrations. Comparator electronics and Schmidt triggers were used to detect and bin analogue pulses representing individual particle output from the CliMET. The PSL calibration curve was used to set threshold levels on the comparator electronics. Particle events in each size bin were counted using an 8 channel pulse counter card (National Instruments, 6602) in the control computer. To confirm the calibration was satisfactory and the pulse counter system was operating correctly, the modified CliMET complete with comparator electronics and pulse counter was compared in the laboratory to a calibrated OPC(Grim Aerosol Tecnik, Dust monitor 1.105, optical scattering size range 0.5 – 20.0 um) and an Aerodynamic Particle Sizer (TSI APS 3321, aerodynamic size range 0.5 – 20 um). A vibrating Orifice Aerosol Generator (VOAG) was used to produce monodisperse aerosol at known sizes (3, 4, 5 um) for use in this comparison. Solutions using NaCl, oleic acid and olive oil were used. Results showed that measured size distributions from all instruments peaked at the same size to within 1um, but that the CliMET exhibited a broader distribution that the other instruments, typically spanning over three size channels, equivalent to the peak size +/-2um. The APS has higher resolution due to many more channels than the CliMET, and the distribution tended to span over ~6 channels, roughly the equivalent to peak size -1/+2um. GRIMM OPC results were harder to discern due to the smaller number of size bins and the staggering of the bin sizes. Typically, peaks spanned two or three size bins, e.g. when measuring 5um, sizes from 3.5 – 7.5um were seen.

Reword to:

The Optical Particle Counter (OPC) used was a CliMET 3100-1158. As the OPC was operated at a lower flow rate (10 LPM) than standard (28.3 LPM) a peak-voltage to size calibration was provided at purchase. The OPC outputs two continuous analogue signals – designated high gain and low gain, from which concentrations in the 1 -2 um particle channels and 3 – 8 um particle channels respectively were derived (channels are named according to channel lower size limit) using inhouse built electronics which sends the counts to the computer (via National Instruments, 6602). The counting efficiency of the high gain channel is reported as 50% +/- 10% for the lowest bin (1um), whereas the low gain channel counting efficiency is reported as 100% +/-10% for the lowest bin (3um). The OPC sampled the total flow from the chamber. Ice particle numbers detected in the total flow are related to the sample flow to derive nucleated IN number concentrations.

To confirm that the electronics, as part of the OPC system, were able to size particles correctly, the modified CLiMET was compared in the laboratory to a calibrated OPC (Grimm Aerosol Tecnik, Dust monitor 1.105, optical scattering size range 0.5 - 20.0 um) and an Aerodynamic Particle Sizer (TSI APS 3321, aerodynamic size range 0.5 - 20 um). A vibrating Orifice Aerosol Generator (VOAG) was used to produce monodisperse aerosol at known sizes (3, 4, 5 um) using NaCl, oleic acid and olive oil solutions. Size distributions from all instruments peaked at the same size (to

within 1um), but that the CliMET exhibited a broader distribution that the other instruments, typically spanning over three size channels, equivalent to the peak size +/-2um. The APS has higher resolution due to many more channels than the CliMET, and the distribution tended to span over ~6 channels, roughly the equivalent to peak size -1/+2um. GRIMM OPC results were harder to discern due to the smaller number of size bins and the staggering of the bin sizes. Typically, peaks spanned two or three size bins, e.g. when measuring 5um, sizes from 3.5 - 7.5um were seen.

19 - Section numbering and manuscript structure:

Beginning with section 4 (Results) the structure of the remaining document is unclear to mean. What is the purpose of having Section 5 (Detection of IN) with subsections which describe the results of the experiments in details when there is already a very brief (summary) section named results? I recommend a more logical re-structuring of these sections.

Response: We agree and have re-numbered the sections.

20 - Figure 3 : please use a log-scale for the activated fraction as in Figures 4 and 5.

Response:

There is now a new version of this graph:



21 - P19290, line 3:

Since the dates of operation were already reported, the first sentence may be removed as it does not fit in this section.

Response: We agree and have removed this sentence.

22 - P19290: line 9:

...here AS THE

Also: please clarify that this is the way you define the ICE activated fraction assuming that all particles above three microns are ice crystals.

Response: The first part of this comment has be changed as noted above. We will add in 'of IN' after 'activated fraction' here to make this clearer.

23 - P19290: line 13:

As already mention before: Can you please plot the initial and final size distributions before and after the transmission correction? It is then easier to discuss possible effects of size on the detection efficiency as you mention later in the discussion or conclusion that this may be a problem. I am wondering how you plan to operate the system in the field then. Do you plan to always measure size distributions in parallel to be able to apply this correction?

Response:

Again, please see the section at the bottom of this document that will be added into the paper., For field work and lab work, aerosol size distributions would always be measured along side other measurements. Measuring aerosol size distributions is standard practise when carrying out aerosol field work.

24 - P19291, lines 6-15:

It would be better and help the structure of the document to discuss differences between the instruments in the appropriate paragraphs in section 2 and only repeat those here briefly which are discussed in connection with the results presented here (e.g. diffusion losses and Nafion drier).

Response: We suggest that the difference discussed here in section 5.2, be moved to section 3 regarding the comparison of the two instruments as part of ICIS-2007

End of section 3:

...... AIDA expansion experiments were carried out for all samples in Table 1. Further information can be found in Mohler et al. 2008.

This will be directly followed by:

This paper reports the results obtained by the MINC instrument described in the previous section. Also shown are comparisons with the CSU-CFDC instrument, an instrument of similar design, but with some notable differences. The CSU-CFDCis the same version (CFDC-1H) which was used in recent laboratory and field studies, e.g. Eidhammer et al. 2010; Richardson et al. 2010.

The CSU-CFDC design differs from the MINC design primarily in the use of an actively cooled evaporation section, and does not have heat-transfer fluid within the inner wall: the copper pipes have been directly attached to the inside wall surface. The CSU-CFDC is also longer than the MINC chamber, with the CSU-CFDC chamber section being 81 cm in length, with a reduced length of non-iced wall below the inlet manifold. Although the function of elements of the inlet section are the same, the physical arrangement and actual components used are different, such that the MINC experiences a lower transmission efficiency than the CSU-CFDC (0.8 - 0.9 for particles >100 nm). This discrepancy in the transmission efficiency between the two instruments is mainly due to the use of the Nafion counter-flow dryer in the MINC system, where the losses are much more extreme than those experienced in diffusion driers which the CSU-CFDC system employs.

The portion of text similar to this on lines 6 - 19 will be removed from what was section 5.2. The sentence starting 'Like MINC, the CSU...' shall also be removed from here.

25 - P19291: lines 27/28 ..and referring to Figure 5:

I cannot follow he conclusion that the corrected CFDC data (for 3 micron in red) fits better to the MINC data than the uncorrected data in black. Visually I would tend to say the opposite. However, it would be better to use mathematical (statistical) methods to proof this. The scatter of both datasets for the two individual measurements is larger than the visual misfit between the datasets.

For this reason it would be interesting to look at the reasons for this (changes in the aerosol). What do the experiment numbers in Figure 5 mean? Ar these different days or experiments and times? As you discuss some of this in the following sentences I would remove this one sentence. It does not fit so well if you read it after reading down to P19292, line 6.

Response:

At the request of the reviewer, we have looked again at this section. Curves were fitted to plots of log(AF) vs RH for each set of data present in both figures 5a and 5b. Then for several RH values, a comparison was made of the corresponding log(AF) value for each of the MINC curves to the other curves in each plot. An example plot is shown below:



It is evident from this plot, and the others like it, that the CSU-3um data is not necessarily closer to the MINC data. The navy squares represent the one-to-one line. On the left hand side of the graph, the green triangles (C17_3785) data looks to be closest, whereas to the right of the plot, the 'corrected' c17_3785 data – orange diamonds, is closer.

As a consequence of these finding, we will change the text so that we do not state that this brings the data closer to the MINC data, but we will still report than the assumed cut-off for detected particles is an important factor when considering measurements of IN with instruments of this type.

Current text:

Results for ATD comparisons are shown in figure 5 for two sample temperatures (~-25 °C and ~-32 °C); figure 5 also shows the CSU-CFDC activated fractions recalculated using an activated ice size threshold of 3 μ m to simulate the MINC detection threshold. CSU activated fractions determined using the 3 μ m threshold are much closer to the MINC data, particularly at higher values of SS_{water}, e.g. at 2 % SS_{water} activated fraction is reduced by 50 %. However, this change was found to make only a modest difference (up to 1%) in the SS values attributed to activation, and is well within measurement uncertainties.

Reword to:

Results for ATD comparisons are shown in figure 5 for two sample temperatures (~-25 °C and ~-32 °C); figure 5 also shows the CSU-CFDC activated fractions recalculated using an activated ice size threshold of 3 μ m to simulate the MINC detection threshold. CSU activated fractions determined using the 3 μ m threshold are closer to the MINC data at higher SS_{water} values, e.g. at 2 % SS_{water} activated fraction is reduced 50 %. However, this change was found to make a modest increase (up to 1%) in the difference of the SS values attributed to activation, and is well within measurement uncertainties.

26 - P19293, line 4:

Does this data in Figure 7 correspond to some of the measurements presented in the previous Figures?

Response: The data in figure 7 corresponds to the ATD data shown in figure 3 and 5b. Reviewer 1 requested that this data be moved to earlier in the text. We hope that this is clearer now.

27 - P19293: line 17-19:

I think you want to make the valid point here that the experiment-to-experiment variation in the aerosol (size distribution but maybe even composition?) was significant and may contribute to the uncertainties between individual measurements with your instrument and the differences between

MINC and CFDC (and others). For this conclusion it would be good if you could specify or explain which data was taken with the same aerosol (same experiment, even when the time was different) and where you are comparing data from different experiments (different aerosol preparations). Were the conditions under which the aerosols were produced always identical? Please re-word this so that these points become clearer.

Response: I think this issue stems from the fact that the legends in the figures have not been explained well enough in the first version on the paper. The legends include this data now and the text has been modified as part of another comment.

At the end of the sentence '... inter-comparison experiments.' Add: Figure 5a clearly shows substantial differences between results from different sample preparations (C-5-3785 and C-6-5875) when using the same instrument and similar sampling conditions.

Add the following to the second paragraph in section 3:

Dust samples were always dispersed in the same way, as described in Mohler et al. 2006, with the cyclone impactor set to remove the larger dust particle fraction. The Snomax sample was dispersed from a water suspension, as described in Mohler et al., 2008.

References:

O. Mohler , P. R. Field, P. Connolly, S. Benz, H. Saathoff1, M. Schnaiter, R. Wagner, R. Cotton, M. Kramer, A. Mangold, and A. J. Heymsfield, 2006, Efficiency of the deposition mode ice nucleation on mineral dust particles, ACP, 6: 3007-3021

O. Mohler, D. G. Georgakopoulos, C. E. Morris, S. Benz, V. Ebert, S. Hunsmann, H. Saathoff, M. Schnaiter, R. Wagner, 2008, Heterogeneous ice nucleation activity of bacteria: new laboratory experiments at simulated cloud conditions, Biogeosciences, 5: 1425-1435

28 - P19293: paragraph starting on line 23:

May this be the result of two different aerosol modes (e.g. size or maybe chemistry or both) to which the two instruments are differently sensitive (due to the losses you describe for your inlet system). See also one of my earlier comment on that (correction for losses).

Response:

As outlined in the text, we think that we are perhaps measuring two different modes (deposition and condensation), but that the MINC instrument in this case is not as sensitive to the deposition mode due to the difference in the two instruments evaporation sections: the passive cooling section versus the active cooling section.

29 - P 19294, lines 5-8.

What kind of trend analysis did you perform? Linear regression? If yes, did you expect a linear trend in these values with temperature? Why?

Response: Yes linear regression. Expected linear trend due to results from previous work – i.e. CSU work (e.g. DeMott et al., 2010, BAMS).

30 - P19294: line 24: Particle_ concentrationS.

Response: Corrected

31 - P19295, line 1:

due to A difference....

Response: Corrected

32 - P19295: lines 3-5, as mentioned earlier: How did you analyze the data to come to this conclusion?

Response:

This is addressed in a comment above.

After '...into closer agreement...', add ' at higher values of Ss_{water}'.

At the end of the above sentence, add in: 'This results illustrates the importance regarding the consideration of the size-cut for what is determined to be an IN, particularly when comparing instruments where this can clearly make a substantial difference in the reported activated fraction.'

33 - P19295: lines 10/11:

To my knowledge: As long as the evaporation section is isothermal and has ice covered walls (= meaning it is ice-saturated), ice crystals should not evaporate in any case. How do you come to the conclusion that it may be sub-satured, the condition that ice crystals may evaporate? If there is a slight temperature difference between the walls you would even have a supersaturation. In the description of the instrument (p.19281,line 27) you state that you keep the section slight supersaturated with respect to ice.

Response:

Whilst we do say in the instrument description that this section should be isothermal, however, we have just reason to assume this not to be the case due to heat gain from the ends of the chamber experienced in the high laboratory temperatures during ICIS-2007. It is also important to note that the polypropylene wall in the evaporation section is not ice-coated in the first version of the CSU instrument and in the later design, with the passive evaporation section like MINC, the polypropylene section warms to 0oC during the icing process and it is not expected to retain any ice coating.

34 - P19296, lines 1-2:

How do you know that you have transient conditions which are problematic? I am not sure that your instrument has these problems...

Response:

As outlined in comment 12 above, transient supersaturations may form if the incoming sample air is colder and more saturated than the sheath air. We experienced issues with the sheath air being well above room temperature during the ICIS-2007 experiments, this was caused by heating from the air pump that was operating continuously over long periods, and the environmental temperature, which was also elevated due to several instruments operating at the same time within close proximity. As such the recirculating air flow was not able to cool down sufficiently before re-entering the chamber, as such we are recommending cooling these lines to prevent this.

35 - P19296:

In general I would move the suggested improvements into the section: Conclusions, since the focus of this paper is an instrument description. But I leave it to the taste of the authors if they agree with that or leave it like it is.

Response:

The sentence 'Several suggested improvements....' shall be removed from the conclusion section, and shall be replaced with the information in the improvements section. After this section has been

added, a new paragraph will be started for the remainder of the conclusion.

36 - P19296, line 20:

I guess you rather wanted to refer to Sect. 6.1 here.

Response: We did, thank you for bringing this to our attention. This has been changed, but it is now part of the conclusion section at the request of comment 35 above.

37 - P19296, line 25...: The requirement to grow ice crystals is still given for most phase discriminating detectors for two reasons: 1. Sensitivity: They might not be able to detect ice crystals AND determine the phase accurately if the particles are too small. More important: How do the unactivated (dust) particles interfere with the phase discrimination for comparable sizes?

Response:

Work in the field of particle counters including depolarisation based phase discrimination is ongoing. It is has recently been demonstrated that it is possible to distinguish between water, ice and marine boundary layer salt particles using backscatter depolarisation measurements, although it is not yet completely clear that there are no other particle types which could confound this technique. Despite this, in the light of recent developments it is believed that depolarisation based phase discrimination of particles does offer promise with regard to the application proposed here, particularly in the elimination of false counts due to supercooled water droplets.

Current version:

"The move to better ice crystal detection by phase detection of smaller particles, if feasible, would seem advantageous for IN detection systems because the need to grow ice crystals to much larger sizes than the largest sampled aerosol particle is not required. Successful phase detection of much smaller particles could remove the need for aerosol selection at the inlet (although problems may arise between misclassification of aerosol particles as ice crystals)"

Reword to:

It is suggested that incorporating the use of phase detection within the counting system would be advantageous for IN detection systems. The use of this technique could remove the need for the impactor stage of the instrument, thus allowing a wider range of atmospheric particles to be tested. It is possible to distinguish between water, ice and marine boundary layer salt particles using backscatter depolarisation measurements, although it is not yet completely clear that there are no other particle types which could confound this technique. Despite this, in light of recent developments (e.g. Nicolet et al., 2010) it is believed that depolarisation based phase discrimination of particles does offer promise with regard to the application proposed here, particularly in the elimination of false counts due to supercooled water droplets.

IODE paper (ZINC counter): <u>http://www.atmos-chem-phys.net/10/313/2010/acp-10-313-2010.html</u>

Appendix A: Inlet aerosol transmission calibration

The sample inlet system is described in section 2.2.3 of the main text. Due to the nature of the system, size dependent losses are expected and so these should be corrected for when considering the ratio of measured ice nucleating particles to the total aerosol population (known as 'activated fraction' (AF)). To calibrate the inlet losses a polydisperse distribution of ammonium sulphate particles was provided by a Topaz aerosol generator. The size distribution of these aerosol

was measured before and after the inlet system using a Differential Mobility Particle Sizer (DMPS, please see Williams et al., 2007 for details on the operation of this instrument). Total aerosol concentrations during these tests were of the order of 10⁶ particles per cubic centimetre. During these tests, the counterflow to the inlet drier was provided by the MINC airflow system as in normal condition of operation, while the inlet flow was provided by a Condensation Particle Counter (CPC, TSI 3025A), part of the DMPS system. Figure A1 shows the measured aerosol size distributions for DMPS scans at the start of the inlet system (direct to the TOPAZ aerosol source), scans after the drier, and then scans after the drier and the impactor. As can be seen from figure A1, for this size range, most of the aerosol losses are due to the counterflow drier.





Using the information given in figure A1, a size dependent transmission curve can be obtained. This is shown in figure A2: the transmission curve is shown for sizes 5 - 500 nm. For sizes 500 - 1000 nm the transmission is assumed to be 40 % as a continuation of the curve in figure A2 levelling off to this value. At sizes >1000 nm, the transmission curve for the impactor is then implemented.



Figure A2: Transmission curve of aerosol particles through the MINC inlet system.

The impactor transmission and 50% cut-off were tested using a similar technique as described above. To test the size range of the impactor, a GRIMM optical particle counter (model 1.108) was used to sample laboratory air directly, and then through the impactor: this data is shown in figure A2. The 50% cut-off was found to be at 1.3um, with 75% transmission at 0.8um and 25% transmission at 1.7um.



Figure A3: GRIMM size distribution for the impactor. Results shown for before and after the inlet system

The low transmission efficiency curve described above is primarily due to diffusional losses of smaller particles in the Nafion counterflow drier, and impaction losses of larger particles in the impactor and also in the connections between inlet parts. Coagulation is not thought to play a role here.

During the ICIS-2007 experiments, the size distribution of the aerosol population in the ACP chamber was not measured continuously, so applying a size dependent correction to the incoming sample aerosol could not be performed. Total aerosol number concentration was measured continuously, so we must provide a correction to this number. This was done by comparing the calculated total number from the APC chamber non-corrected and corrected size distribution information. For each sample, the comparison was made for the size distribution information available across the measurement period. The transmission fraction of aerosol particles was found to be constant for size distribution data collected after approximately one hour had passed since the aerosol was injected. MINC results used in this paper were taken after an hour had passed. Transmission co-efficient used for this study are 0.55, 0.57 and 0.63 for ATD, SD and Snomax respectively.

In future experiments, it would be beneficial to measure the size distribution of aerosol at the inlet to the chamber continuously to allow more accurate results to be obtained.

Appendix B: Temperature calibration

As the temperature of each wall is not measured directly on the inside of the chamber, it is important to know how the recorded temperatures (measured at the back of the copper surface) relate to the ice surface temperatures. It is important to know this as it is these temperatures that are used to calculate the sample temperature and supersaturation conditions.

The instrument has three Platinum Resistance Thermometers (PRTs) on each wall, positioned at the top, middle and bottom of the copper sections (where the top and bottom PRTs are positioned 5 cm from the end of the copper tube). In order to calibrate the temperature difference between where we want to know the temperature (the inside walls of the chamber) and where we actually measure it (on the outside surface of the chamber), calibrated PRTs were placed inside the chamber against the copper wall at corresponding positions to the instrument PRTs. The walls were then cooled/warmed under conditions typical of those used at ICIS-2007. Typical airflow was also used to allow representative results, though the ice coating could not be applied as there was no way to seal the chamber as the calibration PRT wires must go through the chamber outlet. The calibration was performed at three different start temperatures to determine if start temperature has any effect on the results, this is shown in figure B1. Figure B1 has six panels, one for each of the instrument PRTs. It can be seen in each of these plots that a common straight line fit can be used for all three starting temperatures, indicating that starting temperature does not affect the calibration and that a simple correction can be made to each measurement.

Outer wall instrument temperatures agreed well with the calibration temperatures, mostly within 0.5oC. However, the inner wall instrument temperatures reported up to 5oC lower temperatures than the calibration PRTs, especially when operating at very low temperatures. The measurements at the top of the inner wall showed the most difference between instrument and calibration PRT values: 2oC difference at -20oC and 5oC at -40oC. Whereas the middle of the inner wall temperature comparison showed 0oc difference at -20oC and 3oC difference at -40oC.

The other piece of important information that these results show is that there does not appear to be a steady temperature along each of the walls. For example, consider the highest point on each of the outer temperature graphs in figure B1. The middle temperature is lower than the two ends. The same is true when considering the inner wall temperatures. Investigations using additional PRTs along the inside of each wall revealed that wall temperatures were steady along most of each wall, with temperature increasing slightly within ~10cm at each end. This is mainly due to heat gain from the ends of the chamber.



Figure B1: Temperatures as measured by instrument PRTs (x-axis) plotted against temperatures as measured by calibration PRTs* (y-axis) for three different start temperatures (-18oC, -25oC, -30oC)

*Deviations from straight-line fit for outer wall middle temperatures was due to calibration sensor not being firmly fixed to the wall.