Descending Motion of Particle and its Effect on Ozone Hole Chemistry

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Abstract

The descending motion of particles which grow up to a few micrometer under the cold winter Antarctic stratosphere is also important process to accelerate the effect of heterogeneous reactions on ozone hole formations.

1. Introduction

In the winter Antarctic stratosphere, a lot of particulate matter was formed under very cold temperature which decreases to lower than -85 °C at Syowa Station (69° 00'S, 39° 35'E) (e.g., Iwasaka et al., 1985; Iwasaka, 1986). Some investigators suggested that the surface of particulate matter could act as an effective chemical reaction site for ozone-destruction chemical reaction series (e.g., Solomon et al., 1985). It was suggested that the excellent correlation of total ozone content with 100 mb surface temperature during Antarctic spring could be explained if the effect of the heterogeneous chemical reactions including the aerosols was taken into consideration (e.g., Iwasaka and Kondoh, 1987).

Most of previous investigations describing the effect of the particles on chemical reactions payed their attention to the surface area of particles or chemical composition of particles but not the effect of particle descending motion.

The possibility of particle descending in the winter Antarctic stratosphere was suggested on the basis of eye-observation of stratospheric clouds (Stanford, 1973) and lidar measurements (Iwasaka, 1986). McCormick et al. (1985) suggested that the descending motion of the Antarctic aerosol layer observed by SAM II could be associated with a downward air motion. Iwasaka (1986) suggested that the descending motion of the layer was capable when particles grew to a few micron size aerosols. The particle descending motion is important process controlling the redistribution of the chemical species relating 'ozone hole'.

Combination of particle growth, evaporation of particle, chemical adsorption, and particle settling can play as sink or source of chemical species, and it strongly depends on a stratospheric temperature distribution.
Fig. 1. The shaded area means the observed temperature range at Syowa Station (69° S, 40° E) in July 1983 (a) and in September (b). Curve a and b are frost point temperature of pure water. Curve c and d are frost point of HNO₃-H₂O (50% weight) crystal.
2. Particle Growth

Following points are essential for heterogeneous reactions controlling 'Ozone Hole':

1) What kind of gases are used to produce the particulate matter?
2) What kind of reactions can take place on the surface of particles?
   and What is surface area?
3) Is there the possibility of particle descending motion?

Steele et al. (1983) and Iwasaka (1986) discussed growth of ice crystal particle from pre-existing sulfuric acid droplet. In their model, the main gas used to increase particulate matter was water vapor. However, the lidar measurements additionally showed meaningful time delay between particulate content increase and depolarization ratio increase (Iwasaka, 1986). Toon et al. (1986), and Crutzen and Arnold (1986) suggested the possibility that nitric acid vapor condense to solid state HNO₃·3H₂O particles. According to Crutzen and Arnold (1986) the condensation can start at higher temperature (205 + 5° K) than the frost point of water vapor at about 100mb.

Importance of the point 1) is easily understood comparing the amount of gas used to form particles with the remaining gas content. For water vapor, about 10% to 40% of water would be in solid (or liquid) state during the most developed phase of PSC's event. For HNO₃, the ratio of the amount of particulate matter phase will be extremely larger than the case of water vapor. Crutzen and Arnold (1986) emphasized that the depletion of HNO₃ allowed a rapid rise of hydroxyl radical concentrations as the cycle of reactions

\[ \text{OH} + \text{NO}_2 \rightarrow \text{HNO}_3 \quad \text{and} \quad \text{HNO}_3 + \text{OH} \rightarrow \text{H}_2\text{O} + \text{NO}_2 \]

do no longer operate.

The potential influence of surface reactions depends on the surface area of particles.

In Fig. 1, temperature distribution measured in winter at Syowa Station is compared with the frost point temperatures of water and mixture of HNO₃·H₂O (50% weight mixture). These curves suggest that particle production is active near 15km for water vapor, and above about 15km for HNO₃ particles in mid winter.

3. Descending motion of particles

The descending motion of aerosol layer was observed during winter by a lidar at Syowa Station. The particle size would be about a few micrometer or larger than it if the descending motion was due to a gravitational sedimentation (Iwasaka, 1986). As shown in Fig. 1, the condition of super saturation was not always satisfied for pure water vapor or nitric acid vapor even in mid-winter if the density profiles in mid-latitudes is assumed. Therefore the particle which settles to the region of \( P < P_s \), where \( P \) and \( P_s \) are partial pressure of water vapor (or nitric acid vapor) and saturation
pressure or the vapor respectively, evaporates the gases condensed in the particles there.

Iwasaka and Kondoh (1987) showed that ozone depletion rate was largest near 15 km and the second peak was near 10 km and lower than it. The heights of these active ozone loss region do not correspond to the region where particle production rate is large and apparently is lower. In Fig. 2, schematic picture showing the particle sedimentation effect is given. The descending motion of particles to the region where evaporation rate is high seems to accelerate the production rate of Cl₂ and ClO₂H near the tropopause.

4. Summary

The particle descending motion is one possible process which causes ozone loss near the tropopause in Antarctic spring. However, particle size distribution has not been measured yet. The particle settle is an important redistribution process of chemical constituents contained in particles. To understand the particle settle effects on 'Ozone Hole', information on the size distribution and the chemical composition of particles are desired.

Fig. 2. The particles grow at the region where large super saturation occurs in winter and descend to the region where evaporation from particles is active in spring.