

Review of “The Horizontal Ice Nucleation Chamber HINC” (by Larissa Lacher, et al.)

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. 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.

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.

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.

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):

$$\frac{dN}{dt} = -NJV$$

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

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.

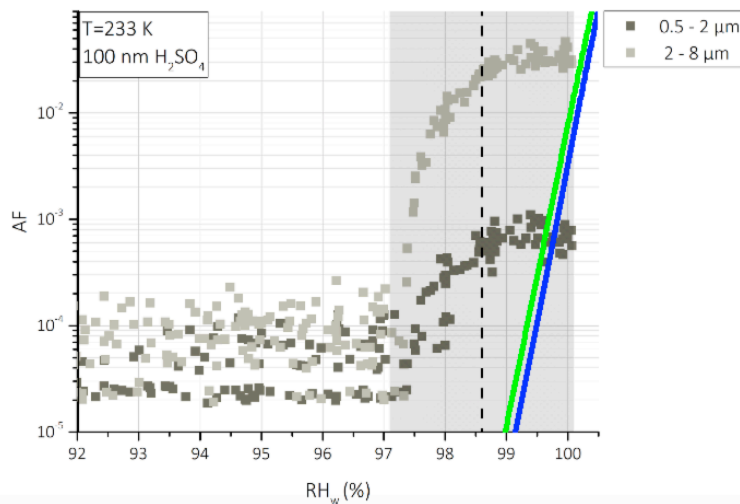


Figure 1. Reproduced Figure 2 and theoretical curves superimposed (blue line assumes a van hof factor of 2, while the green line assumes a Van't Hoff factor of 3).

Furthermore Figure 3 from the paper presents the “warm activation” curve for 200nm dry size H₂SO₄ 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) .

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

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).

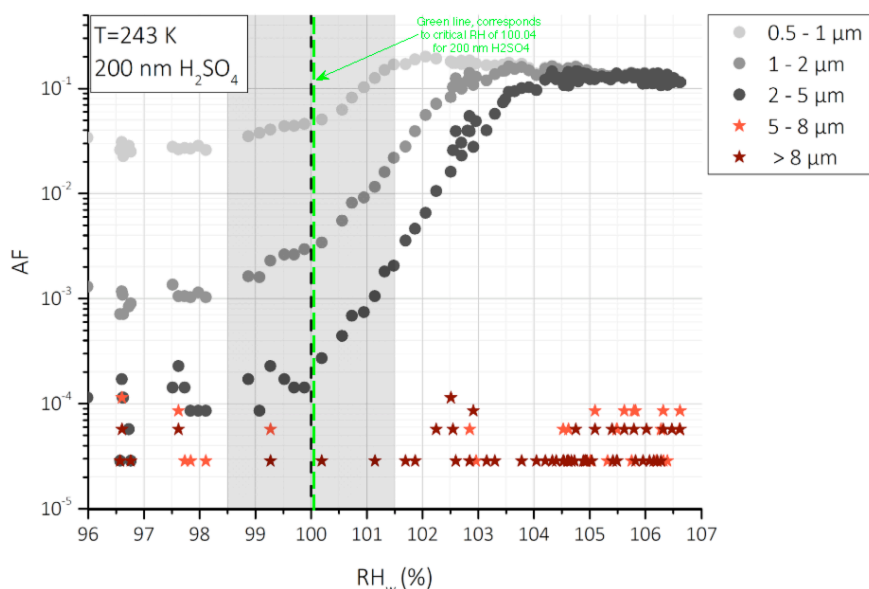


Figure 2. Reproduced from Figure 3 in the paper, with the green line showing the theoretical critical humidity for the activation of 200nm H₂SO₄ particles.

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.

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 $RH > 102 \%$, whereas the “larger than 0.3 micron” curve approaches an activated fraction for $RH > 92\%$ 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 $RH > 80\%$. 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 $RH = 99.5\%$, and not 102% (as in the paper).

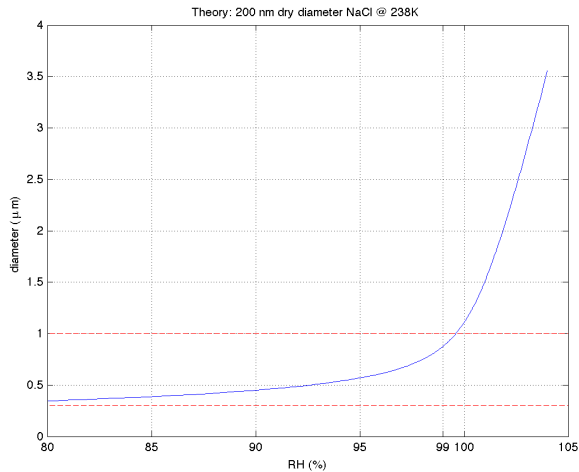


Figure 3. Theoretical calculation of the growth of 200 nm NaCl particles. Bottom red line is $y=0.3$ microns, top red line is $y=1$ micron.

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.

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.