

We thank Minghui Diao for taking the time to carefully read through the manuscript and the generally positive comments. Please find below the reviewer's comments in normal text, with **our responses in blue** and **changes that has been made in the revised version of the manuscript in red**.

RC1: In Figure 7, the occurrence frequency of RH_{ice} in natural cirrus peaks at 95%. But the authors described this figure as the RH_{ice} centers at 100%: (line 460) “In comparison to Fig. 4e, where the frequencies of RH_{ice} in the natural cirrus (SAC-) centre around 100% at temperatures above 225 K (also reported in a global RH_{ice} climatology by Krämer et al. (2020), ...” The reviewer wonders if this suggests that the water vapor measurements or the combination of water vapor and temperature measurements in ML-CIRRUS has a low bias by 5%? The distributions of all in-cloud RH_{ice} for in-situ and remote sensing observations also suggest there may be a low bias for in-situ observations. If this is the case, then the subsaturated conditions for contrail cirrus would be more around 95% instead of 90%. Previously, several studies on US NSF-funded field campaigns analyzed in-situ measurements of RH_{ice} for cirrus clouds. They all showed a peak position at 100% for RH_i distribution.

Figure 12b in Patnaude, R., M. Diao, X. Liu, S. Chu. Effects of Thermodynamics, Dynamics and Aerosols on Cirrus Clouds Based on In Situ Observations and NCAR CAM6 Model. *Atmospheric Physics and Chemistry*, 21, 1835–1859, <https://doi.org/10.5194/acp-21-1835-2021>, 2021

Figure 5 in Diao, M., G.H. Bryan, H. Morrison, and J.B. Jensen, Ice nucleation parameterization and relative humidity distribution in idealized squall line simulations, *Journal of the Atmospheric Sciences*, 74, 2761–2787, <https://doi.org/10.1175/JASD-16-0356.1>, 2017.

Figure 4 in Diao, M., M.A. Zondlo, A.J. Heymsfield, L.M. Avallone, M.E. Paige, S.P. Beaton, T. Campos and D.C. Rogers. “Cloud-scale ice supersaturated regions spatially correlate with high water vapor heterogeneities”, *Atmospheric Chemistry and Physics*, 14, 2639-2656, 2014.

The references were inserted in Line 441.

Can the author look more closely into the time series of the flights, and see if there was possible bias in RH_i measurements? One possible method is to look at RH_{liq} for warm clouds and they should be very close to 100% liquid saturation. Although this method may not work well if the bias from the instrument is temperature dependent (which you should be able to tell from lab calibrations). Did the SHARC instrument participate in any water vapor intercomparison experiment, or lab comparisons with commercial chilled mirror hygrometer such as RHS system (accuracy $\pm 0.1^\circ\text{C}$)? Another possible method is to examine typical cirrus clouds sampled in ML-CIRRUS, and especially the ones mixed with ice supersaturated segments. When the ice crystal regions and clear-sky ice supersaturated regions are intermittently observed, it is often that the ice crystal regions show ice saturation or slight ice supersaturation instead of ice subsaturation. If these segments frequently show ice subsaturation when they are surrounded by clear-sky ice supersaturation, it would be an indicator of possible low bias in RH_{ice} .

The uncertainty of water vapor instrument, temperature probe, and the combined RH_{ice} uncertainty from water vapor and temperature should be added in the description around line 125.

AC1: The SHARC instrument was deployed on board HALO together with the Fast In-situ Stratospheric Hygrometer (FISH) and the Atmospheric Ionization Mass Spectrometer for water vapor (AIMS) during the ML-CIRRUS campaign. The overall uncertainty of SHARC H_2O measurement is 5% relative and ± 1 ppm absolute offset uncertainty (Kaufmann et al., 2018). The nominal accuracies of the BAHAMAS pressure and T_{amb} measurement are 0.3 hPa and 0.5 K (Mallaun et al., 2015; Giez et al., 2017; Kaufmann et al., 2018). The overall accuracy of the in-situ RH_{ice} measurements here is between 10 – 20%, with the respective uncertainties of the temperature, pressure and water vapour measurements considered (Krämer et al., 2016). The description of instrumental uncertainties and accuracies was added to Line 126 in the revised manuscript.

We looked into the flights focusing on natural cirrus. In general, the RH_{ice} in cirrus clouds is higher than neighbouring clear-sky conditions. The RH_{ice} in cirrus clouds is above ice saturation in flight segments when cirrus regions and clear-sky ice supersaturated regions appear intermittently. This holds true during the contrail-dedicated flights, but the overall RH_{ice} is below ice saturation, with some cases where ice crystals and neighbouring clear-sky conditions are observed in ice supersaturated regions. Besides, intercomparisons for SHARC, FISH (calibration before and after flights) and AIMS (in-flight calibration) H_2O measurements showed very good agreement between instruments (Meyer et al., 2015; Kaufmann et al., 2018). No strong bias was found in either water vapour instruments or temperature measurements. Also, in the past years, unpublished work of intercomparisons between SHARC, FISH (the high precision hygrometer from Jülich) and other water vapour instruments during field campaigns suggests SHARC is a very robust instrument.

However, the possibility of a small bias in the in situ RH_{ice} dataset due to a low bias in the Basis Halo Measurement and Sensor System (BAHAMAS) T_{amb} measurement was brought up in Schumann (2021; See page 108). Here, the impact of the low temperature bias of 0.5 K on the RH_{ice} distribution is addressed in Sect. 3.3 starting from Line 491 and more details can be found in Sect. S3 in the Supplement. With the low temperature bias considered, the subsaturated conditions for contrail cirrus would peak at 95%, shifting by 5%, and the peak of natural cirrus RH_{ice} distribution would move closer to 100%. In the revised version, we refer to the discussion about the effect of a possibly low temperature bias already at the beginning of Sect. 3.3.

RC2: The reviewer suggests adding an analysis on the distribution of RH_i for inside contrail cirrus with respect to the cruising altitude. If the author calculate Δz or Δp for each second of flight data with

respect to cruising altitude, and plot RH_{ice} only for inside contrail cirrus (CA + SAC methods), will the RH_{ice} distribution show more ice supersaturation on the higher levels and more subsaturation in the lower levels? This can help verify if these contrails in the sub-saturated conditions happen due to ice crystals sedimenting into lower altitudes with subsaturated conditions, or the contrail ice crystals stay at similar altitudes, but their environmental condition gradually becomes subsaturated.

AC2: As the referee suggested, we plot the RH_{ice} distribution for contrail cirrus (SAC+CA) with Δp (Δp) calculated for each second of flight data during the contrail-dedicated flights, see Fig. 1a. In addition, the distribution of RH_{ice} in relation to temperature and Δp is plotted in Fig. 1b. The ice crystals showing ice supersaturation was appearing more often in the lower part of CA ($p > 222.5$ hPa in Fig. 1a, $T_{\text{amb}} > 212$ K in Fig. 1b), *i.e.*, the ice subsaturation speared more frequently in the upper CA in spite of some ice-supersaturated air masses. From this point of view, it is still difficult to verify from the RH_{ice}-Δp relation vs. pressure p (Fig. 1a) or temperature (Fig. 1b) if ice crystals sedimented in subsaturated region or if the air mass gradually became subsaturated.

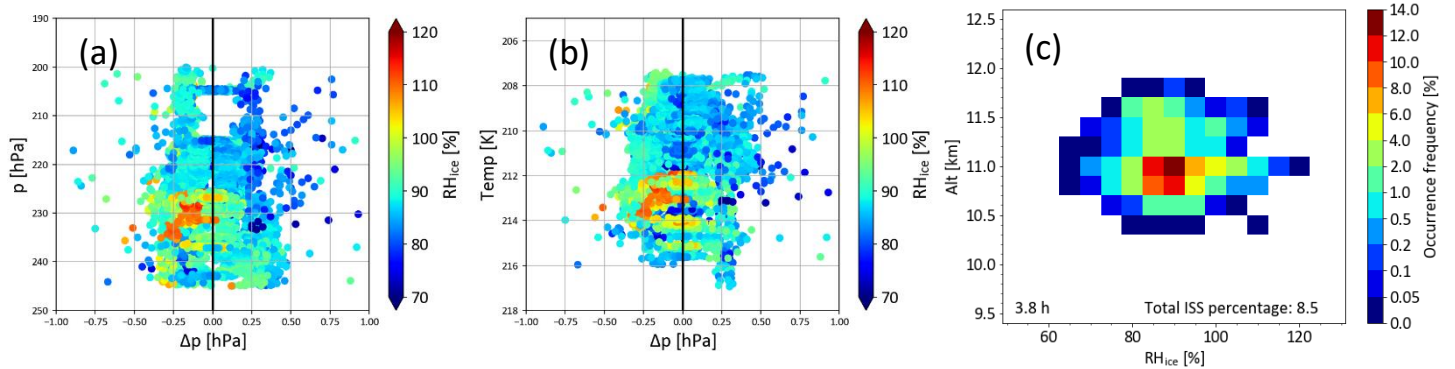


Figure 1. (a): Ambient pressure vs. Δp , color-coded with RH_{ice} for contrail cirrus (SAC+CA) with Δp calculated for each second of flight data during the contrail-dedicated flights. (b): Similar to (a), but for ambient temperature vs. Δp . (c): Altitude vs. RH_{ice} for contrail cirrus (SAC+CA), color-coded with occurrence frequency. The bin widths for RH_{ice} and altitude are 5% RH_{ice} and 200 m, respectively. The total sampling time of the contrail cirrus satisfying SAC and CA is 3.8 h, added in the figure.

Therefore, we plot the RH_{ice} in 5% bins vs. the flight altitude in 200 m bins color-coded by occurrence frequency, shown in Fig. 1c. Here, we can see that the ice supersaturation (ISS, in total 8.9%) was mostly encountered between 10.8 – 11.1 km, the lower altitudes of CA range, where ice subsaturation also occurred most frequently, with the second highest frequency in higher altitudes (11.1 – 11.7 km). It points out that the air mass gradually became subsaturated. The ice-subsaturation might be related to aged contrails, possibly as a result of cirrus sublimation in the environment that gradually becomes subsaturated due to the entrainment of cold and dry ambient air. The ice supersaturation seems to be related to very young contrails

formed at the early stage of the detrainment of hot and humid aircraft exhaust into cold ambient air, because it was mostly detected in warmer temperature regions (212 – 226 K).

RC3: In Figure 3, can the authors add a third row, for N_{ice} versus R_{ice} and RH_{ice} versus temperature (similar to Figure 3 c and d), but categorize the samples into two groups, (1) fulfilling the plume detection criterion or (2) not fulfilling that criterion? It is unclear where the samples fulfilling that plume detection criterion would be distributed, and how they are related to the SAC and CA criteria.

Figure 5 would also benefit from an additional row, illustrating Cirrus: fulfilling SAC, inside CA, and also with restriction to plume detection. The reviewer wonders if applying a third restriction of plume detection criterion to the combined SAC+CA criteria would make a big difference.

AC3: This is a good suggestion. We have included an extra section in the supplement in the revised version. In the new version, another section Sect. S4 and Fig. S4 (shown below) were added into the Supplement, explaining the effect of applying the plume detection algorithm on the separation of contrail and natural cirrus using SAC and CA. And the conclusion of Sect. S4 was inserted in Line 388 in the revised manuscript.

The N_{ice} - R_{ice} distribution for the cirrus fulfilling and not fulfilling the plume detection criteria are plotted in Fig. S4a and c. Figure S4b and d show the corresponding RH_{ice} - T_{amb} relations. Because the plume detection depends greatly on NO_y and aerosol concentrations, and the enhancement signal of the species decays with time, only rather fresh plumes younger than about 4 h can be identified. Therefore, the population of cirrus particles that can be traced back to plumes is rather small, 0.99 h, as shown in Fig. S4a. A large number of the ice crystals found in plume (0.86 h) fulfil SAC (Fig. 5e). Temperature wise, most of the ice particles are found in the CA temperature range (207 – 218 K) with a high occurrence frequency in ice subsaturation, see Fig. S4 b. Most of the cirrus cannot be validated with the plume detection algorithm. They are a mixture of contrail cirrus, in situ- and liquid-origin natural cirrus, spreading in a wide temperature range, see Fig. S4c and d.

The cirrus crystals fulfilling SAC, inside CA and also with restriction to plume detection are shown in Fig. S4e, with their RH_{ice} vs. T_{amb} displayed in Fig. S4f. From Fig. S4e, we can see that the N_{ice} - R_{ice} distribution of ice particles would be represented nearly by the 50th percentile in Fig. 5e. Comparing the median N_{ice} and R_{ice} values represented by Fig. 5e and Fig. S4e (which is also listed in Table 1), we can see that the medians in the dataset using SAC, CA and plume detection are closer to but not significantly different from the ones determined using only the combined SAC+CA. Therefore, it does not make a big difference to add the restriction of plume detection to the combined SAC and CA criteria.

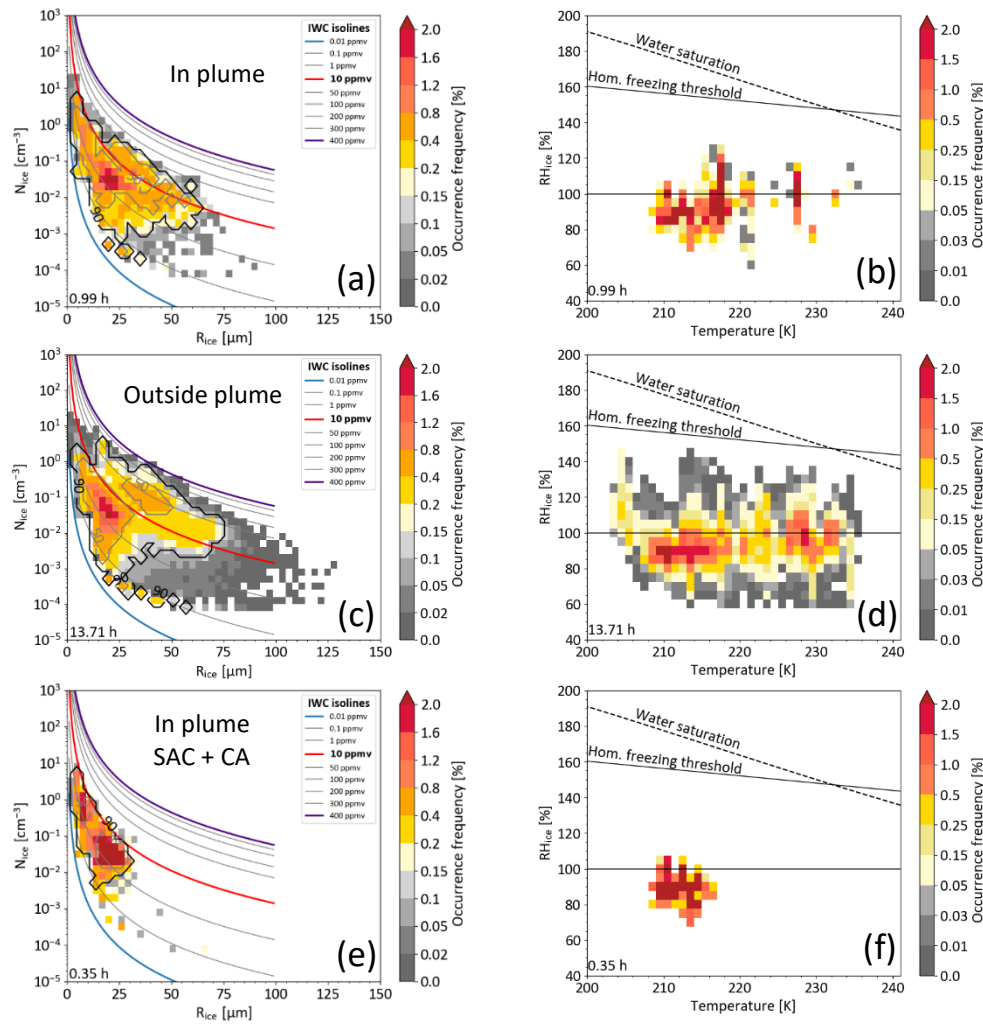


Figure S4. N_{ice} – R_{ice} relations (left) and RH_{ice} – T_{amb} relations (right) color-coded by normalised occurrence frequency, similar to Fig. 3c and d. (a): N_{ice} – R_{ice} relation for the ice particles found in aircraft plumes using the plume detection algorithm (median: $N_{ice} = 0.027 \text{ cm}^{-3}$, $R_{ice} = 23.7 \mu\text{m}$, IWC = 5.0 ppmv and $RH_{ice} = 92\%$). (b): Corresponding RH_{ice} – T_{am} relation. (c): N_{ice} – R_{ice} relation for the cirrus outside aircraft plumes. (d): Corresponding RH_{ice} – T_{am} relation. (e): N_{ice} – R_{ice} relation for the cirrus fulfilling the plume, SAC and CA (ambient pressure 200–245hPa) criteria (median: $N_{ice} = 0.041 \text{ cm}^{-3}$, $R_{ice} = 17.8 \mu\text{m}$, IWC = 4.4 ppmv and $RH_{ice} = 89\%$). (f): Corresponding RH_{ice} – T_{am} relation.

RC4: Line 74, CONCERT 2018 campaign, should this be 2008?

AC4: This is a typo; it should be CONCERT 2008 campaign. It was corrected in the new version.

References

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