Large NAT particle formation by mother clouds: Analysis of SOLVE/THESEO-2000 observations


1Atmospheric and Climate Science, ETH Zürich, Switzerland.
2AWI Foundation for Polar and Marine Research, Potsdam, Germany.
3NASA Langley Research Center, Hampton, VA, USA.
4DLR Oberpfaffenhofen, Wessling, Germany.
5NOAA Aeronomy Laboratory, Boulder, CO, USA.
6Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, USA.

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1. Introduction

[2] Ozone destruction is promoted by PSCs in two important ways. First, their surfaces host heterogeneous reactions which cause chlorine activation. Second, PSCs can remove HNO3 through sedimentation, a process termed denitrification. Based on lidar observations by Browell et al. [1990], Toon et al. [1990] introduced three types of PSCs: type 1a PSCs with low backscatter ratio (BSR) and moderate aerosol depolarization (δaerosol), which they interpreted as containing small number densities of HNO3−4 cm−3 (δaerosol)(I) of spherical particles, mostly nitric acid trihydrate (NAT); type 1b PSCs with moderate BSR and negligible δaerosol, which were later identified as ≈10 cm−3 ternary mixture (HNO3⋅H2SO4⋅H2O) aerosol droplets (Carslaw et al., 1994); and type 2 PSCs with high BSR and δaerosol consisting of water ice particles. Later, Tsias et al. [1999] identified type 1a- enh PSCs as similar to type 1a but with enhanced NAT particle number densities (n ≈ 0.1 cm−3). During the recent SOLVE/THESEO-2000 Arctic stratospheric campaign Fahey et al. [2001] discovered extremely large (r ≈ 3–10 μm) nitric acid hydrate particles, most likely NAT at very low number densities (n ≈ 10−4 cm−3). These so-called “NAT-rocks” play a major role in denitrification which can increase ozone loss.

2. PSC Measurements

[4] We analyzed data from three lidar systems: the ground-based stratospheric aerosol lidar in Ny-Ålesund, Spitsbergen (78.9° N, 11.9° E) operating at two wavelengths (353 nm, 532 nm) [Beyerle et al., 2001]; the NASA LaRC Aerosol Lidar (a piggy-back instrument to the NASA GSFC AROTEL lidar) on board the NASA DC-8 aircraft (532 nm, 1064 nm); and the DLR OLEX lidar system on board the Falcon aircraft (354 nm, 532 nm, 1064 nm) [Flentje et al., 1999]. All three systems measure total backscatter at all wavelengths and depolarization at 532 nm, which allows discrimination between spherical and aspherical (e.g., NAT) particles.

[5] In addition to the lidar data (Figure 1), we use in situ particle measurements from the NO2 instrument on board the NASA ER-2 aircraft [Fahey et al., 2001; Northway et al., 2002]. Two NO2 sampling inlets located on a particle separator body allow Fahey et al. [2001] to distinguish large HNO3 containing particles from NO2 in the gas phase and in small particles (r < 1 μm). According to the day of measurement, the data is labelled D26, D29 and D31. Similarly, model simulations are labelled S26, S29 and S31.

2.1. D26: Ny-Ålesund, Spitsbergen, 26 January 2000

PSCs were observed from the start of the measurements at 03 UTC through the rest of the day (Figure 1a).
Using T-Matrix calculations [Mishchenko, 1991], the cloud layer at 19–21 km with BSR \( \approx 1.1–1.7 \) and \( \delta_{\text{aerosol}} \approx 35–50\% \) can be explained in terms of NAT particles with \( n \approx 0.1–0.01 \) cm\(^{-3}\). Backward trajectories calculated with ECMWF (European Center for Medium-Range Weather Forecasts) analysis data of air masses passing Ny-Alesund at 12 UTC on 26 January reveal a temperature minimum of about 3 K above the ice frost point \( T_{\text{ice}} \) 12–24 h earlier when the air passed Greenland. Two days prior to that, the temperatures were well above the NAT existence temperature \( T_{\text{NAT}} \) (Figure 2a). A numerical hindcast simulation for Greenland with the mesoscale High Resolution Model HRM, initialized at 20 UTC on 25 January, yields lower minimum temperatures around \( T_{\text{ice}} \) (Figure 2b). This temperature history suggests that the NAT clouds observed over Spitsbergen formed about 12 hours earlier in these cold spots over Greenland, possibly on mesoscale ice clouds. The presence of supercooled ternary solution droplets (STS) below the NAT layer (as indicated by an elevated backscatter without depolarization in Figure 1a) shows that the NAT cloud base is adjacent to air with \( S_{\text{NAT}} > 1 \). This is a necessary condition for NAT particles, sedimenting out of the NAT cloud, to undergo growth below the cloud base. Furthermore, forward air trajectories calculated with ECMWF analysis data from the location of the observed PSCs show that they existed in air masses that remained below \( T_{\text{NAT}} \) for more than a week (Figure 2a). This could allow the observed NAT PSC to act as a “mother cloud” for large NAT particles [Fueglistaler et al., 2002].

2.2. D29: DC-8, Greenland, 29 January 2000

The LaRC aerosol lidar system on board the NASA DC-8 measured a large PSC over Greenland at 14–21 km altitude (Figure 1b, geographical extent shown by the thick black line in Figure 3). Low lidar backscatter, BSR(532 nm) \( \leq 1.3 \), combined with significant aerosol depolarization, \( \delta_{\text{aerosol}} \approx 15–30\% \), indicates that the cloud consisted of large particles with low number densities. T-matrix calculations [Mishchenko, 1991] for aspherical PSC particles [cf. Figure 1. (a) D26, Ny-Alesund ground based lidar on 26 January. Fading colors indicate non-depolarizing signal \( \delta_{\text{aerosol}} < 8\% \) from liquid type 1b PSCs. Solid dot: starting point of mother cloud trajectory. (b) D29, LaRC aerosol lidar measurement over Greenland on 29 January. Open circles: location and radius of modelled NAT particles produced by mother cloud trajectory starting at the solid dot in (a). (c) D31, OLEX measurement between North Cape and Spitsbergen on 31 Jan.; particles as in (b).

Figure 2. (a) Temperature history of air passing over Ny-Alesund (D26) at 12 UTC on 26 Jan. on the 460 K isentropic surface (approx. 19.3 km altitude). Temperatures from ECMWF (black) and from a mesoscale HRM (red) simulation. NAT and ice existence temperature are calculated with 5.5 ppmv H\(_2\)O and 8 ppbv HNO\(_3\). D26, D29 and D31 refer to the observations in Figure 1; (b) Temperature at 00 UTC on 26 Jan. on the 460 K isentropic surface from a mesoscale HRM simulation over Greenland. The white line corresponds to the backward trajectory of D26 calculated with HRM data (red line in (a)).

Figure 3. Overview of PSC observations and trajectories (air and particles). D26: Ny-Alesund (ground-based); D29: Greenland (DC-8); D31: North Cape - Spitsbergen (Falcon thick line, ER-2 thin line). Location and date (days of January 2000) shown for 460 K air parcel trajectory (violet) of the mother cloud which was located at 12 UTC on 26 Jan. 2000 over Ny-Alesund and for sedimenting NAT particle trajectories (color-coded for radius) started along the mother cloud trajectory (see text).
suggest that these observations are best explained as NAT particles with radii $r \geq 3 \mu m$ and number densities $n < 10^{-2} \text{ cm}^{-3}$.

2.3. D31: Falcon and ER-2, North Cape–Spitsbergen, 31 January 2000

On 31 January the Falcon flew from the North Cape to Spitsbergen and back [Flentje et al., 2002]. Due to an iced window, the depolarization data is corrupted over a large portion of the flight, but for the short flight segment presented (Figure 1c) the depolarization data is sound. The observed PSC showed similar depolarization to that seen in the LaRC data from 29 January, but a significantly lower BSR, which indicates an even lower particle number density. The ER-2 flew at the same time as the Falcon (flight track shown in Figure 3). NAT particle populations with number densities of $10^{-4}$ cm$^{-3}$ to as low as $10^{-5}$ cm$^{-3}$ and most probable radii of 4.5–5.5 $\mu m$ have been inferred from NO$_y$ in situ measurements along broad segments of the flight track [Northway et al., 2002].

3. NAT Particle Trajectories

Forward trajectories of single sedimenting NAT particles were calculated to test the applicability of the mother cloud/NAT-rock mechanism to the observations D26–D31. The equations describing particle growth and sedimentation have been implemented in the trajectory tool LAGRANTO [Wernli and Davies, 1997]. The HNO$_3$ vapour pressure of NAT was calculated according to Hanson and Mauersberger [1998]. Vortex-averaged gas phase profiles for late January 2000 have been taken from Schiller et al. [2002] for H$_2$O and Kleinbohl et al. [2002] for HNO$_3$ and were kept fixed over time (i.e., the gas phase HNO$_3$ depletion between 26 Jan. and 31 Jan. due to the sedimenting NAT particles themselves was ignored). The available HNO$_3$ for NAT particle growth is derived from the equilibrium partitioning of HNO$_3$ into STS [Carslaw et al., 1995]. Particle trajectories with initial particle radius of 1 $\mu m$ and 3 $\mu m$ in the mother cloud were calculated to represent the lower and upper limit, respectively, of particle size. In both cases growth and sedimentation were calculated for spherical and aspherical (aspect ratio 1:3) particle shape [Fueglistaler et al., 2002]. As all four cases result in very similar patterns of the sedimenting particle ensemble, concerning size, time and location, only trajectories for spherical particles with an initial radius of 3 $\mu m$ are presented here (sensitivity studies are shown in the Supplement).

4. Results

Figure 3 shows the trajectory of the advected mother cloud (D26) as well as NAT particle trajectories started every 6 hours from 26 January 12 UTC to 31 January 00 UTC from the actual mother cloud position. The match in lat./lon./alt. and time of the particle trajectories with the observations is very good, and calculations (not shown) nearby in time and space to D26 yield similar matches with D29/D31, revealing the robustness of this treatment.

Figure 3 shows that air masses passing over Ny-Alesund on 26 January 2000 were at the southern edge of a pool of cold air remaining for more than a week below T$_{NAT}$. Note the good agreement of modeled particle positions and the observed end of the PSC at the West coast of Greenland (D29) and at the southern part of D31. To complement Figure 3, in Panels 1b and 1c white circles indicate where particle trajectories intersect with the lidar measurements. Aside from the horizontal (geographical) match, also the vertical extent of the PSCs on 29 and 31 January could be explained by particles sedimenting from the mother cloud.

From the PSC sedimentation observed by lidar between the outward and incoming flight paths on 31 Jan., Flentje et al. [2002] inferred a particle radius of 8 ± 1 $\mu m$. Based on in situ measurements of the NO$_y$ instrument, Northway et al. [2002] derived particle radii of approximately 5 $\mu m$ near this location (ER-2 cruise levels ≈ 18 km altitude), remarking that even larger particles ($r \approx 10 \mu m$) were observed at 14–15 km altitude. Modelled particle radii of 4 to 7.5 $\mu m$ at altitudes between 19 and 16 km (Figure 1b, c), respectively, compare well with these observations.

Additional trajectory calculations (see Supplement) show that the eastern part of D29 and the northern part of D31 can be matched with particles sedimenting from a mother cloud located on 26 January 12 UTC about 200–300 km north of Spitsbergen. There are no measurements available at this position, but the similar temperatures of backtrajectories from this location and backtrajectories from D26 support the assumption that the mother cloud observed over Spitsbergen extended 200–300 km further north.

Figure 4 shows the results of a 1-D microphysical model [Fueglistaler et al., 2002]. This model neglects effects caused by wind shear, but takes gas phase depletion from the sedimenting particles into account. This enables accurate calculation of particle sedimentation and particle number densities. The simulation started on 26 Jan. 2000 12 UTC with a mother cloud at 19.5–20 km altitude with $n = 0.01 \text{ cm}^{-3}$. The vertical temperature profiles over time are idealized ECMWF temperature profiles taken along the mother cloud trajectory. Using T–Matrix calculations, the microphysical data of the model is converted into a simu-
lated lidar backscatter. After ≈3 days, the mother cloud dissolves and after 5 days the sedimenting particle ensemble reaches a particle number density \( n \approx 10^{25} \text{cm}^{-3} \) in the entire column. Compared with the measurements D26 (Figure 1a), D29 (Figure 1b) and D31 (Figure 1c), the modelled lidar signals S26, S29 and S31 fit both in absolute BSR and decrease in BSR over time. This is consistent with the assumption that the observed decrease in lidar backscatter resulted from a decrease in particle number density in the PSCs.

5. Conclusions and Outlook

[15] PSC observations on 26, 29 and 31 Jan 2000 were matched in latitude, longitude, altitude, and time by forward trajectories of sedimenting NAT particles started from a single advected mother cloud. A 1-D microphysical simulation of particles sedimenting out of the mother cloud reproduces the observed lidar backscatters, and calculated particle radii compare well with in situ measurements. This analysis shows that the suggested mother cloud/NAT-rock mechanism could explain the observed characteristics of large NAT particles without having to invoke a highly selective NAT nucleation mechanism. Instead, the advection of NAT particles within a mother cloud under saturated conditions (\( S_{\text{NAT}} \approx 1 \)) and their subsequent sedimentation into supersaturated regions below the cloud (\( S_{\text{NAT}} \gg 1 \)) with strong growth allows a geographically confined NAT nucleation region to lead to the observed widespread occurrence of large NAT particles far away from the original nucleation area.

[16] As mentioned above, a likely mechanism for the formation of mother clouds is NAT nucleation in ice PSCs. In the Arctic, mountain wave activity over Greenland, Scandinavia and the Urals could lead to enhanced occurrence of ice PSCs [Carslaw et al., 1999]. In the winter 1999/2000, ECMWF analysis data repeatedly showed temperatures below \( T_{\text{Ice}} \) at altitudes of approximately 19 to 25 km, and reported mountain wave activity led to additional ice PSCs [Dörmbrack et al., 2002]. Thus, frequent occurrence of mother clouds at altitudes of 19 to 25 km can be expected and is in agreement with airborne and ground-based lidar observations of PSCs. Fueglistaler et al. [2002] showed that the mother cloud/NAT-rock mechanism requires air parcel temperatures below \( T_{\text{NAT}} \) for at least 2 to 3 days to cause significant denitrification. Due to the non-linear particle sedimentation, each additional day greatly enhances the denitrification potential of a mother cloud. Fueglistaler et al. [2002] estimated that after 5 days, denitrification amounts up to \( \approx 75\% \) directly below the mother cloud, and significant denitrification extends to \( \approx 4 \) km below the cloud. Thus, for the winter 1999/2000, a vortex-wide calculation of denitrification based on the mother cloud/NAT-rock mechanism is expected to show denitrification between 16 and 25 km, with a maximum at altitudes around 20 to 22 km. This is in qualitative agreement with observations: In situ NO\(_2\) measurements on board the ER-2 show denitrification between 16 and 21 km, with a maximum of up to 80\% at 20 to 21 km, and additional balloon-borne data reveal the upper edge of denitrification at about 25 km [Popp et al., 2001].

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