Reply to reviewer #2: 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*.

Larcher et al. describe and characterize a new instrument (HINC) for detecting ice nucleating particles (INPs) in the atmosphere. They then use the instrument to quantify INP concentrations in the deposition mode and immersion mode at a high altitude research station. Concentrations of INPs during two winters are reported and two case studies during high concentrations of INPs are discussed. Since INPs play an important role in climate and the hydrological cycle this topic is well suited for ACP.

The paper is well written, and in most cases the results support the conclusions. The experiments and analyze are also laudable. However, I do have a few major concerns that need to be addressed before I recommend publication.

We would like to thank the reviewer for their comments, and address the major and minor concerns individually as presented below.

Major concerns:

Page 12, Line 11-12. The authors suggest that liquid droplets are not detected in the OPC channel > 5 micrometers because they settle out of the aerosol flow and hence are not sampled by the OPC. Is it possible that some of the ice crystals > 5 micrometers also settle out of the aerosol flow and are not sampled by the OPC? If so, does this mean that HINC only measures a lower limit to INP concentrations?

This is a valid question. We have now performed specific calculations for particle growth and settling taking into account the supersaturation profiles and flow speeds of the operation conditions in HINC as well as growth in the chamber as a function of time, which revealed that particles which activate into cloud droplets and grow to sizes of > 5 µm in the chamber are not lost by gravitational settling. Thus the previous statement on page 12, lines 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 µm. 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, lines 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 µm, 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 µm 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 µm 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."

These calculations also reveal that particles which activate into ice crystal grow to a size > 5 µm within 2 seconds, and can be lost by gravitational settling 7 seconds after activation. Taking into account that not only the particles need time to equilibrate when they are exposed to the supersaturation and temperature in HINC upon entering, which needs 0.2 seconds at the respective conditions, but also that ice nucleation can be a time dependent process, we believe by setting the residence time to 8 seconds, horizontal settling losses should not occur. To further validate this, we performed residence time experiments similar to those described in Kanji and Abbatt (2009), where microcline particles, which are ice active in the given temperature range, with an initial 400 nm dry diameter, were activated to ice crystals at 242 K at RH_w = 104% (Fig. A1 on page 37 in the appendix of revised manuscript, and below). These experiments reveal that the optimum residence time, which is the interplay between giving the INPs enough time to activate and grow to a size > 5µm, is indeed 8 seconds, when the AF reaches its maximum.

This is included now in the appendix, on page 37, line 3 – 8:

"*The optimum residence time for the ice crystal detection at the conditions used in the field experiments was determined by using 400 nm microcline particles which are INPs at this temperature (e.g. Atkinson et al., 2013). The position of the aerosol injector and thus the residence time was chosen accordingly, and the AF of the ice crystal concentration in the OPC size channel > 5 µm was determined (Fig. A1, appendix). The tests revealed the maximum AF at 8 seconds residence time, which gives the aerosols enough time to activate into ice crystals and grow to sizes > 5 µm, at the same time preventing gravitational settling losses.*"

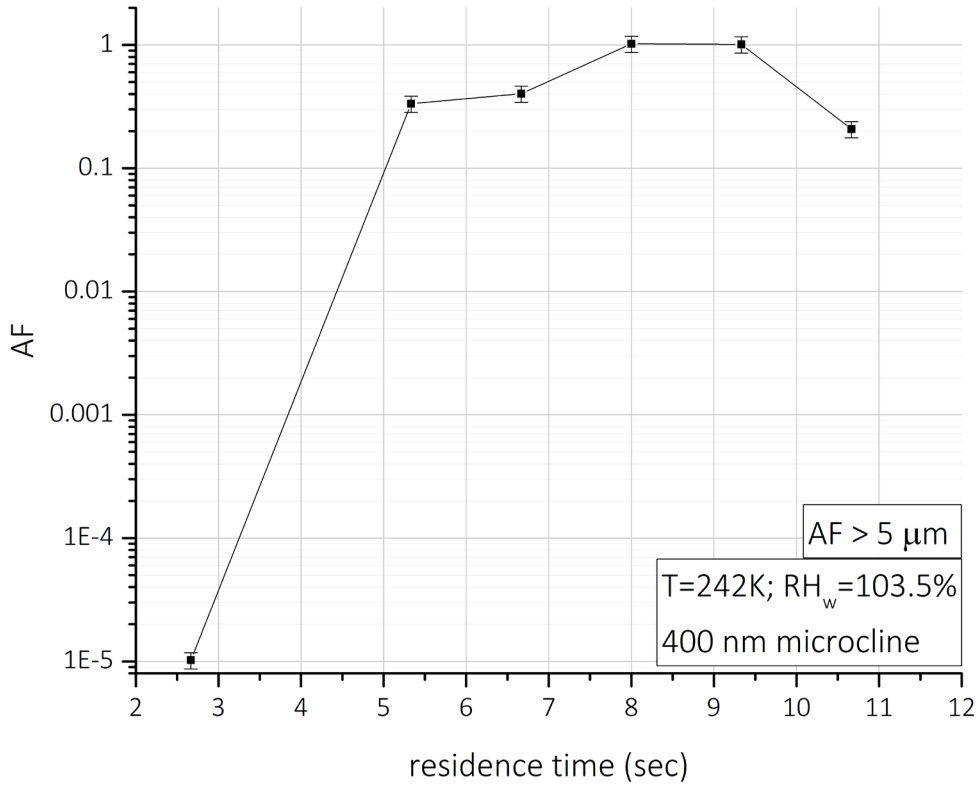

Fig. A1: AF as function of residence time for 400 nm microcline particles at 242 K and RH_w = 104%; data is the average over a total of three experiments at each residence time.

Still, the reviewer raises a valid point that the measured INP concentrations could be slightly underestimated because we cannot completely exclude a minor influence from settling in case a particle immediately activates in to an ice crystal (and settles out in 7 seconds) nor can we exclude the possibility that particles with an extremely high time dependence may not have enough time to grow (see response to reviewer 3). However, from numerous laboratory experiments of different aerosol components, this fraction should be negligible and not affect the presented INP concentrations. We believe that this is of minor importance especially in light of the day to day variability in INP concentrations observed in the field experiments. This is now discussed on page 14, lines 19 - 29:

"Diffusional growth of ice crystals for the respective conditions in HINC reveal that ice crystals can be lost to settling within 7 seconds. Still, residence time experiments with 400 nm microcline particles at 242 K and RH_w = 104% show that the AF is at a maximum value at 8 seconds (Fig. A1, appendix), which informed the 8 second residence time in the field experiments. This discrepancy to the theoretical calculation of 7 seconds is expected due to assumptions in the diffusional growth calculations, such as immediate activation upon entering the chamber and assuming spherical ice crystals. The residence time of 8 seconds therefore should include consideration for the equilibration time of the particles to the center supersaturation (~0.2 seconds), the growth time of ice crystals to > 5 µm (2 seconds) and time dependence for nucleation (up to ~6 seconds). Thus the residence time of 8 seconds should also minimize the number of ice crystals < 5 µm, since ice crystals only need 2 seconds to grow by

diffusion to sizes > 5 µm at this high RH_i = 140%. We believe that undercounting INPs due to ice crystals < 5 µm 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 18, Line 8-9: particle loss for 2 micrometer particles is large (44%). What is the transmission efficiency of unactivated particles > 5 micrometers (i.e. 6-10 micrometer particles)? Could a small percentage of unactivated 6-10 micrometer particles be detected as INPs in your experiments and cause experimental artifacts? As an example, would the conclusions in the paper change, if 1% of the 6-10 micrometer particles are unactivated in the HINC and are detected in the OPC channel > 5 micrometers. Could large (6 to 10 micrometers in diameter) primary biological particles cause experimental artifacts by making it through the HINC unactivated and being detected in the OPC channel > 5 micrometers?

According to particle loss calculations, due to the length of the tubing and position of the tubing upstream of HINC, 14 -50% of 5 – 10 µm particles respectively are lost before reaching HINC. Furthermore we performed tests with ambient particles to determine the transmission efficiency of particles through HINC taking into account all upstream tubing. We found that for ambient particles > 5 µm, 0% were detected downstream of HINC. In addition, the concentration for particles > 5 µm is naturally very low at JFJ (on the order of 0.05 $stdL^{-1}$), since these larger particles are not part of the free troposphere, and increases usually only occurs during SDEs (see comment below) and injections from boundary layer air. Therefore we do not believe that unactivated large particles will influence the INP number we report from HINC measurements. We now clarify this in the revised manuscript on page 19, line 19 – page 20, line 2:

"The experiments revealed a particle loss of 26% for 1 µm particles, and 44% for 2 µm particles, and 100% for particles > 5 µm therefore the OPC channel used to detect ice should not be contaminated with large (> 5µm) unactivated ambient particles."

And on page 20, lines 4 – 7 of the revised manuscript, we update the statement:

"In addition, calculations with the Particle Loss Calculator (von der Weiden et al., 2009) revealed that 0.8 % of 1 µm particles, and 2.6 % of 2 µm particles, and 14 – 50% of 5 – 10 µm particles should be lost in the inlet and tubing upstream of HINC which we consider to be negligible in light of the low abundance of ambient particles > 5 µm (on the order of 0.05 $stdL^{-1}$)."

We appreciate the idea to evaluate the contribution of 1% unactivated particles > 5 µm to INP concentrations, and calculated this based on the size distributions during the campaigns reported here. The highest recorded concentration during the campaigns in the size range > 5 µm, were 0.026 $stdL^{-1}$ and 0.285 $stdL^{-1}$ for winter 2015 and 2016, respectively, both during SDEs. These concentrations are low and do not affect INP concentrations, which are on average 2 – 4 orders of magnitude higher. Implicit in this evaluation is that the larger particles will not activate into ice crystals, which is unlikely.

We include this discussion for winter 2015 in revised manuscript now at page 33, lines 26 – 29:

"A calculation based on the size distribution of ambient particles from the field campaigns at the JFJ reveals that the maximum contribution of 1% aerosol particles > 5 µm remain

unactivated in HINC would be 0.026 stdL^{-1} (0.285 stdL^{-1}) in winter 2015 (winter 2016), during a time when INP concentrations reached 85.5 stdL^{-1} (154.5 stdL^{-1}). Thus a positive bias of larger unactivated particles to INP concentrations should be insignificant."

Page 33, line 11–12: Here the authors indicate that HINC avoids particle losses due to gravitational settling in the horizontally oriented chamber. What size of particles are the authors referring to at this point? I find this statement confusing since earlier they indicated that liquid droplets > 5 micrometers settle out and the transmission efficiency of 2 micrometer particles is low.

We agree, this is confusing. By particle losses here we meant ice crystal losses and we refer to the optimum residence time, which is an interplay between enough time for nucleation growth against too much time such that growing ice crystals are lost due to gravitational settling, as stated in the original manuscript on page 18, lines 3 - 5 (revised manuscript on page 19, lines 14 - 16):

"The injector position was set to an optimal residence time (8 sec) for the aerosol particles, which takes into account prevention of ice crystal losses due to gravitational settling in the chamber but yet allowing for enough growth time to reach an optical diameter of ≥ 5 μm."

The transmission efficiency that the reviewer refers to above for 2 μm being low is for aerosol particles larger than 2μm to enter the chamber given all the tubing and driers upstream of HINC and does not refer to the transmission efficiency and size of ice crystals that nucleate within the chamber.

However, as mentioned in the answer above, we performed now specific calculations and come to the conclusion that at the respective sampling conditions used in the field experiments, which the laboratory experiments aim to validate, no particle settling loss occurs for cloud droplets at 242 K and RH_w = 104% and as such this sentence has been removed (see comment above with Rogers and Yau (1989) reference).

Page 13, line 4-6: I appreciate that the authors have carried out several systematic studies to determine the upper RH limit for ice crystal detection in the immersion mode. However, I am not completely convinced that at T=242K and RHw < 104 % the OPC size channel > 5 micrometers is well-suited to reliably detect ice crystals in ambient conditions without experimental artifacts. The authors suggest that these experimental conditions are appropriate based on measurements with 200 nm sulfuric acid and 200 nm ammonium sulfate particles, as well as ambient particles.

However, perhaps the results from these test cases are not applicable for all air masses encounter at Jungfraujoch. For example, do the results from the test cases apply to 50 nm secondary organic aerosol and 50 nm sea spray particles? What about 800 nm particles? Since the authors have not investigated the effect of particle size or chemical composition (other than sulfuric acid, ammonium sulfate and one ambient situation), I do not know the answer to this question. Also, how representative were the measurements with the ambient particles shown in Figure 4? Were the ambient measurements shown in Figure 5 only carried out on one day or one type of air mass? At T=242K and RHw < 104 % perhaps the OPC size channel > 5 micrometers is not well-suited to reliably detect ice crystals in air masses

influenced by marine origin. Additional discussion and possibly additional results are needed to address these questions.

To clarify and support our results, we include now the above mentioned calculations for diffusional growth of cloud droplets (on page 14, lines 2 - 9), indicating that for initially 200 nm dry size particles water droplets grow to a size > 5 µm only at a RH_w > 107%, when they can be misclassified as ice. However, our measurements should not be affected by this conditions since we conduct our experiments at $RH_w \leq 104\%$. Using sulfuric acid and ammonium sulfate represents the upper limits in hygroscopicity thus we can be confident that water droplets growing from less hygroscopic ambient particles will not contaminate the 5 µm OPC size channel.

Based on the growth calculations, particles with an initial diameter of 200 nm exposed to 242K and RH_w = 104% can grow to 4.038 µm in diameter, and with an initial particle diameter of 50 nm and 800nm to 4.035 µm and 4.08 µm, respectively, showing that these differences in the initial size are not relevant.

The size and chemistry of the aerosol particles are crucial for the cloud droplet activation, with aerosol size being the dominant factor (as found by Dusek et al. (2006)), however our calculations reveal that the initial particle size is of minor importance for the diffusional growth of the cloud droplets to a size of 5 µm in diameter. Hence the water droplets surviving into the OPC ice detection channel at the given sampling condition is not changing by applying a 50 nm, 200 nm or a 800 nm initial dry diameter, since neither of them can grow by diffusion to 5 µm for our sampling conditions. The chemical effect on activation and subsequent diffusional growth of cloud droplets is negligible, because we run the chamber at RH_w = 104% thus for droplet activation a difference between 50 nm secondary organic aerosol and 50 nm sea spray particle should not be of concern because at these high RH both should activate. In addition, even very hygroscopic particles like sulfuric acid should not reach a 5 µm diameter as discussed in the manuscript.

The same insensitivity of diffusional growth on the initial particle size is found for the diffusional growth of ice crystals: At the same conditions of 242 K and RH_w = 104%, ice crystals growing from particles with initial sizes of 200 nm, 50nm and 800 nm reach a final size of 6.8 ± 0.1 µm. Here the chemistry might play an important role as well, but as it was found in a previous study by Petters et al. (2009), investigating the ice nucleation of biomass burning particles, that these less efficient INPs activated within a residence time of 4 - 5 seconds. This gives us thus confidence by applying a residence time of 8 seconds in our ice chamber, that we are able to detect INPs with a range of efficiencies.

For clarification we add to the revised manuscript on page 14, lines 9 – 15:

"These calculations also reveal that the diffusional growth of the activated cloud droplets, and hence the final size of the cloud droplets of interest, which is 5 µm for the discussed field experiments, are insensitive to the initial dry diameter of the aerosol particles, since the final droplet size at 242K, RH_w = 104% and a residence time of 8 seconds of an initial 50 nm, 200 nm and 800 nm is 4.035, 4.038 and 4.08 µm, respectively. Also effects of the particle chemistry are assumed to be negligible for the droplet activation, since we conduct our experiments at $RH_w \geq 104\%$ where a variety of aerosol chemical compositions should activate into droplets. Thus water droplets contaminating the 5 µm channel should not occur even with varying hygroscopicities and sizes of aerosol populations."

By referring to the insensitivity of the final size of cloud droplets to its initial dry diameter, we believe that our results for ice crystal particle concentrations are not biased by any sampled aerosol population, since none of them can grow to a size of > 5µm, and cannot be falsely classified as ice.

Minor concerns:

Abstract, Page 2, Line 17-19: The evidence for marine aerosol acting as INPs is circumstantial. Hence, I think "indicating" should be replaced by "possibly indicating" or "consistent with". In the abstract, the authors should also point out that during the event influenced by marine air, they cannot rule out contributions from anthropogenic or other sources.

Agreed and changed into *"suggesting"* (now in abstract, page 2, line 20).

The contribution from anthropogenic or other sources to the marine air mass event is now mentioned in the abstract (page, 2, line 17):

"The contribution from anthropogenic or other sources can thereby not be ruled out."

Page 4, Line 6-8: This sentence refers to reports more than five decades ago, but then references a paper published in 2014. Either remove the 2014 reference or modify the sentence for consistency.

We removed "*Cziczo and Froyd, 2014"* as a reference (revised manuscript, page 4, lines 7 – 8).

Figure 2: Indicate in the figure caption that the legend on the right hand side refers to different size channels of the OPC.

Included now in the caption of Fig. 2 on page 10, line 2 - 3:

"…in the size channel 0.5 – 2 µm (dark grey) and 2 – 8 µm (light grey)."

Page 20, Line 18: please indicate the purity of the nitric acid solution.

We added *"(65%)"* to the revised manuscript on page 22, line 3.

Page 23, Line 29: The authors state that the median concentrations are less than or equal to 0.2 stdL-1 during winters 2015 and 2016. This statement appears to contradict Fig. 8, where the median values are greater than 0.5 stdL-1 for winter 2015. Am I missing something here?

This value is the median of 2015 and 2016 together, not to the individual field campaigns. It refers to table 2, which is now explicitly mentioned on page 24, line 21:

"… for winters 2015 and 2016 (Table 2, third row)."

Page 27, line 19-21: Could primary biological particles also be important for land not covered by snow.

Due to the fact that the ambient temperatures at JFJ were below -20°C at the 2nd February, it is unlikely that biological particles contributed to the measured peak INP concentrations, since they would have been activated at warmer temperatures during transport to the JFJ and subsequently fallen out prior to arrival at JFJ (Conen et al., 2015;Stopelli et al., 2015). This is mentioned in the revised manuscript on page 5, lines 14 – 16:

"…previous INP activation and subsequent fall-out from advected air masses prior to reaching the JFJ, leaving an air mass which is depleted in INPs upon arrival to the JFJ (Conen et al., 2015; Stopelli et al., 2015)."

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