

Reply to reviewer #1: 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*.

The paper presents a new ice nucleation counter (HINC) for measurements of ice nucleating particles at conditions relevant for mixed-phase clouds at the Jungfraujoch. Generally the paper is well written with few typos – I have hardly any issues with the approach and style of presentation.

We thank Reviewer 1 for the comments.

However, I have some concerns about what the measurements actually show and how they can be used quantitatively, so I have focussed on these points in my review. If left unaddressed I feel that the paper will potentially lead to a great deal of confusion between various groups working in similar areas.

The reviewer’s comments and calculations are appreciated, and we believe that responding to these will help clarify the manuscript. We hope by responding to concerns and making the corresponding changes stated below, especially with regard to the validation experiments, that confusions on interpretation will be completely circumvented.

Firstly figures 2, 3, 4, and 5 present scans from the HINC instrument. The y-axis is labelled AF, which I assume is short for “activated fraction”; however, I do not think this is a good description because they each refer to different things: for example, figure 2 is number of ice crystals nucleated; figures 3 and 4 are fraction of water drops above a certain size (although a small fraction could still be ice crystals); figure 5 is refers to particles that grow above a certain size and eventually activate. Below 100% particles will not be activated in any case, so these should not be referred to as activated fraction.

The activated fraction is defined in the revised manuscript on page 8, lines 2 – 3:

“The activated fraction (AF) which is the ratio of aerosol particles which activated into cloud droplets or nucleated ice crystals, respectively, to the number of total particles...”

Thus we believe that it is suitable to describe Fig. 2, showing the fraction of nucleated ice crystals, and Fig. 3 and 4, which refers to the activation of water droplets and possibly nucleated ice crystals. Indeed, the term should not be used for Fig. 5, which shows deliquescence and subsequent growth. The term in the original manuscript on page 8, line 18 “AF” has been changed to (revised manuscript, page 9, lines 9 - 10)

“The observed increase in the particle fraction due to deliquescence and hygroscopic growth compares well to literature results reported to be $RH_w = 77 \pm 2.5 \%$ (Koop et al., 2000b).”

as well as in the caption and y-axis labelling of Fig. 5 (revised manuscript page 13, line 2).

Figure 2 from the paper presents the homogeneous freezing curve of 100nm dry diameter H₂SO₄ particles at 233K (-40C), which according to Koop et al. (2000) will freeze when the RH increases and the particles take up water and become dilute enough. I have reproduced the figure from the paper below (Figure 1) and superimposed the fraction of ice crystals nucleated (calculated with a model that uses the equations for homogeneous nucleation rate by Koop et al. 2000).

The model assumes that there is a ramp in RH that occurs over 100 seconds. This time period was purely arbitrary for demonstration purposes, although with a shorter ramp in RH (8 seconds) the onset of nucleation occurs even later and has a lower peak.

Here we would like to point out that the particles going through the chamber are in continuous flow, so the particles themselves are not exposed to a ramp but more to a step function between dry conditions prior to entering the chamber (~ RH < 2%), and upon entering the chamber to the respective conditions of RH in the chamber (as indicated on the x-axis of Fig. 2 in the manuscript). The usual time of a relative humidity ramp in HINC is ~20 minutes, but particles continuously flow in and out of HINC thus being exposed to a constant RH for only 8 seconds residence time, i.e. the same particles are not being exposed to a ramp over 8 seconds, but instead are exposed to a constant RH over 8 seconds as such there could be a time limitation for homogeneous freezing.

For clarification, we added to the original manuscript on page 8, line 4 the following sentence (page 8, lines 8 - 11, revised manuscript):

“In the respective experiments the RH_w conditions in HINC were increased at an approximate rate of 0.5% min⁻¹, so that an increase in RH_w of ~10% was achieved over a total time of 20 minutes, which implies that during the 8-second aerosol residence time in HINC, the particles experienced constant RH conditions.”

In the calculations at 99% RH the physical size of the particles is approximately 0.4 microns diameter, corresponding to a diameter growth factor of 4 and a liquid water volume of approximately 3e-20 m³.

According to the Koop et al. (2000) paper, the nucleation rate under these conditions is approximately 1.51e+16 m⁻³ s⁻¹, so we may estimate the fraction of droplets frozen in 1 second as:

$$f = 1 - \exp(-1.51 \times 10^{16} \times 3 \times 10^{-20} \times 1)$$

which is around 5e⁻⁴ for the activated fraction.

The above equation for frozen fraction is derived from the usual 1st order rate equation used to approximate homogeneous nucleation (J is the nucleation rate and V is the drop volume):

$$dN/dt = -NJV$$

The full results of this time-dependent calculation are shown in Figure 1, below. The blue line assumes that the Van't Hoff factor is 2, while the green line assumes it is 3 to span a range relevant to H₂SO₄). As can be seen there is a marked difference between the theoretical curve and the measurements.

The measurements suggest that the ice crystals nucleate starting at around 97% RH, while the theory suggests it is 99% RH.

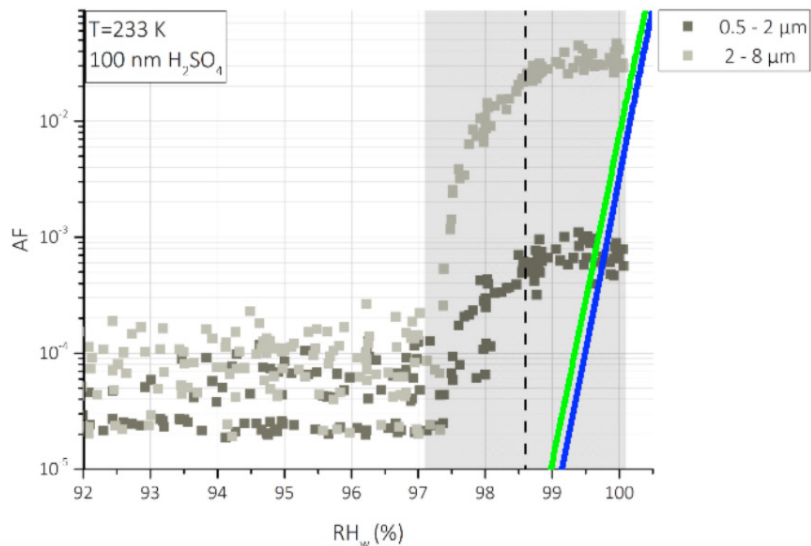


Figure 1. Reproduced Figure 2 and theoretical curves superimposed (blue line assumes a van hof factor of 2, while the green line assumes a Van't Hoff factor of 3).

As shown by the vertical dashed line in Fig. 2, we have indicated the value based on Koop et al. (2000a), for the expected RH for nucleation of 100 nm solution droplets, which suggests the homogeneous freezing threshold for these conditions at $RH \geq 98.6\%$, which is consistent with the $RH = 99\%$ suggested by the reviewer. We assumed a fixed nucleation rate of $10^{10} \text{ cm}^{-3}\text{s}^{-1}$ (Koop et al., 2000a), which is now added to the revised manuscript on page 8, lines 13 - 14:

“... based on a fixed nucleation rate coefficient of $10^{10} \text{ cm}^{-3}\text{s}^{-1}$ (Koop et al., 2000a), resulting in a RH_w of 98.6%...”

The RH reported for the chamber on the x-axis of Fig. 2 is a nominal centre RH in the chamber (original manuscript, page 14, lines 11 - 13; revised manuscript page 16, line 12 – page 17, line 1):

“...there is a temperature variation of $\pm 0.4 \text{ K}$ 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 $\pm 1\%$ ($RH_i \pm 2\%$). This translates into a calculated total uncertainty of $RH_w \pm 2\%$ ($RH_i \pm 3\%$) at 242 K and $RH_w = 104\%$.”

As such a fraction of the aerosol particles experience a higher RH than the nominal center value reported. In order to visualize this variation in RH including the uncertainty, we plotted the grey area as the total uncertainty at 233 K and $RH_w = 98.6\%$, which is $RH_w \pm 2\%$ ($RH_i \pm 3.5\%$) to present both the variation in RH and the uncertainty in RH from temperature in the aerosol layer.

In Fig. 2 of the revised manuscript, the grey area is the associated variation/uncertainty in RH that particles would experience if the chamber were set to the theoretical RH of 98.6% (as indicated by the vertical dashed line). As such it is very likely that the increase in AF seen at 97.5% is indeed due to a fraction of particles within the aerosol layer that are in fact exposed to higher RH of 98-99% required for homogeneous freezing as shown by the vertical dashed line and the lines supplied by the reviewer. We include this discussion now in the revised manuscript on page 8, lines 14 - 20:

“The reported RH_w on the x-axis in the figure represents the nominal conditions at the center line of the chamber, which is the center of the aerosol layer. Due to the width of the aerosol layer, the particles are exposed to a variation in $RH_w \pm 1\%$, and to an uncertainty in $RH_w \pm 1\%$ due to the temperature uncertainty. The grey shading in Fig. 2 represents this total calculated uncertainty of $RH_w \pm 2\%$, for a prescribed $RH_w = 98.6\%$. When the chamber is set to an $RH_w = 98.6\%$, the aerosols can be exposed to a range of $96.6 - 100.6\%$. Our experiments reveal an increase in the AF of particles between $2 - 8 \mu\text{m}$ starting at 97.5% , and reaching a plateau value at 99.5% , which is in excellent agreement to the expected range of freezing within the aerosol layer.”

Also the caption of Fig. 2 (revised manuscript on page 10, lines 4 - 5) is updated accordingly:

“... and the shaded region indicates the calculated range of RH_w and uncertainty to which the particles in the aerosol layer in HINC are exposed to.”

There are also differences in the shape and height of the frozen fraction curve.

Moreover, the RH variation in the aerosol layer also explains the difference in shape of the AF curve i.e., a quasi-step function (i.e. more gentle slope) and progressive AF as a function of RH_w rather than a steep step function of RH_w , since the particles are exposed to a distribution of RH, and not a discrete value. Thus some particles activate earlier, at the upper end of the RH_w variation and start growing into a detectable size range of the OPC, whereas a fraction of the particles appear to activate delayed that are exposed to the lower end of the variation in RH_w .

There are discrepancies between the observed AF in HINC and the reviewer's modelled AF. The reviewer calculated AFs based on e.g. a physical particle size of e.g. 400 nm at $RH_w = 99\%$ and a nucleation rate of $1.5\text{E}+10 \text{ cm}^{-3}\text{s}^{-1}$, according to Koop et al. (2000a), and a nucleation time of 1 second.

However, the reported AF in Fig. 2 in the manuscript is measured after a maximum nucleation time of 8 seconds, which is the residence time in HINC. Using 8 seconds, we expect at the given conditions a higher AF than the $5\text{E}-4$ suggested by the reviewer.

In order to verify this, we performed the same calculations as performed by the reviewer. For the physical size at the respective humidity conditions we use Koehler theory as calculated with the E-AIM (<http://www.aim.env.uea.ac.uk/aim/aim.php>) and find a physical size of the initial $100 \text{ nm H}_2\text{SO}_4$ at 99% of $0.33 \mu\text{m}$ in diameter, which might explain some of the discrepancies compared to the value of $0.4 \mu\text{m}$ calculated by the reviewer.

The AF results from using the drop diameter of $0.33 \mu\text{m}$, homogeneous nucleation rate coefficient of $1.5\text{E}+10 \text{ cm}^{-3}\text{s}^{-1}$ and a nucleation time of 1 and 8 seconds are shown in the figure below. We indeed observe differences in the AF between the measured and the modelled AF at 1 second nucleation time, but a good agreement for a nucleation time of 8 seconds. Thus the nucleation time of 1 second is not representative for our measurements in HINC, and explains the observed differences by the reviewer.

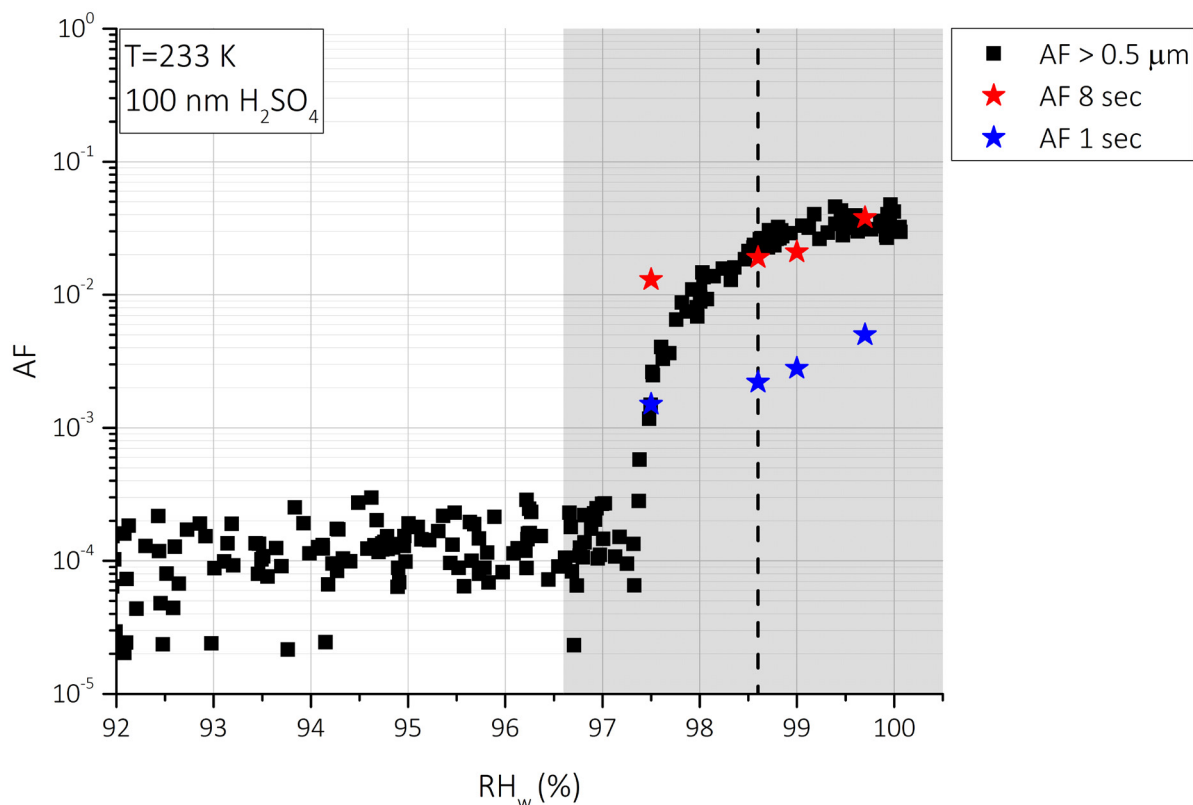


Fig. 1.2: AF as a sum as function of RH_w at 233 K for initial 100 nm H_2SO_4 ; black markers refer to observed AF in OPC size channel $> 0.5 \mu m$ in HINC; blue stars are based on a nucleation time of 1 second, and red stars on 8 seconds; grey area refers to the calculated RH variation and uncertainty.

This is now discussed in more detail in the revised manuscript on page 8, lines 20 - 24:

“As a result of the RH variation, the shape of the AF is not as steep a function of RH_w as the theoretical lines, but rather a steady increase within the range of RH_w to which the aerosol layer is exposed to. According to theoretical calculations (Koop et al., 2000a) at 233 K and 98.6% (99%) RH_w and resulting nucleation rates, we expect for initial 100 nm H_2SO_4 particles at a residence time of 8 seconds an AF of 0.03 (0.04), which is in agreement with the observed AFs in Fig. 2.”

I would suggest that these points needs some discussion, otherwise it will lead to confusion in the literature between measurement and models. It is not clear to me whether this is a problem with the theory by Koop et al. or with the measurements presented in the paper.

Considering the above discussion, the observed differences between theory and experiments presented here are explained by the variation in T and RH conditions in the aerosol layer, and by the nucleation time available for the particles.

Furthermore Figure 3 from the paper presents the “warm activation” curve for 200nm dry size H_2SO_4 particles. The temperature of 243K (-30C) is high enough so that it is not affected

too strongly by homogeneous freezing, so the data represent the growth of aerosol particles into cloud drops. The plot is reproduced below in Figure 2 and shows the activated fraction of 200nm H₂SO₄ particles given different assumptions about the size of an activated particle.

One can also do a theoretical calculation to infer the critical humidity where particles grow into droplets by finding the maximum value of the Koehler curve (note we have used the assumption of ideality for the activity of water term).

$$RH_{eq} = \frac{n_w}{n_w + \nu n_s} \exp\left(\frac{4M_w \sigma}{RT\rho D}\right)$$

for $n=3$; $n_s=7.86e^{-17}$ moles; $s=0.084 \text{ Nm}^{-1}$ (at 243K); $M_w=0.018 \text{ kg mole}^{-1}$; $R=8.314 \text{ J mole}^{-1} \text{ K}^{-1}$; $T=243 \text{ K}$, $r=1000 \text{ kg/m}^3$, a minimisation routine finds that $n_w=1.11e^{-11}$ moles; $D=7.26e^{-6} \text{ m}$ and $RH_{eq}=1.00038$.

So for dry 200nm (spherical) H₂SO₄ particles the critical humidity is approximately 100.04%, much less than the >102% suggested by the plot in Figure 2. The above theoretical values are more consistent with text books (see Figure 6.3, page 175, of Pruppacher and Klett, 1997, for dry NaCl for example).

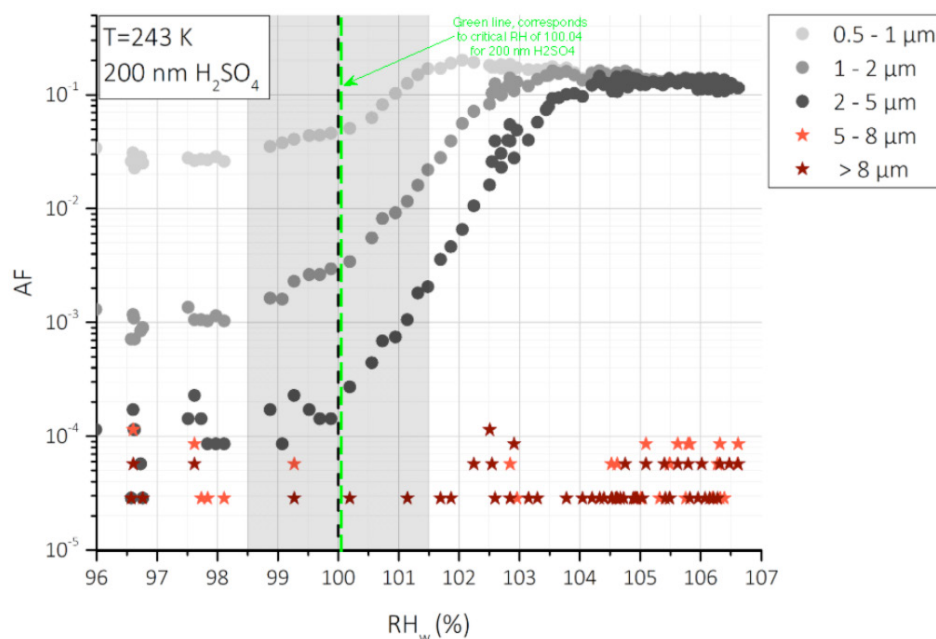


Figure 2. Reproduced from Figure 3 in the paper, with the green line showing the theoretical critical humidity for the activation of 200nm H₂SO₄ particles.

The experiments with 200 nm H₂SO₄ particles aims to determine the onset of cloud droplet formation, which should occur slightly above 100% RH_w based on Koehler theory and indicated by the vertical dashed black line in Fig. 3 of the manuscript (Fig. 2 above) and confirmed by the calculations of the reviewer (onset at 100.04% RH_w). The RH of 102 %, to which the reviewer refers to, is the RH at which the maximum in the AF is reached, and not the RH at which the particles start to activate into cloud droplets and grow to detectable sizes. Because of the RH variation (and uncertainty) in the chamber, we expect a fraction of the particles to activate later than the nominal RH indicated on the x-axis (see discussion above), but what is crucial is that a change in the size is observed for a fraction of the particles which are indeed exposed to the nominal centre RH (indicated on the x-axis). To clarify this confusion, we modified the manuscript on page 8, line 26 - 27:

“We thereby refer to the onset of cloud droplet formation, hence the first observed increase in the AF at a given size which represents cloud droplets.”

Also, captions of Fig. 3, 4 and 5 are updated accordingly by adding (revised manuscript, page 11, lines 3 - 4; page 12, lines 3 - 4; page 13, line 3):

“Vertical dashed line represents expected onset for cloud droplet formation; ...”

In Fig. 3 an increase of particle concentrations in size channels $> 0.5 \mu\text{m}$ occur at $\text{RH}_w = 99\%$, with a steeper increase at $\text{RH} = 100\%$. This is expected as a fraction of the particles in the aerosol layer in HINC are exposed to a RH higher than 99% when the nominal RH of the chamber is set to 99% because of the variation in RH that the aerosol layer is exposed to (as discussed above). We now make this clear in the discussion on page 8, lines 29 - 31:

“The grey shaded area in Fig. 3, 4 and 5 also includes the calculated variation and uncertainty in RH_w for the given temperature of 242 and 243 K, respectively. As discussed above, the exposure of the aerosol particles to this variable RH lead to deviations from the theoretically calculated critical RH.”

Also the captions of Fig. 3, 4 and 5 (revised manuscript, page 11, line 4; page 12, line 4; page 13, lines 4 - 5) were updated accordingly

“...grey area refers to the calculated variation and uncertainty of RH in the aerosol layer.”

Given the above discrepancies for size selected particles it is difficult to know how to interpret figure 4 from the paper, which is based on passing ambient poly-disperse particles through the instrument.

Fig. 4 aims to determine the onset of cloud droplet formation for ambient particles, which should occur slightly above 100% RH_w based on Koehler theory as stated in the manuscript and confirmed by the calculations of the reviewer (onset at 100.04% RH_w) and as indicated by the dashed vertical line.

We note that there is a slight increase in particle concentration in the smaller size channels ($0.5 - 1 \mu\text{m}$) between RH_w 99-101% but a more steep increase starting at a RH_w of 101% for the OPC size channel $0.5 - 1 \mu\text{m}$, followed by an increase in the $1 - 2 \mu\text{m}$ and $2 - 5 \mu\text{m}$ channel. The increase in particle activated cloud droplet fraction over the range can be explained by the RH variation and uncertainties in the aerosol layer. Furthermore, we expect to have a significant fraction of particles $< 100 \text{ nm}$ in the ambient air, and also particles with hygroscopicities lower than that of sulphuric acid, which supports the slightly higher observed RH for activation into cloud droplets at this temperature. To make this point clear, the respective discussion is updated in the revised manuscript on page 8, line 31 – page 9, line 6 (original manuscript page 8, lines 9 – 15):

“We note that an increase in the AF of initial 200 nm H_2SO_4 is observed prior to $\text{RH}_w = 100\%$ in the 0.5 and 1 μm channels (Fig. 3) which is to be expected due to hygroscopic growth of the H_2SO_4 particles. Therefore, the increase in size for $\text{RH}_w < 100\%$ is only observed in the smaller size channels occurring prior to droplet activation at $\text{RH}_w = 100\%$, while an increase at $\text{RH}_w = 100\%$ in the $> 2 \mu\text{m}$ channel is observed due to cloud droplet activation. On the other hand, the ambient particles show droplet activation in the $> 0.5 \mu\text{m}$, $> 1 \mu\text{m}$ and $> 2 \mu\text{m}$ channels at $\text{RH}_w = 101.5\%$ (Fig. 4). This is likely due to the lower hygroscopicity of the ambient particles compared to H_2SO_4 and due to a larger fraction of the sampled ambient particles being $\ll 100$

nm, requiring higher RH for the droplets to activate and grow to detectable cloud droplet sizes at this temperature.”

The following sentence on page 8, line 15 (original manuscript)

“... but could also be compounded by RH uncertainties (see sect. 2.3).”

is updated in the revised manuscript on page 9, lines 6 – 7:

“In addition, the experiments could also be influenced by RH uncertainties (see sect. 2.3).”

Moreover, prompted by this review, we re-visited performed calculations for particle settling, also taking into account the supersaturation profiles and flow speeds at respective conditions in HINC, which reveal that cloud droplets will only grow to $> 5 \mu\text{m}$ if $\text{RH}_w \geq 107\%$. Thus the increase at RH_w 104 - 105% observed in Fig. 4 in the size range $> 5 \mu\text{m}$ is believed to arise from a small fraction of ambient particles that act as INP and form ice crystals heterogeneously at these conditions, since water droplets do not grow to this size at $\text{RH} = 104\%$. 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 $> 5 \mu\text{m}$. 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, 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 $4 \mu\text{m}$ in HINC at 242 K, $\text{RH}_w = 104\%$ for a residence time of 8 seconds (conditions used for field experiments reported here), giving us confidence that droplets are not detected in the $5 \mu\text{m}$ channel. Only at an RH_w of 107% cloud droplets grow to $> 5 \mu\text{m}$, and therefore by conducting our experiments at $\text{RH}_w = 104\%$, we only detect ice crystals in the $5 \mu\text{m}$ 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 $\text{RH}_w = 104 - 105\%$ 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.”

Finally, figure 5 in the paper shows a humidity scan at 238K for 200 nm NaCl particles. This scan has two traces – one for particles larger than 1 micron and the other for particles larger than 0.3 microns. The “larger than 1 micron” curve approaches an activated fraction for $\text{RH} > 102\%$, whereas the “larger than 0.3 micron” curve approaches an activated fraction for $\text{RH} > 92\%$ or so. A theoretical calculation (Figure 3) shows that the physical size of 200 nm NaCl particles will always be greater than 0.3 microns for humidities $> 80\%$. If this is the case, the figure 5 should have the activated fraction for 0.3 micron particles equal to 1 for all $\text{RH} > 80\%$. Additionally, the physical size of these particles exceeds 1 micron for an RH of 99.5%, not 102% (as suggested by the data in figure 5). Hence, the activated fraction curve should go to 1 for $\text{RH} = 99.5\%$, and not 102% (as in the paper).

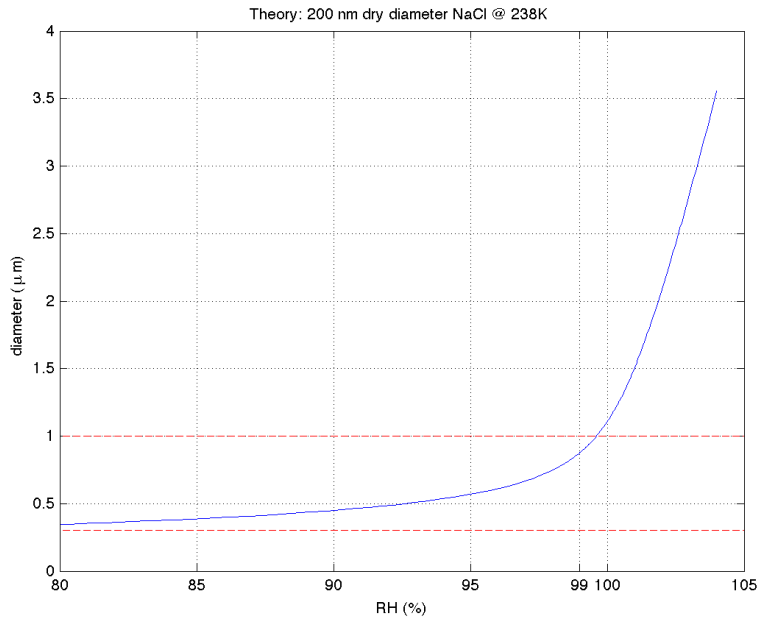


Figure 3. Theoretical calculation of the growth of 200 nm NaCl particles. Bottom red line is $y=0.3$ microns, top red line is $y=1$ micron.

The uncertainty in size selection with the DMA is $\pm 3.5\%$, resulting in a size distribution of size selected particles in the range of 200 ± 7 nm. However, in reality this should be much larger, because NaCl particles are not perfectly spherical, thus one can expect a pseudo-mono disperse population. In addition, since the particles are charged prior to passing through a DMA, there exists a substantial doubly or triply charged population of particles (8%, 2%, respectively) meaning that 10% of particles can be in the size range 320-440 nm of the population of NaCl sampled.

Because there is a breadth in the size distribution (as already acknowledged by the reviewer) in addition to the multiply charged particles, a fraction of NaCl particles can grow to sizes larger than $1 \mu\text{m}$ which is demonstrated by the increase already at $\text{RH}_w = 80\%$ (in the particles $>1 \mu\text{m}$ trace), with a gradual increase in AF for this trace as the progressively smaller particles in the size distribution grow hygroscopically to sizes larger than $1 \mu\text{m}$. This is also true for the particles $> 0.3 \mu\text{m}$ trace. The fact that the increase to an AF of 1 is not a step function like the increase observed at $\text{RH}_w \sim 80\%$ suggests that the particle properties are different i.e. the breadth in the size distribution (and the variation in RH experienced by the aerosol layer). The RH_w variation in HINC can only account for a small delay $\sim 2\%$, which would explain why complete activation as the reviewer states is only observed at $\text{RH}_w \sim 102\%$. In addition, the OPC sizing and counting accuracy at sizes as small as $0.3 \mu\text{m}$ is of a lower accuracy, since the wavelength of the laser (780 nm) is similar to the size of the particles and thus the absolute values in this case should be less significant and it is more crucial to observe a change in the signal.

Since the purpose of this experiment is to show that we can observe growth at $\text{RH}_w = 80\%$, and this would only be possible due to the deliquescence RH being surpassed at $\text{RH}_w = 77\%$. I.e. the fact that we don't see the onset of the growth (and hence preceding phase change) at 90% or already at 70% suggests that the deliquescence of NaCl should have taken place in the expected range of 77% which could only then have allowed for hygroscopic growth and increase in size that was detected by HINC at $\text{RH}_w = 80\%$.

The goal is to demonstrate that within uncertainties, we can prescribe the RH in HINC. As shown, complete activation of the particles is only achieved at $RH_w > 100\%$ (and not at $RH_w = 100\%$) which also informs our decision to perform our immersion/condensation freezing experiments for INP at $RH_w = 104\%$, to be above water saturation for the entire aerosol lamina. We do not suggest that HINC should be used to accurately determine deliquescence RH, but rather use the deliquescence concept to infer that HINC is able to achieve with reasonable accuracy the RH we expect in the center by setting the wall temperature.

This discussion is now included in the manuscript on page 9, lines 11 - 28:

“We observe a first strong increase in the particle fraction > 0.3 and $> 1 \mu\text{m}$ at 80 - 81%, followed by a gradual increase in the particle fraction to unity (within uncertainties) at $RH_w \approx 94\%$ for the $> 0.3 \mu\text{m}$ trace. Deliquescence is a phase change and not a growth process, and a delay as compared to the literature value (dashed line Fig. 5) is expected, since the deliquesced particles need to grow to a size $> 0.3 \mu\text{m}$ to be detected in the OPC. In theory we would expect all particles to grow to sizes larger than $0.3 \mu\text{m}$ at $RH_w \geq 80\%$ since the deliquescence and growth threshold has been reached. However, we note that due to an uncertainty in sizing of up to 3.5% in the DMA, particles between 193 and 207 nm for a nominal size of 200 nm will be sampled. In reality, we expect an even broader size distribution because dried NaCl particles are aspherical and result in larger sizing errors (Ardon-Dryer et al., 2015). Due to the size selection method with the DMA, a non-negligible fraction of larger particles (10%) between 320 - 440 nm (from double and triple charged particles) will also be sampled by HINC. This breadth in size distribution may explain the initial increase in particle fraction at $RH_w = 80\%$ arising from the multiply charged particles followed by a progressive increase in the particle fraction up to $RH_w = 94\%$ where all the particles grow to sizes $> 0.3 \mu\text{m}$. The same can be said for the $> 1 \mu\text{m}$ trace. Note that complete activation in this trace occurs at $RH_w > 100\%$, which is expected from the variation in RH in the aerosol layer. Finally, we note that the goal of this experiment is to demonstrate that HINC can achieve prescribed RH conditions with reasonable accuracy by controlling the wall temperature as is seen by the onset in growth at $RH_w = 80\%$ in Fig. 5. Additionally, we acknowledge that the fraction of particles $> 0.3 \mu\text{m}$ reaches a maximum at higher RH_w than theoretically expected, which can also be attributed to the sizing and counting uncertainty of the OPC, which is most pronounced at these small particle size, when the wavelength of the laser (780 nm) is similar to the diameter of detectable particles.”

I understand that size-selected particles will also have a distribution in size, and this may affect the results slightly. It would be worth discussing the breadth of the size-selected distribution to allow readers to better understand the measurements being presented.

We agree with the reviewer's comment and by including the above mentioned statement on page 9, lines 11 - 28 we hope to have addressed this point.

However, at present, given the above difficulties my suggestion is for a major revision of the manuscript to either calibrate any systematic biases, or to give clear reasons for the apparent inconsistencies.

We have now given clarifications for the parts that were found to be confusing for the reviewer.

References:

Ardon-Dryer, K., Garimella, S., Huang, Y. W., Christopoulos, C., and Cziczo, D. J.: Evaluation of DMA Size Selection of Dry Dispersed Mineral Dust Particles, *Aerosol Science and Technology*, 49, 828-841, 10.1080/02786826.2015.1077927, 2015.

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, *Nature*, 406, 611-614, 10.1038/35020537, 2000a.

Koop, T., Kapilashrami, A., Molina, L. T., and Molina, M. J.: Phase transitions of sea-salt/water mixtures at low temperatures: Implications for ozone chemistry in the polar marine boundary layer, *J. Geophys. Res. Atmos.*, 105, 26393-26402, 10.1029/2000JD900413, 2000b.

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