Widespread polar stratospheric ice clouds in the 2015/2016 Arctic winter – Implications for ice nucleation

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Abstract. Low planetary wave activity led to a stable vortex with exceptionally cold temperatures in the 2015/2016 Arctic winter. Extended areas with temperatures below the ice frost point temperature T_{ice} persisted over weeks in the Arctic stratosphere as derived from the 36-years temperature climatology of the ERA-Interim reanalysis data set of the European Center for Medium Range Weather Forecast ECMWF. These extreme conditions promoted the formation of widespread polar stratospheric ice clouds (ice PSCs). The space-borne Cloud-Aerosol Lidar with Orthogonal Polarization CALIOP instrument onboard the CALIPSO satellite (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) continuously measured ice PSCs for about a month with maximum extensions of up to 2×10^6 km^2 in the stratosphere. On 22 January 2016, the WALES (Water Vapor Lidar Experiment in Space - airborne demonstrator) lidar onboard the High Altitude and Long Range Research Aircraft HALO detected an ice PSC with a horizontal length of more than 1400 km. The ice PSC extended between 18 and 24 km altitude and was surrounded by nitric acid trihydrate (NAT) particles, supercooled ternary solution (STS) droplets and particle mixtures. The ice PSC occurrence histogram in the backscatter ratio to particle depolarization ratio optical space exhibits two ice modes with high or low particle depolarization ratios. Domain filling 8-days back-trajectories starting in the high-depol ice mode are continuously below the NAT equilibrium temperature T_{NAT} and decrease below T_{ice} ~10 hours prior to the observation. Their matches with CALIPSO PSC curtain plots demonstrate the presence of NAT PSCs prior to high-depol ice suggesting that the ice had nucleated on NAT. Vice versa, STS or no PSCs were detected by CALIPSO prior to low-depol ice. In addition to ice nucleation in STS potentially with meteoric inclusions,

we find evidence for ice nucleation on NAT in the Arctic winter 2015/2016. The observation of widespread Arctic ice PSCs with high or low particle depolarization ratios advances our understanding of ice nucleation in ~~cold~~ polar ~~and tropical~~ latitudes. It further provides a new observational data base for the parameterization of ice nucleation schemes in atmospheric models.

5 **1 Introduction**

While synoptic-scale ice PSCs commonly occur in the Antarctic winter stratosphere (Solomon et al., 1986), widespread ice PSCs extending over several thousand km² have rarely been observed in the Arctic. Even since enhanced observational coverage of the polar regions by the CALIOP instrument (Pitts et al., 2009; Pitts et al., 2011) onboard the CALIPSO satellite, synoptic-scale ice PSCs were detected only occasionally in the Arctic (Pitts et al., 2013; Engel et al., 2013; Achtert

10 and Tesche, 2014; Koshrawi et al., 2016). ~~Similarly only S~~sporadic evidence for synoptic scale ice PSCs in the Arctic is also derived from infrared emission measurements from space with the Michelson Interferometer for Passive Atmospheric sounding (MIPAS) (Spang et al., 2017). Generally Arctic stratospheric temperatures are above the ice frost point temperature T_{ice} (Murphy and Koop, 2005) on synoptic scales and hence limit the formation of extended ice PSCs. The reason for the warmer temperatures in the Arctic compared to the Antarctic is a more alternated land-ocean contrast in the northern

15 hemisphere, supporting the generation of planetary waves, which disturb and hence weaken the Arctic polar vortex due to inmixing of warmer mid-latitude air (Solomon, 2004). In addition, radiative heating of the displaced and elongated vortex contributes to warmer Arctic vortex temperatures.

Ice PSCs exist at cold conditions with temperatures below T_{ice}, while other PSC types prevail at higher temperatures. These can contain NAT particles (Voigt et al., 2000a; Fahey et al., 2001), STS (Dye et al., 1992; Carslaw et al., 1994; Schreiner et

20 al., 1999a; Voigt et al., 2000b) and particle mixtures. Other nitric acid containing solid particle types such as nitric acid dihydrate NAD (Stetzer et al., 2006), or nitric acid water condensates (Thornberry et al., 2013; Gao et al., 2015) have been measured in laboratory. However, robust atmospheric evidence for NAD is missing so far (Höpfner et al. 2006a). Complementary to in-situ particle composition measurements (e.g. Schreiner et al., 1999b, Northway et al., 2002), lidar measurements led to a cloud type classification based on the optical properties of solid or liquid PSC particles (Toon et al.,

25 2000). PSCs with depolarization below 0.04 at 532 nm wavelength were classified as STS (Pitts et al., 2009). PSCs with higher depolarization were labelled Mix1 and Mix2, probably NAT clouds with lower or higher NAT particle number densities, respectively, and some amounts of STS. Finally depolarizing PSCs with backscatter ratios above 5 were classified as ice. Later studies (Pitts et al., 2011; Pitts et al., 2013; Achtert and Tesche, 2014) explored a more detailed differentiation of PSC types. In addition or in combination with lidar measurements, infrared emission measurements allow for advanced

30 atmospheric PSC composition analysis and a classification in PSC type fractions (e.g. Höpfner et al., 2006a&b, Lambert et al., 2012).

Large ice crystals in PSCs may sediment down and transport water vapor to lower altitudes (Fahey et al., 1990; Schiller et al., 2002). Further, due to their large surface areas, ice PSCs very efficiently process halogen compounds (Hanson and Ravishankara, 1992) and hence contribute to ozone loss (Toon et al., 1989). Processing of halogenated reservoir gases on PSC particles leads to a release of unstable chlorine and bromine species, which become activated by sunlight in polar spring

5 and effectively destroy ozone in catalytic cycles (Farman et al., 1985; Crutzen and Arnold, 1986). Ozone depletion is stopped, when the activated chlorine and bromine species react with nitrogen dioxide to reform stable reservoirs. ~~The deactivation of chlorine species can be delayed under denitrified conditions, when the nitric acid concentration in the stratosphere is reduced through sedimenting NAT particles or nitric acid containing ice particles.~~ In the absence of ice at warmer temperatures, other PSC types can provide surfaces necessary for heterogeneous chlorine processing (Grooß et al.,

10 2005; Manney et al., 2011; Drdla and Müller, 2012). E.g. in the Arctic winters 2009/10 and 2010/11 (Manney et al., 2011; Sinnhuber et al., 2011; von Hobe et al., 2013), polar ozone loss ~~was~~ may have largely been driven by STS ~~aerosol~~ (Wohltmann et al., 2013) and NAT (Nakajima et al., 2016). ~~Using multi-year global modeling, Kirner et al. (2015) suggest that also in the Antarctic, a major fraction of ozone loss results from chlorine processing on liquid aerosol.~~ In these cases, the role of ice as transporter for nitric acid enhancing denitrification and slowing down ozone loss may gain importance.

15 Ice PSCs may nucleate homogeneously in liquid STS aerosol or heterogeneously by the aid of solid particles (Toon et al., 1989; Peter, 1997; Zondlo et al., 2000). Homogeneous ice nucleation in STS may occur at high cooling rates as observed in localized mountain wave events (e.g. Dörnbrack et al., 2002). On synoptic scales in the Arctic, the ice saturation ratio S_{ice} rarely increases above the homogeneous ice nucleation threshold of ~1.6 for supercooled ternary solution droplets (Koop et al., 2000). Temperatures rarely decrease 3 to 4 K below T_{ice}, therefore homogeneous ice nucleation ~~rates~~ (Koop et al., 2000)

20 might play a minor role in the Arctic on the synoptic scale. ~~generally are too low to allow for homogeneous ice nucleation in the Arctic.~~ Heterogeneous ice nucleation was investigated in the laboratory by Hoose and Möhler (2012) for various ice nuclei down to temperatures of -60°C. In the polar stratosphere, heterogeneous ice nucleation in STS with meteoric dust (Cziczo et al., 2001; Curtius et al., 2005; Weigel et al., 2014) aided by small scale temperature fluctuations helped to explain the formation of a synoptic-scale ice PSC in the Arctic observed in January 2010 (Pitts et al., 2013; Engel et al., 2013).

25 Here we present new measurements of large scale ice PSCs in the Arctic winter 2015/2016 and discuss ice nucleation pathways. In addition to ice nucleation in STS with meteoric dust inclusions as proposed by Engel et al. (2013), we suggest that ice nucleation on pre-existing NAT might be required to explain a second branch in the ice PSC occurrence histogram. First, we describe the instrumentation and methods. Then we give an overview of the meteorological conditions, which led to a strong cooling of the polar vortex in the Arctic winter 2015/2016. Ice PSCs were observed over elongated periods by the

30 spaceborne CALIOP lidar as described in section 4. On 22 January 2016, differential absorption lidar measurements of an elongated ice PSC were performed onboard the HALO research aircraft during the POLSTRACC (Polar Stratosphere in a Changing Climate) campaign. Based on the PSC occurrence histogram in the backscatter ratio to depolarization optical space, we define a threshold for the $1/R_{ice}$ threshold for ice and investigate the effect of different thresholds on ice PSC occurrence in sensitivity studies. The PSC histogram shows two branches in ice PSC occurrence. We calculate back-

trajectories starting in each of the two branches and investigate possible ice formation pathways for each branch. Finally we discuss the implications of the suggested ice formation pathway on NAT for Arctic PSCs and tropical ice clouds and suggest a way forward for PSC modeling.

2 Instrumentation and methods

5   In this study, we use lidar measurements from the research aircraft HALO and from the CALIPSO spacecraft in combination with meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) numerical weather prediction model to investigate occurrence and formation of ice PSCs in the Arctic winter 2015/2016 with a special focus on 22 January 2016.

10   **2.1 WALES lidar measurements on HALO during the POLSTRACC campaign**

From 7 December 2015 till 20 March 2016, the international field campaign PGS (POLSTRACC/GW-CYCLE/SALSA) was conducted with the High Altitude and Long Range Gulfstream G550 research aircraft HALO out of Oberpfaffenhofen, Germany (48°N, 11°E) and Kiruna, Sweden (68°N, 20°E). The POLSTRACC campaign particularly focused on investigating polar stratospheric chemistry and dynamics in the 2015/2016 Arctic winter. With a ceiling altitude of 15 km

15   HALO is perfectly suited to study the evolution of the lower part of the polar vortex and to provide specific in-situ PSC information at these altitudes.

PSC abundance above HALO flight altitudes was detected with the WALES lidar instrument (Wirth et al., 2010; Groß et al., 2014). The WALES lidar as configured during POLSTRACC has backscatter channels at 532 nm and 1064 nm wavelengths and additionally a high spectral resolution lidar (HSRL) channel and a depolarization channel at 532 nm wavelength for

20   particle detection. The HSRL capability allows the retrieval of the extinction corrected backscatter coefficient of clouds at 532 nm without assumptions about the phase function of the particles (Esselborn, 2008). The backscatter ratio R is the ratio of the un-attenuated (extinction corrected) total (unpolarized) backscatter coefficient and the molecular backscatter. The extinction correction is done by the HSRL channel using a molecular reference profile calculated from pressure and temperature data from ECMWF operational analyses at ~ 16 km horizontal resolution (6 h temporal resolution) and short

25   term forecasts (1 h steps) to interpolate between the analyses. To further distinguish between particles of different type we use the depolarisation of the linear polarised laser light caused by scattering on non-spherical particles. The depolarisation caused by molecular scattering is removed from the signal, following the method outlined by Freudenthaler et al. (2009). The quantity used further is therefore called linear *particle* depolarisation ratio. The relative sensitivity of the two polarized channels is recalibrated regularly during flight to guarantee reliable depolarization values. The particle depolarisation ratio is

30   sensitive to the particle shape and size. Spherical particles do not depolarize and particles much smaller than the wavelength of the laser light also show unmeasurable low values. But in general there is no simple relation between depolarization and

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particle shape, size or composition, see for example Reichardt et al. (2002) for a more detailed discussion of this topic. Nevertheless, cloud regions which show distinct depolarisation ratios point to a significantly different shape or size distribution. In combination with the backscatter ratio R, the particle depolarisation has been successfully used to discriminate different PSC types from ground based, airborne and spaceborne lidar measurements (e.g. Pitts et al. (2009, 2011, 2013), Achtert and Tesche (2014) and references therein).

2.2 Spaceborne CALIOP lidar data

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar on board the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite measures backscatter at wavelengths of 1064 nm and 532 nm, with the 532 nm signal separated into parallel and perpendicular polarization components. The general performance of CALIOP and calibration of the CALIOP data are discussed in Hunt et al. (2009) and Powell et al. (2009). The PSC results in this paper are based on the Version 2.0 CALIOP PSC detection and composition discrimination algorithm (Pitts et al., 2018), which uses night time-only profiles from 8.2 to 30 km altitude of CALIOP V4.10 Lidar Level 1B 532 nm data smoothed to a uniform 5 km horizontal (along track) by 180 m vertical resolution grid.

Following the methodology of Pitts et al. (2009; 2013), PSCs are detected as statistical outliers relative to the background stratospheric aerosol population in either 532 nm perpendicular backscatter (β_{perp}) or 532 nm scattering ratio R, which is the ratio of total backscatter to molecular backscatter. Successive horizontal averaging (5, 15, 45, and 135 km) is also used to ensure that strongly scattering PSCs (e.g., fully developed STS and ice) are found at the finest possible spatial resolution while also enabling the detection of more tenuous PSCs (e.g., low number density liquid-NAT mixtures) through additional averaging. CALIOP PSC composition classification is based on comparing CALIOP data with temperature-dependent theoretical optical calculations of β_{perp} and R for non-equilibrium mixtures of liquid (binary H_2SO_4-H_2O or STS) droplets and NAT or ice particles. The assumption of NAT instead of nitric acid dihydrate (NAD) particles is based on Höpfner et al. (2006), who found no spectroscopic evidence for the presence of NAD from MIPAS observations of PSCs over Antarctica in 2003. The main improvement in the Version 2.0 PSC algorithm in regard to the present paper(Pitts et al., 2018) is that the threshold value of R separating ice and NAT mixture PSCs (R_{ice}) is calculated as a function of altitude and time based on the observed abundance of nitric acid and water as estimated from nearly coincident Aura MLS measurements (Manney et al., 2016). Thus in the Version 2.0 R_{NAT} R_{ice} (or 1/$R_{NAT}$$R_{ice}$) take into account the impact of denitrification/dehydration on the optical signature of ice and NAT mixture clouds. In mid-late January 2016, 1/R_{NAT} R_{ice} range between 0.2 and 0.4 in the 18-24 km altitude region. The CALIPSO PSC areal coverage is estimated as the sum of the occurrence frequency (number of PSC detections divided by the total number of observations) in ten equal area latitude bands spanning 90°-50° N, multiplied by the area of each band. We assume that the CALIOP observations from the approximately 15 daily orbits are representative of the PSC coverage within each latitude band. The data are aggregated on daily time scales and smoothed over 7 days to reduce noise.

2.3 Meteorological data sets

We use meteorological data from two operational analyses of the Integrated Forecasting System (IFS) of the European Centre for Medium-Range Weather Forecasts (ECMWF) to describe the meteorological conditions of the polar stratosphere in winter 2015/2016. From December 2015 to March 2016, the IFS produced the operational analyses cycle 41r1 (T_L1279L137) with a horizontal resolution of about 16 km (0.~~25~~14° × 0.~~25~~14° at the equator) and the experimental IFS cycle 41r2 (T_C1279L137) simultaneously with a higher resolution of about 8 km (0.~~125~~07° × 0.~~125~~07° at the equator). The later IFS cycle became operational after 8 March 2016 (Holm et al., 2016). The enhanced horizontal resolution was achieved by changing from linear to cubic spectral truncation and introducing an octahedral reduced Gaussian grid (Malardel and Wedi 2016). Here, we show data in both resolutions for the 1 December 2015 to 8 March 2016 period to investigate the effects of a higher resolution on the meteorological data set, after 8 March 2016 the high resolution data are presented. We give 6-hourly operational analysis and use 1-hourly forecast data to interpolate between the time steps.

In addition, to compare to previous years, we use 6-hourly ERA-Interim reanalysis data (Dee et al. 2011) retrieved at a horizontal resolution of (1° × 1°). ERA-Interim is a global atmospheric reanalysis from 1989 to today. The data assimilation system used to produce ERA-Interim data is based on the release of the IFS cycle 31r2 (T_L255L60) in 2006. The system includes a 4-dimensional variational analysis with a 6-hour analysis window. The spatial resolution of the data set is ∼ 80 km horizontally on 60 vertical levels from the surface up to 0.1 hPa. ERA-Interim data can be downloaded from the ECMWF public datasets web interface or from MARS archive. A detailed documentation of the ERA-Interim data archive is given by Berrisford et al. (2011).

We derive the minimum temperature T_{min} (K) between 65° and 90°N at the 30-hPa pressure surface from the ERA-Interim reanalysis and from the two operational analyses of the IFS. We further calculate the area A_{ice} with temperatures below the ice frost point T_{ice} using Murphy and Koop (2005) and the area A_{NAT} with temperatures below the NAT equilibrium temperature T_{NAT} using Hanson and Mauersberger (1988) for 4.6 ppmv H_2O and 7 ppbv HNO_3, as measured by MLS in the Arctic vortex in January 2016 (Manney and Lawrence, 2016).

3 Temperature evolution of the Arctic stratosphere in the 2015/2016 winter

The temperature evolution in the Arctic winter stratosphere is influenced by planetary wave activity. In early winter 2015 a strong tropical tropospheric temperature anomaly reinforced the meridional temperature gradient from the tropics to the poles which led to adverse conditions for the propagation of planetary waves (Matthias et al., 2016). Weak planetary wave activity measured as low meridional heat flux thus enforced the formation of a strong and stable polar vortex and caused extremely low temperatures in the Arctic winter 2015/2016 (Dörnbrack et al., 2016).

Therefore, stratospheric temperatures decreased dramatically as derived from the meteorological data of the IFS of the ECMWF numerical weather prediction model. Figure 1A shows the evolution of T_{min} at latitudes > 65°N and at 30 hPa in the Arctic winter 2015/2016 in two resolutions. In December 2015, T_{min} decreased below T_{ice} and then remained below T_{ice} from

late December 2015 till end of January 2016. Within the first cold phase, T_{min} down to 182 K were detected in the operational IFS analysis with 16 km horizontal resolution. In February 2016, three minor stratospheric warmings influenced the vortex and led to warmer conditions in a coherent but slightly displaced polar vortex (Manney and Lawrence, 2016). Then, the final stratospheric warming resulted in a split of the vortex by mid-March and the subsequent dissipation of the vortices associated with a temperature increase of more than 20 K at 30 hPa in a few days.

The exceptional coldness of the Arctic winter 2015/2016 also reflects in the fact that by the end of December 2015, T_{min} dropped even below the minimum temperatures $> 65°N$ at 30 hPa ever obtained by the ERA interim data record extending from 1989 to 2016 (Dee et al., 2011). Throughout the Arctic winter 2015/2016, T_{min} was continuously lower than the mean temperature of the 36-years ERA interim data record (Voigt et al., 2016), see Figure 1.

To illustrate the effect of mesoscale temperature fluctuations on T_{min}, we also show T_{min} derived from the IFS cycle 41r2 at ~ 8 km horizontal resolution compared to the cycle 41r1 at ~ 16 km resolution at 30hPa for the December 2015 to 8 March 2016 time period where both data sets are available. Temperature deviations up to 7 K between the higher and lower resolution data sets occurred during the first cold phase with $T_{min} < T_{ice}$ in December and early January as shown in Figure 1A. At higher resolution, minimum temperatures reach down to 179 K on the 30 hPa level. In the second cold period at the end of January 2016, temperature deviations up to 3 K are found in the higher resolution data set. Mesoscale gravity wave activity in December and January 2016 is better covered in the higher resolution data and could have caused this temperature difference (Dörnbrack et al., 2016). On 8 March 2016, the two data sets merge and from then on, ECMWF operational analysis are given at ~ 8 km resolution.

At 30 hPa, the area A_{ice} with $T < T_{ice}$ extended over regions up to 3.6×10^6 km^2 (see Figure 1B), as derived from IFS cycle 41r1 analysis at 16 km resolution. From 18 January till the end of the month, A_{ice} was continuously more than one order of magnitude larger than the 36-years average and larger than the maximum of the 36-years ERA interim data record. In addition the area A_{NAT} with $T < T_{NAT}$ in January 2016 continuously reached the maximum of the 36-years ERA interim data set (Figure 1C). Generally A_{ice} and A_{NAT} are lower than the mean Antarctic conditions as given by the dashed gray line in Figure 1B and C (shifted by 6 months to account for seasonality).

These extremely cold stratospheric winter conditions in the Arctic set the stage for synoptic-scale PSC formation in winter 2015/2016.

4 Occurrence of ice PSCs in the Arctic winter 2015/2016 derived from CALIOP observations

The CALIOP lidar onboard the CALIPSO spacecraft detected PSCs from December 2015 to January 2016 (Figure 2). On 28 January 2016 the CALIOP science data acquisition was suspended due to a spacecraft anomaly. The problem was subsequently resolved and data acquisition began again on 14 March 2016.

Ice PSCs were measured by CALIOP continuously for a month from late December 2015 to late January 2016 (Figure 2A). The ice PSCs were observed at altitudes between 15 and 26 km during the period with extremely cold temperatures inside

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the Arctic vortex. The maximum extension of ice PSCs derived from CALIOP $A_{ice,max}$ of $(1.75 – 2.0) \times 10^6$ km^2 is reached on 30 December 2015. In this phase, the ice PSC formation was <u>predominantly</u> triggered by mountain wave activity and spread out to synoptic scales, see Dörnbrack et al. (2016). Considering the uncertainties in A_{ice} retrieval from the CALIOP data set interpolated to latitude bands, the maximum A_{ice} derived from CALIOP observations agrees reasonably with

5   maximum A_{ice} derived from weather forecast IFS data of 2.1×10^6 km^2 at 30 hPa (~21.6 km). Also the second peak in ice PSC occurrence A_{ice} on 24 January 2016 is captured by the CALIOP routine though with a smaller peak amplitude. A decrease of water vapor concentrations and dehydration due to falling ice crystals has been observed by MLS (Manney and Lawrence, 2016) at these altitudes throughout January 2016. This may explain lower A_{ice} derived from CALIOP data at the end of January compared to A_{ice} derived from in the IFS data set using a fixed H_2O mixing ratio of 4.6 ppmv to calculate

10   A_{ice}.

NAT <u>mixtures</u> and STS PSCs were present in the CALIOP data set from early December 2015 till the end of the observation period (Figure 2B). Summarized, we find strong evidence for the unprecedented existence of widespread ice PSCs in the Arctic winter 2015/2016.

5 Optical properties of the PSC measured on 22 January 2016

15   PSCs were detected with the WALES lidar inside the vortex on all 6 HALO flights between 22 January and 29 February 2016. Before that <u>date</u>, the lidar on HALO was not operational. Due to the strong temperature increase at the end of January and measurement locations of HALO in warmer parts of the vortex, extended ice PSCs were observed by WALES solely on 22 January 2016.

5.1 Extension of the PSC on 22 January 2016

20   A large synoptic scale PSC was measured by WALES during a flight from Kiruna to the northern tip of Greenland on 22 January 2016 as shown in Figure 3. The PSC extended between 14 and 25 km altitude over a horizontal distance of 2200 km. It was continuously observed within in the Arctic vortex from 72°N to the outermost return point at 86°N. High backscatter ratios are indicators for the presence of large particle surfaces and high depolarization ratios suggest the presence of solid particles. The co-located measurements of the particle backscatter ratio and depolarization ratio thus allow for a cloud

25   classification into different PSC types.

5.2 Classification of the PSC measured on 22 January 2016

Figure 4 shows the joint occurrence histogram of the inverse backscatter ratio 1/R and particle depolarization for the PSC measurements in Figure 3. The histogram bin size is 0.02×0.02 and the color scale indicates the number of cloud observations (4 km horizontal by 100 m vertical) falling within each bin. Overlaid are the regions which correspond to

30   different PSC types following Pitts et al. (2011) with two modifications. First the sub-classification of Mix2 into Mix2-

enhanced and normal Mix2 is dropped. Second, the $1/R_{ice}$ threshold for ice and NAT regions is set to 0.3. Without the latter change, a substantial part of the branch connecting STS and fully developed ice clouds would have been counted as NAT Mix2 instead of ice, while ice is the more obvious interpretation of the lower branch in the ice class given the form of the joint histogram. The $1/R_{ice}$ threshold for ice and NAT regions is discussed in more details below.

We find two modes in the joint histogram of the ice class, a mode with high particle depolarization ratio (high-depol ice) connected to the NAT Mix2 regime and a mode with lower particle depolarization ratio at the same backscatter ratio (low-depol ice) connected to the STS class. Given several ten thousands individual measurement points within the ice class, these two different ice modes can clearly be distinguished. The NAT Mix2 regime with enhanced NAT particle concentrations is strongly populated. In contrast, no observation falls into the wave ice class forming in strong mountain waves.

We ~~now~~ use the classification by Pitts et al. (2011) and the threshold of $1/R_{ice} = 0.3$ of ice versus NAT Mix2 to classify the PSC from Figure 3. A large ice PSC (red areas in Figure 5) extends in a cloud layer with a vertical thickness up to 6 km between 18 and 24 km altitude over distances of over 1400 km. The ice PSC is measured twice for more than 1.6 h during the outbound flight leg between 10:34 and 12:30 and during the inbound flight leg between 13:50 and 15:32 UTC. The synoptic-scale ice PSC is observed mainly at temperatures below T_{ice} as indicated by the dashed contour lines. Here T_{ice} is calculated based on temperature data from the integrated forecast system IFS (cycle 41r1) of ECMWF and measured water vapor mixing ratios. In the northern part of the cloud between 12:30 and 13:50 UTC, the ice PSC is surrounded by NAT Mix2 cloud layers (yellow area) extending above and next to the ice PSC to the North-East. Within the ice PSC, NAT can be masked by the higher optical signal from ice, but may be present. ~~The combination of a high depolarization ratio with a quite low color ratio (ratio of the backscatter coefficient for the 532 nm channel and the 1064 nm channel, not shown here) of 1.5 at the bottom of the PSC (green area) points to larger NAT particle sizes, thus suggesting sedimenting NAT particles in the lowest part of the cloud. In addition,~~ The southernmost part of the PSC observed between 10:15 and 10:34 and again between 15:15 and 16:00 UTC consists of non-depolarizing liquid STS droplets (blue region). The STS layer extends towards the South and below the ice layer.

The ice, NAT Mix and STS occurrence histogram of this PSC is plotted versus temperature difference to T_{ice} in Figure 6. The peak in ice occurrence is located 0.5 K below T_{ice}. The peak in NAT Mix2 (NAT Mix1) occurrence appears at 0.5 K (3 K) above T_{ice}, while STS occurrence peaks 0.5 to 1.5 K above T_{ice}. The occurrence of ice at temperatures below T_{ice} and of NAT Mix and STS at temperatures above T_{ice} is consistent with their expected temperature range, see also Pitts et al. (2013). The $1/R_{ice}$ threshold of 0.3 best matches the PSC measurements in the low-depol ice mode (see Figure 4). Also, the PSC area classified as ice agrees best with the ice area derived from numerical weather predictions. For this threshold, the peak of the NAT Mix2 occurrence is located above T_{ice}, while it is located near and below T_{ice} for $1/R_{ice}$ of 0.2. Therefore we use the threshold $1/R_{ice}$ of 0.3 for the WALES observations on 22 January 2016. The analysis of CALIPSO measurements throughout the winter 2015/16 with changing HNO_3 and H_2O concentrations requires a variable $1/R_{ice}$ threshold as used by Pitts et al. (2018). A detailed investigation of the $1/R_{ice}$ threshold is given in the supplementary material S1.

The PSC was also measured by CALIPSO on 22 January 2016 between 10:42 and 10:47 UTC. Figure 7 shows the CALIPSO curtain plot measured at the CALIPSO footprint near the HALO flight track shown in Figure 8. The classification by Pitts et al. (2018) is used to determine the PSC types from CALIPSO. PSC observations by WALES on the slower flying HALO aircraft lead to the time differences of up to 2 h between the collocated PSC measurements. Similar to the WALES observations, the CALIOP lidar detected an ice PSC between 18 and 24 km altitude with a horizontal extension of about 1200 km. An STS layer is located below the ice cloud and predominantly NAT Mix1 PSCs are measured to the North-West. Few NAT Mix2 PSCs were measured by CALIPSO, mainly located at the edge of the ice cloud. Differences in the flight paths of HALO and the CALIPSO footprint explain the lower number of NAT Mix2 PSCs observed by CALIPSO compared to WALES. WALES measured NAT Mix2 mainly north-west of the CALIPSO footprint. Also a fraction of the low-depol ice mode was located predominantly north-west of the CALIPSO PSC curtain and therefore merely covered by CALIPSO.

6 Discussion of PSC formation

We now discuss PSC formation in two steps based on the WALES lidar and CALIPSO observations combined with trajectory analysis. First we investigate the formation of the ice, NAT and STS layers of the PSC observed on 22 January 2016 using 8 days back-trajectories starting in the different PSC layers at 21.5 km altitude every 2 minutes. Then we separate the ice PSC into a mode with high particle depolarization (high-depol ice mode) and a mode with low depolarization (low-depol ice mode) as derived from the backscatter ratio to depolarization occurrence histogram in Figure 4 and discuss ice formation pathways of each ice mode based on domain filling Lagrangian back-trajectory calculations and their matches with PSC measurements by CALIPSO up to 5 days prior to the ice. We start with the discussion of the formation of the ice, NAT and STS layers of the 22 January 2016 PSC.

6.1 Temperature history of trajectories starting in ice, NAT **Mix2 and** STS PSC layers

The trajectory calculations were performed using the Hybrid Single-Particle Lagrangian Integrated Trajectory dispersion model HYSPLIT (Draxler, 1998) with operational forecast of the deterministic IFS (cycle 41r1 interpolated to $0.25 \times 0.25°$) from ECMWF at 3-hourly time steps as meteorological input. The HALO flight track and ECMWF temperatures at 30 hPa on 22 January 2016 are shown in Figure 7. We calculate 8 days backward-trajectories starting in the PSC observations every 2 min along the HALO flight track (corresponding to approximately 24 km horizontal spacing) at 21.5 km altitude (near 30 hPa) to investigate PSC formation. We further calculate T_{NAT} based on Hanson and Mauersberger (1998) for an altitude dependent climatological HNO_3 profile and use H_2O, T_{ice} and the ice saturation ratio S_{ice} from the meteorological data set to discuss ice nucleation pathways. Further we perform sensitivity studies every 2 minutes at higher and lower altitudes (19, 21 and 23 km) to account for particle sedimentation, which is not included in the simplified trajectory calculations shown here. As a rough estimate, a 10-µm sized ice crystal sediments about 1 km in a day (Fahey et al., 2001). For aspherical particles

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the sedimentation rates are even lower (Woiwode et al., 2014; Weigel et al., 2014; Woiwode et al., 2016), hence generally our simplified calculations cover the altitude range of sedimenting PSC particles.

In Figure 8 we show temperatures of backward-trajectories starting every 2 min in the PSC at 21.5 km altitude at the WALES cross section of the outbound flight leg to the North (corresponding to the section from 10:30 to 12:34 in Figure 5). The air mass trajectories circulate around the pole within these 8 days, the innermost trajectories (NAT Mix2 at the observation) stay near the Arctic cold pool and remain cold (T < 190 K) for 8 days while the outermost trajectories (STS at the time of observation) encounter higher temperatures (T > 198 K) outside of the cold pool. The innermost NAT Mix2 trajectory 4 bypasses Greenland and circulates within the vortex core, while the more southern trajectories are slowly lifted above Greenland. The slow lift contributes to the synoptic cooling within the Arctic cold pool and induces a transient low amplitude mountain wave temperature disturbance in the lee of the Greenlandic island (dark blue areas in Figure 8). The synoptic cooling induces the formation of a large scale ice PSC above and to the East of Greenland within the cold pool of the polar vortex. Teitelbaum et al. (2003) previously investigated the important role of synoptic scale dynamics for PSC formation.

To investigate PSC formation, 4 trajectories representative for STS starting at 10:38 UTC (label 1, circle, blue line in panel B and C), low-depol ice at 11:36 UTC (label 2, filled diamond, red line), in high-depol ice at 12:14 UTC (labels 3, open diamond, orange line) and NAT Mix2 at 12:34 (label 4, square, green line) are selected to represent the temperature histories of the back trajectories of the different PSC types. The numbering of the trajectories is in chronological order during the outbound flight leg and the color coding in panel (B) and (C) corresponds to the PSC type classification in Figure 5, with a blue line for STS, a green line for NAT Mix2 and red line for low-depol ice at the time of the WALES PSC observation. The orange line is used to distinguish the trajectory starting in the high-depol ice mode from the low-depol ice mode (red line). Small symbols denote 48-h time steps along the 8 days backward trajectories.

Starting with the outermost STS trajectory, its temperature varies between T_{NAT}-2K < T < T_{NAT}+11K for 6.5 days as the trajectories circulate mainly out of the Arctic cold pool. Then, the temperature decreases below T_{NAT} ~36 h and below T_{ice} ~16 h prior to the observation before increasing to T_{ice} +2K at the time of STS detection. The temperature at the time of observation is within the STS temperature range.

Slightly further to the North, the low-depol ice mode back trajectory (red line) is located at the edge of the cold pool, with temperatures oscillating between T_{NAT}-6K < T < T_{NAT}+6K for 7 days. The temperatures decrease below T_{NAT} ~36 h and finally below T_{ice} ~16 h prior to the observation. At the time of observation of the low-depol ice mode, the trajectories' temperatures are near and below T_{ice} explaining the detection of ice PSCs. The ice saturation ratio S_{ice} increases to 1.4 and varies between 1.4 and 0.92 in the last 16 h prior to observation. While S_{ice} < 1.4 does not allow for homogeneous ice nucleation (Koop et al., 2002; Murphy and Koop, 2006), heterogeneous nucleation (e.g. Hoose and Möhler, 2012) could explain ice formation in this case. Thereby meteoric material (Czicio et al., 2001; Voigt et al., 2005; Curtius et al., 2005) probabaly embedded in STS could serve as ice nuclei. This has been suggested by Engel et al. (2013) to explain the

observation of an Arctic ice PSC in 2010. Small scale temperature fluctuations might be present in our case, but cannot be resolved in the meteorological model.

In contrast, the northernmost NAT Mix2 trajectory (4, green line) circulates within the cold pool at temperatures T_{NAT}-8K < T < T_{NAT}-4K for 8 days without passing over Greenland. The temperatures remain above T_{ice} throughout that period and T > T_{ice} at the time of observations, consistent with NAT. Homogeneous NAT nucleation rates (Knopf et al., 2002) are too low to support homogeneous NAT nucleation in this case. In contrast, NAT nucleation rates on meteoric dust (Voigt et al., 2005; Grooß et al., 2005; Hoyle et al., 2013; Grooß et al., 2014) may explain the formation of NAT within few days and are consistent with the observations of NAT.

Similarly, the trajectory of the high-depol ice mode (3, orange line) circulates within the polar vortex for 8 days at temperatures between T_{NAT}-10K < T < T_{NAT}. However, when passing over Greenland, the high-depol ice trajectories decrease below T_{ice} ~10 h prior to the observation. The temperature is T_{ice}-2K at the time of observation of high-depol ice and S_{ice} increases up to 1.2. This temperature history supports heterogeneous ice nucleation. Laboratory measurements of heterogeneous ice nucleation (Hoose and Möhler; 2012) were performed for a suite of different ice nuclei at temperatures down to 213 K, hence above PSC formation temperatures. Meteoric dust could potentially serve as ice nuclei in the stratosphere at S_{ice}~1.2, however surface area densities of meteoritic material are significantly smaller than those of NAT PSCs, therefore we investigate the possibility of ice nucleation on NAT and NAT serving as ice nuclei.

The temperature history of the high-depol ice trajectory also remains below the existence temperature of NAD (T_{NAD} ~ T_{NAT}-2.3 K, Voigt et al., 2005) for about a week. The formation of NAD from binary nitric acid water solutions has been observed in the laboratory (e.g. Knopf et al., 2002; Wagner et al., 2005; Stetzer et al., 2006), but homogeneous nucleation rates are too low to explain denitrification (Knopf et al., 2002). Also, pseudo-heterogeneous nucleation rates (Knopf, 2006) cannot explain atmospheric nitric acid hydrate particle number densities. Möhler et al. (2006) suggest that ambient supersaturations with respect to NAD of 8 to 9 are required over days to explain NAD particle nucleation at number densities, which can be detected with current instrumentations. Furthermore, a phase change from NAD into NAT or a nucleation of NAT on NAD under atmospheric conditions is not supported by laboratory experiments (Tizek et al., 2004; Stetzer et al., 2006). Observational evidence for the presence of NAD in the atmosphere is missing so far, e.g. Höpfner et al. (2006) found no spectroscopic evidence for the presence of NAD from MIPAS observations of PSCs over Antarctica. Therefore, we focus the discussion on ice nucleation on NAT in our study.

6.2 Ice nucleation pathways

The existence of the two ice modes with high and low particle depolarization ratios (Figure 4) in combination with the trajectory analysis suggests two different ice nucleation pathways. (1) Ice nucleation in STS may account for the low-depol ice mode. Inclusions of meteoric material in STS as suggested by Engel et al. (2013) are below the detection limit of the WALES lidar but may be present. (2) A second ice nucleation pathway is required to explain the formation of the high-depol ice mode. The long time period below T_{NAT} prior to the observation and the similarity of the NAT Mix2 and the high-depol

ice trajectories (except for the last hours where the high-depol ice trajectories are ice supersaturated) points to ice nucleation on pre-existing NAT particles.

The hypothesis of ice nucleation on NAT ice is investigated in more detail using a Lagrangian match approach of 5 days back trajectories matched with CALIPSO PSC measurements similar to the method outlined by Santee et al. (2002). Domain filling trajectories starting in the WALES ice PSC observations are calculated every 2 minutes along the flight path from 10:30 UTC until 16:00 UTC (corresponding to ~24 km distance) at 15 different altitude levels with 500 m spacing between 17 and 24 km. Thus a total of 2490 trajectories are derived. We match the Lagrangian trajectories with all CALIPSO PSC curtain plots measured north of 60°N within 5 days prior to 22 January 2016. Pitts et al. (2018) is used to classify the PSC types within the CALIPSO curtains. A match is defined as a point along a trajectory lying within a given horizontal distance limit from one of the CALIOP footprints at the same time. To yield a statistically significant number of matches we set a distance limit of 100 km. A threshold of this order is justified by the limited accuracy of the trajectory calculations, too. In addition, to make the spatial resolution comparable with the initial spacing of the trajectories' starting points, 15 CALIPSO PSC data points with 3×180 m vertical and 5×5 km horizontal distance are grouped to smooth variations in the PSC type classification in nearby data points at the edge of PSC layers. A match point is classified as ice, NAT or STS, if > 50% of the CALIPSO pixel array is classified as the respective PSC type. The match method is selected to investigate whether a different PSC type was present before the high-depol and low-depol ice PSC observation by WALES on 22 January 2016. If so, the analysis indicates, which PSC type has been detected prior to high-depol or low-depol ice.

The trajectories are classified using the following criteria: (1) NAT has been measured in the CALIPSO curtain–trajectory match point prior to ice measured by WALES, or prior to the last CALIPSO match with ice on the same trajectory, (2) STS has been observed in the CALIPSO curtain–trajectory match point prior to ice, (3) no PSC (nil) has been observed in the match point prior to ice, or (4) no match (nom) has occurred along the trajectories.

The result of the match analysis is presented in Figure 9 overlaid on the ice PSC contour of the 22 January PSC. The result is clear: NAT has been measured by CALIPSO on all match points directly prior to the high-depol ice mode. Thus the CALIPSO-trajectory match analysis delivers strong evidence for ice nucleation on NAT. Ice could have nucleated on pre-existing NAT particles, which could also explain the high particle depolarization ratio of the ice mode. In contrast, predominantly STS or no PSC has been observed on the match points prior to the low-depol ice mode. We note the possibility that due to fast evaporation times, STS might have existed along the trajectories classified is nil, but was not present at the time of the CALIPSO overpass. Only few trajectories have no match points with CALIPSO.

Summarized, we find strong evidence for ice nucleation on NAT. Ice nucleated on NAT may produce ice particles with higher particle depolarization ratios. In contrast, the low-depol ice mode can be explained by ice nucleation in STS, possibly with solid meteoric inclusions as suggested by Engel et al. (2013). Ice nucleation in STS may lead to lower particle depolarization ratios of the ice mode. These two ice formation pathways would be consistent with the CALPSO and WALES observations combined with results from domain filling trajectory analysis.

7 Conclusions and outlook

Extremely low temperatures existed in the Arctic stratospheric winter 2015/2016 because low planetary wave activity resulted in a stable vortex. Synoptic-scale ice PSCs formed in late December 2015 and persisted throughout January as observed by the CALIOP lidar onboard the CALIPSO satellite. The sedimentation of the ice PSC particles led to significant dehydration (Koshrawi et al., 2017). From January to early March 2016, water vapor data MLS data show severe dehydration between 400 and 500 K potential temperatures (Manney and Lawrence, 2016). In addition, ice PSCs can serve as efficient transporters for nitric acid incorporated into ice. Large ice particles sediment faster than smaller NAT particles and therefore ice can lead to efficient denitrification at PSC altitudes. Massive denitrification has been measured by MLS with an onset in mid-December 2015 throughout the Arctic winter (Manney and Lawrence, 2016; Khosrawi et al., 2017).

We use high resolution WALES lidar measurements of a large-scale ice PSC observed on 22 January 2016 in combination with domain filling back-trajectories matched to CALIPSO PSC observations to investigate ice nucleation pathways. Two distinct modes with high and low particle depolarization in the ice PSC occurrence histogram suggest different particle size distributions and shapes in the two modes, possibly linked to two ice formation pathways. Ice nucleation in STS, possibly with meteoric inclusions as suggested by Engel et al. (2013), may lead to the ice mode with low particle depolarization ratios. In addition, NAT has been detected by CALIPSO on trajectory match points prior to high-depol ice. Thus, ice formation on NAT could lead to the ice mode with higher particle depolarization ratios. While the low-depol STS-ice branch is frequently populated in space-borne CALIOP lidar data, the high-depol NAT Mix2-ice branch less frequently observed by CALIOP in Arctic or Antarctic PSC measurements (Pitts et al., 2013; Pitts et al., 2018). Larger noise in the satellite data with CALIPSO travelling at an orbit near 700 km and a higher detection limit of the CALIOP lidar data compared to the WALES measurements on aircraft cruising at 14 km altitude may contribute to this difference.

Lambert et al. (2012) investigate PSC occurrence in the Antarctic in early winter from 11 to 30 June 2008 based on CALIOP and MLS observations. They detect a NAT Mix2 type PSC linked to the high-depol ice regime. They also note that the conservative estimate by Pitts et al. (2013) of the threshold $1/R_{ice} = 0.2$ may lead to a miss-classification of the ice PSC with respect to the NAT MIX2 regime. The threshold of 0.3 provides a better fit to their observation. Similarly to the Arctic case, the NAT Mix2 trajectories remain 3 to 5 K below T_{NAT} for 5 days, suggesting heterogeneous NAT nucleation. Ice formation is not investigated in detail for this Antarctic PSC, although ice is present. The low-depol ice mode is sparsely populated supporting the hypothesis of ice nucleation on NAT in the Antarctic June 2008 PSC. Further high resolution lidar observations are required to estimate the global occurrence of the high-depol ice mode. These observations could then help to evaluate the importance of the suggested NAT-ice nucleation pathway in other regions of the atmosphere. In addition, combined lidar and in-situ observations from aircraft (e.g. Fahey et al., 2001; Northway et al., 2002) could help to answer the question on the abundance of the NAT Mix2, which might include aspherical NAT particles as observed by Molleker et al. (2014) and Woiwode et al. (2016).

The NAT crystal has a stoichiometry of 1 $HNO_3 \times 3$ H_2O molecules and exists in $\alpha-$NAT or $\beta-$NAT crystal structure (Iannarelli and Rossi, 2015). Compared to liquid aerosol or meteoric particles, the NAT crystal structure is more similar to that of ice, and therefore NAT readily nucleates on ice as frequently observed in laboratory experiments (Hanson and Mauersberger, 1988; Iannarelli and Rossi, 2015; Gao et al., 2016; Weiss et al., 2016) and in mountain wave ice PSCs

5   (Carslaw et al., 2002; Fueglistaler et al., 2002; Luo et al., 2003; Voigt et al., 2003). Vice versa, we propose here that ice may nucleate on NAT. This pathway has been suggested in early PSC studies (e.g. Peter, 1997) and has later been neglected. Here, we find evidence for ice nucleation on NAT and elucidate its importance in the polar lower stratosphere. Direct laboratory measurements of the nucleation rate of ice on NAT are missing (Koop et al., 1997) and would be required to provide further evidence of the suggested ice nucleation process.

10   Ice nucleation on NAT may be responsible for the early onset of ice PSC formation in December 2015. Further, iIce nucleation on NAT may be important in the tropical tropopause region, where the existence of a NAT belt and cirrus has been detected by in-situ measurements (Popp et al., 2007, Voigt et al., 2008). Also CALIPSO measurements showed indications for the existence of NAT in the tropics (Chepfer and Noel, 2009), although Pitts et al. (2009b) argue that mixtures of liquid aerosols and thin cirrus instead might have been misinterpreted as NAT-like particles in the tropics. Based

15   on the observational data set, the ice nucleation rate on NAT could be parameterized and implemented in a large scale model in order to assess the global relevance the different ice nucleation pathways in different regions of the atmosphere.

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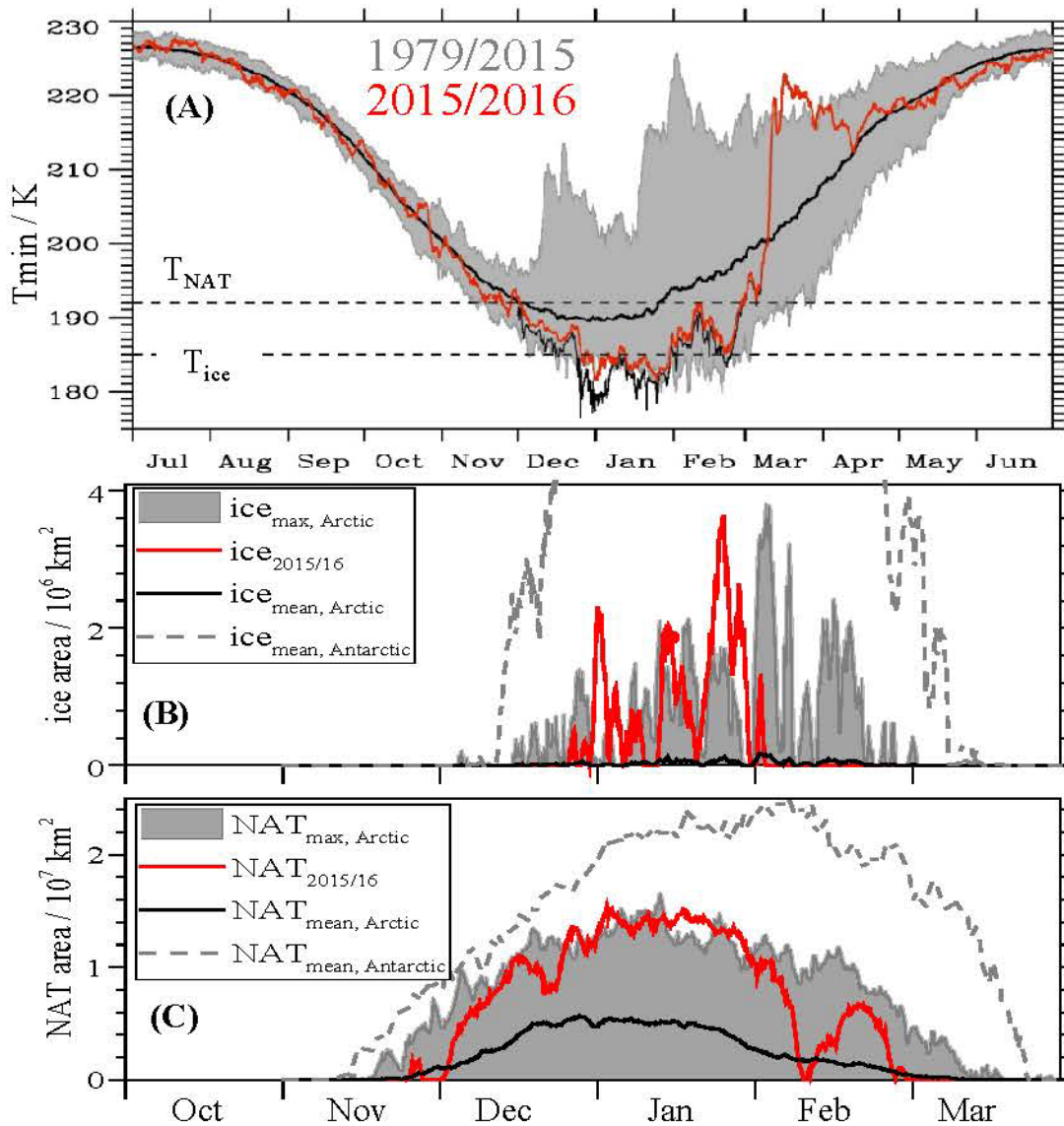
Figure 1: Temperature evolution of the 2015/2016 Arctic winter stratosphere (A) 6-hourly ECMWF ERA interim reanalysis (Dee et al. 2011) data retrieved at a horizontal resolution of 1°: minimum temperature T_{min} (K) between 65° and 90°N at the 30-hPa pressure surface. The thick black line denotes the mean values of T_{min} averaged during 1979 to 2015, and the shaded areas encompass the minimum

5   and maximum values of T_{min} between 1979 and 2015. The red line marks the evolution of T_{min} from operational analyses of the IFS cycle 41r1 until 8 Mar 2016. The thin black line indicates T_{min} from the IFS cycle 41r2 in the preoperational phase from 1 December 2015 to 8 Mar 2016 retrieved at a resolution of 0.125°. After 8 March 2016, the black line continues as red curve of the operational IFS cycle 41r2. T_{ice} (Murphy and Koop, 2005) and T_{NAT} (Hanson and Mauersberger, 1988) are calculated using 4.6 ppmv H_2O and 7 ppbv HNO_3, relevant for the 2015/2016 Arctic vortex conditions (Manney and Lawrence, 2016). (B) Evolution of the vortex area with temperatures below T_{ice}.

10   The black line marks the mean area below T_{ice} (A_{ice}) at 30 hPa pressure (~21.6 km) between 1979 and 2015. The gray shading indicates maximum and minimum A_{ice} in the same time period. The red line shows the evolution of A_{ice} at the 30 hPa pressure surface in the Arctic winter 2015/2016 of the IFS cycle 41r2. The gray dashed line gives the mean area below T_{ice} at 30 hPa south of 65°S from 1979 to 2015, shifted by 6 months to account for seasonality. (C) Same data for NAT.

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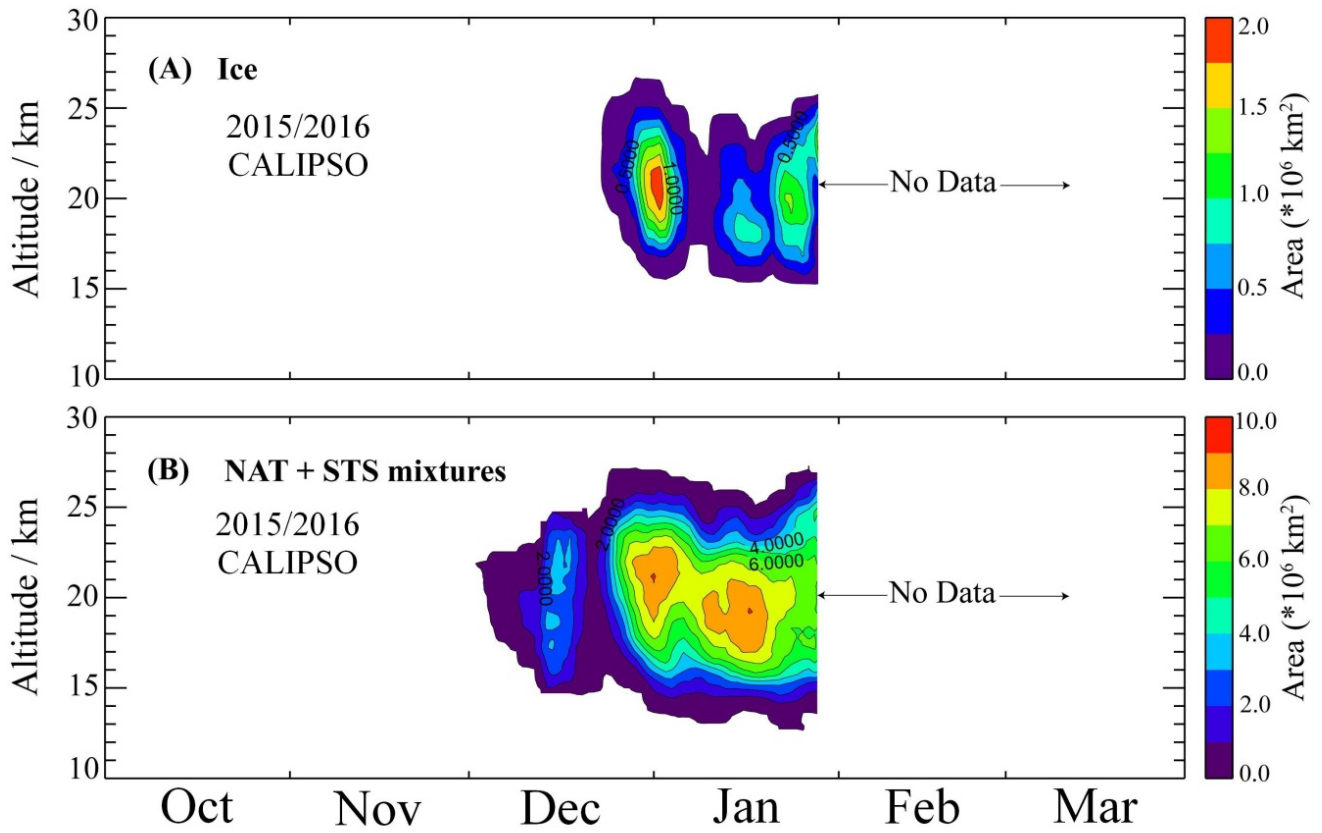
5   **Figure 2: Curtain plot of the areal occurrence of PSCs in the winter 2015/2016 detected by the CALIOP lidar** onboard the CALIPSO satellite using the classification from Pitts et al. (2018). (A) Curtain plot of ice PSC occurrence. (B) Curtain plot of NAT and STS mixtures PSC occurrence. ~~Synoptic I~~ice PSCs were observed by CALIOP from ~~mid~~end of December 2015 till end of January 2016.
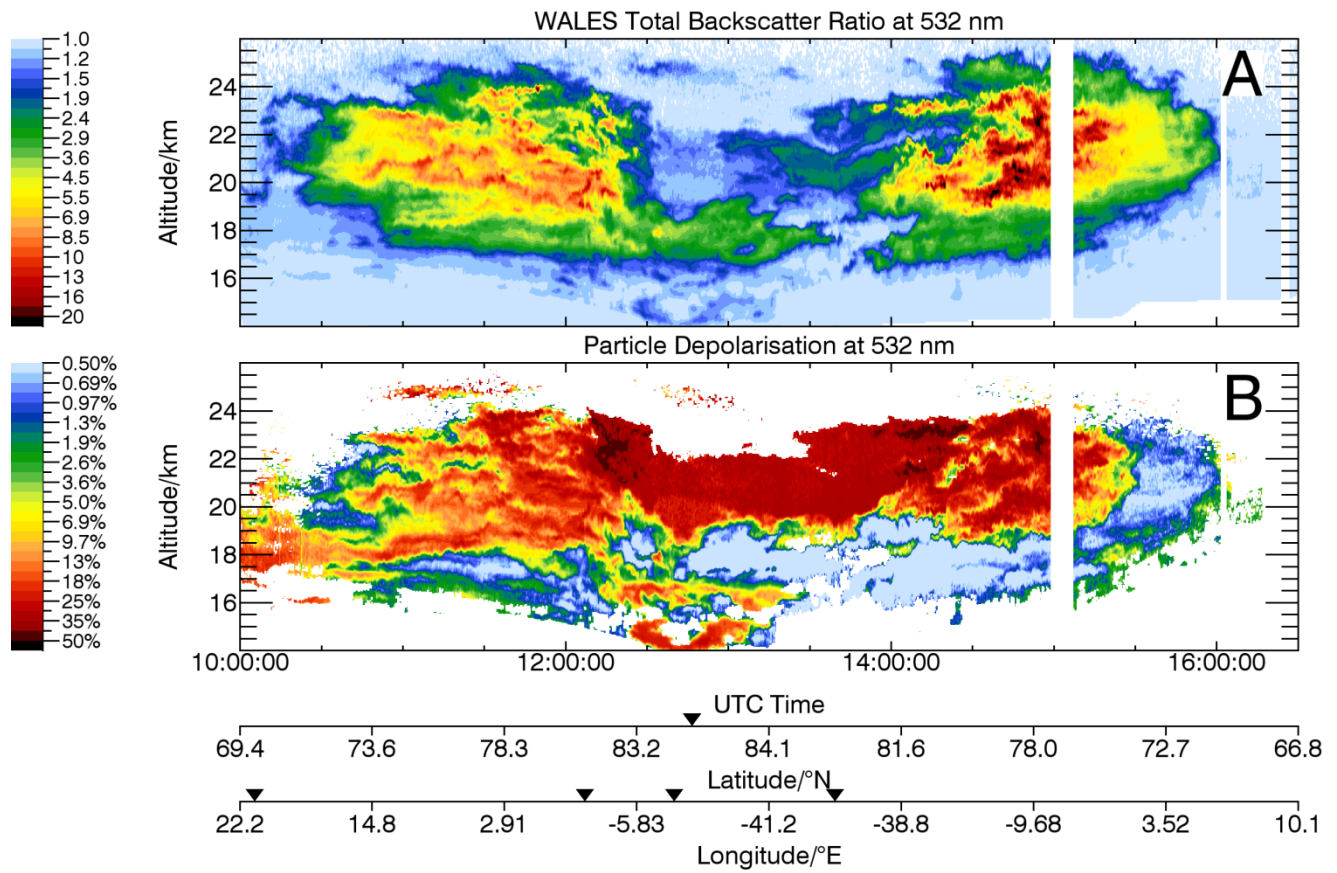
Figure 3 Lidar observation of a synoptic-scale polar stratospheric cloud observed on 22 January 2016 (A) Backscatter ratios from the WALES lidar (Wirth et al., 2009) at 532 nm wavelengths during a HALO flight into the Arctic vortex and (B) particle depolarization. The time of the flight, as well as latitude and longitude of the HALO flight path are indicated. Turning points of the HALO are marked by triangles.
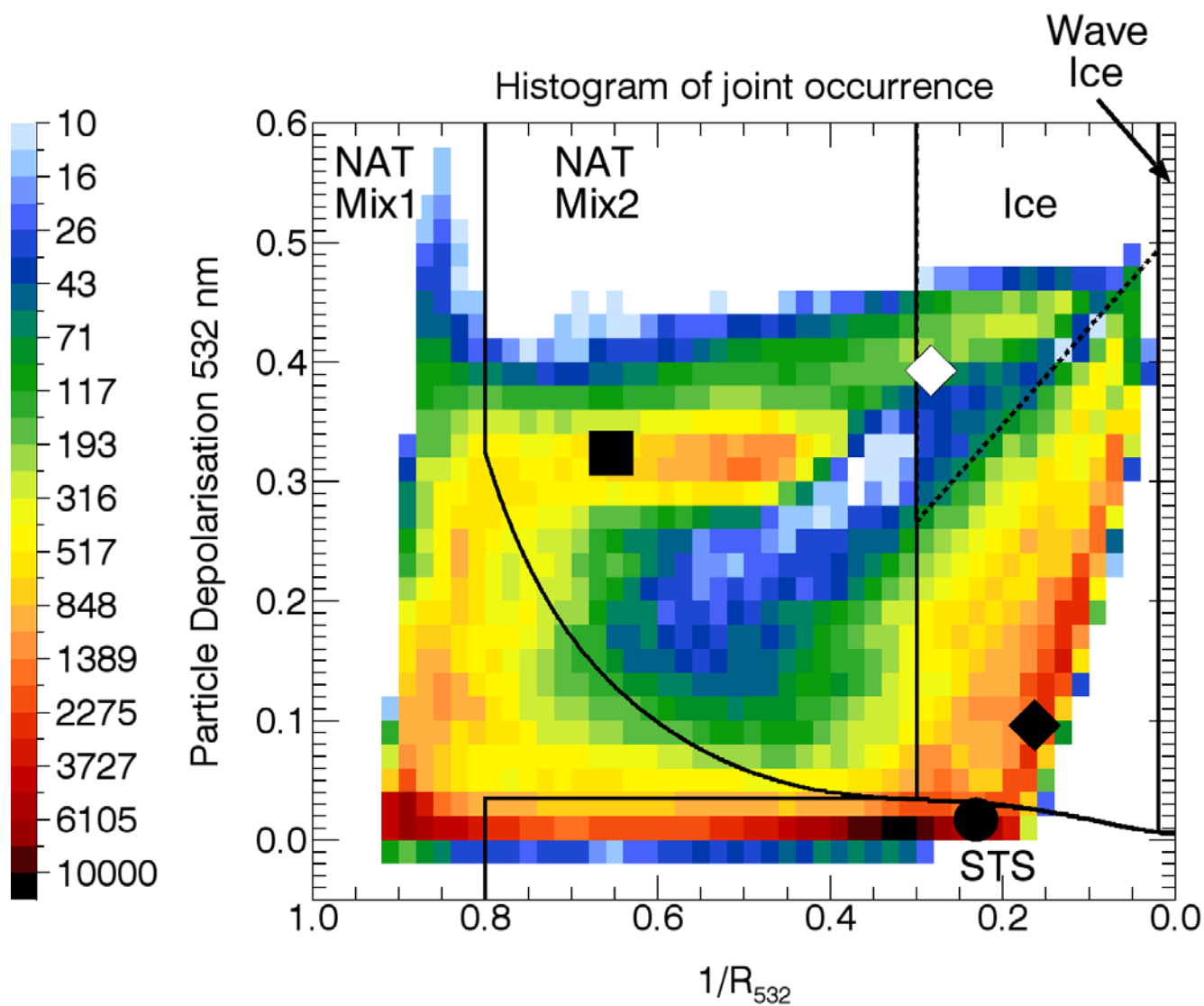
Figure 4 Composite 2-dimensional <u>occurrence</u> histogram of the PSC <u>observed</u> on 22 Janaury 2016 shown in Figure 3 in the 1/backscatter ratio versus ~~aerosol~~particle depolarization ratio coordinate system. The solid black lines denote the boundaries of the PSC types defined by Pitts et al. (2011) with the threshold between ice and NAT Mix2 $1/R_{ice}= 0.3$. The dotted line separates the <u>high-depol and low-depol ice modes. The symbols indicate the starting points of trajectories within different PSC types (square: NAT Mix2, hollow diamond: high-depol ice, filled diamond: low-depol ice and circle: STS).</u>
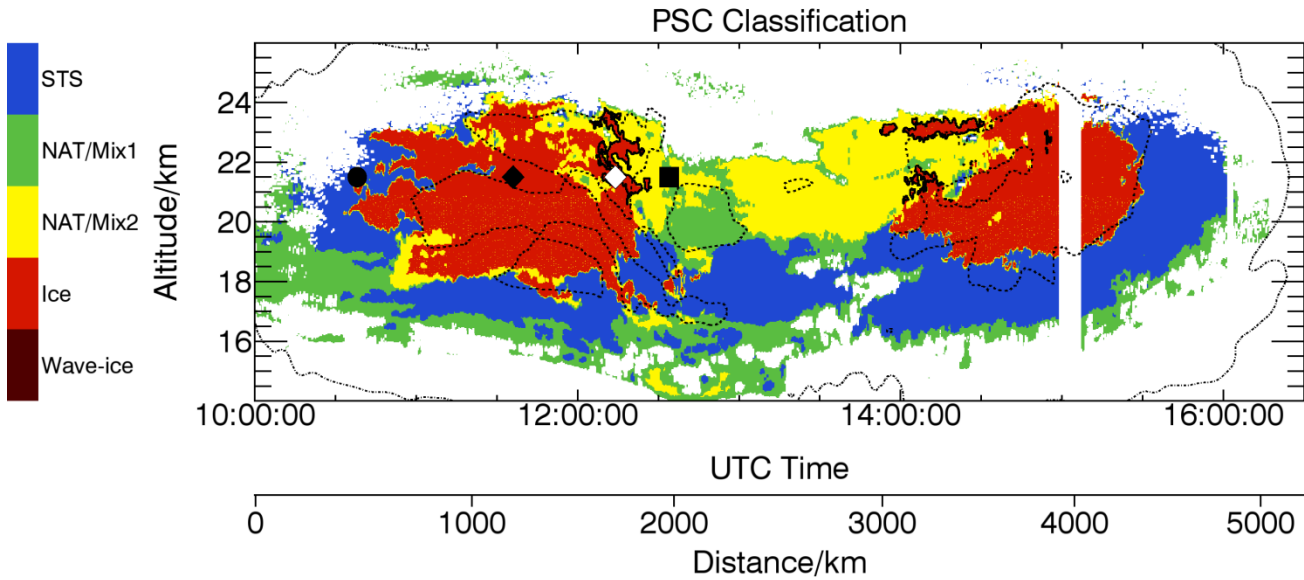
Figure 5 Classification of the synoptic-scale polar stratospheric ice cloud on 22 January 2016 using the classification given in Figure 3. A synoptic ice PSC (red area) extends over several hundred km above the HALO flight track. The thick black line encloses the low-depol ice mode (see Figure 4). To the North-West, the ice PSC is embedded in NAT layers (yellow: NAT Mix2, green: NAT Mix1). A liquid STS layer (blue) is located below and to the South-East of the ice PSC. The wave-ice class is not populated. The ending points of the trajectories for ice (high-depol ice (hollow diamond) and low-depol ice (filled diamond)), NAT (square) and STS (circle) given in Figure 8 are marked. The dashed line shows the T_{ice} contour and the dotted line the T_{NAT} contour lines derived from 6-hourly IFS operational weather analysis (cycle 41r2) interpolated to 1-hourly time steps using meteorological forecast data and the water vapor field measured by WALES as well as the HNO_3 field from MLS.
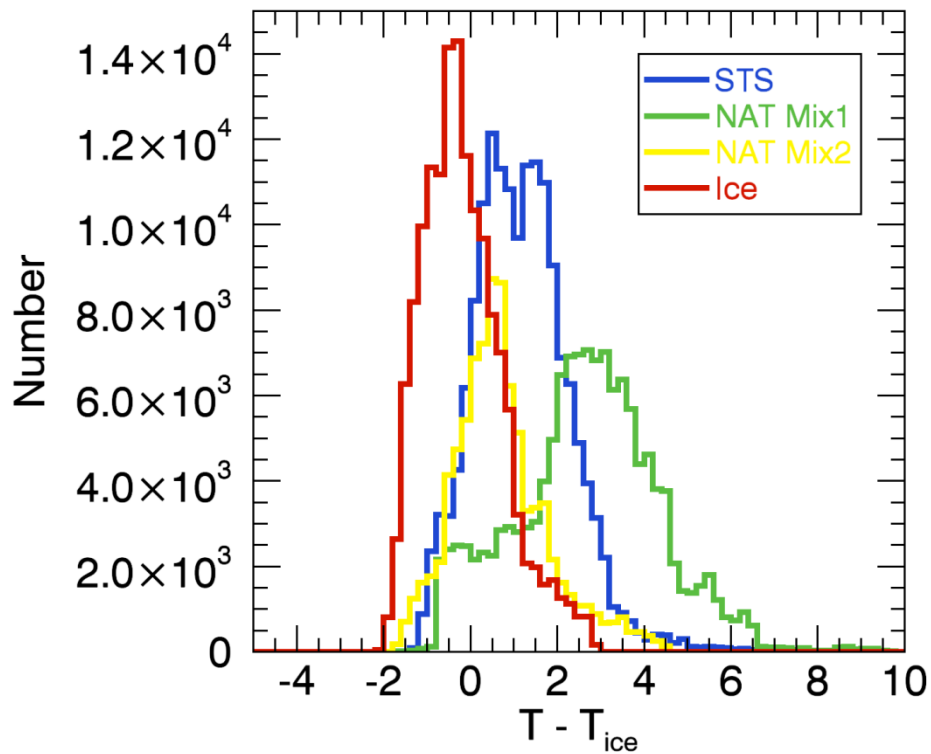
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Figure 6 Occurrence histogram of Ice, NAT Mix2, NAT Mix1 and STS of the 22 January 2016 PSC (Figure 5) versus temperature difference to T_{ice}.
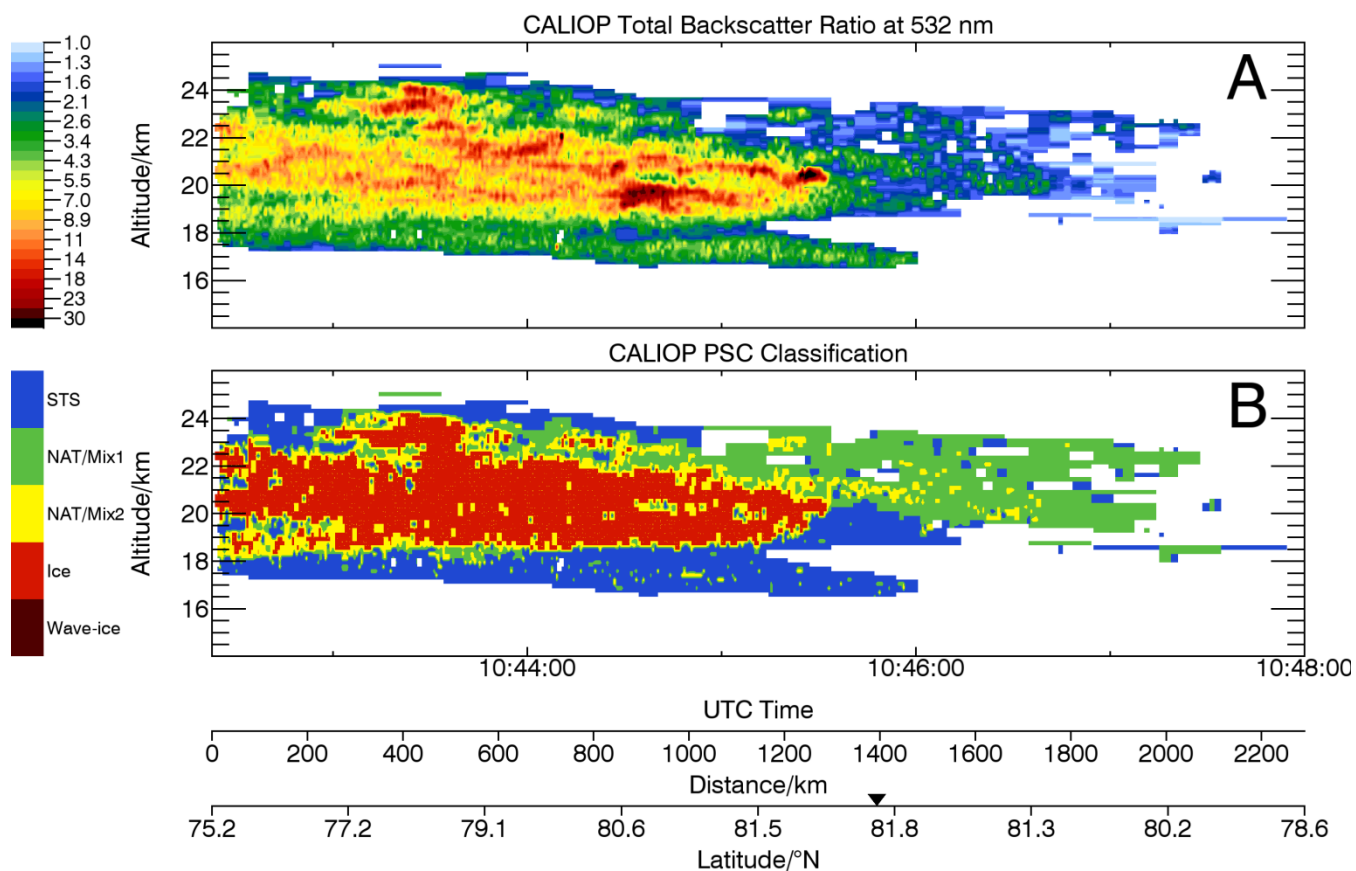
Figure 7 CALIPSO PSC measurements on 22 January 2016 near the HALO flight path (A) Backscatter ratios from CALIPSO at 532 nm wavelengths and (B) PSC type classification from Pitts et al. (2018). The time of the overpass between 10:42 and 10:47 UTC, as well as the horizontal distance and the latitude of the CALIPSO footprint are shown. The northernmost turning point of the CALIPSO footprint is marked by the triangle. The CALIPSO footprint is shown in Figure 8.
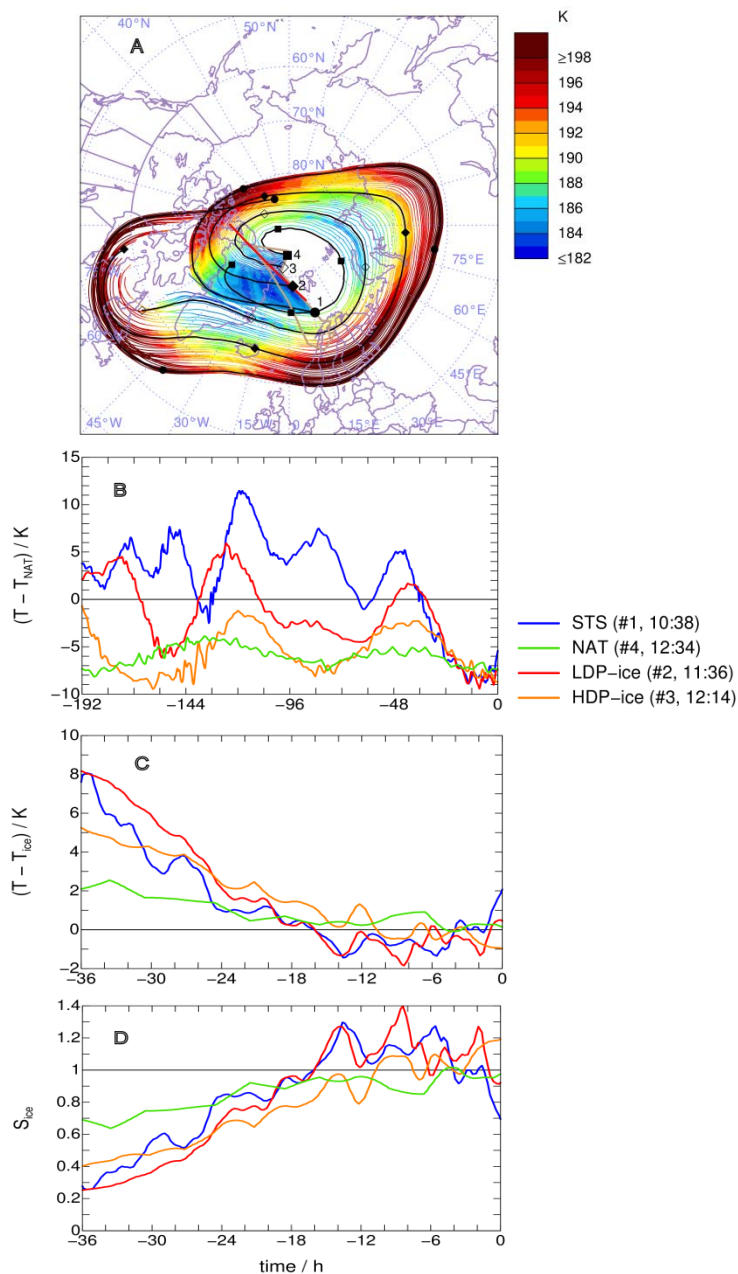
Figure 8 8-days back-trajectories starting in the different PSC layers measured on 22 January 2016 (A) Temperatures (colour coded) of the back-trajectories derived from the IFS operational analysis (cycle 41r1) starting every 2 min at 21.5 altitude in the PSC on 22 January 2016. Four back-trajectories starting in STS (1, circle), low-depol ice (2, LDP-ice, filled diamond), high-depol ice (3, HDP-ice, open diamond) and NAT Mix2 (4, square) are shown as black lines with small symbols marking every 48 hours. The HALO flight track is shown as brown line and the CALIPSO foot print as red line. The starting points of the 4 trajectories are also given in Figures 4 and 5. The temperature evolution along the 4 trajectories (STS: blue, NAT: green, LDP-ice: red and HDP-ice: orange) is shown in panel (B) as temperature difference to T_{NAT} for 8 days and (C) as the temperature difference to T_{ice} for 30 h prior to the observation. The ice saturation ratio S_{ice} is given in panel D. T_{NAT} is calculated from Hanson and Mauersberger (1988) for altitude dependent climatological HNO_3 profile, H_2O, T_{ice} and S_{ice} is calculated from the meteorological data set.

31
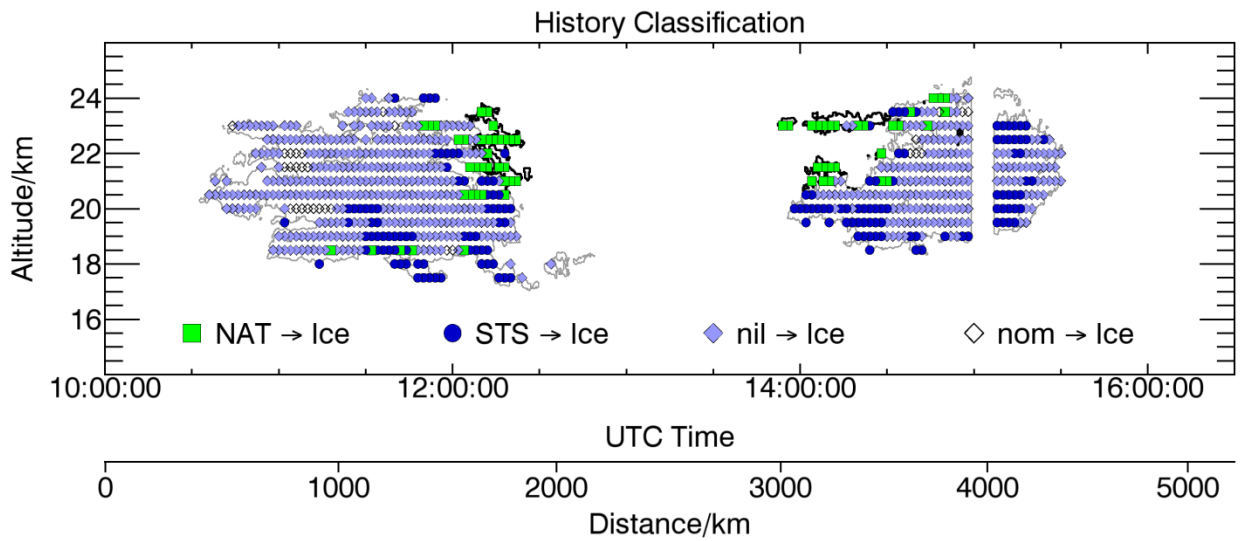
Figure 9 PSC type measured by CALIPSO prior to ice on match points with 5 days back-trajectories. Green squares indicate that NAT has been measured by CALIPSO prior to ice, blue circles indicate STS prior to ice, light blue diamonds (nil) indicate that no PSC has been observed and (nom) that no match point of the trajectories and the CALIPSO curtain exist within 5 days. NAT has been detected by CALIPSO prior to the high-depol ice mode measured by WALES (thick black contour line) and predominantly STS or no PSC have been detected prior to the low-depol ice mode measured by WALES (thin gray contour line). UTC times of the WALES ice PSC observations and the horizontal distance are given.

Atmospheric
Chemistry
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Discussions

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

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 ∼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.

C11

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 space-borne 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 thresh-

old? 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.
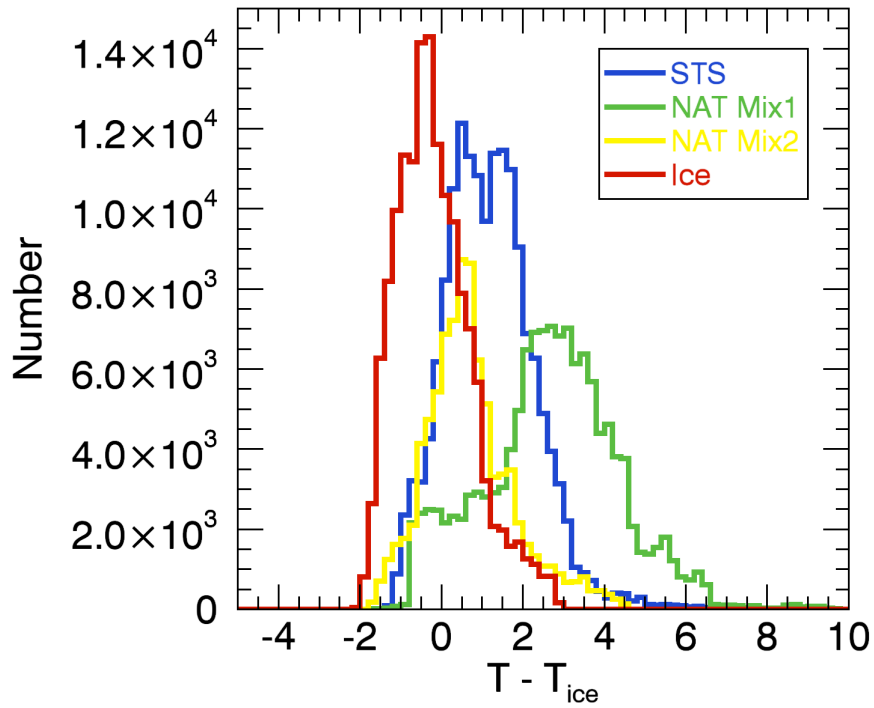
Fig. 1. Occurrence histogram of the 22 Janaury 2016 PSC types with respect to temperature difference to T_ice for the 1/R_ice threshold of 0.3 used in this study
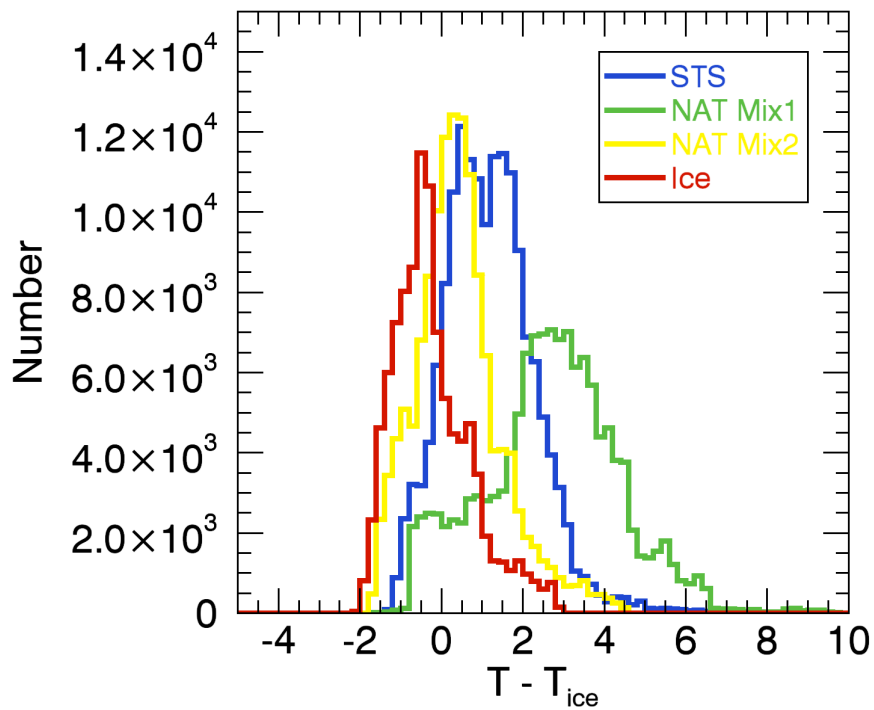
Fig. 2. Occurrence histogram of the 22 Janaury 2016 PSC types with respect to temperature difference to T_ice for the 1/R_ice threshold of 0.2.

Atmospheric
Chemistry
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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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Received and published: 5 July 2018

Reply to Anonymous Referee #2

We thank referee 2 for the positive evaluation of the "uniqueness of the dataset on polar stratospheric clouds", their quality and their scientific importance. We agree that the data provide "new and important insight into the structure of PSC clouds and allows for conclusions on PSC microphysical processes which still are still uncertain or unknown in many respects."

We thank referee 2 for insightful comments, which helped to improve the manuscript.

C1

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

Main comments:

C1: The authors argue at the beginning of section 6.2 that there are "two branches in the ice regime linked directly to STS or NAT/mix2 regimes respectively as shown in Figure 4. "... Later in the discussion of Figure 5 it is shown that the latter is also spatially located to the NAT/mix2 regime. This is a very nice result, but the spatial collocation alone does not necessarily point to the role of the solid nitric acid hydrates for the nucleation of ice observed in these areas. Also the trajectory analysis shown in Fig. 7 is not really conclusive in this direction.

R1: We thank the referee for this comment. We agree that the spatial collocation of NAT Mix2 and high-depol ice alone does not necessarily point to the role of solid nitric acid hydrates. Therefore, we now added a Lagrangian match study to the manuscript, using domain filling trajectory calculations starting in the ice PSC matched with all CALIPSO PSC curtain plots detected up to 5 days prior to the 22 January 2016 PSC. Using a statistical approach we investigate whether PSCs were present on the match points before ice, - and if so, which PSC type has been detected by CALIPSO on the match points. The result is convincing: NAT (and only NAT, no other PSC type) has been measured on the match points for the upper ice branch, now called high-depol ice mode, prior to high-depol ice. On the contrary, STS or no PSC has predominantly been measured on the match points for the lower ice branch, now named low-depol ice mode. The comprehensive Lagrangian study therefore adds new observational evidence for ice nucleation on NAT Mix2 for the high-depol ice mode. In addition, it supports ice nucleation in STS for the low-depol ice mode. We added a section and a new figure of PSC composition prior to ice to the manuscript. We excuse the lack of detail that the referee is missing in the previous version of the manuscript.

C2: The second comment is on the evolution of STS with decreasing temperatures par-

allel to the existence of NAT: But now one may ask what happens to the STS particles when the temperature of an STS regime drops below T_NAT and nitric acid hydrates start to nucleate? Will all STS particles nucleate? This will probably not be the case even if homogeneous nucleation occurs. …But what about their role in forming ice after the temperature drops below Tice?

R2: Binary sulfuric acid/water (BS) droplets grow by condensation upon cooling, initially mainly by uptake of H2O from the gas phase and then upon further cooling to temperatures 3 to 4 K < T_NAT by uptake of H2O and HNO3, forming ternary solution particles STS. Thereby the STS equilibrium temperature T_STS is approximately T_NAT-3.5 K (Carslaw et al., J Phys Chem, 1995). If NAT already exists, then due to the presence of NAT which determines the HNO3 and H2O partial pressures, T_STS decreases and the liquid droplets remain binary down to lower temperatures and grow more slowly. At temperatures a few K above T_ice, STS and NAT can coexist, that is why NAT is called NAT Mix2 to indicate the potential coexistence of STS (or BS) and NAT. If NAT nucleates in the presence of STS, the STS particles will release HNO3 and H2O to the gas phase and will decrease in size. Gasphase HNO3 and H2O condense on NAT due to the lower HNO3 and H2O partial pressures of NAT compared to STS. Depending on their size, due to the Kelvin effect the binary solution particle might be too small to get activated into sizes that allow for ice nucleation at the ice frost point T_ice and further cooling is required to enable their growth. So the meteorological conditions, temperature, H2O, HNO3 (and H2SO4) mixing ratios and the time scales for ice / NAT nucleation determine equilibrium or non-equilibrium conditions and respective PSC particle compositions.

Regarding ice formation in the high-depol ice regime on 22 January 2016, at the time of ice nucleation the STS droplet population with particle depolarization ratios < 0.03 would have to jump into the high-depol ice mode to particle depolarization ratios > 0.3, which is less obvious than ice nucleation on NAT. In the latter case the particle depolarization ratio changes only slightly.

C3: Many laboratory studies formulate homogeneous and heterogeneous ice nucleation as a function of the ice supersaturation or saturation ratio. In the discussion of ice nucleation modes, the temperature difference to Tice should also be expressed as an ice saturation ratio. Also the supersaturation thresholds for heterogeneous and homogeneous ice and NAT nucleation should be discussed if available and reference to respective laboratory work should be given.

R3: We thank the referee for this comment and now show the temperature different to T_ice along the trajectories and include the ice saturation ratio S_ice in the discussion of ice formation pathways in order to facilitate the comparison to laboratory measurements. Further include the supersaturation thresholds for heterogeneous and homogeneous ice and NAT nucleation in the manuscript, discuss them and include the respective references from the laboratory (e.g. hom ice nucleation: Murphy and Koop, Q. J. R. Meteorol. Soc., 2005; heterogeneous ice nucleation: Hoose and Möhler, Atmos. Chem. Phys., 2012; homogeneous and heterogeneous NAT nucleation: Knopf et al., Atmos. Chem. Phys., 2002; Knopf, J PhysChem, 2006; Hoyle et al., Atmos. Chem. Phys., 2013; and references therein). We note that small scale temperature fluctuations add a significant uncertainty to the to the large scale saturation ratios derived from the meteorological model.

C4: Sentences like in line 32 of page 7 ("As soon as temperatures decrease below Tice ice PSCs are present." are misleading because also heterogeneous ice formation usually requires some supersaturated conditions to occur at significant rates.

R4: We excuse that inadequate wording and take this sentence out in the revised version of the manuscript.

C5: The manuscript is not very clear in defining and using terms like "STS regime", "NAT/mix2 regime", "NAT regime". For instance, is there a "NAT regime" in PSCs? I recommend to stay with the term "NAT/mix2" throughout the manuscript, or is there independent evidence that the PCS discussed in this manuscript included pure NAT

clouds?

R5: We are sorry for this inconsistency pointed out by the referee and now use the PSC nomenclature defined and presented in Figure 4 more consistently throughout the manuscript.

C6: And what about the question of NAT vs. NAD?

R6: As mentioned in the manuscript, the formation of NAD from binary nitric acid water solutions has been observed in the laboratory (e.g. Knopf et al., ACP, 2002; Wagner et al., J Phys Chem, 2005; Stetzer et al., ACP, 2006; Möhler et al., ACP, 2006), but homogeneous nucleation rates are too low to explain denitrification (Knopf et al., ACP, 2002). In addition, pseudo-heterogeneous nucleation rates of NAD and NAT (Knopf, J PhysChem, 2006) are too low to explain observed nitric acid hydrate PSC number densities. Observational evidence for the presence of NAD in the atmosphere is missing so far, e.g. Höpfner et al. (2006) found no spectroscopic evidence for the presence of NAD from MIPAS observations of PSCs over Antarctica. The Lidar measurements cannot distinguish between NAT and NAD, additional information e.g. from infrared spectroscopy is required.

We now discuss the laboratory studies in more detail in sight of the atmospheric measurements. However, for reasons of consistency with the lidar classification of NAT Mix1 and NAT Mix2, we prefer to use this nomenclature consistently throughout the manuscript as suggested by the reviewer in the previous comment.

C7: Is there any independent microphysical or instrumental explanation for the need to shift the 1/R532 value in order to separate the NAT/Mix2 from the ice regime?

R7: The 1/Rice value of 0.3 is used to include the major fraction of ice with low particle depolarization ratios (low-depol ice) into the ice mode (linked to STS). This is evident in Figure 4. We further give now additional evidence for this threshold by showing the ice, NAT Mix2, NAT Mix1 and STS occurrence versus temperature to T_ice in a new

figure. We also show the T_ice contour in Figure 5. We discuss the choice of the 1/Rice threshold for the WALES observations in more detail in Section 5.2 and in the supplementary material S1. We note that "The analysis of CALIPSO measurements throughout the winter 2015/16 with changing HNO3 and H2O concentrations requires a variable 1/Rice threshold as used by Pitts et al. (2018)."

C8: The introduction could be shortened for the heterogeneous chemistry part and extended for a more thorough introduction to the PCS classification from lidar data which is more relevant to this manuscript. How is "depolarisation" defined here? And how does it depend on particle size, shape, and mixtures? What explains the higher depolarisation ratio of the upper so-called "NAT ice" branch in Figure 4?

R8: We renamed the NAT ice branch to "high-depol ice mode". Further we now give extended information on the particle depolarization in the instrumental section 2.1. The particle depolarisation ratio is sensitive to the particle shape and size. Spherical particles do not depolarize and particles much smaller than the wavelength of the laser light also show unmeasurable low values. But in general there is no simple relation between depolarization and particle shape, size or composition, see for example Reichardt et al. (2002) for a more detailed discussion of this topic. Nevertheless, cloud regions which show distinct depolarisation ratios point to a significantly different shape or size distribution.

The higher depolarization of the high-depol ice mode could be explained by differences in the shape of the size distribution of the high-depol and the low-depol ice mode. Ice nucleation on NAT might lead to fewer and larger ice crystals and larger asphericity of the ice particle nucleated on solid NAT compared to the ice crystals nucleated in STS.

C9: Check for proper use of English and use of clear definitions.

R9: We checked for the proper use of English and explain the type of histogram in the abstract. We further use high and low temperatures and the frost point temperature T_ice.

C7