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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Widespread polar stratospheric ice clouds in the 2015/2016 Arctic winter – Implications for ice nucleation" *by* Christiane Voigt et al.

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Reply to Referee #1 (Mike Fromm)

We thank the referee for his judgement on the relevance of the topic and appropriateness of this paper for ACP.

His comments motivated the extensive exploitation of CALIOP data in the revised version of the manuscript. We now performed an advanced Lagrangian analysis of ice PSC formation using domain filling trajectories matched with CALIOP PSC observations. We found independent and convincing evidence for the proposed ice nucleation

pathways. The novel evaluation is integrated in the revised version of the manuscript and helps to progress the scientific knowledge on ice nucleation and ice properties in the polar stratosphere.

In summary, we are grateful to referee 1, whose comments helped to significantly improve the scientific quality of the manuscript.

In the following we abbreviate and enumerate the reviewer's comments and the replies (comment 1: C1; Reply 1: R1)

Major comments:

C1: The referee suggests to reinforce the use of CALIPSO data and to extend the trajectory analysis in order to address the substantial science question regarding ice nucleation.

R1: We thank the reviewer for this comment.

We additionally performed domain filling back trajectory calculations starting in the PSC observation on 22 January 2016 and match the trajectories with vertical PSC cross sections of CALIPSO classified with respect to PSC type. We then mark the matches according to the PSC types found at the interception point and perform a sensitivity study of the results. Indeed, NAT PSCs (and only NAT PSCs, no other PSC types) are detected by CALIPSO on the match points of the PSC trajectories, which start in the ice mode with high particle depolarization. Thus according to CALIPSO, a NAT PSC was observed prior to the high-depol ice PSC. This gives independent evidence for ice nucleation on NAT and strongly supports our hypothesis of ice nucleation on NAT. In contrast, predominantly STS or no PSC has been detected on the matches of those trajectories, which start in the ice mode with lower particle depolarization. Hence this new analysis gives compelling evidence for the proposed ice nucleation pathways (1) of ice nucleation on NAT for the high-depol ice regime and (2) of (heterogeneous) ice nucleation in STS clouds for the low-depol ice regime. We have added a new figure and text to the manuscript to present the results of the match study and changed the abstract accordingly.

C2: The referee suggests using the orbit of CALIPSO on 22 January 2016 that passes between northern Scandinavia and northern Greenland close in space and time to the HALO flight track for analysis.

R2: The HALO flight track 22 January 2016 was indeed planned as a match with CALIPSO. We now show the CALIPSO PSC observation path #7 close to the HALO flight leg and use the PSC classification from Pitts et al., ACPD, 2018, to consolidate the ice PSC observation. The CALIPSO observations are presented in a new figure in order to monitor the spatial extension of the ice PSC and the other PSC layers. In both observations from satellite and aircraft the general structure of the ice PSC is shown and the STS layer below and the NAT Mix1/Mix2 layers to the north-east of the ice PSC are measured. Within the temporal and spatial variability of PSCs, the PSC observations from the aircraft and the spaceborne lidar are consistent within the collocated part of their flight paths‘.

However, the CALIOP flightpath (leg 7 at 10:44 UTC on 22 January 2016) missed significant fraction of the specific high-depol ice mode, which is of interest for our study. The high-depol ice mode was mainly measured north of 80°N and less covered by CALIOP observations on leg 7. CALIOP mainly passed south west of the high-depol ice regime (see Figure 8 in the revised manuscript). (In contrast, the proceeding CALIOP pass #6 at 9:07 UTC crossed through the high-depol ice cloud and ice was detected by CALIOP.) Therefore ice particle properties in the high-depol ice regime of WALES and CALIOP cannot be compared directly on leg 7 and the detailed intercomparison of CALIOP and WALES data is beyond the scope of our study. However, the CALIOP observations were exploited in the back-trajectory study to investigate ice PSC formation pathways. Results from domain filling trajectory calculations in the two ice regimes were successfully matched to previous CALIPSO PSC observations and give additional evidence for the ice formation pathways, as detailed in R1.

A more detailed instrument intercomparison of the CALIPSO and WALES lidar is beyond the scope of ACP and is planned for a different journal on measurement techniques.

C3: Auth refer to “branches” between non-ice and ice regimes in this 2D space as suggestive of ice-nucleation pathways. While this may be true, it has not been established here or in prior literature that this 2D construct is to be interpreted in this manner. It is perhaps equally likely that the particular patterns (auth’s “branches” as well as other, unnamed definable features) of Figure 4 are just an artifact of static sampling of a broad cloud.

R3: As given in Figure 4, high resolution WALES lidar observations show several ten thousand data points in the ice regime, this allows for a decent statistical analysis of the ice regime. Few thousand individual data points are measured in the “upper ice branch” with high particle depolarization, now named high-depol ice mode and several ten-thousands data points lie in the “lower ice branch” or ice regime with low particle depolarization (for the same backscatter ratio), now named low-depol ice mode. In between these two regions, there is a region which is less populated, depicted by the dotted line in Figure 4. Referring to the formation history analyzed using matches of trajectories with CALIPSO curtains, NAT clouds were observed prior to ice in the high-depol ice mode and STS (or no PSC) were observed prior to ice in the low-depol ice mode.

While the representation of PSC types in the histogram plot in Figure 4 does not present a Lagrangian view of particle formation, it still allows for the interpretation of phase transitions. Assuming as an example ice nucleates in STS droplets with inverse backscatter ratios of 0.2 and particle depolarizations near 0.01. The nucleated ice core then leads to slight increases in particle depolarization and in backscatter ratio, therefore the particles’ optical properties pass the STS-ice threshold and traverse into the ice regime. Similarly assuming ice nucleates on NAT particles with inverse backscatter ratios slightly below the NAT-ice threshold of 0.3. Then, ice nucleation on NAT particles leads to particle growths and therefore slight increases in backscatter ratio and particle depolarization. Therefore the particle properties cross over the NAT-ice threshold into the ice regime. Again, the phase boundary indicates a region for phase transitions.

C4: (2) Auth state, but do not show, where the points from which they launch back trajectories show up in Figure 4.

R4: Thank you, we now show the starting points of the trajectories also in Figure 4.

C5: (3) In the final section of the paper auth discuss NAT nucleation on ice as well as ice nucleation on NAT. If both of these pathways indeed exist, the NAT-to-ice “branch” they identify could represent both directions along that pathway. Yet their assumptions appear to be that the Fig. 4 branches imply only transformations to ice from other compositions.

R5: The trajectory analysis shows that temperatures are below T_ice at the point of the ice PSC observation and higher temperatures prior to T_ice. This is indicative for ice nucleation on NAT. The opposite pathway - NAT nucleation on ice - would appear in the NAT regime and therefore could potentially represent a pathway for NAT particle formation. This pathway has often been observed in the lee of mountain wave ice PSCs and therefore is not within the primary scope of our study. However its existence further supports the proposal of the opposite process of ice nucleation on NAT.

C6: (4) The trajectory analysis, which is rightly presented to build on the 2D histogram space’s patterns, is inconclusive in my view. . ..

R6: We thank the reviewer for his comment and added domain filling back-trajectory analysis to our study (see R1). To this end, we calculate > 2500 individual 8 days back trajectories starting every 2 min in the PSC event at altitudes between 16 and 25 km. We show temperatures of back-trajectories at 21.5 km every 10 min throughout the complete PSC event. We now explicitly calculate T_NAT and T_ice along the back trajectories with altitude dependent H2O and HNO3 profiles. We refrain from showing

STS temperatures, as they are not required for our analysis. For reasons of clarity, we further show the temperature difference to T_NAT and the ice saturation ratio S_ice for 4 “arbitrarily” selected back trajectories starting in the different PSC types to shine light on ice formation pathways. We correct the inconsistency in ice symbols in Figures 5 and 7.

C7: Given that auth employ back trajectories to determine ice-nucleation transitions, they have not availed themselves of at least two very critical papers employing trajectories and satellite data in similar quests. One is Teitelbaum et al. (2003) . . .Second, Santee et al. (2002). . .

R7: We excuse this incompleteness and now discuss our results in sight of these references.

C8: Abstract and Conclusion section: The "tropical" aspect is not developed at all in this paper. It is only mentioned in summary, speculative fashion at the very end of the "Conclusion" section. Given the critical concerns mentioned above, I contend that the link with tropical cloud nucleation does not belong in this paper.

R8: Conditions are favorable for the widespread occurrence of NAT in the tropics, and indications were found for a tropical NAT belt (e.g. Voigt et al., ACP, 2007; Chepfer and Noel, GRL, 2009; reply by Poole et al., 2009,. . ..). Hence the proposed ice nucleation pathway on NAT might be of importance for the tropics. We discuss this aspect in more detail in the discussion section of the manuscript and remove the tropical aspect from the abstract.

C9: Section 5.2: Auth predicate much of their paper on the two-dimensional histogram of lidar-based PSC optical properties developed/refined by CALIPSO scientists and co-authors Mike Pitts and Lamont Poole. . . . To my understanding, there was no aspect of phase-transition built into this construct.

R9: See R3

C10: Section 5.3: Auth experiment with Pitts et al.'s criteria for a NAT Mix2/Ice boundary. On what basis is this determination made? ...

R10: The decision on the NAT-ice boundary for the WALES observations from 22 January started with previous work by Pitts et al., 2011, as detailed in the text. The occurrence histogram of PSC types (Figure 4) shows an artificial separation of the ice and the NAT phases for a phase boundary of 0.2, which is significantly reduced using the phase boundary of 0.3. Indeed the NAT-ice boundary of 0.2 will classify a significant fraction of the ice particles as NAT (see Figure 4, low-depol ice mode). Independent support for this phase boundary for the WALES observations from 22 January 2016 comes from the temperature analysis, which shows that the major fraction of ice is measured at temperatures below T_ice and the major fraction of NAT is measured at temperatures above Tice and blow T_NAT when using 0.3. We now support the investigation of the phase boundary by replacing Figure 6 with a new Figure showing the occurrence histogram of PSC type versus temperature difference to T_ice. The peak NAT Mix2 and NAT Mix1 occurrence is located above T_ice for the phase boundary of 0.3 and ice occurrence peaks slightly below Tice. In contrast, the peak in NAT Mix2 occurrence is at T_ice for the phase boundary of 0.2. The novel classification by Pitts et al., ACPD, 2018 suggests a H2O, HNO3 and time dependent PSC type classification. The boundary of 0.3 is close to the new classification by Pitts et al., (2018) for 22 January 2016.

Substantial Concerns

C11: Section 5.2: Auth give a brief discussion of 1064/532 nm color ratio. If the color ratio data are to be discussed, and speculation made regarding sedimentation, a figure is called for.... The color ratio analysis should be presented in more exacting detail, or dropped.

R11: We now drop the analysis of the color ratio according to the reviewer's suggestions.

C12: Section 6.1 and 6.2: Auth explicitly invoke Greenland and its orographic role in PSC formation and phase change within the WALES lidar's sampling of a synoptic-scale PSC. While the orographic influence of Greenland has been convincingly documented in prior papers, all the evidence here (PSC observations and air mass trajectories based on reanalysis data) point to synoptic-scale drivers for the cloud and parcel temperature/height excursions....

R12: As shown in Figure 7, panel C in Voigt et al., ACPD, (2017) a synoptic scale lift from ∼20.5 to ∼22 km altitude occurs for air masses passing over Greenland (panel A). This slow synoptic lift leads to adiabatic cooling by ∼6 K over 30 hours prior to the observation and eventually to ice formation. Trajectories missing Greenland (e.g. trajectory 5 in Figure 7 in Voigt et al., ACPD, 2017) remain near 22 km altitudes and temperatures vary by only 2K. This led to the assumption that the orography of Greenland contributed to the synoptic lift of the trajectories passing over Greenland. Due to its large size, Greenland can be regarded as one of the synoptic scale drivers of air ascent and respective temperature excursions. In Figure 8 of the revised manuscript we show he temperatures along domain filling trajectories, which clearly show the formation of gravity waves with moderate temperature excursions in the lee of Greenland. We now moderate the discussion and indicate that in synoptic scale drivers including the elevation of Greenland contribute to the temperature decrease and the temperature oscillations within the last 30 h prior to the observation.

C13: Section 6.2, Page 11, Line 14: "Summarized, the trajectory analysis supports our hypotheses of ice nucleation in STS with meteoric inclusions . . ." I do not see how the trajectories of 1-3 support this hypothesis. .... If not, please alter this discussion suitably.

R13: We again thank the referee for comment 1 and refer to R1 for this discussion.

C14: Section 7, Page 12, Line 5: Here auth discuss the sensitivity differences between CALIPSO and WALES. This was not explored herein, and should have been if this point is to me part of the conclusions. Moreover, involving CALIPSO in the case study is natural and essential. My suggestion is to redo the case with an integrated CALIPSO/WALES analysis.

R14: As suggested by referee 1, we redid the case with an integrated CALIPSO/WALES analysis for the trajectory study and investigated of ice formation pathways. We show the CALIPSO cross section, which was matched by HALO. Regarding other aspects we refer to R1 and R2.

Minor Concerns

R: We note that the there is an inconsistency in the referees line numbers (and content) with respect to the latest version of the submitted manuscript.

C15: Section 6.1, Page 10, Line 11-12: This statement is inconsistent with the mapped trajectories...parcel 5 does pass over Greenland âĹijday 8.

R15: In the manuscript, we state (Section 6.1, Page 10, Line 17-18): For the ice layers, the trajectories' temperatures decrease below Tice $\sim$10 h prior to the observations during a slow uplift over Greenland. In contrast, the typical NAT trajectory circulates within the inner vortex at temperatures below 188 K for 7 days without passing over Greenland. We now show 2 min trajectories along the outbound flight leg at 21.5 km altitude in Figure 8 to monitor their position with respect to Greenland and their temperature history.

C16: Section 6.1, Page 10, Line 12: "Therefore" implies that something stated in the prior sentence(s) is the determinant for why "temperatures stay above Tnat" It's not clear what that link is or that one even exists. Please clarify.

R16: We removed therefore.

C17: Section 6.1, Page 10, Line 13: The PSC temperature of 5 and 6 are both within the envelope supporting both STS and NAT. However, their lidar-based compositions are clearly different. How do temperature histories help us understand why one is NAT

and one is STS?

R17: Trajectory 5 (ending in NAT) stays below T_NAT for 10 days prior to the observation of NAT, so NAT can nucleate and grow slowly at low nucleation rates within at least 10 days. Trajectory 6 (ending in STS) only decreases below T_NAT 28 h prior to the observation and is above TNAT before. While STS formation and growth is a fast process, which takes place within few hours, NAT nucleation rates are low and merely contribute to particle formation within a day. The trajectories are consistent with the observations of NAT and STS. In the new manuscript we show 2 min trajectories along the outbound flight leg and highlight 4 trajectories, one for each PSC particle type.

C18: Section 6.2, Page 10, Line 24: Because auth are relating the points in Fig 5 and 7 to the histogram space, it would be essential to show the six symbols in their respective locations within Fig. 4.

R18: We now show the starting points of the trajectories in Fig.4.

C19: Section 6.2, Page 10, Line 25: The black line in the figure is extremely difficult to see. Moreover, there are more than one black enclosures in the figure. Can the line be plotted more boldly? Should auth point out the multiple locations of the black enclosures and discuss them?

R19: We now plot the line thicker and refer to the multiple locations of the black enclosures.

C20: Section 6.2, Page 10, Line 27: The diamond color convention is inconsistent between Figs 5 and 7. Hence the text is confusing. Please correct the figures and make the discussion consistent.

R20: We now use the diamond for ice, the square for NAT and the circle for STS throughout the manuscript.

C21: Section 6.2, Page 10, Line 29: Again, what s the significance of Greenland? These synoptic-scale variations in height/temperature are a marker of stratospheric synopticscale dynamics. All the evidence that I have assembled (e.g. tropoause-height analyses, total ozone maps) indicate that the cold pool here is enhanced due to a tropospheric anticyclone forcing a bulge in stratospheric isentropes. The fact that it is near Greenland is probably inconsequential. If auth agree, please clarify the discussion. If not, please explain Greenland's influence.

R21: We refer to comment 12 and reply 12. We moderate the discussion of the influence of the orography of Greenland.

C22: Section 6.2, Page 10, Line 30: The temperature history of 4 in the time frame immediately preceding observation (i.e. within the preceding 5 days) supports a previous composition of STS as well (T<Tsts and Tnat). Hence this definitive conclusion here is not supported.

R22: We now use independent observations from CALIOP observations to assess the particle composition prior to ice formation. For trajectories 1 and 2 we find either STS or no PSC in the matches. As STS is forming and evaporating fast without nucleation barrier, thus CALIOP observations provide further evidence for the existence of STS prior to ice. (New) trajectory 4 was below T_NAT for 8 days and CALIOP detected NAT on the trajectory matches before ice, so this is consistent with ice formation on NAT. We now in addition show results of domain filling trajectory calculations matched to CALIOP observations to strengthen the analysis.

C23: Section 6.2, Page 10, Line 34: Presumably auth are referring to Figure 1 here. If so, they should state that. But even so, Fig. 1 only shows a combination of NAT and STS, so there is no indication within this paper that the history of NAT and STS in January allows them to make this conclusion. If auth agree, please clarify the discussion.

R23: We now calculate T_NAT and T_ICE and discuss the temperature history with respect to these thresholds and in addition with respect to laboratory measurements of ice and NAT nucleation rates.

C24: Section 6.2, Page 11, Line 9: There is no evidence presented that orography is implicated in the dynamical signals in the T/z data. Orography by itself does not play a direct role in the lagrangian reference frame here.

R24: We refer here to reply 12 and 21. We moderate the discussion of the influence of the orography of Greenland. And we refer to Teitelbaum et al. (2001) to point to the synoptic scale cooling. However, please note that those trajectories are lifted by 1 to 2 km which pass over Greenland and those trajectories which miss Greenland are merely lifted (Figure 8).

C25: Section 7, Page 11, Line 28-29: What does this discussion of large-particle sedimentation have to do with this paper's analysis or main point?

R25: Ice PSCs are of importance for polar chemistry due to particle sedimentation and redistribution of trace species. Thereby, ice PSCs lead to dehydration and denitrification. In fact, these are major roles of ice PSCs, in addition to chlorine activation. Large ice particles sediment faster than smaller NAT particles and therefore ice can lead to efficient denitrification at PSC altitudes.

C26: Section 7, Page 12, Line 11: This claim about a specific “branch” in the 2D histogram space is not supported herein or by other papers. I suggest removal of this statement.

R26: Two branches are clearly visible in the ice regime in Figure 4, based on the occurrence of PSC types. This is evident in Figure 4 without taking any further information into account. However, we rename the specific regimes high-depol ice mode and low-depol ice mode, where appropriate.

C27: Section 7, Page 12, Line 14: “NAT nucleation on ice. . .” That process was not discussed or examined here. However, by mentioning it, auth acknowledge the inherent weakness of the “branch” interpretation of Figure 4. I.e. the various “branches” in Figure 4 could signify phase transitions in opposite directions. This makes clear that the analysis presented herein is incomplete if the aim is to constrain “ice nucleation” as embodied in the paper’s title.

R27: As stated in R5 NAT nucleation on ice will populate the NAT regime not the ice, which is not within the focus of the paper. However this process supports the hypothesis of a reverse nucleation scheme, namely ice nucleation on NAT.

C28: Section 7, Page 12, Line 21: Here auth briefly speculate on the implications for tropical ice clouds. More recent, and arguably more relevant papers are not cited here. Chepfer, H., and V. Noel (2009), A tropical “NAT-like” belt observed from space, Geophys. Res. Lett., 36, L03813, doi:10.1029/2008GL036289. Also a comment on the above paper: Poole, L. R., M. C. Pitts, and L. W. Thomason (2009), Comment on “A tropical ‘NAT-like’ belt observed from space” by H. Chepfer and V. Noel, Geophys. Res. Lett., 36, L20803, doi:10.1029/2009GL038506. The conclusion I discern is that some of the co-authors of this paper have reservations about how to transfer PSC lessons based on the optical 2D histogram space to other realms. Please consider removing the tropical thread of this paper, if it cannot be more fully established.

R28: We refer to R8. We further note that all co-authors fully support the content of the present manuscript.

C29: References, Page 18, Line 4: “Poole, L. R., and M. C. Pitts, pers. comm.” Please give an update on this. It does not show up in AMT-D as of Jan 13 2018.

R29: We give the update on this manuscript: Pitts, M. C., Poole, L. R., and Gonzalez, R.: Polar stratospheric cloud climatology based on CALIPSO spaceborne lidar measurements from 2006–2017, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-234, in review, 2018.

C30: Figure 4: The dotted line needs to be much bolder.

R30: We increased the font size of the dotted line.

C31: Figure 6 caption: How are MLS measurements used to justify the sloping threshold? I cannot find an explanation, and it is not self evident.

R31: We now refer to the reference Pitts et al., ACPD, 2018.

C32: Figure 6 caption: What is “ISF”?

R32: Integrated forecasting system IFS as given in Section 2.3 l. 23.

C33: Figure 7: Why is a Tsts line not plotted. I think this is a natural and essential item to include here.

R33: We now include S_ice and T_NAT and T_ice. These thresholds are more important than Tsts for the purpose of our manuscript. We aim to avoid dublicating information in plots, and we give information on T_STS in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-1044, 2017.

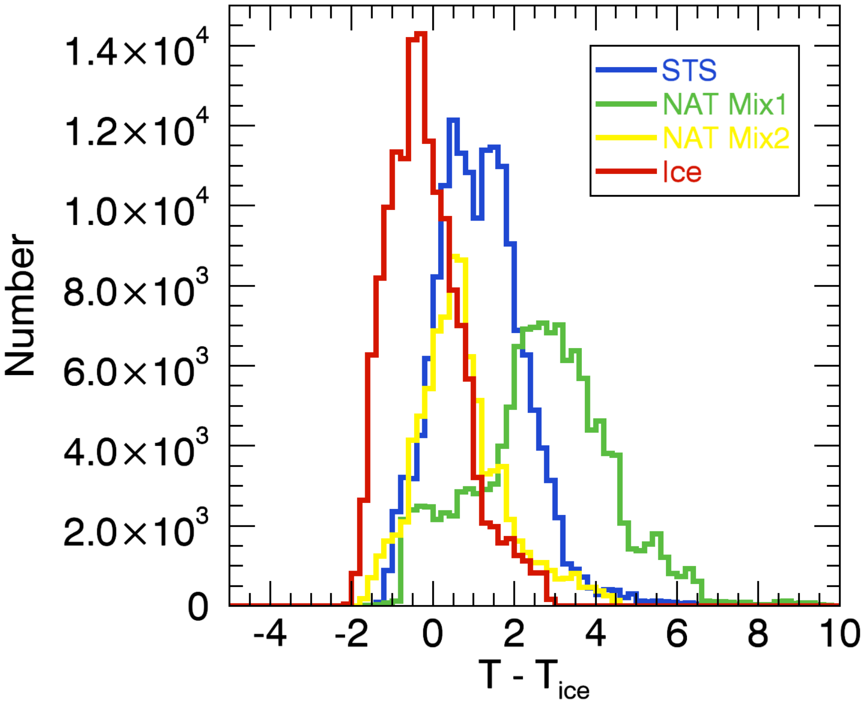

Fig. 1. Occurrence histogram of the 22 Janaury 2016 PSC types with respect to temperature difference to T_ice for the 1/R_ice threshold of 0.3 used in this study

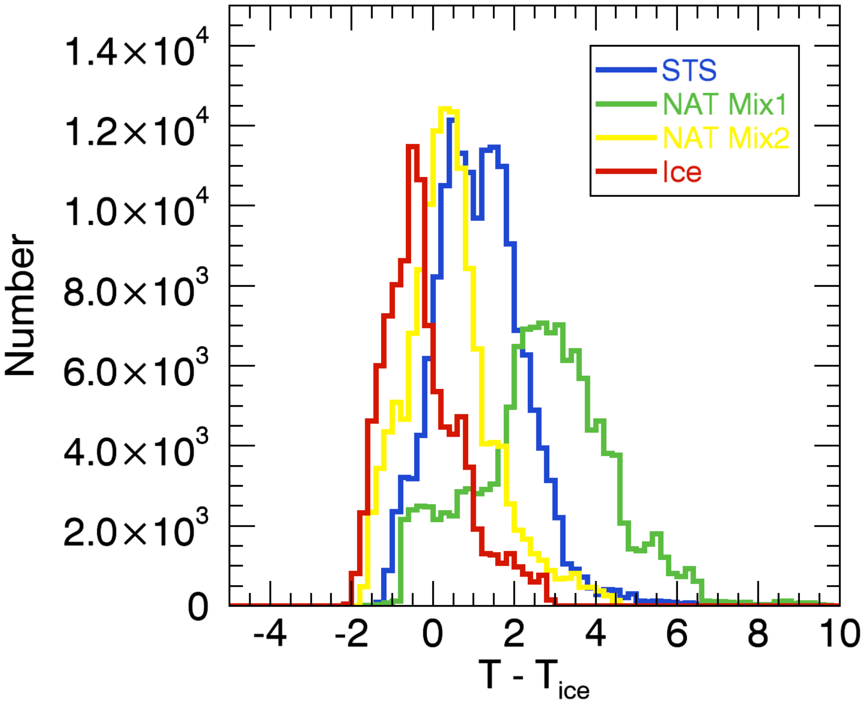

Fig. 2. Occurrence histogram of the 22 Janaury 2016 PSC types with respect to temperature difference to T_ice for the 1/R_ice threshold of 0.2.