Response to comments from Referee #1

We thank the Referee for their comments. Response is given in black and respective changes to the manuscript in *italics*. The Referee comments are reproduced in *blue*.

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

Summary and general comments

This study presents a quantitative evaluation of the ice nucleation (IN) abilities of seven types of aerosol particles measured by four IN counters co-located at TROPOS, Leipzig. Based on the results obtained, the authors address the importance of the inter-comparison workshop with co-deployed instruments, uniform aerosol dispersion procedure and size segregation method. From my point of view, the difference in the ice crystal threshold sizes of PINC and SPIN is well justified (Sect. 3.2). Not employing upstream impactors to minimize biases of particle losses throughout this inter-comparison work was wise (P5L2-4).

Besides the suggestions for future studies made by the authors (P22L28-33), a comparative validation workshop of atmospherically representative ambient samples or in-situ field comparison of IN techniques (DeMott et al., 2017, ACPD) is also an important assignment for the IN research community. Finding a universal calibrant that can be used for validating any IN instruments at home bases should be kept as an alternative approach especially for those who may join this research field in the future.

I support publication of this manuscript after some minor comments below are properly addressed. Given the technical nature of the manuscript, it may be better published as a technical note in ACP or AMT than a regular research article according to the journal guidance:

www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html.

I will leave this discussion to the authors and the editor.

The authors thank the Referee for their comments on suggestions and provide responses to their comments and questions below:

We acknowledge the reviewer's comment to move this paper to AMT due to the technical nature of some parts. However, we suggest retaining the paper in ACPD/ACP because the instruments inter-compared in this paper are of a common design/identical to instruments already used to publish results in ACP and future publications with similar instruments (e.g. SPIN) are anticipated in ACP. So it would be unusual that a paper addressing comparisons of instruments whose results have been and will be published in ACP is itself published in AMT.

Secondly, the inter-comparison is based on results of non-technical aspects, which are

1. The investigation of the ice nucleation ability of birch pollen washing waters (biological) using continuous flow diffusion chambers for experiments in the deposition and condensation freezing regime
2. A study on nitric and sulfuric acid treated microcline for which the acid has been completely removed prior to the experiments. The results address conditions of immersion freezing (nitric/sulfuric acid treatment), as well as condensation freezing (nitric acid treatment).

Minor/technical comments

Section 3.3: It is not conclusive that the observed difference between FF and AF is due to different ice nucleation modes (that is, immersion vs. condensation) or technical artifacts/limitations (e.g., different methods in ice detection and IN efficiency estimation).

The following sentences have been added to the manuscript: *"Possible reasons for observed differences, such as technical artifacts or differences in the ice nucleation modes, are discussed. For simplicity, FF is used for experiments exclusively performed in the immersion mode. In contrast, for ice nucleation chambers measuring in the condensation mode, which is in this case not explicitly* *distinguishable from immersion freezing, data is presented as AF in the following figures." (p. 19, line 15 ff).*

Is it really fair to say what the authors present in this particular section is immersion vs. condensation? One can presume that the technical artifacts, such as ice detection, IN efficiency estimation (e.g., FF vs. AF), misalignment of particle stream in the chamber and inhomogeneous distribution of particles in individual droplets, play substantial role on potentially explaining the observed difference amongst the compared techniques. For this matter, the sub-title of this section should be named differently?

We agree with the reviewer and have changed the name of section 3.3 to "*Apparent differences between immersion and condensation freezing*". (p. 19, line 5)

P1L10: Better read with "the whole range of atmospherically relevant thermodynamic conditions"

The suggested change has been made (p. 1, line 10).

P2L19: The dominance of immersion freezing (P18 L2-3) may be better discussed in here. The extended discussion may be helpful to the reader.

We accepted this valuable suggestion. The explanation (p. 18, line 2-3 in the initial manuscript) has been moved and implemented as follows: *"Further, Marcolli (2014) suggested that deposition nucleation might in fact be immersion freezing (or homogeneous freezing for T < 235 K) of water trapped in pores and cavities at water subsaturated conditions. 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). It has been suggested that immersion mode is the dominant heterogeneous freezing pathway under mixed-phase cloud conditions (e.g., Ansmann et al., 2008, de Boer et al., 2011, Westbrook and Illingworth, 2011)."* (p. 2, line 19 ff)

P4L5-6: "…droplet containing a single aerosol particle…" - how good is this assumption? In reality, multiple particles might be in a droplet when aerosols were made using a suspension (Emersic et al., 2015, ACP; Baydoun et al., 2016, ACP; also your own statement in P9L4-5). This may be RH & droplet size dependent. Would this factor be important to interpret the difference between FF and AF?

Indeed particles from suspensions may contain agglomerates, particularly when comparably large droplets are generated directly from suspensions. These are the kind of droplets that are referred to in the literature you cite above. However, in our case, the suspension is atomized and the resulting droplets are then dried and the residual dry particles are size selected. Based on measurements done earlier in other experiments, we know from dispersion of dry particles from the mineral dust samples, that small enough particles in mineral dust samples are not available so as to form agglomerates amounting to the sizes studied in this work (200 – 500 nm). However, as this cannot be ruled out completely and to clarify this, we added the following: "*For insoluble materials such as kaolinite or feldspar, such a particle could consist of an agglomerate of smaller primary particles. However, the number concentration of primary particles in the dry sample strongly decreases with size for the mineral dust samples and the size range used in this study, which makes the presence of dust agglomerates unlikely."* (p. 5, line 1 ff).

For the birch pollen samples it has been shown that the suspension contains small macromolecules, in which case agglomerates of molecules are produced. The following sentence has been added to the manuscript: *"For suspension of birch washing waters containing small macromolecules, an agglomerate of molecules is produced, which is referred to as a (single) aerosol particle after size selection in this work…".* (p. 5, line 4)

Ice nucleation instruments obtain AF and FF, which are based on particle counting after suspension, drying and size selection, as such single particles (or similar agglomerates of molecules for birch washing waters) can be assumed to be investigated in both cases. If they exist, any agglomeration should be systematically biasing the variables in FF and AF and therefore cannot be used to explain the difference between AF and FF. We have reworded the mentioned sentence and it now reads *"…droplets containing single-immersed aerosol particles…".* (p. 4, line 9-10)

P14 L12: Sullivan et al., 2010, GRL – authors perhaps meant to cite the following paper?

Sullivan, R. C. et al. (2010, ACP), Irreversible loss of ice nucleation active sites in mineral dust particles caused by sulphuric acid condensation, Atmos. Chem. Phys., 10 ,11,471–1 1,487, doi :10.5194/acp-10-11471-2010.

Sullivan et al. (2010, GRL) demonstrated that condensation/diffusion of hygroscopic materials could make particles ice active in immersion/condensation mode (which may be discussed anyway in the paper separately…).

The reviewer correctly pointed out that the Sullivan et al., (2010) paper in ACP was to be cited here and was accidently cited as the GRL 2010 manuscript by the same first author. In the revised manuscript Sullivan et al. (2010, GRL) has been replaced by Sullivan et al. (2010, ACP). (p. 14, line 10).

P14L24-25: "The brichN particles are the most hygroscopic…" - based on what? CCNC? Either data or reference is missing.

Indeed, this statement is based on CCNC measurements, which were continuously conducted in parallel to the INP measurements. For birchN, already at SS = 0.1% (i.e., RH_w = 100.1%, the lowest supersaturation sampled) all particles with a diameter of 300 nm were activated to droplets (CCN/CN = 1), while for all other samples, particle hygroscopicity could be derived. The information about the source of our statement was added to the text: *"The birchN particles are the most hygroscopic particles of the samples examined in this work, which was deduced from CCNC measurements where 300 nm particles fully activated at a supersaturation of 0.1% (i.e., the lowest supersaturation sampled), while for all other samples, particle hygroscopicity could be derived, i.e. 50% active fraction was achieved at a higher supersaturation."* (p. 15, line 7 ff).

P21L4-5: "…particles of uniform composition such as microcline" - I disagree with this statement. The authors state that their microcline sample contains bi-components (P4L10-12).

We agree with the reviewer and have deleted the respective sentence in the manuscript.

P22L7-10: I suggest separating into two sentences to improve the clarity of the statement - e.g., "Treatment of the microcline sample with either sulfuric or nitric acid…permanently in immersion freezing. In addition, the nitric acid treatment lead to…between 233 K and 243 K.".

The suggested change has been made and the section reads as follows: *"Treatment of the microcline sample with either sulfuric or nitric acid, followed by washing off the acid, destroyed the ice nucleation ability of the microcline permanently in immersion freezing mode. In addition, the nitric acid treatment led to a significantly reduced AF in deposition nucleation and condensation freezing conditions between 233 K and 243 K."* (p. 22, line 19 ff)

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