## **Response to comments by Editor Martina Krämer**

## Comments to the author:

Étienne Vignon and co-authors,

I'm please to accept your very interesting study for publishing in ACPD. I have some questions (listed below), which can be discussed in the open discussion phase of the paper, because at the current stage of the review process (quick access review) only technical issues should be considered. But of course you could take the comments into account already now if you want.

With kind regards, Martina Krämer

Dear Martina Krämer, Thank you very much for this enthusiastic general comment and for publishing our paper in ACPD. We also thank you for your very constructive comments. Please find herebelow our response.

- How do you avoid ice particles to enter the intake, evaporate and add humidity ?

This point was already raised by a reviewer of the first paper presenting the hygrometers (Genthon et al. 2017, see discussion here :

https://acp.copernicus.org/articles/17/691/2017/acp-17-691-2017-discussion.html

We particularly emphasized that blowing-snow particles and snow flakes are not expected to subtantially impacts the RH measurements. Moreover, we underlined the fact that above -50°C, the RH data from this instrument compares very well with RH measurements from a frost-point hygrometer

Regarding more specifically diamond dust and fog cristals, note that the two inlets from which the air enters the system are oriented downward, which prevent sedimenting particles from directly falling into the instrument. (see Fig 2 of Genthon et al 2017, www.atmos-chem-phys.net/17/691/2017/). The latter is not equipped with any filters or with another system that block the advection of particles from below (due to turbulent motions or because the air is mechanically aspirated by the fan). However in the present study cases, the aspiration of thin-fog particles (if any) is not expected to substantially affect our measurements.

If we assume a fog formed by homogeneous nucleation under the typical cooling rate that are observed, the number concentration of ice particles at a temperature of ~220 K would be of the order of 5 10<sup>-1</sup> cm<sup>-3</sup> (according to Fig 1b in Baumgartner et al. 2022). Let's further assume that the mean particle dimension is around 10 microns (which is the order of magnitude of diamond dust and fog particles collected by Santachiara et al. 2016, see their Figs 1 and 2). One can thereby calculate the tendencies of water vapor partial pressure de and temperature dT (and saturation vapor pressure desl from Clausius Clapeyron's equation) if all condensates sublimate in the intake.

The change in relative humidity wrt liquid RHl (quantity measured by the HMP) associated with the sublimation of fog particles can then be calculated as : dRHl=(esl<sup>-2</sup>)(esl de- e desl) The small python code that was used to estimate dRHl is available here : <u>https://web.lmd.jussieu.fr/~evignon/paper\_fog\_acp/</u>

For an ambient RHl value of 60 %, our calculations give a dRHl value of 1.8 %. This value is not negligible but generally lower than the measurement uncertainties given in Appendix A.

It is also worth remembering that we have assumed spherical ice particles and that all of them sublimate in the intake. Those two assumptions lead to an overestimation of the mass of ice that sublimates and the dRHl value given above is therefore probably overestimated.

In the revised version of the paper, we have added the following paragraph to explain this point :

'It is worth mentioning that the intakes are oriented downward such that sedimenting ice crystals cannot directly fall into the measurement system. Nevertheless, if the instrument is embedded in a foggy air, some ice particles may enter from below due to turbulent eddies and the mechanical aspiration. We have therefore analytically estimated the effect of sublimation of thin-fog ice particles – with a typical size of about 10 mum (Santachiara et al. (2016)) and a typical number concentration of 0.5 cm<sup>-3</sup> (Baumgartner et al. 2022) - on the RHI measurements (not shown). The obtained RHI change values are of the order of a few percents at most depending on the ice crystal shape assumption, on the ambient temperature and relative humidity values, and on the fraction of particles that sublimate. In any case, the obtained values are lower than the instrumental uncertainties calculated in Appendix A. The sublimation effects can therefore be deemed second order with respect to the intrinsic temperature and RHI measurement uncertainties.'

Baumgartner, M., Rolf, C., Grooß, J.-U., Schneider, J., Schorr, T., Möhler, O., Spichtinger, P., and Krämer, M.: New investigations on homogeneous ice nucleation: the effects of water activity and water saturation formulations, Atmospheric Chemistry and Physics, 22, 65–91, https://doi.org/10.5194/acp-22-65-2022, 2022.

Genthon, C., Piard, L., Vignon, E., Madeleine, J.-B., Casado, M., and Gallée, H.: Atmospheric moisture supersaturation in the near-surface atmosphere at Dome C, Antarctic Plateau, Atmos Chem Phys, 17, 1–14, doi:10.5194/acp-17-1-2017, 2017.

Santachiara, G., Belosi, F., and Prodi, F.: Ice crystal precipitation at Dome C site (East Antarctica),<br/>AtmosphericResearch,<br/>167,108–117,<br/>167,108–117,<br/>https://doi.org/https://doi.org/10.1016/j.atmosres.2015.08.006, 2016

- A similar instrument as yours to measure humidity exist in an airborne version, here is a reference in case you want to cite it: https://amt.copernicus.org/articles/8/1233/2015/ and here you can see where it is operated:

https://www.iagos.org/iagos-core-instruments/

Thank you for mentioning this airborne instrument that we did not know. The design and the concept of the their instrument and ours are not exactly the same but in both cases the relative humidity has to be estimated from a relative humidity measurement at a 'sensor' temperature that differs from the ambient air temperature. We have added a reference to Neis et al. (2015) in our paper.

- How did you calculate the homogeneous freezing threshold? Explicitly or as an approximation?

Thank you for raising this point that we have not detailed in the paper. As a first approximation, we used the analytical fit of Koops et al's (2000) results derived in Ren and MacKenzie 2005 : Scr=2.349-T/259. This fit assumes that particles have a typical radius of 0.25 µm and that they freeze homogeneously within 1 min (see also Kärcher and Burkhardt 2008). In this equation, the solution droplets are assumed to be in equilibrium with the environment which is reasonable for

most atmospheric situations (Koop 2015) and particularly for temperatures > 205 K (which is the case for our two fog events at Dome C).

Values of Scr depend on the size of the particle, on the composition of the particle and on the formulation - and related uncertainties - of water activities and saturation vapor pressure (see thorough discussion in Baumgartner et al. (2022)). Individually, those effect make Scr vary by about 0.01 to 0.05 (see Baumgartner). An envelop of 0.05 has also been added around the Koop's curve in our Figs.4, 8 and B1. The inclusion of this shading is only intended as a rough indicator of the uncertainty and to guide the eye. Moreover, we have added the following paragraph in Section 2.2 :

'To detect the possible occurrence of homogeneous freezing of solution aerosols, we will compare our RHi measurements with the so-called Koop et al. (2000)'s threshold. In the approach of Koop et al. (2000), solution particles spontaneously freeze when RHi exceeds a threshold value that primarily depends on temperature. As a first approximation, we calculate the RHi threshold value (RHi<sub>T</sub>, in %) using the analytical fit of Koop et al. (2000)'s experimental results derived in Ren and Mackenzie (2005):

 $RHi_T = (2.349 - T/259) \cdot 100$ 

where T is the temperature in Kelvin. This fit has been performed for solution particles in equilibrium with the ambient vapor that have a typical radius of 0.25  $\mu$ m and that can freeze homogeneously within 1 min (see also Kärcher and Burkhardt, 2008). The exact value of the threshold also depends on the size of the particle as well as on the composition thereof and on theformulation and uncertainties of water activities and saturation vapor pressure. Individually, those effects make RHi<sub>T</sub> vary by about 1 to 5 % (see Baumgartner et al., 2022). An envelop of 5 % has therefore been added around the Koop's curve in our graphs. This envelop is only intended as a rough indicator of the uncertainty and to guide the eye.'

Baumgartner, M., Rolf, C., Grooß, J.-U., Schneider, J., Schorr, T., Möhler, O., Spichtinger, P., and Krämer, M.: New investigations on homogeneous ice nucleation: the effects of water activity and water saturation formulations, Atmospheric Chemistry and Physics, 22, 65–91, https://doi.org/10.5194/acp-22-65-2022, 2022.

Kärcher B. and Burkhardt U. : A cirrus cloud scheme for general circulation models, Q. J. R. Meteorol. Soc. 134: 1439–1461 (2008)

Koop, T.: Atmospheric Water, in: Water: Fundamentals as the Basis for Understanding the Environment and Promoting Technology, edited by: Debenedetti, P. G., Ricci, A., and Bruni, F., IOS, Amsterdam, Bologna, 45–75, https://doi.org/10.3254/978-1-61499-507-4-45, 2015

Ren C, MacKenzie AR. 2005. Cirrus parameterization and the role of ice nuclei. Q. J. R. Meteorol. Soc. 131: 1585–1605.

I ask because the RHi of the measurements is close to but does not exactly meet the threshold. Or could it be that the freezing threshold is a bit lower at the warmer cirrus temperatures, as observed in the lab by Schneider et al. (2021), ACP (https://acp.copernicus.org/articles/21/14403/2021/ ), and also discussed in Baumgartner et al. (2021)?

Note that for the second event, the threshold is met at z=18 m. For the 1st event, note that the difference between the homogeneous freezing threshold and the measured RHi peak is lower than the measurement uncertainty and that the Koop' threshold uncertainy as well (we now specify it in

the text). Those uncertainties prevent us from discussing subtle variations in the freezing threshold or from questioning its exact value as in Schneider et al. (2021) and Baumgartner et al. (2022).

Also, I think it could be worth to discuss the long times where the ice fog exist in slight subsaturation.... or could it be that the humidity has a slight dry bias ?

Following your recommandation, for the first event we have adapted the text as follows.

For the first event :

'Fig. 2a shows that the depth of the fog layer gradually increases from 0600 LT, 8 March up to about 80 m at 1800 LT, 8 March, as the daytime convective boundary layer deepens in  $\sqrt{t}$  (Stull et al. 1990). The growth of the fog is possible in the higher part of the boundary layer as its top is supersaturated wrt ice (Fig 2c). Ice crystals can hence grow by vapour deposition and sediment down to the near-surface layers where they probably partly sublimate (Fig. 2 and 4). Concurring with Genthon et al. 2022 (see their Fig. 8), the near-surface air becomes subsaturated wrt ice particularly during daytime when the near-surface air warms by convective mixing.'

For the second event :

'From the evening of the 25 August, RHi remains slightly below saturation at 3 and 18 m probably owing to a net flux of vapor towards the surface in the very shallow boundary layer. Ice crystals at these two heights can therefore not grow very close to the surface but their detection at 18 m by the lidar may be rather explained by the sedimention from higher layers.'