

Reply to reviewer #3: Interactive comment on “The Horizontal Ice Nucleation Chamber HINC: INP measurements at Conditions Relevant for Mixed-Phase Clouds at the High Altitude Research Station Jungfraujoch” by Larissa Lacher et al.”

Reviewer comments are reproduced in **bold** and our responses in normal typeface; extracts from the original manuscript are presented in *red italic*, and from the revised manuscript in *blue italic*.

Authors present a new instrument to measure INP concentration particularly relevant to mixed-phase clouds. They validate the instrument performance using standard chemical compounds for which the thermodynamic properties are well known. They deploy the instrument at the field site to further validate the performance. Results look promising, and I recommend publication after addressing following minor comments. The main concern is how ice crystals are distinguished from droplets, and the discussion over this topic is not convincing.

We would like to thank the reviewer for the comments and address the minor comments individually below.

Can aerosol movable injector perturb the flow conditions within the main chamber?

The critical value for transition between laminar and turbulent flow is given by a Reynolds number of 2000. At given chamber dimensions and at 242 K, the Reynolds number for the total flow in the chamber is 66, and the Reynolds number for the disturbance by the aerosol injector is 880. Since both values are below the critical number, no turbulence is expected within the ice chamber. Furthermore, after the perturbation of the flow by the injector, it is expected that the sheath flow, due to its sandwiching role, stabilizes the flow conditions again, and the initial Reynolds number of 66 is re-established. The flow regime has also been discussed in Kanji and Abbatt (2009) at 223 K, and we now include the calculations at 242 K and for the aerosol injector in the revised manuscript on page 6, line 16 - 21.

“The outer diameter of the injector is 6.35 mm (inner diameter 3.175 mm) which ensures that at a flow rate of 2.8 stdL min⁻¹, and at 242 K, the turbulence regime is not encountered by air flowing over the aerosol injector (Reynolds number 880, well below the threshold of 2000 for turbulent flow). Furthermore, given the chamber dimensions, before and after the injector, the Reynolds number is 66, well below the critical number. It is expected that the minor disturbances in the flow by the injector will not result in transitioning from the laminar to turbulent regime.”

The injector object is obstructing the sheath flow.

The sheath flow can pass above and below the injector as it is placed directly in the center of the chamber. The laminar flow should be maintained as discussed in the comment above.

The length of the injector is defined but did not find the diameter of the injector. Do the injector has slots to inject aerosols? What are the dimensions of these slots? Do all these slots deliver aerosol particles evenly and how this is calculated?

The inner (outer) diameter of the aerosol injector is 3.175 (6.35) mm (now included in the manuscript, see comment above), and aerosols are released through a slit which has a cross sectional area that is slightly smaller than the area of the entrance to the injector, leading to a small overpressure in the injector in front of the aerosol slit, promoting an equal distribution of the particles over the width of the slit. We include this point for a complete description of the instrument now on page 6, line 14 - 16:

“The cross section area of the slit is smaller than the cross section of the inner diameter of the injector, which creates a small overpressure at the particles exiting through the slit promoting an equal distribution of the aerosols over the width of the slit.”

The distance between the warm and cold plates is 20 mm. Typically within CFDC, it is around 10 mm. Is the gap is maintained at 20 mm because to increase the particle residence time? Would this affect the supersaturation profile?

This is correct, the wider distance was chosen in order to have a wider range of residence times when combined with a variety of injector positions.

The exposure of the aerosol layer to the supersaturation profile at the given chamber dimension of 20 mm depends on the sheath-to-aerosol flow. When maintaining the sheath to aerosol ratio at 12:1, which is a typical setup in the field, this results in an exposure of the aerosol layer to a variation of $RH_w \pm 1\%$ ($RH_i = 2\%$), which is updated in the revised manuscript on page 16, lines 12 – 13 :

“...there is a temperature variation of ± 0.4 K across the aerosol layer for the temperature conditions (242 K) used in the field measurements presented here. The variation in temperature causes a variation in RH_w of $\pm 1\%$ ($RH_i \pm 2\%$).”

Figures 2 and 3: It is not clear why different size acid droplets were used.

Different initial sized particles were chosen according to the aimed experiment: For the activation experiments in Fig. 3 at temperatures > 235 K, larger sized acid particles were selected in order to be closer to the mode size of ambient particles. In Figure 2, the goal was to perform homogeneous freezing experiments to validate the RH and T in HINC, as such it was not necessary to be similar to the conditions we use in the field experiments. Thus we chose 100 nm since that was most convenient for the production and size selection of the particles in our aerosol instruments.

In figure 2 ice can be detected in the OPC size bin 2 – 8 μ m. In figure 3, where we don't expect ice particles, the droplets can be seen in three bin channels that range from 0.5 – 5 μ m. The size channels overlap between figure 2 and 3. This is confusing to understand. Is this means one cannot use size channels only to distinguish between ice and supercooled droplets?

Different OPC size channels were chosen according to the aim of the performed experiments: In the homogeneous freezing experiments we do not expect water droplets in any channels above 0.5 μ m, thus all channels above 0.5 μ m of the OPC can be used to detect ice.

Fig. 3 refers to cloud droplet activation at 243 K, using H_2SO_4 , where we expect only cloud droplets to be measured and no ice crystals (homogeneous freezing is insignificant for these

conditions) in any size channels. In order to see the relative growth of cloud droplets to certain sizes, the respective size channels are shown individually. This is specifically done to assess which channels *cannot* be used for detecting ice at 242 K and $RH_w = 104\%$ (if ice were to be present) merely by demonstrating that water droplets could also grow in to these larger size channels. It is from Fig. 3 that we can say water drops will not contaminate the 5 μm channel thus we can use it to confidently detect ice crystals. But it is also from this type of an experiment that we can say at 242 K and $RH_w = 104\%$ we cannot use the 3 and 4 μm channel to detect ice crystals for ambient aerosol particles (whose pure composition is not known) as these channels may be contaminated with droplets.

We update the statement in the revised manuscript on page 7, line 21 – page 8, line 2:

“For the deliquescence and cloud droplet activation experiments, 200 nm particles, and for homogeneous freezing experiments 100 nm H_2SO_4 were used. Results from these experiments are presented for different size channels which show the growth of the different particles at different RHs to various sizes. E.g. for the homogeneous freezing experiments at 233K, ice particles are observed in size channels $> 0.5 \mu\text{m}$ at $RH_w > 97.5\%$, while in the experiments at 242/243 K and at $RH_w 100 - 107\%$ cloud droplets are measured in size channels $< 5 \mu\text{m}$, but not in the size channel $> 5 \mu\text{m}$ which is thus used to detect ice crystals.”

This leads to other questions regarding figure 4. How one can determine the INP concentration using this data? On page 13 it says one can use size channel $> 5 \mu\text{m}$, but on page 12 (line 11) it says larger particles ($> 5 \mu\text{m}$) may settle and one cannot see any particles in this channel. Both statements are confusing.

Fig. 4 is not used to determine INP concentrations, but to observe the survival of water droplets in the OPC channels which are used to determine ice crystals. The settling comment was made with regards to water droplets and not ice crystals (due to the difference in densities). But we clarify this in the revised manuscript.

Still, the reviewer addressed a valid point of inconsistencies regarding the particle settling losses previously mentioned in the manuscript. We have now performed specific calculations of water droplet growth in HINC for the conditions reported here (242 K; up to $RH_w 107\%$) and we find that 200 nm SA particles grow to 5 μm only at $RH_w > 107\%$. Furthermore there is no settling of cloud droplets expected at 242 K and 104% RH_w , based on our diffusional growth calculations.

However, since we do see a small signal at 105% in the 5 μm channel for the ambient case (Fig. 4), we believe that this is strong evidence for ice crystals in the ambient case because only ice crystals are expected to grow to sizes larger than 5 μm under these conditions, since water droplets (as shown by the lab experiments with extremely hygroscopic particles) do not reach 5 μm in size below $RH_w = 107\%$. As such we are confident for a RH of 104% we do not detect water drops but only ice crystals in the size channel $> 5 \mu\text{m}$.

Thus the previous statement on page 12, line 10 – 12 in the original manuscript

“An example of an increase in RH_w to $> 106\%$ at 243 K is shown in Fig. 3, where WDS is not observed in the OPC channel $> 5 \mu\text{m}$. This is likely due to settling of the larger liquid droplets out of the aerosol flow, which grow to sizes too large to be sampled by the OPC due to the hygroscopic nature of H_2SO_4 .”

is replaced by (revised manuscript, page 14, line 2 - 9):

“Based on diffusional growth calculations (Rogers and Yau, 1989) activated cloud droplets of an initial diameter of 200 nm can grow to a size of 4 μm in HINC at 242 K, $RH_w = 104\%$ for a residence time of 8 seconds (conditions used for field experiments reported here), giving us confidence that droplets are not detected in the 5 μm channel. Only at an RH_w of 107% cloud droplets grow to $> 5 \mu\text{m}$, and therefore by conducting our experiments at $RH_w = 104\%$, we only detect ice crystals in the 5 μm OPC channel. As a confirmation, no counts in the size channel $> 5 \mu\text{m}$ were observed for H_2SO_4 particles (Fig. 3) even up to an RH_w of 107%, and only with ambient particles an increase in AF for particles $> 5 \mu\text{m}$ at $RH_w = 104 - 105\%$ is observed (see Fig. 4), which can be caused by ice crystals forming heterogeneously, since water droplets cannot grow to this size at the respective conditions in HINC.”

Page 13, lines 4-6: The experiment was conducted at these conditions and results are shown in Figure 4. Here it was shown that there are no particles observed in channel $> 5 \mu\text{m}$. What if all these ambient particles were Organics or pure inorganic salts; one can see the data shown in figure 4. But if there are small dust particles, they might induce ice nucleation and grow to size $< 5 \mu\text{m}$. In this case, droplets and ice particles co-exist. Discussion along these lines is necessary. Limitations imposed on INP concentrations by these conditions (page 13, lines 4-6) must be discussed.

If there are dust particles that form ice, they will activate upon entering the chamber and grow within the residence time to sizes larger than 5 μm . After nucleation, at the conditions used in the field, particles will grow to 5 μm rather quickly (2 s), compared to cloud droplet growth, because of the difference in RH_i and RH_w (142% and 104%, respectively). As such we should have a negligible contribution from undercounting ice crystals due to particles that nucleate ice but don't grow to 5 μm .

This is now included in the manuscript on page 14, lines 26 - 29:

“... the residence time of 8 seconds should also minimize the number of ice crystals $< 5 \mu\text{m}$, since ice crystals only need 2 seconds to grow by diffusion to sizes $> 5 \mu\text{m}$ at this high $RH_i = 140\%$. We believe undercounting INPs due to ice crystals $< 5 \mu\text{m}$ should not significantly influence the INP concentrations reported especially given the day to day variability in INP concentrations found at the field site studied in this work.”

Page 17: Authors use field measurements to validate the performance. However, there is an assumption made (although not mentioned in the manuscript) that aerosol properties (size, composition, morphology) have remained constant over all the years throughout winter. This may not be true and cannot use such data to validate the performance of the chamber. I suggest rephrasing the discussion to say that INP measurements results were comparable (Fig 9) with previous measurements but using different instruments.

We agree with the reviewer, and rephrased the revised manuscript on page 24, line 23 – 24:

“INP concentrations were comparable in different years, given that the minimum and maximum INP concentrations below water saturation overlap.”

To improve the readability I also suggest moving some of the text from section 2, 3 and 4 to supplementary. For example, section 2.3 can be shortened. Also, section 3.3 and 4.2.

We believe that the mentioned sections are required to understand the presented results in a complete format, and necessary to understand how the chamber works, as such we have opted to retain it in the main section of the manuscript.

References:

Kanji, Z. A., and Abbatt, J. P. D.: The University of Toronto Continuous Flow Diffusion Chamber (UT-CFDC): A Simple Design for Ice Nucleation Studies, *Aerosol Sci. Tech.*, 43, 730 - 738, 10.1080/02786820902889861, 2009.

Rogers, R. R., and Yau, M. K.: *A Short Course in Cloud Physics*, Pergamon, 1989.