Polar Stratospheric Ice Cloud above Spitsbergen

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Abstract
Within the extremely cold and stable polar vortex of the winter 2004/2005, a polar stratospheric ice cloud was observed from Ny-Ålesund (Spitsbergen) on 26 January 2005. The observation of a cloud with backscatter ratios up to 23 and volume depolarization larger than 50% is unique in our 15-year ground based lidar data record. Simultaneous balloon-borne water vapor measurements indicate the presence of mesoscale ice clouds nearby. Normally, low horizontal wind speeds inside the inner vortex prevent vertical wave propagation. However, the rare coincidence of different meteorological processes occurring during a poleward breaking Rossby wave event caused favorable conditions for the vertical propagation of mountain waves excited by the flow past Spitsbergen. Detailed meteorological analysis shows that the ice particle formation processes on 26 January 2005 were most likely provoked by mesoscale stratospheric temperature anomalies, leading to a local reduction in water vapor.
1. Introduction

Every winter, the large-scale radiative cooling of the air inside the southern hemispheric polar vortex enables the existence of stratospheric ice clouds, so called polar stratospheric clouds (PSCs) of type II. Heterogeneous chemical reactions on the ice PSC particles convert effectively inert reservoir gases to photo-chemically active species that drive the catalytic ozone destruction [Solomon, 1999]. Additionally, ice PSCs remove trace gases such as water (H₂O) and nitric acid (HNO₃) temporarily from the gas-phase by condensation and freezing. The ice cloud particles may grow to larger sizes and, eventually, transport the particle bound trace gas components downwards due to gravitational settling. Hence, PSCs of type II can irreversibly remove H₂O and HNO₃ from higher altitudes (dehydration and denitrification, respectively). Under denitrified conditions, ozone depletion can be prolonged and the springtime ozone loss is enhanced [Rex et al., 1997; Waibel et al., 1999]. In the Antarctic, dehydration is regularly observed on a large scale [Nedoluha et al., 2000; 2002] as vortex temperatures are low enough over a long period for the widespread occurrence of ice PSCs every winter. Distinctively, in the Arctic, dehydration is found to a much lesser extent and is commonly linked to single events [e.g. Vömel et al., 1997; Herman et al., 2002; Schiller et al., 2002].

The majority of Arctic ice PSC observations by ground-based, balloon-borne and airborne instruments is documented for Scandinavia [Hansen and Hoppe, 1997; Carslaw et al., 1998a; Wirth et al., 1999; Kivi et al., 2001, Dörnbrack et al., 2002; Fueglistaler et al., 2003; Reichardt et al., 2004]. In fact, Arctic ice PSCs have only been observed in regions of additional mesoscale cooling caused by internal gravity waves. Gravity waves with amplitudes large enough to reduce the temperature significantly below the ice frost point T_{ice} were predominantly excited orographically [e.g. Dörnbrack et al. 1999]. Since strong and nearly unidirectional winds throughout the troposphere and stratosphere favor the upward
wave propagation, mountain wave-induced stratospheric temperature fluctuations leading to PSC formation are enhanced at the inner edge of the polar vortex [Dörnbrack and Leutbecher, 2001]. Alternatively, PSCs can be induced by jet-stream instabilities in breaking Rossby waves and by shear instabilities in the tropopause region [Teitelbaum et al., 2001; Hitchman et al., 2003; Buss et al., 2004].

In this paper, we present a unique lidar observation of an ice PSC at the German Koldewey station in Ny-Ålesund (78.9° N, 11.9° E), Spitsbergen, in combination with nearly simultaneous balloon-borne stratospheric water vapor measurements. During the time of observation on 26 January 2005, the polar vortex was extremely cold. Temperatures of 1 to 2 K below the ice frost point $T_{\text{ice}}$ were achieved in the cold pool by radiative cooling as well as by the adiabatic cooling of ascending air parcels in a planetary wave [as described in Teitelbaum and Sadourny, 1998]. However, as synoptic-scale temperatures were close to $T_{\text{ice}}$, additional cooling through mesoscale processes might be required to explain the formation of the ice PSC particles.

A thorough analysis of the meteorological conditions discusses large-scale cooling processes inside the Arctic vortex. In addition, we investigate whether mesoscale atmospheric processes as inertia-gravity and mountain waves induced favorable formation conditions for ice particles locally above Spitsbergen. In the next section, we present the ice PSC observation and related measurements in Ny-Ålesund on 26 January 2005. Section 3 describes the large-scale meteorological situation in the stratosphere whereas Section 4 investigates the impact of smaller scale meteorological processes on the stratospheric temperature distribution by means of mesoscale numerical simulations. Section 5 discusses different formation scenarios for the observed ice PSC and, section 6 concludes the paper.
2. PSC Observations

2.1 The Ny-Ålesund lidar data record of PSC observations

The lidar system at the German Koldewey station in Ny-Ålesund is operational since the winter 1988/1989. Stratospheric aerosol backscatter and depolarization measurements at a wavelength of 532 nm have been regularly performed every winter since 1991 whenever the tropospheric cloud cover allowed for [Beyerle et al., 1994; Biele et al., 2001; Massoli et al., 2006]. In total, the Ny-Ålesund lidar record represents the longest continuous lidar PSC record in the Arctic region. Figure 1 shows the backscatter ratio $R_{532\text{nm}}$ and the volume depolarization $\delta_{532\text{nm}}$ of all PSC data retrieved between 1991/1992 and 2004/2005. The standard 10-minute integration profiles with 150 m vertical resolution limited to $R_{532\text{nm}} > 1.2$ result in more than 50000 PSC data points. The observation of the 26 January 2005 (180 PSC data points) clearly stand out against all our previous measurements.

Recently, a representative subset of these data has been classified [Massoli et al., 2006]. The majority of our observations (37%) constituted of liquid PSCs characterized by low depolarization, while 25% of the PSCs were identified as consisting of depolarizing solid nitric acid trihydrate (NAT) particles [Voigt et al., 2000] with small backscatter ratio $R_{532\text{nm}} < 1.56$. A large group of PSCs (31%) was composed of mixed (liquid/solid) particles associated with their intermediate backscatter and depolarization characteristics. Only 4% of all PSC data were so called NAT enhanced PSCs [Tsias et al., 1999], being solid NAT particles with high backscatter ratio $R_{532\text{nm}} > 1.56$. However, a PSC with a backscatter ratio of $R_{532\text{nm}} > 10$ and volume depolarization $\delta_{532\text{nm}} > 50\%$ had never been identified in the complete data record prior to 26 January 2005 (Fig. 1). Before we discuss this unique PSC observation in detail, the radiosonde temperatures are presented next.
2.2 Temperature measurements on 26 January 2005

The ambient stratospheric temperature $T$ was measured with a F-Thermocap sensor by a standard RS-92 radiosonde launched from Ny-Ålesund on 26 January 2005 at 2300 UTC. The uncertainty of the temperature observation was $\pm 0.3$ K in the lower stratosphere. Figure 2a shows the stratospheric temperature measured during ascent near Ny-Ålesund at 2330 UTC. Temperatures as low as 183 K have been detected at about 22 km altitude. A radiosonde measurement 12 hours earlier (grey line in Fig. 2a) shows a temperature minimum of 184 K below this level but up to 4 K warmer temperatures compared to the 2330 UTC measurement in the altitude range between 19 and 22 km.

The temperature difference $T-T_{\text{ice}}$ is shown in Fig. 2b whereby the ice frost point $T_{\text{ice}}$ was calculated applying a formula for the saturation water vapor pressure recently developed by Murphy and Koop [2005] and assuming a water vapor profile as sketched by the dashed line in Fig. 2d. This undisturbed H$_2$O-profile may represent the conditions upstream of Spitsbergen and near the location of PSC nucleation. Additionally, the temperature difference to $T_{\text{ice}}$ has been calculated using the observed undisturbed H$_2$O-profile (solid lines in Fig.2b and d). The water vapor measurements will be discussed in more detail in section 2.4. Combined, the temperature profiles reveal a cold layer with $T < T_{\text{ice}}$ between 18.5 and 22.5 km altitude. Inside this cold layer, the observed temperatures dropped up to $\approx 3$ K below $T_{\text{ice}}$ near 20.8 km and 22 km altitude, respectively.

Please note that the region with $T < T_{\text{ice}}$ is capped by a narrow inversion layer with a local maximum of the buoyancy frequency $N = (g/\Theta \cdot \partial \Theta/\partial z)^{1/2} \approx 0.04$ s$^{-1}$, where $g$ is the gravitational acceleration and $\Theta$ denotes the potential temperature. A mixing layer with strongly reduced values of $N \approx 0.005$ s$^{-1}$ is located beneath this capping inversion. Here, we can only speculate that this structure constitutes the signature of overturning gravity waves [e.g. Dörnbrack, 1998].
2.3 Lidar measurements of an ice PSC

Vertical profiles of the backscatter ratio $R_{532\text{nm}}$ and the volume depolarization $\delta_{532\text{nm}}$ of 40 minutes of lidar measurements on 26 January 2005 between 1855 and 1934 UTC are shown in Figure 2c. Unfavorable weather conditions with low cloud cover before and after that period prevented longer PSC measurements on that day (readers interested in the tropospheric weather situation are referred to Appendix A).

A thick polar stratospheric cloud with $R_{532\text{nm}} > 2$ was measured between 17 and 22 km altitude. Volume depolarization values $\delta_{532\text{nm}} < 1.44\%$ classify the lower part of this cloud ($z < 19$ km) as a PSC composed of liquid particles. Volume depolarization values larger than 35% were measured in three distinct regions around 19.5 km, 20.5 km, and 22 km altitude, together with $R_{532\text{nm}} > 5$, respectively. Maximum values of $\delta_{532\text{nm}} \approx 70\%$ and $R_{532\text{nm}} \approx 23$ were attained between 20 and 21 km altitude. Using the PSC classification schemes by Browell et al. [1990] and Toon et al. [1990], we unambiguously identify this middle layer as type II PSC or polar stratospheric ice cloud. This ice layer was detected in a temperature range up to 3 K below $T_{\text{ice}}$, and thus under conditions of ice supersaturation.

Although the both neighboring layers revealed lower backscatter values, their respective depolarization values of $\delta_{532\text{nm}} \geq 50\%$ were unusually large. These layers may be classified as NAT enhanced PSCs. Such clouds were frequently observed downwind of ice PSCs [Carslaw et al., 1998b; Hu et al., 2002] or in their close vicinity [Wirth et al., 1999]. Here, however, temperatures near and below $T_{\text{ice}}$ in concert with optical calculations (B. Luo, pers. communication, 2005) and the water vapor measurements (Fig. 2d) suggest that these layers might be composed of ice particles (thin PSCs of type II). Ice PSCs are rarely found in thermodynamical equilibrium (i.e. at $T = T_{\text{ice}}$), but were measured at a range of ice supersaturations [Luo et al., 2003, Fueglistaler et al., 2003]. Hence, we do not assume a fully developed and thick ice cloud. Instead, the cloud layers around the unambiguously identified ice PSC may consist of growing or evaporating ice particles depending on their temperature.
history, the ambient meteorological conditions, and the availability of trace gases. In the following, we concentrate on the discussion of the ice PSC layer at around 20 km altitude.

2.4 Water vapor observation

Around 2000 UTC, a balloon-borne Lyman-α hygrometer [Yushkov et al., 1998; 2001] measured the stratospheric water vapor profile with an uncertainty of ±8% and a vertical resolution of about 150 m in the PSC altitude range (Figure 2 d). To avoid contamination effects caused by tropospheric remnants in the plume of the balloon, only the descent data about 100 km southeast of Ny-Ålesund are shown. Between 16 and 24 km altitude, the water vapor mixing ratio \( q_{H_2O} \) gradually increases with altitude as expected for inner vortex conditions. In three distinct layers between 19.5 and 22.5 km altitude, water vapor was depleted by up to 1.5 ppmv below the undisturbed values.

While we cannot completely exclude that the minima in the water vapor concentration represent permanent removal of water vapor, we argue that the laminated structure of the \( H_2O \) concentration may reflect the local partitioning of water into ice particles, or measurements in an ice PSC. This view is supported by the strong correlation between the three maxima in the backscatter profile and the three minima in the water vapor profile. The vertical shift of about 500 m between the lower two water vapor minima and the maxima in the backscatter ratios might be explained by the drift of the hygrometer balloon, hence, the detection of the water vapor profile about 100 km downwind southeast of the lidar PSC observation above Ny-Ålesund.

Altogether, the measurements of temperature, backscatter ratio, volume depolarization and water vapor concentration show that stratospheric ice clouds were detected at temperatures below \( T_{ice} \). In the following, we discuss the meteorological situation in winter 2004/2005 and, particularly, we concentrate on 26 January 2005 in order to explain this unique observation of an ice PSC above Spitsbergen.
3. Synoptic-scale analyses

In winter 2004/2005, the Arctic polar vortex was exceptionally stable and cold from its early formation onward [e.g. Kleinböhl et al., 2005; Manney et al., manuscript submitted to Geophys. Res. Lett., 2005]. At higher stratospheric levels (\(\Theta > 550\) K, \(p < 30\) hPa, \(z > 22\) km), temperatures allowed the existence of NAT PSCs already in late November 2004. Temperatures on the 475 K isentropic surface (\(p \approx 50\) hPa, \(z \approx 19\) km) were below the NAT threshold from December 2004 onwards. In addition, the stratospheric temperature fell below the ice frost point on several single days around the turn of the year, at the end of January, and in the second half of February 2005. Most prominent, one week period end of January 2005 revealed the largest potential ice PSC area in the Arctic ever calculated from ECMWF analyses so far (P. v. d. Gathen, pers. communication, 2005).

In this particular period, the polar vortex was kidney-shaped and mostly barotropic. A displacement between the geopotential height and the temperature field (disturbance of the Arctic vortex by a planetary wave) occurred between Greenland and the Norwegian Sea and was caused by an underlying anticyclone elevating the tropopause and the stratospheric levels above. Based on T511/L60 operational ECMWF analyses valid on 26 January 2005, the very cold area with \(T < T_{\text{ice}}\) extended from the North Pole to about the latitude of Sodankylä, Finland (\(~ 67^\circ\) N) reaching from 30° W to about 65° E. The Spitsbergen area was characterized by a rather uniform temperature field of \(~188\) K at 50 hPa and \(~185\) K at the 30 hPa level, both levels being about 1 K below \(T_{\text{ice}}\) (Fig. 3).

In order to investigate the temperature history of the air mass sampled at Ny-Ålesund, backward trajectories were calculated, with starting points on different pressure levels located at 79°N and 12°E on 26 January 2005, 2100 UTC. The applied trajectory scheme LAGRANTO [Wernli and Davis, 1997] was run with meteorological data provided by T511/L60 operational ECMWF data interpolated on a regular 0.5°×0.5° latitude-longitude
grid every 3 hours (6 hourly operational analyses at meteorological standard times plus $t_0+3\text{h}$ and $t_0+9\text{h}$ predictions at the intermediate steps based on the forecasts initialized at $t_0 = 0000 \text{UTC}$ and $t_0 = 1200 \text{UTC}$, respectively), see Figure 3.

All backward trajectories passed the Atlantic in the first 12 hours and crossed Greenland’s east coast at about 78° N. Note that the mesoscale stratospheric temperature anomalies above Greenland’s east coast and downstream occurred predominantly at lower latitudes. Thus, air parcels arriving at Ny-Ålesund were not strongly influenced by the extreme low temperatures further south with an absolute minimum of 176.5 K at 30 hPa. Instead, the most prominent cooling of the air parcels by about $\Delta T = 8 \text{K}$ occurred slowly during the 36 hours prior to the observation. Inspection of the pressure change along the trajectories suggests that this slow cooling is caused by large-scale adiabatic lifting of air induced by an underlying tropospheric high pressure ridge. However, only about 12 hours prior to the observations the temperature drops below $T_{\text{ice}}$, with $T-T_{\text{ice}}$ always less than 2 K.

In the next section we investigate the possible impact of mesoscale processes on the stratospheric temperature field.

4. **Mesoscale perturbations of the stratospheric temperature field**

4.1 **Inertia-gravity waves**

The large-scale synoptic situation before and during the PSC observation was characterized by a so-called "P2-type" breaking Rossby wave event [Peters and Waugh, 1996]: a broad extrusion of subtropical tropospheric air spreading northeast and wrapping itself up anti-cyclonically, forming a large anticyclone over the Atlantic which extended up to a latitude of 80°N on 26 January 2005. On the ECMWF tropopause maps [Morgan and Nielsen-Gammon, 1998] shown in Figure 4, the anticyclone with an elevated tropopause is marked by large values of potential temperature $\Theta$. Sharp $\Theta$-gradients are associated with the
tropopause jet forming a $\Omega$-shaped ring of high horizontal wind speeds around the anticyclone. Exceptionally, Spitsbergen is located directly at the edge of this tropopause jet.

A strongly anti-cyclonically curved jet is not geostrophically balanced. In order to maintain this state, the relationship between the mass and the velocity field is adjusted by radiating inertia-gravity waves [“Rossby adjustment problem”, see Gill, 1982; Plougonven et al., 2003]. Subsequently, these waves propagate vertically and horizontally and are able to modulate the temperature field in the upper troposphere [e.g. Spichtinger et al., 2005] and lower stratosphere [e.g. Hitchman et al., 2003].

In order to investigate if the jet stream located west of Spitsbergen (cf. Fig. 4) excited inertia-gravity waves on 26 January 2005, we inspected horizontal and vertical sections of the divergence of the horizontal wind speed based on operational T511/L60 ECMWF analyses as suggested by Plougonven and Teitelbaum [2003]. The vertical cross section perpendicular to the jet axis, shown in Figure 5, exemplifies the situation: above the eastern flank of the jet stream (maximum $V_H \approx 80$ m/s) alternating bands of positive and negative divergence mark the presence of inertia-gravity waves. Their wave fronts are aligned parallel to the jet axis, and typical vertical and horizontal wavelengths as determined from the ECMWF analyses amount to $\lambda_z \approx 3$ km and $\lambda_H \approx 200$ km, respectively. Above Spitsbergen and about 200 km east of the jet axis, the local temperature within the stratospheric cold layer was reduced by about $\Delta T = 2$ K to $T = 184$ K ($\approx 2$ K below $T_{icw}$). Maximum values of the horizontal divergence, i.e. of inertia-gravity wave activity occurred in the period before local noon on 26 January 2005.

In order to simulate inertia-gravity wave excitation and propagation with higher horizontal and vertical resolutions, we performed a set of mesoscale numerical experiments using the non-hydrostatic weather prediction model MM5 (for the setup see the Appendix B). The mesoscale model MM5 has been successively used to simulate the dynamics of inertia-gravity waves [e. g. Dörnbrack et al., 1999; 2002; Zhang, 2004; Zülicke and Peters, 2005].
Figure 6 shows the simulated stratospheric temperature distribution at 20 km altitude at 0600, 1200, 1700, and 2100 UTC (+18, +24, +29, and +33 h simulation time after MM5 initialization), respectively. The horizontal section at 0600 UTC reveals a significant perturbation of the stratospheric temperature field due to inertia-gravity waves, visible as temperature fluctuations with fronts nearly parallel to the axis of the jet stream, above and east of Spitsbergen, $\lambda_H$ being 200 km close to the jet axis and becoming shorter toward east. The peak-to-peak amplitude of the temperature fluctuations $\Delta T \approx 4$ K is reduced with increasing distance from the jet axis in association with the decreasing horizontal wind speed toward the center of the polar vortex. The mesoscale simulations confirm that Spitsbergen is mainly influenced by inertia-gravity waves in the morning of 26 January 2005. After this time, the simulated wave fronts became gradually perpendicular to the jet axis and the temperature fluctuations grew significantly at all stratospheric levels (Fig. 6). This change of the orientation of the wave fronts indicates another source of wave-induced stratospheric temperature fluctuations, namely, mountain waves.

4.2 Mountain waves

The mesoscale simulation shows that mountain waves were excited during the entire day until about 1800 UTC due to a strong post-frontal northwesterly wind passing Spitsbergen. However, favorable propagation conditions for waves (i.e. nearly unidirectional winds in troposphere and stratosphere) occurred just after about 0600 UTC, when the anticyclone extended very far north and the axis of the associated tropopause jet shifted above the western flank of Spitsbergen (cf. Fig. 4). Eventually, the north-westerly wind direction of the jet stream was aligned with the near-surface winds as displayed by the wind vectors parallel to the contour lines of geopotential height at 900 hPa (see Fig. 6). During the day, the 900 hPa-contour line separation became wider (corresponding to weaker winds) and, finally, at about 1800 UTC the lower tropospheric wind turned to south-westerlies after the passage of
the ridge axis, in agreement with the observed wind of the 2300 UTC radiosonde measurement. At this time, mountain wave excitation was reduced, and the diminishing tropospheric wind speed as well as the large-scale descent in the anticyclone degraded the favorable wave propagation conditions. Thus, mountain waves exclusively modulated the stratospheric temperature field during the afternoon of 26 January 2005.

Figure 7 displays this modulation by jet-parallel vertical cross sections of T, Θ, and the vertical wind speed at the corresponding times as in Fig. 6. At 1200 UTC, the waves just commenced to modulate the cold stratospheric layer (see Figs. 6 and 7). The mountain waves have typical characteristics of vertically propagating hydrostatic gravity waves in the non-rotating limit: \( \lambda_H < 50 \text{ km} \) and \( \lambda_z \approx 6\ldots8 \text{ km} \). The horizontal temperature field at stratospheric levels consist of numerous patches of localized cooling whereby \( T_{\text{MIN}} \approx 184 \text{ K} \) (see Fig. 6, 1700 UTC). These fast-propagating waves seem to be excited at individual ridges of the model orography. Linear wave theory predicts a vertical group velocity \( c_{gz} = V_H^2 k_H / N \) [Gill, 1982] of about 5.65 ms\(^{-1}\), where \( k_H = 2\pi / \lambda_H \) is the horizontal component of the wave vector, i.e. to reach a height of 22 km these relatively short waves only needed about 1 hour assuming mean values of \( V_H = 30 \text{ ms}^{-1} \) and \( N = 0.02 \text{ s}^{-1} \).

It is interesting to note that the mountain waves seem preferentially to propagate into the direction of higher horizontal wind speed, see horizontal sections at 1200 and 1700 UTC in Figure 6, respectively. This again is in accordance with linear wave theory which predicts the amplitude of internal gravity waves being proportional to the magnitude of the horizontal wind speed. Thus, the impact of the mountain waves on the stratospheric temperature field is indeed maximal on the high wind-speed side of the aligned tropopause and stratospheric jets (or, in other words: at the inner edge of the polar vortex). During the following hours, the wave and temperature fields changed dramatically: horizontally longer waves with smaller \( c_{gz} \) values dominated and the peak-to-peak amplitude of the temperature fluctuations grew to values of \( \Delta T \approx 12 \text{ K} \) leading to \( T_{\text{MIN}} \approx 181 \text{ K} \) (see Fig. 6, 2100 UTC). Whereas the vertical
velocity field displays wave propagation throughout the troposphere and stratosphere between 1200 and 1700 UTC, the 2100 UTC vertical section clearly shows the absence of wave modes excited by the flow across Spitsbergen (Fig. 7).

Starting at about 1700 UTC, isentropic surfaces began to steepen and gravity wave breaking was simulated between 18 and 22 km altitude in the subsequent period; see the detailed stratospheric temperature fields at 1700 and 2100 UTC in Figure 8. At 1700 UTC, two regions with localized mountain-wave induced cooling below 183 K were located upstream of Ny-Ålesund between 20 and 21.5 km altitude, respectively. Subsequent wave overturning and the attenuated wave forcing due to the weaker tropospheric wind reduced the stratospheric mesoscale temperature anomalies upstream of Ny-Ålesund until 2100 UTC.

Given the strongly non-linear wave breaking in concert with the time-dependent wave forcing as well as the limited stratospheric data, it is overly ambitious to compare quantitative details of the observed and modeled gravity waves. Furthermore, inaccuracies of the simulated flow (e.g. parameterized mixing might have changed the simulated temperature field irreversibly in an unrealistic manner) produce numerical artifacts in disguise of physical effects.

We conclude that the mesoscale numerical simulations unambiguously demonstrated the short-term presence of mountain-wave induced stratospheric temperature anomalies with localized temperatures low enough to explain the formation of ice PSCs above Spitsbergen.

5. PSC formation scenarios

In order to investigate whether homogeneous freezing was responsible for the PSC occurrence, we calculated the ice saturation ratio $S_r = p_w / p_{sat}(T)$, where $p_w$ is the partial pressure of water vapor and $p_{sat}$ is the saturation vapor pressure over a plane ice surface, calculated after Sonntag [1990] (see also Murphy and Koop [2005]) along backward trajectories. Furthermore, the freezing threshold saturation ratio $S_{cr}$ for homogeneous nucleation of ice
crystals from supercooled aqueous solution droplets [Koop et al., 2000] was computed using
the approximation by Kärcher and Lohmann [2002]. In the temperature range between 184 K
and 190 K, $S_{cr}$ is nearly constant and amounts to about 1.7, i.e. homogeneous freezing requires
large ice supersaturation.

Figure 9 depicts the ratio $S_i/S_{cr}$ both for synoptic-scale as well as mesoscale backward
trajectories during the last 24 h and 3 h, respectively. The mesoscale backward trajectories
were computed using the half-hourly output of the MM5 simulations. Although the synoptic-
scale temperature decreases gradually by about 4 K during 24 h in the relevant altitude range
from 50 hPa (~19 km) to 30 hPa (~22 km) and $S_i$ is larger than 1, the ice saturation ratio $S_i$
ever exceeds the threshold $S_{cr}$ (Fig. 9a). Air parcels released 3 h later at the same locations
experienced a similar temperature history and $S_i/S_{cr}$ was less than one all the time. Only when
we decreased the temperature along the backward trajectories artificially by 2 K, the air
parcels released at 30 hPa and 40 hPa achieved $S_i \approx 1.1 \times S_{cr}$ during the first 3 h after departure.
Thus, the T511/L60 ECMWF analyses had to be wrong by 2 K to allow homogeneous
nucleation by synoptic-scale cooling.

In contrast, $S_i$-values calculated along mesoscale backward trajectories exceeded the
threshold for ice nucleation $S_{cr}$, reaching $S_i \approx 1.4 \times S_{cr}$ due to adiabatic cooling of up to -15 K/h
in the uplift regions of the mountain waves, see Fig 9b. Altogether, mesoscale trajectories
were released at 121 horizontal locations in an 80 km $\times$ 80 km grid (see Fig 7 at 1700 UTC)
and on 10 vertical levels every hour between 1600 UTC and 2000 UTC. The temporal
evolution of $S_i/S_{cr}$ and $T$ of all mesoscale trajectories launched close to Ny-Ålesund showed
similar characteristics: (i) $S_i$ never exceeded $S_{cr}$ along trajectories for $p < 30$ hPa as well as for
$p > 55$ hPa; (ii) the maximum supersaturation was reached for parcels released between 45
and 50 hPa (~19 to 20 km altitude), the actual height of maximum supersaturation was usually
500 to 800 m higher due to wave-induced uplift; (iii) maximum values of $S_i/S_{cr}$ were achieved
for trajectories released at 1800 UTC and 1900 UTC, while $S_i/S_{cr} > 1$ is rarely achieved for
trajectories released before and after; (iv) the typical simulated time of freezing conditions $S_i/S_{cr} > 1$ amounts to about 10 to 20 min and took place during the first 30 min (see Fig. 9b).

Backward trajectories released further downstream of Ny-Ålesund at locations close to the balloon observations revealed a much more complex history as the air parcels passed several mesoscale temperature maxima and minima. Almost all stratospheric levels experienced conditions where the critical value $S_{cr}$ was exceeded for several times in the first three hours after release. Thus, the lidar as well as the H$_2$O observations show probably growing or decaying ice PSC layers depending on the actual temperature and the temperature history along the trajectories. It is interesting to note that the possibility of precipitating ice particles from upper layers to the layers below 30 hPa can be excluded, as all upper trajectories never attained values of $S_i/S_{cr} > 1$.

Microphysical calculations of the H$_2$O depletion by growing ice crystals have been performed. As the large water vapor depletion of 1.5 ppmv could not have been caused by NAT particles due to the limited availability of HNO$_3$ in the order of a few ppbv, the calculations were limited to the growth of water ice particles.

At time $t = 0$, a number density $n_i = 1, 5, 10, 50, 100, and 500$ ice crystals per cm$^3$ with an initial mass of $10^{-16}$ kg were assumed to freeze at $T_{HOM}=182.6$ K for $q_{H2O} = 5$ ppmv and $T_{HOM}=183.6$ K for $q_{H2O} = 6$ ppmv background water vapor concentration; the values of $T_{HOM}$ correspond to the temperature threshold for homogeneous freezing. Subsequently, the diffusive growth of the ice crystals reduces the ambient water vapor. The growth rates for the ice crystals were calculated according to Pruppacher and Klett [1978, Chapter 13.3]. In order to simulate the maximum observed depletion of up to 1.5 ppmv, number densities $n_i > 10$ cm$^{-3}$ are necessary. In case of homogeneous nucleation, these high number densities are generated under cooling rate conditions that are equivalent to vertical wind velocities $w > 0.1$ m s$^{-1}$ (see Fig. 3 in Kärcher and Lohmann, 2002). These values correspond closely to our typical simulated mesoscale values of $|w_{\text{MAX}}| = 0.5$ m s$^{-1}$; exceptionally, maximum
values \( w > 1.2 \text{ ms}^{-1} \) were calculated along individual trajectories. The calculation shows that 1.5 ppmv of water vapor can indeed be reduced in less then 45 min.

Summarizing, the adiabatic cooling and homogeneous freezing in mesoscale mountain waves could explain the formation of the ice PSCs around Ny-Ålesund whereas homogeneous nucleation of ice particles due to synoptic-scale cooling along trajectories is very unlikely.

Heterogeneous freezing requires generally lower ice supersaturations \( S_i \) in the temperature range of about 185 K [deMott et al., 2003]. Assuming \( S_i \approx 1.3 \), similar microphysical simulations as for the homogeneous freezing, whereby \( n_i \) now represents the initial concentration of activated ice nuclei, reveal that even higher number concentrations \( n_i > 50 \text{ cm}^{-3} \) and much longer times spans would be necessary for reducing the water vapor by 1.5 ppmv. As recent in-situ observations of total number concentrations of aerosol particles show \( n_i \approx 10 \text{ to } 20 \text{ cm}^{-3} \) in an altitude range between 18 and 20 km, the requirements are hardly met. Yet, the enhanced fraction of non-volatile particles (meteoritic dust) inside the polar vortex might increase heterogeneous nucleation rates [Curtius et al., 2005]. These particles or micrometeorites have been recently suggested to be suitable candidates for the nucleation of PSCs consisting of low number densities of large NAT particles [Voigt et al., 2005]. However, scientific knowledge on the actual heterogeneous freezing processes in the stratosphere is limited, hence, we leave it open whether this process might explain the formation of the observed ice PSC.

6. Conclusion

On 26 January 2005, an ice PSC with backscatter ratio \( R_{532\text{nm}} > 10 \) and volume depolarization \( \delta_{532\text{nm}} > 50\% \) was observed at Ny-Ålesund, Spitsbergen, under exceptional meteorological conditions. This fortunate event is unique within the 15 year data record of the German lidar observations and confirms the documented rareness of NAT enhanced PSCs [Massoli et al., 2006]. Moreover, simultaneous balloon-borne measurements of depleted
layers of stratospheric water vapor indicate that freezing produced a sufficiently large number of ice particles, hence, the presence of ice PSCs further downstream of Ny-Ålesund. However, the paucity of sufficient experimental information on the upstream stratospheric conditions and on the availability of ice nuclei does not allow a final conclusion about the actual ice PSC formation processes.

Even so, the synoptic-scale and mesoscale temperature analyses revealed that the ice PSCs most likely formed locally and shortly before the observation. In addition, the fine-structure in the observed stratospheric profiles of $R_{532\text{nm}}$, $\delta_{532\text{nm}}$, $q_{\text{H}_2\text{O}}$, and temperature indicate the presence of small-scale processes in the PSC region such as mountain waves. Thus, we favour the scenario of ice PSC formation provoked by mesoscale temperature fluctuations. In this scenario, the prevailing planetary wave activity provided the necessary background conditions that brought the stratospheric temperature below the frost point $T_{\text{ice}}$.

The exceptional synoptic situation responsible for this event was characterized by the combination of three closely linked processes: (i) the passing tropospheric cold front (see Appendix A) with strong post-frontal northwesterly flow past Spitsbergen’s mountains excited the mountain waves, (ii) the unusual location and orientation of the tropopause jet due to the northward extending high-pressure ridge behind the frontal system lead to an alignment with the lower tropospheric winds and favored vertically upward gravity waves propagation into the stratosphere, and (iii) finally the presence of the vortex cold pool in the baroclinically deformed polar vortex above Spitsbergen (see schematic sketch in Fig. 10).

Although we cannot exclude that the long-lasting mountain wave event above Greenland caused large-scale dehydration south of Spitsbergen and in other parts of the polar vortex, we find that the observed $\text{H}_2\text{O}$ reduction above Spitsbergen was a local event.

Monitoring the Arctic water vapor distribution by satellite-borne instruments in conjunction with spatially high-resolved meteorological analyses might answer the question
whether mesoscale events can influence the water vapor budget of the Arctic stratosphere significantly.

Appendix A

The two days before the lidar observations were characterized by strong gusty winds and low level clouds associated with a storm system. This low developed in the period starting on 24 January 2005 where an explosive cyclogenesis (surface pressure drop of 30 hPa in only 24 hours) occurred over the Atlantic between Greenland and Spitsbergen. The low-pressure system with a minimum surface pressure of 973.5 hPa passed the station one day before the observation on 25 January 2005 with the subsequent cold front passage at about 2200 UTC. The Koldewey meteorological ground station registered a sharp drop of the ground temperature from about -2.5°C to about -10°C within 1.5 hours.

Thus, on 26 January 2005, cold Arctic air dominated the tropospheric conditions in Ny-Ålesund. The ground pressure rose constantly during the day due to the approaching high pressure ridge while the surface wind speed decreased gradually from 15 ms\(^{-1}\) at 0000 UTC to about 2 ms\(^{-1}\) at the time of the observation at 1900 UTC. Very low clouds with snow showers were present all morning and afternoon, while the ground temperature (about -12°C) and wind direction (from north-west) remained more or less constant. Furthermore, the 1100 UTC radiosonde measurements revealed a wind direction roughly uniform from north-west throughout the troposphere and lower stratosphere and the thermal tropopause being located at 6.5 km (not shown).

At the evening of 26 January 2005, Ny-Ålesund was located east of the ridge axis and nearly westerly winds dominated the near-surface flow field. Due to the descent under high pressure conditions, the formerly uniform low-level clouds broke up and allowed the lidar measurements. In the lower part of the troposphere, the 2300 UTC radiosonde measurements revealed much weaker wind and its direction turned to south-westerlies. The thermal
tropopause was now located at an altitude of 9 km and, in contrast to the 1100 UTC sounding, it was very sharp as usually observed under anticyclonic conditions.

Appendix B

The mesoscale meteorological fields are calculated with the non-hydrostatic weather prediction model MM5-version 3.4 [Dudhia, 1993; Dudhia et al., 2001]. The outer model domain is centered at (78.5° N, 17° E) with an extension of 1092 km × 1092 km. In this domain a horizontal grid size of Δx = 12 km is used. A local grid refinement scheme with a nested domain of 4 km horizontal resolution is applied to resolve most of the horizontal wave number spectrum of gravity waves excited either by the orography or by jet stream instabilities. In the vertical direction 150 levels up to the model top at 10 hPa (Δz = 200 m) are applied. Turbulent and moist processes are accounted for by standard schemes as turbulence parameterization [Hong and Pan, 1996], Grell's cumulus parameterization [Grell et al., 1994] and Reisner's microphysical scheme [Reisner et al., 1998]. The initial condition on 25 January 2005 at 12 UTC and the boundary values of the model integration were prescribed by operational analyses of the ECMWF model with a horizontal resolution of 0.5° in latitude and longitude and 15 pressure levels between 1000 hPa and the 10 hPa pressure level.

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Figure 1: Aerosol backscatter ratio at 532 nm and volume depolarization of all PSC data points from 10-minute integration profiles retrieved by the German lidar in Ny-Ålesund, Spitsbergen, between 1991/1992 and 2004/2005. The total number of PSC data points from 10-minute integrated profiles with 150 m vertical resolution (gray points) is larger than 50,000, while the observation on 26 January 2005 (black points) accounts for 180 data points.
Figure 2: Measurements on 26 January 2005 in Ny-Ålesund. (a): Radiosonde temperature at 1100 UTC (gray line) and 2300 UTC (black line); (b) Temperature difference $T - T_{\text{ice}}$ ($T_{\text{ice}}$ calculated after Murphy and Koop, 2005) for the 2300 UTC radiosonde temperature and the water vapor profiles shown in panel (d): for the FLASH-B measurement (black line) and the anticipated $q_{\text{H}_2\text{O}}$ profile (dashed gray), respectively; (c): Aerosol backscatter ratio (black line, lower axis) and volume depolarization (gray line, upper axis) at 532 nm, integrated between 1855 and 1934 UTC. (d): Water vapor mixing ratio $q_{\text{H}_2\text{O}}$ as measured by FLASH-B hygrometer at 2000 UTC. The straight gray line indicates the anticipated mean increase of $q_{\text{H}_2\text{O}}$ under undisturbed conditions. The height of the $q_{\text{H}_2\text{O}}$ profile was calculated by integrating the hydrostatic equation using a mean value of the temperature taken from the 1100 UTC and 2300 UTC soundings.
Figure 3: Temperature (color shaded, K), $T_{\text{ice}}$ calculated with $q_{\text{H}_2\text{O}} = 5$ ppmv (dashed white contour line), and geopotential height (solid lines, dam) at the 30 hPa and 50 hPa pressure levels (upper and middle panel, respectively), valid on 26 January 2005 at 1800 UTC. The 10-day backward trajectories launched at 2100 UTC are plotted in white, a dot every 12 hours. Lower Panel: $T-T_{\text{ice}}$ for the trajectories released at 30 hPa (solid) and 50 hPa (dashed).
Figure 4: ECMWF analyzed tropopause maps valid on 26 January 2005 at 1200 UTC: Potential temperature (left, K) and horizontal wind speed (right, m/s) at the dynamical tropopause defined by the 2 PVU surface; 1 PVU = $10^{-6}$ m$^2$ s$^{-1}$ K kg$^{-1}$ (e.g. Holton et al., [1995] – PV-definitions of the dynamical tropopause in the literature range from 1.5 to 3.5 PVU).
Figure 5: Vertical cross section perpendicular to the jet axis. Magnitude of the horizontal wind speed \( V_H \geq 10 \text{ m/s} \) (thick black lines, m/s; \( \Delta V_H = 10 \text{ m/s} \)), potential temperature (gray solid lines, K; \( \Delta \Theta = 10 \text{ K} \)), and divergence of the horizontal wind (blue/red contour lines, \( 10^{-5} \text{ s}^{-1} \); lowest contour lines at \( \pm 1 \times 10^{-5} \text{ s}^{-1} \), respectively, the contour increment is \( 1 \times 10^{-5} \text{ s}^{-1} \)). The stratospheric cold layer with \( T < 190 \text{ K} \) is shaded with superimposed contour lines of absolute temperature (K, \( \Delta T = 2 \text{ K} \)). Data: T511/L60 ECMWF operational analyses valid on 26 January 2005 at 1200 UTC.
Figure 6: Simulated mesoscale temperature (color shaded, K) and horizontal wind speed (m/s; wind flags: small barbs 5 m/s, long barbs 10 m/s) at 20 km altitude. The solid contour lines mark the geopotential height of the 900 hPa surface indicating the near-surface flow conditions. Valid times: 26 January 2005 at 0600 and 1200 UTC (upper row: left and right), and 1700 and 2100 UTC (lower row: left and right). Numerical results from the innermost nested domain with $\Delta x = 4$ km.
Figure 7: Vertical cross section parallel to the jet axis along the baseline sketched in Fig. 6. MM5 simulated vertical wind (blue/red contour lines, cm/s; \(\Delta w = 10\) cm/s), potential temperature \(\Theta\) (black, K; \(\Delta \Theta = 10\) K). The stratospheric cold layer is color shaded with superimposed contour lines of absolute temperature (K, \(\Delta T = 1\) K). The location of the dynamical tropopause is indicated by the dark gray band enclosing the 1.5 and 2.5 PVU surfaces. Times as in Figure 6; numerical results from the innermost nested domain with \(\Delta x = 4\) km. Ny-Ålesund is located at about 160 km.
Figure 8: Enlargement of the stratospheric temperature structure between 15 and 25 km from Fig 7: at 1700 UTC (+29 h since MM5 initialization; top panel) and at 2100 UTC (+33 h since MM5 initialization; bottom panel).
Figure 9: Ratio of the ice saturation ratio $S_i$ to the threshold value $S_{cr}$ where homogeneous nucleation occurs and temperature (K) along synoptic-scale (a) and mesoscale (b) backward trajectories released at Ny-Ålesund on 26 January 2005 1800 UTC (T511/L60 ECMWF, a) and 1900 UTC (MM5, b). The pressure at the departure levels for ECMWF backward trajectories is 30 hPa (solid lines), 40 hPa (dashed lines), and 50 hPa (dash-three dots) and for the mesoscale backward trajectories 32.2 hPa (solid line), 36.8 hPa (dotted lines), 41.6 hPa (dashed lines), 46.5 hPa (dash-dot lines), and 51.4 hPa (dash-three dots lines), respectively.
Figure 10: Schematic sketch of the synoptic situation on 26 January 2005, illustrating the conditions for inertia-gravity waves induced by the jet stream (left) and for mountain gravity waves (right).