Response to comments from Referee #2

Referee comments are reproduced in *blue*. Response is given in black and respective changes to the manuscript in *italics*.

Review of "Leipzig Ice Nucleation chamber Comparison (LINC): Inter-comparison of four online ice nucleation counters" by Burkert-Kohn et al.

General Comment:

This manuscript presents the results of the direct inter-comparison of four cloud chambers (i.e., The Leipzig aerosol cloud interaction simulator (LACIS), the Portable immersion mode cooling chamber coupled to the portable ice nucleation chamber (PIMCA-PINC), the Portable ice nucleation chamber (PINC), and the spectrometer for ice nuclei (SPIN)). Different size-selected (200, 300, and 500 nm) uncoated (microcline, kaolinite, birch pollen) and coated (microcline/H2SO4, microcline/HNO3) aerosol types were used to determine the performance of these INP counters. Given the versatility of the used cloud chamber, three out of the four heterogeneous ice nucleation modes were also studied (i.e., deposition nucleation, condensation and immersion freezing).

Although this manuscript does not present a completely novel idea given that this approach was introduced/presented in earlier studies, it is the first time that these four instruments are directly compared. This is of high value for the ice nucleation community, especially because the SPIN is a commercial instrument that will be used in several studies. There is an urgent need to understand what different cloud chamber measures, and this study provides very useful information that helps cloud chamber users to interpret their results. The manuscripts is very well written and the figures are easy to follow. The experiments were carefully designed and the results are accurately interpreted. This manuscript can be accepted in ACP after the following minor corrections are taken into account.

The Authors thank the Reviewer for the comments and address the minor comments below:

Minor comments:

1. *I am wondering if the authors can comment about the following question. Out of the four instruments investigated in this study, what is the most reliable? Is there a "standard" instrument that can be used to validate a newly designed INP counter?*

In this study, we address the inter-comparison of four ice nucleation chambers with their similarities and differences, including three chamber configurations that have been used in the field for sampling ambient INPs. We refrain from endorsing one particular chamber precisely because what we demonstrate in the manuscript is, that a number of factors need to be considered to interpret the data and inter-compare. Our goal here is to bring to attention to the community these different aspects and factors that need to be considered. Endorsing one INP counter also depends on what the research goals are for a given field or laboratory experiment. As has been pointed out recently in Cziczo et al. (2017) that a variety of instruments and methods would be necessary to cover the dynamic T and RH range and particle sizes for investigating heterogeneous ice nucleation in the atmosphere.

2. *I like Figure 2 but it took me sometime to follow it. It may be more useful to use filled and open symbols here.*

We agree with the reviewer. In the current version symbols only distinguish PIMCA-PINC experiments (circles) and LACIS (crosses) for clarity and different colors have been used for different aerosol samples investigated as in the previous version of the manuscript. We choose crosses for LACIS data rather than open symbols due to the number of (partly overlapping) data points in particular in Panel A (Fig. 2 and Fig. B1). We think the use of a different shape improves the readability of the plot.

3. *I did not understand why the LACIS modeled results by the SMB reached a plateau but not the PIMCA-PINC? Is it the model instrument specific?*

The SBM model is not observed to be instrument dependent as has been previously shown by Peckhaus et al. (2016) where microcline data from a cold stage has successfully been modelled based on Niedermeier et al. (2015) data. Also, already in Niedermeier et al. (2015), SBM calculations based on LACIS measurements were found to agree well with data from Atkinson et al. (2013), another cold stage method. The increase in FF of the PIMCA-PINC data has been reproduced well by the SBM, however, SBM produces a plateau (constant AF as a function of decreasing T) whereas PIMCA-PINC observations show a levelling-off in the slope of the FF (small increase in AF as a function of decreasing T). The reason for this discrepancy is currently unclear and a plateau as modeled by the SBM has also not been observed with similar instrumentation from other previous studies (see p. 9, line 16-17).
The following statement has been added to the manuscript: *"In a recent study by Peckhaus et al. (2017) the SBM was successfully used to reproduce other ice nucleation data from a cold stage experiment. In principle, a contact angle distribution describes the ice nucleation ability of a material, and is then combined with classical nucleation theory in the SBM. Thus the SBM is not believed to be instrument specific."* (p. 9, line 22 ff).

Specific comments:

Page 2, line 3: Do the authors mean: "its importance"?

The sentence has been changed the following: *"The importance of ice nucleation mechanisms and the properties of aerosol particles acting as so-called ice nucleating particles (INPs),…"* (p. 2, line 4-5).

Page 2, line 3: I don't think this is the only paper showing this. Please add more references here.

We agree with the reviewer and have added two more references here (Boucher et al., 2013, Mülmenstädt et al., 2015 (p. 2, line 3).

Page 2, line 4: Spell-out INP.

Done.

Page 2, line 6: Add references after "climate".

The following references have been added in the suggested location: DeMott et al. (2010) and Phillips et al. (2013). (p. 2, line 7)

Page 2, line 19: Replace "which" with "what".

The sentence has been changed as follows: "Which ice nucleation pathways exist and under what conditions they are relevant in the atmosphere is not fully understood, but has been speculated and discussed (e.g., Kanji et al., 2017). (p. 2, line 20-22)

Page 2, line 20: Add references after "understood".

We have added a reference as suggested by the reviewer (Vali et al., 2015, Kanji et al., 2017, p. 2, line 5-6).

Page 2, lines 20-22: I don't understand what the authors want to communicate here. Please clarify it.

To avoid confusion the following sentence has been deleted from the manuscript*: "However, there is no reason to believe that in the atmosphere, and particularly in mixed-phase clouds, a difference in the freezing mechanism might be relevant, as ice formation in these clouds generally proceeds via the liquid phase."*

Page 2, line 25: hourly? daily?

The sentence has been rephrased using *"hour-to-day timescales"* when referring to online ice nucleation measurements (p. 2, line 28).

Page 2, line 33: DeMott et al., 2016 could be add it to the list.
The suggested reference has been added (p. 3, line 2).

Page 3, line 10: Please state what type of CFDC the authors refer to. CSU?
We have added "CSU-CFDC" to be clear which CFDC we refer to in all relevant locations (p. 3, line 4, 5 13 and 15; p. 19, line 33. The acronym "CFDC" without further distinction is used throughout the manuscript for all continuous flow diffusion chambers and for statements concerning typical CFDCs (including e.g., PINC and/or SPIN).

Page 3, line 12: Please state what type of mineral dust was used.
We have now specified in the revised manuscript the sources of the dust used (see p. 3, line 15-16).

Page 3, line 33: Add references after "activity".
We have slightly modified the preceding sentence and added a reference (see p. 3, line 35 ff) to read as follows: "*During long-range transport of aerosol particles in the atmosphere, internal mixing with organics and inorganic constituents can cause a temporary or permanent change in the physicochemical properties of the particles and can decrease their ice nucleation activity as discussed in Kanji et al., 2017.*"

Page 4, line 23: I found the pore size quite big. Is there any chance that small fragments of the pollen grains could go through the filter pores?
The pore size of the filters is indeed comparably high. However, we like to refer the reader to Pummer et al. (2012) who have investigated the same pollen type as used in this study. They examined the pictures of an electron microscope (dried pollen extracts) to make sure that no submicron particle fragments are left in their solution and the ice active components were much smaller than the filter size. In addition, experiments in this study were performed for size-selected particles in the submicron range, thus any larger particles would not have had any importance on the results.

Page 4, line 31 and Figure 1: I think it should be "Nuclei" instead of "Nucleus".
We agree and have made the corresponding changes in the revised manuscript (p. 5, line 8 and Fig. 1 caption).

Page 5, line 12: "parallel" is out of place.
The sentence has been rephrased by replacing "*parallel*" by "*simultaneously*". It reads as follows in the revised manuscript: *"Typical particle concentrations during the ice nucleation experiments measured simultaneously with a CPC were 240±70 cm^{-3} for the presented measurements and were diluted to 25-40 cm^{-3} for PIMCA-PINC measurements."* (p. 6, line 8)

Page 6, line 12: Please state what diameter the authors are referring to.
We refer to the particle size threshold to distinguish unactivated aerosol particles from ice crystals. The sentence has been changed and reads as follows: *"For data analysis in this study an ice crystal size threshold of 2 µm (diameter) is used."* (p. 7, line 7 ff)

Page 7, line 29-30: Based on Garimella et al. (2016), the SPIN has a depolarization optical detector. I am wondering why the authors did not use this to discriminate between ice particles and droplets, instead to focus on their size only?
The reviewer is correct in pointing out that SPIN has a depolarization detector to distinguish between ice particles and droplets. The depolarization signal was not used in the current study due to two reasons:

a) To make the comparison of SPIN and PINC data more direct (PINC uses an OPC only measuring particle size).

b) Monodisperse particles were used for the experiments allowing reliable distinguishing of ice from dry particles. The limiting factor for the discrimination from a depolarization detector is the growth of ice crystals that are large enough (which is the case for LACIS and PIMCA-PINC). The depolarization signal of very small ice is not distinct from the dry particle signal and therefore could be prone to errors for smaller ice crystals as expected to be formed in PINC and SPIN in this study.

Page 8, line 6: Delete Hartmann et al., 2011.
Change made.

Page 10, line 25: I am wondering why the authors did not perform an experiment with 500 nm microcline particles as done for kaolinite and Birch pollen.
The particle types and sizes were chosen based on the following criteria and considering available experiment time of the study:

a) The general selection of particle types and sizes has been made to allow instrument inter-comparison for the maximum range of operational conditions for the instruments to evaluate the instruments performance in comparison with each other (see p. 4, line 4-5).
b) Technical limitations to produce a high enough particle concentration for the measurements with all participating instruments. For example for microcline particles a sufficient particle concentration for the measurement was not achieved for a particle size of 500 nm size-selected particles.

We have added this explanation in methods section and reads as follows: "*The specific particle sizes were chosen to allow measurements in the whole range of detectable frozen/activated fractions for all instruments and for comparison with literature data. A limiting factor for larger particle sizes was the particle generation system, which did not produce a sufficiently high particle concentration for simultaneous measurements with all instruments, thus for example microcline was not tested for sizes larger than 300 nm.*" (p. 6, line 4 ff).

Page 11, line 1: Add references after "INPs".
Two references (Archuleta et al., 2005, Welti et al., 2009) have been added to the manuscript (p. 11, line 19).

Page 11, line 16: "note" is out of place.
The sentence has been rephrased in the revised manuscript as follows: *"Note, that measurements with PIMCA-PINC using the same particles show a significant reduction in the ice activity with a T_{50} of ~2 K lower for wet generated kaolinite (Fluka) particles for measurements conducted at ETH Zurich in succession to LINC (see Appendix C for more details)."* (p. 11, line 34 ff)

Page 12, line 18-19: This is a bit unclear. Please clarify it.
The sentence has been rephrased and now reads as follows: "*This is observed in the PINC data, which indicates that the most active particles are found at 233 K with initial onset of ice formation at RH_w of 82-86% corresponding to RH_i of 121-127%.*" (p. 13, line 14-15)

Page 14, line 28-32: Given that these experiments differ from the others (i.e., they are not directly comparable), I am wondering if this should be removed for consistency.
The data on nitric acid treated microcline for the PINC and SPIN instruments are shown here for completeness. All aerosol particles were prepared and produced in the same manner with the same aerosol generation instruments and both data sets were taken individually by each instrument. There are no limiting factors to exclude the data from this study except that measurements were not performed in parallel, from which a difference was not to be expected. The explanation has been revised as follows: "*Note that measurements on nitric acid treated microcline were performed on two different batches of nitric acid treated samples, i.e. PINC and SPIN did not measure in parallel for this*

aerosol type for which a discrepancy was not expected. It is possible that the observed difference between SPIN and PINC data is based on the ice active material in the PINC batch which may not have been as thoroughly deactivated during acid treatment compared to the batch measured with SPIN." (p. 15, line 12 ff).

Page 17, line 11-12 and Figure 7: Add "W" to RH.

The authors would like to keep "*RH*" without the index "w" in this respect. The reason is that the RH here refers to both, RH_w and/or RH_i (p. 18, line 10-11).

Page 18, line 3: I don't think this is the only paper showing this. Please add more references here.

The sentence: *"Recently, CFDCs have often been used for field measurements of INP concentration at water supersaturated conditions to represent immersion freezing (e.g., Kanji et al., 2017 and references therein"* has been deleted from the manuscript.

We rephrased the sentence, which reads as follows in the revised version: *"In many field measurements CFDCs have been used for measurements of INP concentration at water supersaturated conditions (e.g., DeMott et al., 2010, 2016, Tobo et al., 2013, Boose et al., 2016, Lacher et al., 2017) and sometimes are used to represent immersion freezing (e.g., DeMott et al., 2017)."* (p. 19, line 6 ff)

Table 1. Why is microcline twice here?

The two lines for microcline have been used to distinguish between 200 nm and 300 nm size-selected aerosol particles. This distinction has been made more clear by adding *"(200 nm)"* and *"(300 nm)"* to the current manuscript (Table 1, first column, rows 1-2).

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