Size and Number Concentration of Liquid PSCs: Balloon-Borne Measurements at Ny-Ålesund, Norway in Winter of 1994/95

By M. Hayashi, Y. Iwasaka,1 M. Watanabe, T. Shibata
Solar-Terrestrial Environment Laboratory, Nagoya University, Nagoya 464-0814, Japan

M. Fujiwara
Laboratory of Applied Physics, Fukuoka University, Fukuoka 814-0180, Japan

H. Adachi, T. Sakai, M. Nagatani
Solar-Terrestrial Environment Laboratory, Nagoya University, Nagoya 464-0814, Japan

H. Gernandt, R. Neuber
Alfred Wegener Institute for Polar and Marine Research, Potsdam D-14401

and

M. Tsuchiya
Shonan Institute of Technology, Tsujido-Nishikaigan, Fujisawa 251-8511, Japan

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Abstract

Size (radius: 0.15–1.8 μm) and number concentration of polar stratospheric clouds (PSCs) were observed with a balloon-borne particle counter at Ny-Ålesund, Norway (79°N, 12°E) in winter of 1994/95 (December 18, 1994 and January 17, 1995). The measurements suggested that PSCs actively formed in the cold winter stratosphere. During the balloon-borne measurements, the depolarization ratio of PSCs was monitored with a lidar near the balloon observational site to see the phase (liquid or solid) of PSCs. Both measurements suggested that many sub-layers were contained in the PSCs.

Particle size distribution of the early PSCs event (December 18, 1994) was different from that observed on January 17, 1995, after active PSCs events. This difference possibly suggests that particle formation processes were largely disturbed by redistribution of chemical constituents relating with PSCs formation and growth in the polar winter stratosphere. Lidar measurements suggested active formation of liquid phase PSCs in addition to solid phase PSCs. The number-size distribution pattern showed enhancement of the number of particles with their radius larger than 1.8 μm in the solid state sub-layer of PSCs. On the other hand, not only large particles (r > 1.8 μm) but also smaller particles (r = 0.25–0.6 μm) were enhanced in the liquid state PSCs.

1. Introduction

Polar stratospheric clouds (PSCs) have been believed to play an important role in large ozone depletion in the polar stratosphere through heterogeneous reaction including PSCs. The nature of PSCs is a large concern because PSCs convert inert chlorine to active chlorine.

Extensive research in the past 10 years has produced a outline of PSCs structure and composition (McCormick et al., 1985; Iwasaka et al., 1985, 1986; Hanson et al., 1988; Browell et al., 1990;
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size distribution and concentration of PSCs parti-
However, lidar observations cannot obtain the exact
layer was accompanied by a solid-
lidar measurements made at Ny-Alesund, Norway
Shibata et al. (1997) suggested, on the basis of the
1997). Then, type I PSCs were classified into two
experiments (e.g., Hanson and Mauersberger, 1988),
which showed that particles of nitric acid trihydrate
(NAT; HNO3·3H2O) would be stable under the po-
sharacteristic conditions. Some investigators sug-
ggested that background H2SO4·H2O droplets froze
in the polar winter stratosphere and subsequently
as sites for NAT nucleation and growth (e.g.,
Poole and McCormick, 1988; Fahey et al., 1989).
Despite the widespread acceptance of solid PSCs
(NAT and ice) in the decade since discovery
of the ozone hole, the freezing mechanism of
the background H2SO4·H2O droplets remains un-
certain.
Some observations contradicted the particle for-
mation model for type I PSCs by solid NAT (Dye
et al., 1992; Toon and Tolbert, 1993; Carslaw et al.,
1997). Then, type I PSCs were classified into two
sub-types, called type Ib of solid particles and type
Ib liquid particles (Dye et al., 1992). Most recently
Shibata et al. (1997) suggested, on the basis of the
lidar measurements made at Ny-Alesund, Norway
(79°N, 12°E), that a relatively large size droplet
PSCs layer (called type X in the paper) appeared
frequently and the layer was accompanied by a solid-
state PSCs layer in the Arctic winter stratosphere.
However, lidar observations cannot obtain the exact
size distribution and concentration of PSCs parti-
even though they are very important to under-
stand the role of PSCs on the ozone depletion and
the mechanism of PSCs development.
There are, although a great deal of observational
work continues in the Arctic region, few reports de-
scribing PSCs particle size distribution measured
with a particle counter, since performing balloon-
borne measurements in the Arctic winter was tech-
ically difficult. Fortunately, simultaneous mea-
urements of PSCs with a lidar and balloon-borne
particle counter were made at Ny-Alesund, Nor-
way (79°N, 12°E) in winter of 1994/1995 as part
of cooperative measurements between Japan (Solar-
Terrestrial Environment Laboratory, Nagoya Uni-
versity and Laboratory of Applied Physics, Fukuoka
University) and Germany (Alfred Wegener Institute
for Polar and Marine Research). Here, we showed
the size and number concentrations of various types
PSCs on the basis of the measurements.

2. Observation and results

2.1 Balloon-borne measurements on particle size
and number concentration
The main specification of the balloon-borne optical
particle counter used here is summarized in Table
1, and the optical system of the particle counter is
schematically shown in Fig. 1. The details of the
counter were described in a paper by Tsuchiya et al.
(1996). Particle size discrimination is made at 0.15,
0.25, 0.4, 0.6, and 1.8 μm (in radius) on the basis
of a laboratory experiment for the refractive index
of particulate matter, n = 1.4 + 0i (for sulfuric acid
droplet of about 70 wt. %, which is referred from the
calculated values of Steele and Hamill (1981), and
the observational values of Baumardner et al. (1996)
in the stratosphere). The sizing range of the counter
will be easily modified when we treat other aerosol
materials that have different refractive indices.
Atmospheric particle size and number concentra-
tion were observed with a balloon-borne particle
counter at Ny-Ålesund, Norway (79°N, 12°E) on De-
cember 18, 1994 and January 17, 1995 (Fig. 2). The
statistical uncertainty in counting is about 3, 10,
32, and 100 % for particle concentrations of 1, 0.1,
0.01, and 0.001 cm⁻³, respectively. Therefore, the
relative error range of number concentration of the
particles with radius > 0.25 μm can be estimated,
for example, as about 4 % at 18 km where noticeable
peak of number concentration was found in the mea-
surements of December 18, 1994. However, the error
range becomes larger than about 56 % above about
21 km, since the number concentration becomes very
low. The mass mixing ratio of particulate matter,
calculated using Eq. (1), is also shown in Fig. 2.

\[
MR = ((N_0 - N_2) \times 4/3 \times \pi \times r_{12}^3) + (N_2 - N_3) \times 4/3 \times \pi \times r_{23}^3 + (N_3 - N_4) \times 4/3 \times \pi \times r_{34}^3 + (N_4 - N_5) \times 4/3 \times \pi \times r_{45}^3 + N_5 \times 4/3 \times \pi \times r_5^3 \times d \times 10^9 / \rho,
\]

where \( MR \): mass mixing ratio of particulate
matter ranging from 0.15 μm to 1.8 μm in radius (ng/g),
August 1998  M. Hayashi, Y. Iwasaka, M. Watanabe and et al. 551

Table 1. Specification of Balloon-Borne Optical Particle Counter.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume:</td>
<td>30 x 35 x 35 cm</td>
</tr>
<tr>
<td>Weight:</td>
<td>5.5 kg (including battery)</td>
</tr>
<tr>
<td>Wave length of source light:</td>
<td>(\lambda = 810) nm (laser-diode)</td>
</tr>
<tr>
<td>Detection:</td>
<td>Photodiode detector;</td>
</tr>
<tr>
<td></td>
<td>forward-scattering light</td>
</tr>
<tr>
<td></td>
<td>(13° ~ 44° from the beam axis)</td>
</tr>
<tr>
<td>Air sampling flow rate:</td>
<td>50 cm³/sec</td>
</tr>
<tr>
<td></td>
<td>(under the condition of standard</td>
</tr>
<tr>
<td></td>
<td>atmospheric temperature and pressure)</td>
</tr>
<tr>
<td>Sampling-time interval:</td>
<td>20 sec for 1st measurement, and 24 sec for</td>
</tr>
<tr>
<td></td>
<td>2nd measurement</td>
</tr>
<tr>
<td>Wave frequency for data transmission:</td>
<td>400 MHz (Vaisala radio-sonde)</td>
</tr>
</tbody>
</table>

The observations indicate the existence of particle sub-layers in which mass mixing ratio was enhanced; peak height of layers were 22 km (A) and 18 km (B) in the measurements of December 18, 1994. Below the enhanced sub-layers, the broad particle layer (possibly the background sulfate layer) was found from about 12.5 to 16 km in altitude. In the measurements of January 17, 1995, aerosol sub-layers (A, B, and C) were identified because of following reasons. Sub-layer B (around 18 km) is characterized by enhancement of particle concentration whose radius is larger than 0.4 \(\mu\)m. Sub-layer A (around 20 km) and sub-layer C (around 15.5 km) are characterized by enhancement of mass mixing ratio and/or particle concentration whose radius is larger than 1.8 \(\mu\)m.

Temperature distributions of December 18, 1994 and January 17, 1995 are compared with the frost point of nitric acid trihydrate (NAT: Type I PSCs) in Fig. 3. The frost point temperatures of nitric acid vapor on the surface of NAT and water vapor on the surface of ice are estimated, under the assumed

\[N_n: \quad \text{integrated particle number concentration for channel } n \text{ (cm}^{-3}\text{),}\]
\[r_{nm}: \quad \text{mean radius for channel } n \text{ and } m \text{ (cm),}\]
\[r_n: \quad \text{radius for channel } n \text{ (cm),}\]
\[d: \quad \text{specific density of particulate matter, assumed as 1.5 (g/cm}^3\),\]
\[\rho: \quad \text{air density (g/cm}^3\),\]
\[\pi: \quad \text{the circular constant; 3.1416.}\]
Fig. 2. Vertical profiles of particle concentration of particle radius larger than 0.15, 0.25, 0.4, 0.6, and 1.8 μm measured with a balloon-borne particle counter at Ny-Ålesund, Norway (79°N, 12°E). Profiles of mass mixing ratio of particulate matter, calculated by Eq. (1) (see text), are also shown in right panels. (a) profiles observed on December 18, 1994. Polar stratospheric clouds contained sub-layers A and B showing enhancement of number concentration and/or mass mixing ratio. (b) profiles observed on January 17, 1995. Polar stratospheric clouds contained sub-layer A, B, and C.
atmospheric condition that mixing ratio of HNO₃ and H₂O are 10, 5, 1 ppbv and 5, 3 ppmv respectively, on the basis of the laboratory experiments by Hanson and Mauersberger (1988), Jaecker-Voirol et al. (1990), and Luo et al. (1995). These values are referred from observational values of stratospheric water vapor (e.g., Kelly et al., 1990; Pruvost et al., 1993; Overlez and Overlez, 1994), and of nitric acid vapor (e.g., Clarmann et al., 1993; Oelhaf et al., 1994; Murcray et al., 1994; Hofner et al., 1996). From the temperature distributions, it is suggested that stratospheric temperature decreased the expected threshold temperature of PSCs (Type I) formation in those days.

Comparing the stratospheric temperature of December 18, 1994 and January 17, 1995, the stratosphere of January 17 was apparently colder (Fig. 3). Assuming the equilibrium condition, larger mass content of particulate matter were expected during the lower temperature (January 17) rather than the higher one (December 18). However, sub-layer B of December 18 had the largest mass content of 5.7 ng/g in the measurements (Fig. 2), which suggested that PSCs growth was affected by not only cold temperature, but also some other factors.

### 2.2 Lidar measurements

The scattering ratio (defined by Eq. (2)), depolarization ratio (defined by Eq. (3)), and Angstrom exponent (defined by Eq. (4)) are obtained by the lidar to know temporal and spacial changes in concentration, phase, and size distribution of PSCs, respectively. Detailed description of specification of the lidar used here was given by Shibata et al. (1997).

The scattering ratio and depolarization ratio are defined here by the following relations: (2) \( R = \frac{\beta_1}{\beta_2} = \frac{(\beta_1 + \beta_2)}{\beta_2} \) and (3) \( \delta = \frac{\beta_{2\perp}}{(\beta_2/)} \), where \( \beta_1 \) and \( \beta_2 \) are backscattering coefficient of atmospheric molecules and atmospheric aerosol particles, respectively. Total backscattering coefficient \( \beta \) contains both components of molecules and aerosols, and suffixes // and \( \perp \) mean parallel and perpendicular components, respectively, of backscattering light to the optical plane of emitted laser light. Therefore, the value of \( (R - 1) = \beta_2/\beta_1 \) can be recognized as the mixing ratio of particulate matter measured by a lidar.

The depolarized component in backscattering light from Rayleigh scattering of atmospheric molecules is extremely low (\( \delta = \text{about 0.4%} \),...
and the depolarized component of backscattering light from non-spherical particles becomes noticeably high. Therefore it is easy to detect non-spherical particles such as crystalized PSCs (e.g., Shibata et al., 1997) or tropospheric soil particles (Iwasaka et al., 1988) from the measurements of depolarized ratio of particulate matter.

Angstrom exponent $\alpha$ is defined here by following relation: $(4) \alpha = -\ln(\beta_{M1}/\beta_{M2})/\ln(1064/532)$, where $\beta_{M1}$ and $\beta_{M2}$ are aerosol backscattering coefficients at 1064 and 532 nm, respectively. The larger value of $\alpha$ shows more rich for smaller particle in the size distribution of aerosols, and values of $\alpha$ for background stratospheric aerosols is about 1.4.

Temporal changes in scattering ratio and depolarization ratio are shown in Fig. 4. It is clear that aerosol scattering ratio and depolarization ratio become frequently larger than the background value ($S = 1.3$ and $\delta = 0.4\%$), which was extremely enhanced during December 1994 and January 1995 (Fig. 4).

3. Discussion

Recently great interest has been focused on the variety of types of PSCs in the Arctic winter stratosphere on the basis of lidar measurements (Browell et al., 1990; Toon et al., 1990; Carslaw et al., 1997; Shibata et al., 1997), since the variety is caused by differences of particle size, phase, and materials, which affect ozone destruction processes and development of PSCs. Nevertheless, information on the size of PSCs has been extremely limited, since few measurements have been simultaneously made with a lidar and a particle counter. We successfully made simultaneous measurements of PSCs with a lidar and a balloon-borne particle counter at Ny-Alesund, Norway on December 18, 1994 and January 17, 1995.

Atmospheric temperature distribution in winter of 1994/1995 was monitored on the basis of routine measurements of meteorological sondes at Ny-Alesund (Fig. 5). Stratospheric temperature rapidly decreased in mid December of 1994 corresponding to the polar vortex formation in the Arctic stratosphere. Then, the cold region where the temperature was lower than the expected threshold of NAT formation was frequently observed from mid-December 1994 to the end of January 1995. Variation of temperature in the stratosphere was not so large from mid-December 1994 to the end of January 1995 and become large after the end of January 1995. This shows that the polar vortex had been stable and covering over Ny-Alesund from mid-December 1994 to the end of January 1995.

Enhancement of scattering ratios (larger than 1.4 around 18 km and 1.1 above 20 km in altitude) was frequently observed from mid-December 1994 to the end of January 1995, according to decreases in stratospheric temperature, suggesting active formation of PSCs in the Arctic stratosphere (Figs. 4 and 5). Enhancement of depolarization ratio larger than about 1.0 % was also observed frequently, however it did not correspond to the enhancement of scattering ratio. Enhancement of the scattering ratio with a low depolarization ratio was sometimes observed, for example around 18 km in altitude on January 10, 1995. A similar feature was suggested on the analysis of lidar measurements in 1989 AASE (Airborne Arctic Stratospheric Expedition) (Browell et al., 1990). They revealed two sub-classes of Type I PSCs (crystals of Type Ia and droplets of Ib), both appearing at temperature at, or below, the equilibrium threshold point of NAT, but higher than ice frost point. Shibata et al., (1997) suggested that liquid phase PSCs containing large size particles frequently formed, on the basis of the lidar measurements of developed PSCs with high scattering ratio in mid-January, 1995, at Ny-Alesund. They suggest that there are distinct types of liquid PSCs (called type Ib and type X in their paper) with different size distributions. Therefore, at least, three types of nitric acid PSCs were observed during the PSCs event, from mid-December 1994 to the end of January 1995 over Ny-Alesund. The most active PSCs with a high scattering ratio (larger than 5.0) was observed in mid-January. Therefore, the balloon-borne observation of December 18, 1994 was carried out at an early step of PSCs events, and that of January 17, 1995 was carried out at the end of a stable polar vortex condition after active PSCs events.

In Fig. 6 we show the vertical distribution of the scattering ratio, depolarization ratio, and Angstrom exponent measured on December 18, 1994 and January 17, 1995 when balloon-borne particle counters were sounded. Two sub-layers are identified for December 18. The upper one is characterized by high depolarization ratio (larger than 1.0%), and another one is characterized by high scattering ratio (larger than 2.0) and low depolarization ratio. On January 17, three sub-layers are identified. The middle one has a high scattering ratio (larger than 2.0), and the upper and lower ones have a high depolarization ratio (larger than 1.0%).

Two sub-layers PSCs, found in size-number concentration and/or mixing ratio of December 18, 1994 (Fig. 2), show good correspondence to the peaks of scattering ratio measured simultaneously, but not completely to peaks of depolarization ratio (Fig. 6). From the profiles of the depolarization ratio of December 18, it is suggested that sub-layer B was mainly composed of liquid phase particles, since values of $\delta$ showed a noticeable minimum and/or low (0.4–0.8 %) near the peak height of sub-layer B. On the other hand, it is suggested that sub-layer A contains non-spherical particles, by high values of $\delta$ (larger than 1.0%).

In the sub-layers found in size-number concentra-
Fig. 4. Scattering ratio (a) and depolarization ratio (b) measured with the lidar at Ny-Ålesund, Norway (79°N, 12°E) in winter of 1994/95. Enhancement of scattering ratio and depolarization ratio occurred in the middle of December and continued until the end of January. In early and mid-January 1995, the scattering ratio was extremely enhanced at about 20 km height.
Fig. 5. Changes in atmospheric temperature over Ny-Ålesund, Norway (79°N, 12°E) in winter of 1994/95. Launchings of balloon-borne particle counter are shown with arrows.

Fig. 6. Vertical profiles of scattering ratio (R; solid line), depolarization ratio (δ; broken line), and Angstrom exponent (α; dotted line) observed with a lidar, simultaneously observed with a balloon-borne particle counter. Sub-layers of PSCs are shown by arrows (see text). a: 13:04-14:03 UT (thick line) and 14:06-15:05 UT (thin line) on December 18, 1994. b: 10:49-11:36 UT (thick line) and 11:14-13:06 UT (thin line) on January 17, 1995.
tion and/or mixing ratio of January 17, 1995 (Fig. 2), we identified the layer showing good correspondence to peaks of scattering ratio or depolarization ratio (Fig. 6). The sub-layer B shown in Fig. 2a has high scattering ratio (about 2.0) and low depolarization ratio (less than 0.4 %), suggesting that this layer also is composed of liquid PSCs particles like the sub-layer B of December 18, 1994. On the other hand, the sub-layers of A and C of January 17 showed existence of particles with \( r > 1.8 \mu m \), even though such large particles are not normally found in the stratosphere. Corresponded sub-layers observed with the lidar (Fig. 6b) show a very small value in scattering ratio (1.2–1.3), and the value of \( \delta \) showed noticeable peak (larger than 1.0 %), suggesting that those sub-layers were composed of solid state particles.

In Fig. 7 the size-number concentration pattern and integrated particle concentrations of the sub-layers of PSCs are compared. A noticeable difference was found between particle size-number distributions of December 18, 1994 and those of January 17, 1995. For example, concentration of super micron particles larger than 1.8 \( \mu m \) of the sub-layers of December 18, 1994 were a little larger than that of January 17, 1995. As described previously, the mixing ratio of aerosols observed on 18 December, 1994 was also larger than that observed on 17 January, 1995. One possible interpretation for depletion of large particle concentration and mixing ratio is the descending motion of the particles with large size. As shown in Fig. 7, all of the sub-layers included super micron particles larger than 1.8 \( \mu m \) in radius. The descending speed of the particles with radius of about 2 \( \mu m \) is expected to be about 1.0 mm/sec at about 20 km (about 1 km descent in half month), and they are expected to contain lots of HNO3 and/or H2O. Then, the particles formed early in PSCs events may descend from the main PSCs layer during the PSCs events, transporting HNO3 and/or H2O. Therefore, it is reasonable to consider that the densities of HNO3, H2O, and others can be largely disturbed through descent of particulate matter from (or on) which those vapors evaporate (or condense) during the particle descending motion.

In the patterns of particle size distribution of sub-layers (Fig. 7), liquid phase sub-layers of PSCs (curves of 12/18 B and 1/17 B) show an interesting feature. In those two liquid phase sub-layers, particle concentrations were larger than those of background sulfate aerosol not only in sub-micron size,
but also in super micron size. In addition, the distribution pattern of 12/18 B shows a larger concentration of size range from 0.3 to 1.8 μm in radius, comparing to that of 1/17 B. Shibata et al. (1997) suggest that PSCs sometimes contained large-size droplets (type X PSCs), although the exact size of particles cannot be deduced from lidar measurements. Sub-layers observed with a lidar (Fig. 6) corresponding to sub-layers 12/18 B and 1/17 B observed with balloon-borne measurements are classified into type X (α: less than 0.5) and type Ib (α: larger than 0.5), respectively. Then, it is suggested that curve 12/18 B shows one example of size distribution of type X PSCs and curve 1/17 B that of type Ib.

In all of solid state sub-layers, sub-layers A of December 18 and A and C of January 17, had the size distribution showing a high concentration of particles larger than 0.4 μm in radius. In addition, they show typical bimodal patterns, running parallel to the horizontal axis at radii larger than 0.4 μm. This shows an existence of large particles of 1.8 μm in radius, and lack of sub-micron particles ranging from 0.4 to 1.0 μm in radius. On the other hand, the concentration of supermicron particles (r > 1.8 μm) in sub-layers observed on January 17, 1995 was less than that on December 18, 1994 (Fig. 7), even though the temperature on January 17 was lower than that on December 17 (Fig. 3). The present measurements of size distribution of solid state PSCs sub-layers suggest that supermicron particles composing early step PSCs had already fallen by January 17, by descending motion of NAT or ice particles, as described above.

In the present study, we measured particle concentration only at a size range from 0.15 μm to 1.8 μm. It is necessary to know the density distributions of the particles with radius smaller than those in order to better understand PSCs formation. In addition, it is necessary to know larger particle concentration in order to understand transport of constituents by PSCs descending motion. Information of temporal variation of PSCs size distribution also is necessary, and therefore balloon sounding should occur quite frequently.

4. Summary

Balloon-borne measurements of stratospheric aerosol size, made in winter of 1994/95 at Ny-Ålesund, showed the following features of PSCs;

i) In mid-December, 1994 polar stratospheric clouds appeared corresponding to a decrease in stratospheric temperature.

ii) From the measurements of vertical profiles of size-number concentration, the enhanced aerosol layer under the cold atmospheric temperature showed multi-layer structure, and those layers corresponded well to the peaks of scattering ratio and/or depolarization ratio of particular matter observed simultaneously with a lidar.

iii) The number-size distribution patterns of PSCs measured in December 18, 1994 noticeably differed from those of January 17, 1995, and suggested that PSCs particle growth rate and/or production rate were largely disturbed during the PSCs event, possibly due to decrease (or increase) of gases relating with PSCs.

iv) The number-size distribution pattern of liquid sub-layer of PSCs showed enhancement of concentration of particles with in sub-micron range. Concerning super-micron range particles, large enhancement was found in December 18, 1994 and small enhancement in January 17, 1995. Those enhancement in super-micron particles basically agreed with the suggestion that large size droplets were detected with a lidar (Shibata et al., 1997).

v) The number-size distribution pattern of solid state PSCs showed enhancement in supermicron particles and therefore showed good agreement with the suggestion that crystalized type I PSCs had relatively large size (Browell et al., 1990). However, at the end of January, redistribution of nitric acid seemed to occur through the descending motion of NAT or ice particles during PSCs events.

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### Japanese Text

液滴極成層雲粒子の粒径と数濃度：ノルウェー領ニーダーセンにおける

1994/95 冬季の気球観測

林 敦彦・岩坂泰信・渡辺征春・柴田 隆

（名古屋大学太陽地球環境研究所）

藤原卓夫

（福岡大学理学部）

足立 宏・酒井 哲・長谷正博

（名古屋大学太陽地球環境研究所）

H. Gernandt・R. Neuber

（Alfred Wegener Institute for Polar and Marine Research, Potsdam D-14401）

土屋正義

（湘南工科大学）

気球搭載型のバーティクルカウンターを用いて、1994 年から 1995 年にかけての冬の期間、ノルウェーのニーダーセン（北緯 79 度、東経 12 度）において、極成層雲のサイズ（半径 0.15 ～ 1.8 μm）と数濃度を観測した。観測結果は、寒冷な冬の期間 PSCs が活発に生成されていることを示していた。気球観測期間中、放球場所の近くでライダーによって PSCs の相（液相か固相か）を観測した。双方の観測から PSCs がいくつかの層（サプレイヤー）から成っていることが示された。

1994 年 12 月 18 日（PSCs 形成の初期）に観測された数ーサイズ分布は、活発な PSCs 活動を経た後の 1995 年 1 月 17 日のものとは異なっており、PSCs の活動が極成層雲の化学組成、特に PSCs に関係のある組成、の分布を乱している可能性を示している。ライダー観測からは、固相の PSCs に加えて液相の PSCs が発生していることが考えられる。固相の PSCs の数ーサイズ分布は、1.8 μm 以上の粒径に粒子濃度の増大がみられた。一方で、液相の PSCs の数ーサイズ分布は、1.8 μm 以上の粒径に加え、より小さいサイズ (0.25 ～ 0.6 μm) にも増大が見られた。