

A - 3.1.2 Modeling of Particle Formation and Loss

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Abstract Numerical model suggested that Type I PSCs (Polar Stratospheric Clouds) which formed in the upper part of PSCs layer descended to the troposphere within about 5~6 days, but the particles forming in the mid- and low PSCs layer evaporate during their descending motion. According to the radiative model including effect of spherical atmosphere and PSCs scattering, scattering effect of PSCs particles can activate photo dissociation of Ozone and other minor constituents in the winter polar stratosphere.

Key Words Polar Stratospheric Ozone, Polar Stratospheric Clouds (PSCs), Denitrification of the Stratosphere, Scattering Effect of PSCs

1. Introduction

Formation of Polar Stratospheric Clouds (PSCs) largely disturbs ozone chemistry through chemical reactions on the surface of PSCs and denitrification of the polar stratosphere through condensation of nitric acid vapor on PSCs. Surface reactions dissociate inert chlorine compounds and convert them to active chlorine, and ozone destruction by ClO_x catalytic reactions are extremely activated. Growth of PSCs through condensation of nitric acid vapor can give rise to denitrified atmospheric condition under which atmospheric ozone is easily destroyed by ClO_x since formation of ClONO₂ is depressed. There is the possibility that the grown up PSCs descend to the troposphere and it activates denitrification of the polar stratosphere. Here particle behavior in the polar stratosphere is discussed using numerical model.

The scattering effect of PSCs has been neglected since in the polar winter stratosphere there is no effective solar radiation. But this is based on the plane parallel atmosphere model and the contribution of effect of the spherical atmosphere has not been considered.

2. Detection of Nitrate in the Aerosol Materials Collected in the Polar Stratosphere

Electron microscopic observation of individual particles collected in the winter Arctic stratosphere (Sweden, Kiruna, January 31, 1990) with a balloon borne impactor showed that the layer of particles containing NO₃⁻ ion (possibly Type-I PSCs: nitric acid trihydrate particles) expanded on the background sulfate aerosol layer.

As shown in Fig. 1, two different type particles were identified in nitrate particles collected on nitron surface. One is the particles having the structure like disk in the center (type A nitrate) and another does not have it (type B nitrate). From only this measurements, it is hardly possible to give exact physical meaning, but we can speculate that the particle showing disk structure (type A nitrate) may be liquid phase particle and spread like disk on film surface when the particle impacted on nitron film, or be the structure of the NAT particles rarely growing through selective nucleation of Type-I PSCs.

The formation of liquid phase PSCs was suggested from the measurements of polarization property of PSCs. Lidar measurements also showed enhancement of particulate matter with low depolarization ratio in the early stage of PSCs development.

The observation suggested the possibility of Type I PSCs, however it is impossible to discuss motion of the particles from the observations.

3. Condensation Growth of PSCs

PSCs particles grow (or shrink) through condensation (or evaporation) of nitric acid vapor but contribution of coagulation with other particles to the growth of PSCs particles is neglected since the growth rate of coagulation is extremely smaller than the rate of condensation of HNO₃ and H₂O vapor. PSCs particles start to actively move to the lower region through gravitational sedimentation when particles grow to several μ m. If the particle descend to the region having higher (or lower) mixing ratio of H₂O and HNO₃ vapors, condensation growth (or evaporation decay) rate can increase (or decrease).

Condensation grow rate is given by following relations assuming spherical particle;

$$dn/dt = 4\pi r^2 (C_i - C_s) V X$$

where

$$X = [4/\alpha + r^2 V / \{ (r + \Delta) D \}]^{-1}$$

$$\Delta = 1 \text{ for } r < 1, \quad 2l/3 \text{ for } r > 1$$

n : number of molecules in a particle

r : radius of particle

α : sticking coefficient of vapor

l : mean free path of vapor molecule

V : mean speed of vapor molecule

D : diffusion coefficient

C_s : concentration of vapor on particle surface

C_i : concentration of vapor in the surrounding atmosphere

Temperature, concentration of water vapor, and concentration of nitric acid vapor used here are shown in Fig. 2. In Fig. 3, trajectory of test particles in the polar stratosphere is shown.

From the results shown in Fig. 3, three stages of growth are suggested—in early stage particles keep their initial altitudes, in the second stage the settling speed of particles becomes larger and particles start active descending motion, and the third stage particles descend to the evaporation region. In the third stage, the particles having large descending speed can across the evaporating region in short time, can reach the tropopause.

Most of particles which start to grow in the mid- and low-PSCs region shrink during their descending motion.

Computer simulation suggests that solar radiation can reach to the lower stratosphere in winter through scattering effect of polar stratospheric clouds and of the non-plane parallel atmosphere. Photo dissociation of stratospheric minor constituents may cause ozone destruction even in winter season when photochemical processes and ozone loss due to photochemistry has not been expected.

Many observations established that the ozone decline occurs largely during spring and the effect of solar radiation to the ozone hole chemistry in the spring atmosphere was frequently discussed. However, the contribution of solar radiation to the chemistry in polar winter season has been neglected since it is speculated that solar radiation does not reach the lower stratosphere on the basis of the plane parallel atmosphere model.

Here, we estimate solar radiation in the winter Antarctic stratosphere with a numerical model including the effect of solar radiation scattering by PSCs and the contribution of scattering in

the non-plane parallel atmosphere, and discuss possible photodissociation reactions in the winter polar stratosphere.

4. Polar Stratospheric Clouds (PSCs) and Radiative Transfer in the Polar Winter Stratosphere

The size distribution function of PSCs particles and background aerosol particles is given according to balloon measurements at Antarctica by Hoffman et al. [1989],

$$n(r)dr = (N_0 / 2\pi) \exp(-\alpha^2/2) d\alpha$$

$$\alpha = [\ln(r/r_0)] / [\ln(\sigma)]$$

where N_0 is the total number density of particles per unit volume (cm^{-3}), $n(r)$ the differential particle number density ($\text{cm}^{-3} \mu\text{m}^{-1}$), r_0 the characteristic size (μm), and $\sigma = 1.20$ for PSCs and 2.50 for background aerosols.

In the present calculation, two different PSCs vertical profiles are examined to discuss the effect of height variation to the scattering of solar radiation.

The difference of radiation estimated on the basis of the plane-parallel atmosphere model and spherical atmosphere model is shown in Fig. 4.

The photo dissociation rate of O_3 in the atmosphere containing PSCs is estimated on the basis of spherical atmosphere model, and the value of $[J_{\text{psc}} - J] / J \times 100$ is shown in Fig. 5, where J_{psc} is photodissociation rate in the atmosphere containing PSCs and J in the PSCs free atmosphere.

Apparently, in the upper PSCs layer photo dissociation rate of ozone increases due to the scattering effect of PSCs. Considering that the ozone layer peak is frequently observed in 15~25km, the effect of the enhancement of photo dissociation of ozone seems to disturb ozone chemistry in the winter polar region.

5. Conclusion

The descending loss of type I PSCs from the stratosphere to the troposphere is demonstrated by numerical model, and this process is an important process as denitrification of the polar stratosphere in winter season. The scattering effect of PSCs is revealed by radiative transfer model including the effect of spherical atmosphere. The scattering effect of solar radiation by PSCs may activate ozone loss in winter season.



Fig. 1 Electronmicrogram of particles collected on nitron thin film in the winter polar stratosphere.

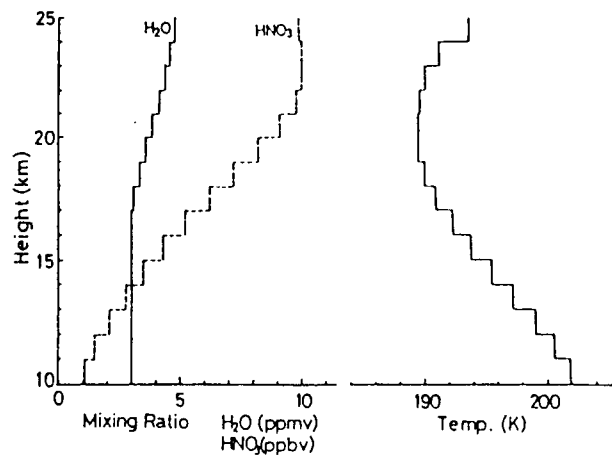


Fig. 2 Temperature, water vapor mixing ratio, and nitric acid vapor mixing ratio used for numerical experiment (see text).

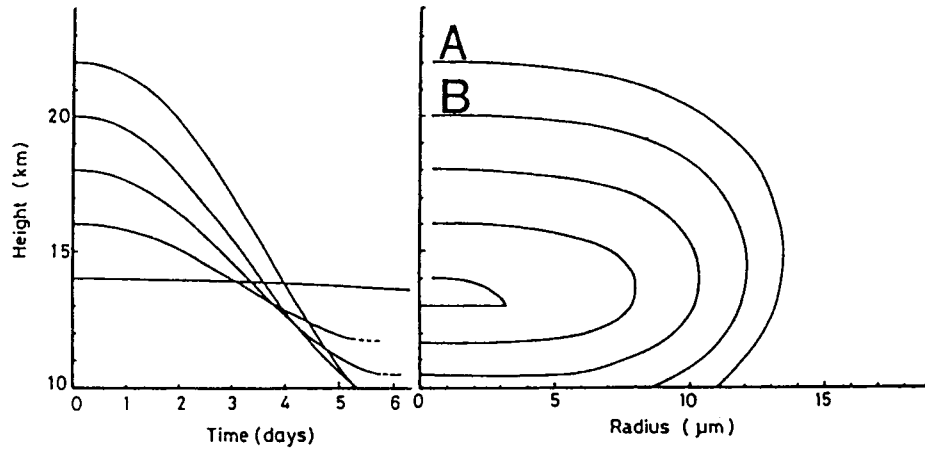


Fig. 3 Trajectories of test particles of NAT on height-time plane (right) and height-radius plane (left).

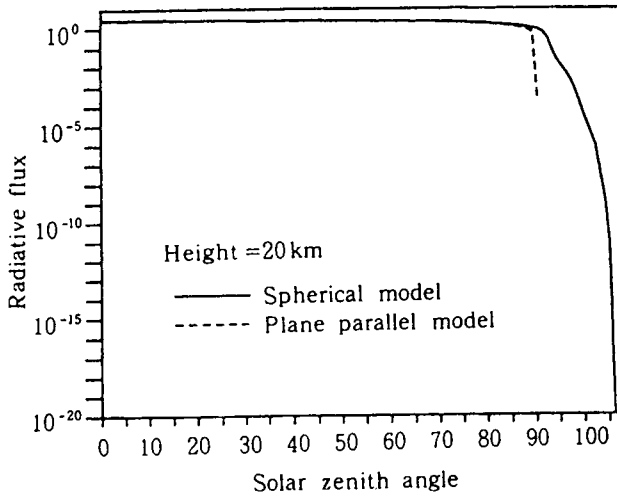


Fig. 4 Actinic flux (direct + diffuse) in wavelength of 410nm and altitude of 20km for all solar zenith angles. Solid line is the result for spherical model and the dot line is that for plane parallel model.

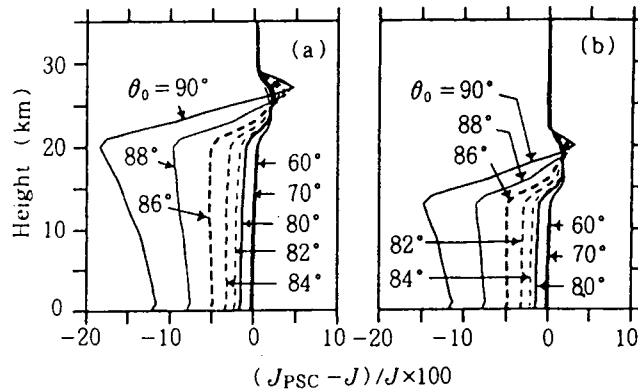


Fig. 5 The percent difference (as defined in Fig. 8) in the photodissociation rate of O_3 Chappius bands of some solar zenith angles. (a) the first stage. (b) the second stage.