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COMPARISON OF STRATOSPHERIC CLOUDS IN THE ANTARCTIC AND THE ARCTIC

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Abstract. Balloonborne measurements of stratospheric clouds using optical particle counters in the Arctic and the Antarctic are compared in terms of particle size distribution. There appear to be two distinct classes of particles, a small mode ($r \geq 0.2$ μm) in which at least half of the available condensation nuclei have grown and a large mode ($r \approx 2$-3 μm) in which fewer than 1% of the available condensation nuclei have grown. The latter particles appear in thin layers (0.1 to 0.3 km) while the former appear in relatively thicker layers (2 to 5 km). Temperatures dictate a nitric acid trihydrate composition for most of the layers and the inferred HNO_3 vapor in the large particles is comparable to that available. While the small particle mode is probably related to fast cooling events such as those associated with mountain lee waves, the origin of the thin layers of large particles is not apparent.

Introduction

It is now generally accepted that polar stratospheric clouds (PSC) are largely composed of nitric acid and water, probably in the form of the crystalline trihydrate [Toon et al., 1986; Crutzen and Arnold, 1986]. Laboratory measurements indicate that nitric acid trihydrate (NAT) will form at temperatures about 5°C higher than the frost point [Hanson and Mauersberger, 1988] and thus should be much more prevalent than clouds containing pure ice particles. We here report recent measurements of PSC formation both in the Antarctic and the Arctic. In the latter case, observations were made at the end of January 1989, following the coldest January on record for the north pole at 30 hPa.

Observations and Discussion

Measurements were made with balloonborne optical particle counters similar to those used by the University of Wyoming since 1971 [Hofmann et al., 1989a] except that the air sampling rate has been increased by a factor of about 12 in order to resolve low concentrations of particles and the air sample is kept at ambient temperature to avoid particle evaporation. With this instrument, concentrations of volatile ice crystals as low as 10^{-3} cm^{-3} can be measured. Data are collected in 7 size ranges, $r \geq 0.20$, 0.25, 0.30, 1.0, 2.0, 3.0 and 5.0 μm. A condensation nuclei (CN) counter is used to obtain the total aerosol ($r \geq 0.01$ μm) concentration.

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Figure 1 shows PSC layers at McMurdo, Antarctica (78°S) on September 9, 1988 (12-16 km), and at Kiruna, Sweden (68°N) on January 23, 1989 (18-23 km), in which the concentration of $r \geq 0.20$ μm particles exceeded 50% of the CN concentration. Normal stratospheric sulfate profiles (September 4 and January 30, respectively) are shown for comparison. The small particle clouds are indicative of a high degree of vapor supersaturation caused by fast cooling (FC) events since a sizable fraction of CN grow to form the relatively small ($r \approx 0.2$ μm) cloud particles. The temperature profiles suggest that wave disturbances and/or vertical motions may be associated with the particle growth phenomenon [Hofmann, 1989; Hofmann et al., 1989b]. Temperatures in the FC events were close to the frost point (see Table 1) and either a water or NAT composition is possible. In fact, in the 10 to 11.5 km region in the Antarctic data, temperatures reached -85°C where large ($r \geq 5.0$ μm) particles, presumably ice crystals, were observed [Hofmann, 1989].

All profiles in Figure 1 display enhanced thin layers of large cloud particles at concentrations <1% of the available CN in the 15-27 km altitude range. Large particles observed previously in Antarctica were believed to be formed during slow cooling (SC) events, i.e., time scales of weeks to months, in which preferential growth on large ($r \approx 0.3$ μm) sulfate particles occurred [Hofmann et al., 1989a]. Figure 2 shows temperature and particle concentration profiles for the thin layers, first observed at McMurdo in September 1988 and later at Kiruna in January 1989 [Hofmann et al., 1989b]. The thin clouds observed on January 30 in the Arctic are more numerous and they formed at higher altitudes than in the Antarctic. They were also observed at Kiruna on January 23 but less frequently at lower concentration (see Figure 1).

Apparent near-adiabatic layers (potential temperature approximately constant with altitude) occur in both the Arctic and Antarctic cases in Figure 2 but superpositions of smaller height scale, wavelike temperature variations are present, especially in the Arctic case. These temperature variations represent changes of as much as 5°C in 200 m. The Vaisala temperature sensor, mounted on top of the gondola, has a response time of 0.3 s at 1000 hPa. It's stratospheric response is not known. The data points in Figure 2 are at 10 s intervals (≈ 30 m) and are believed to be representative of the true temperature variations.

Also given in Figure 2 are frost point and NAT condensation temperature curves [Hanson and Mauersberger, 1988] for what are believed to be representative H_2O mixing ratios for the two polar regions at the respective times (3 ppmv in the Antarctic spring, 5 ppmv in the Arctic winter [Rosen et al., 1988; 1989]). Although the small scale temperature variations occur in the temperature range where NAT may form for expected HNO_3 mixing ratios (2-5 ppbv in the Antarctic

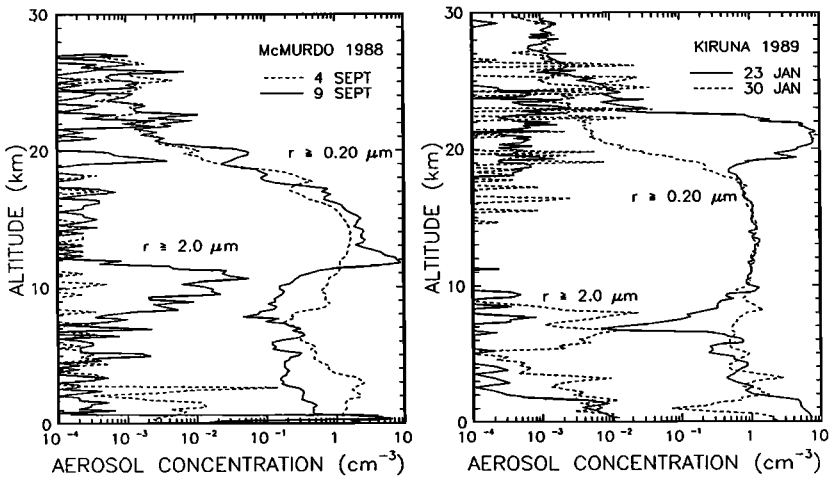

Fig. 1. Aerosol concentration profiles for 2 size ranges at McMurdo, Antarctica in September 1988 and at Kiruna, Sweden in January 1989.

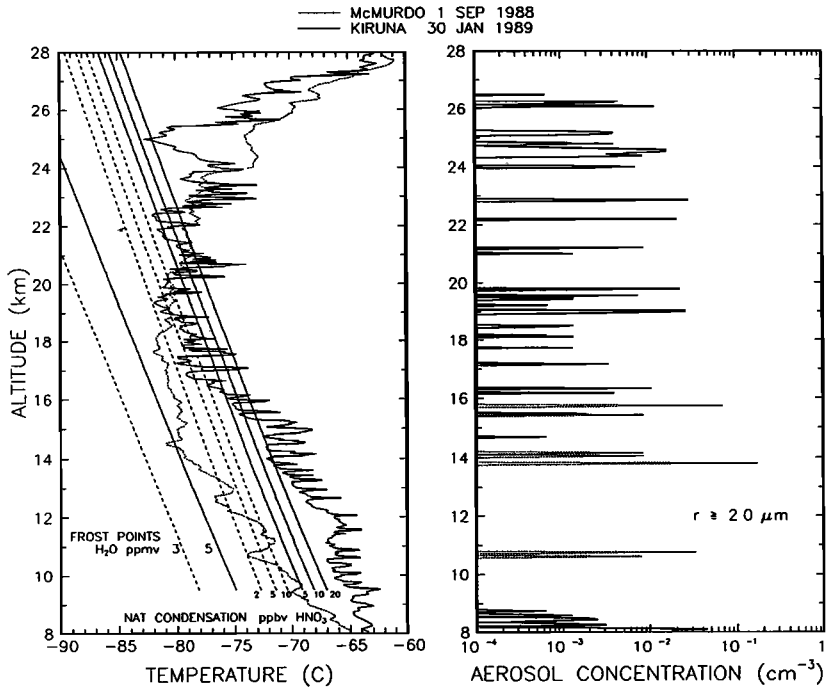

Fig. 2. Temperature and aerosol concentration profiles in selected height intervals containing PSC particle distributions at McMurdo Station, Antarctica in September 1988 and at Kiruna, Sweden in January 1989. The smooth curves are the water frost point and NAT condensation temperature curves for the indicated H_2O and HNO_3 mixing ratios.

spring, 10-20 ppbv in the Arctic winter), there is not a consistent correlation between cloud appearance and regions of expected NAT formation. Clouds do not appear in the Antarctic at 16-18 km, where it is sufficiently cold for NAT to form suggesting substantial denitrification by the end of August 1988, while clouds appear at high altitude in the Arctic in regions too warm for condensation.

The large particle layers on January 30 occurred between temperatures of -74 and -82°C with a correlation coefficient of only -0.20. However, restricting the data to concentrations >0.01 cm^{-3}, for $r \geq 2.0$ μm, gives a correlation coefficient of -0.78 even though the 9 data points span the same temperature range. Comparing temperature lapse rate with concentration, the correlation coefficient is only -0.12, although 78% of the particle data was observed with a negative lapse rate.

Figure 3 shows size distributions for FC events and for the 13 to 15 km sulfate layer under cloud-free conditions, both for the Antarctic and the Arctic. The FC cloud particle size distributions are very similar even though the clouds were at much different altitudes. For the McMurdo cloud layer at 12 km, the CN (total aerosol) concentration was about 20 cm^{-3} with about 1 cm^{-3} having radii greater than 0.2 μm [Hofmann, 1989], while for the Kiruna cloud layer at 21 km, the CN concentration was about 15 cm^{-3} with no particles having radii greater than 0.2 μm. The size

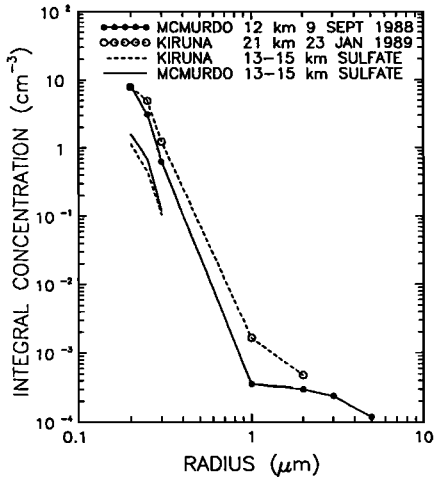

Fig. 3. Thick layer PSC and sulfate layer particle integral size distributions at McMurdo Station, Antarctica in September 1988 and at Kiruna, Sweden in January 1989.

distributions are very similar because condensation occurs on most of the CN in an FC event and the concentrations of these were similar at the two altitudes. We see a minor enhancement in the large particle mode ($r \geq 1$ μm) in both clouds. These are probably associated with SC events as discussed earlier.

Figure 4 shows the size distributions of the thin, large particle cloud layers observed both at McMurdo and Kiruna. Below about 20 km, where there is a sizable sulfate component, the size distributions are bimodal with both a small sulfate and a large cloud particle mode present. Above 20 km the size distributions are essentially unimodal with only the large particle mode present. Because the concentrations are low and particle sizes large in this mode, one should classify these as SC events; however, in SC events the cooling may occur over weeks to months and mixing would disperse the layers which formed. Since these layers are only 30 to 300 m thick they

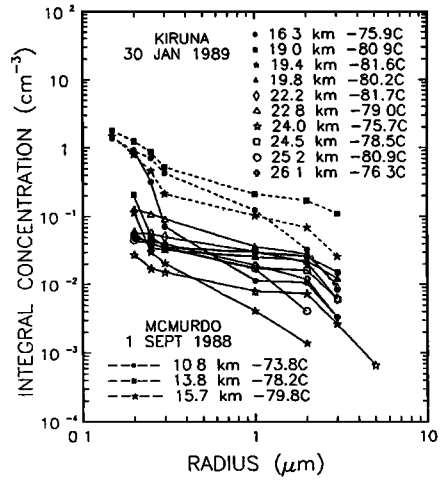

Fig. 4. Thin layer PSC particle integral size distributions at McMurdo Station, Antarctica in September 1988 (dashed curves) and at Kiruna, Sweden in January 1989 (full curves).

could not have existed for very long, probably only hours to at most a day, under the assumption that they are growing. For rapid growth a high degree of vapor supersaturation is required with concomitant condensation on all available CN, which is not observed. These thin layers of large particles thus appear to defy description by any standard condensational growth mechanisms. They may be associated with the fine scale temperature structure discussed earlier; however, the rapid variation in temperature with altitude may not be indicative of the cooling experienced during particle formation. In fact, as will be discussed later, one could also interpret the data as indicating particle evaporation.

Fitting lognormal distributions to the size data in Figures 3 and 4 results in the parameters for the total concentration, median radius and width of each mode listed in Table 1 along with the ice saturation mixing ratio for each layer. The latter is considerably greater than the H_2O vapor mixing ratio which could be available for the January 30 case at Kiruna, especially for the thin layers of large particles above 20 km suggesting an NAT composition. Table 1 also gives the volume mixing ratios of H_2O and HNO_3 vapor calculated from the aerosol mass, assuming an NAT composition in the latter case. Particle sphericity has been assumed which could cause considerable errors in mass estimates for large particles if they are highly aspherical. This uncertainty is larger for large water ice crystals than for denser (1.62 gcm^{-3}) NAT particles whether in crystalline (orthorhombic) or solid solution form.

The mass in the September 1 Antarctic clouds indicate that they are composed of water ice as

TABLE 1. PSC Size Distributions and Mass

Location Date	Alt. (km)	T (°C)	H_2O_s[a] (ppmv)	N_o (cm^{-3})	r_o (μm)	σ	H_2O[b] (ppmv)	HNO_3[c] (ppbv)
McMurdo	10.8	-73.8	8.2	.20	1.2	1.70	.03	6.5
880901	13.8	-78.2	6.7	.22	3.0	1.73	.80	194*
	15.7	-79.8	7.1	.12	2.2	1.52	.13	30*
McMurdo	12	-80.0	3.4	20	0.19	1.28	.004	1.0
880909	12	-80.0	3.4	.0004	3.3	2.17	.005	1.25
Kiruna	21	-85.0	6.5	.15	0.20	1.36	.01	4.6
890123	21	-85.0	6.5	.002	1.5	1.60	.001	0.4
Kiruna	16.3	-75.9	13*	.012	2.6	1.30	.01	3.3
890130	19.0	-80.9	9.4	.03	3.0	1.53	.14	33
	19.4	-81.6	9.0	.01	0.84	2.22	.008	2.0
	19.8	-80.2	12*	.025	2.95	1.40	.094	22.6
	22.2	-81.7	14*	.03	2.35	1.35	.08	18.4
	22.8	-79.0	25*	.036	2.50	1.33	.12	29.5
	24.0	-75.7	52*	.008	3.1	1.40	.08	18.3
	24.5	-78.5	37*	.018	2.7	1.28	.10	22.7
	25.2	-80.9	28*	.02	1.5	1.46	.03	6.4
	26.1	-76.3	68*	.02	2.2	1.39	.09	21.7

[a] H_2O_s = ice saturation mixing ratio
[b] H_2O mixing ratio for H_2O aerosol composition
[c] HNO_3 vapor mixing ratio for NAT aerosol composition, density = 1.62 g cm^{-3}
*Too large for available vapor mixing ratio

the implied HNO_3 vapor mixing ratio for NAT is too high. Most of the thin layers of large particles observed at Kiruna on January 30, 1989 can be interpreted as being composed of NAT and contain most of the available HNO_3 vapor with a mixing ratio of about 20 ppbv. While this value is higher than previously assumed Arctic values, it is in general agreement with results of remote sensing measurements of HNO_3 vapor and insitu measurements of NO_y (Y. Kondo, personal communication, 1989) conducted on the January 23 Kiruna flight and insitu measurements of HNO_3 vapor [Arnold et al., 1989] on the January 30 flight.

The thin large particle layers may be evaporative remnants of nacreous clouds (ice crystals) formed during mountain lee wave events upwind of Kiruna and McMurdo as large ice crystals may not evaporate rapidly in undersaturated conditions [Toon et al., 1989]. Synoptic conditions in the stratosphere (flow over upwind mountain ranges to the west with speeds as high as 50 ms^{-1}) favor such an interpretation as does the Kelvin effect (preferential evaporation of the small particles). In addition, since large particles are concentrated near the center of a cloud, earlier evaporation of the smaller particles at the edges would tend to narrow the cloud layer.

It is possible that if the high altitude layers are evaporative remnants, the forerunner was a nitric acid-water cloud in which large NAT crystals have survived. Evaporation of solid solution NAT prior to the crystalline form was proposed by Rosen et al. [1989] to explain thin scattering layers observed in the Arctic stratosphere in January 1989. Although the two regions of maximum cloud appearance at Kiruna (19-20 km and 24-25 km) were colder than their surroundings, the lower layer is supersaturated with respect to NAT condensation, for expected levels of HNO_3 vapor, while the upper is highly undersaturated. Simultaneous insitu measurements of HNO_3 vapor indicate depletion only at the lower altitude suggesting the possibility of condensation or denitrification in that region [Arnold et al., 1989]. For the 24-25 km layer, evaporation is probably more likely returning HNO_3 vapor to the gas phase and, with subsequent mixing, might explain the apparent absence of correlative thin HNO_3 depletion layers at high altitude. However, without detailed knowledge of the air parcel's temperature history, it is impossible to evaluate such nonequilibrium evaporative mechanisms.

Summary

Balloonborne measurements of PSC particles in the Antarctic and the Arctic were used to discuss the nature of the clouds and their possible formation mechanisms. Most of the cloud particles appear to be consistent with an NAT composition but there appear to be two distinct size distributions: thick layers of numerous small particles, associated with fast cooling episodes such as might occur in association with large scale mountain lee waves, and thin layers of sparsely populated large particles which cannot be explained by a slow cooling history because of their apparent youth. If composed of NAT, these thin layers appear to contain all the available nitric acid vapor with values of about 20 ppbv although considerable uncertainty exists without knowledge of the shape of the large particles. The thin layers appear to be associated with fine scale structure in the vertical temperature profile; however, preferential condensation on only 1 in 10^3 or so of the available condensation nuclei cannot be explained in the framework of classical nucleation theory. In the warmer high altitude region the layers may be remnants of clouds undergoing evaporation.

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