# STRATOSPHERIC "CIST" AND WATER VAPOR BUDGET IN THE STRATOSPHERE

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*Abstract*: The growth condition of ice crystal particles in the winter season of the polar stratosphere is discussed. Numerical calculation supports the idea that the formation of ice crystal particles in the polar stratosphere (stratospheric "Cist" cloud particles) can affect the stratospheric water vapor budget on a global scale.

# 1. Introduction

In this paper, we should like to describe the ice crystal particle growing processes in the polar stratosphere on the basis of numerical calculations, and discuss the effect of so-called "freezing-out process" on the water vapor budget of the stratosphere. That the Antarctic winter stratosphere may serve as a sink for stratospheric water vapor is suggested by the following evidence: (1) the lowest naturally occurring temperatures on Earth have been observed there, (2) persistent clouds (termed stratospheric "cirrostratus", stratospheric "Cist") were observed there from June or July to the beginning of October in 1950 and 1951 by LILJEQUIST (1956), and (3) available observations of stratospheric humidity show a north-to-south gradient (MCKINNON and MEREWOOD, 1970).

There are a number of reasons for believing that the Antarctic winter stratosphere should be colder than that of Arctic region, and thus the temperatures are more likely to reach below  $-80^{\circ}$ C required to create a sink for stratospheric water vapor.

Recently, STANFORD (1973, 1977) has presented the results of careful analysis of the original Maudheim radio sonde data, together with a comparison of the Maudheim cloud logs, and proposed that the Antarctic winter stratosphere provides a frost-trap sink for stratospheric water vapor.

## 2. Growing of Ice Crystal Particles in the Antarctic Winter Stratosphere

A necessary condition for the growth of ice crystal particles is that the water vapor pressure must exceed the local saturation.

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The saturation vapor pressure over plane surface  $p_i$  can be found from thermodynamic considerations, and is given by the Clausis-Clapeyron equation,

$$\mathrm{d}p_i/p_i = \mathrm{d}T/T^2[L_v m_w/k], \qquad (1)$$

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where

 $L_{v}$ : the latent heat of vaporization,

 $m_w$ : the mass of a water molecule,

k: Boltzmann's constant,

T: temperature.

Integrating eq. (1), we find

$$T_{s} = T_{0} \ln(p_{i}/p_{i0}) [1 - kT_{0}/(L_{v}m_{w})], \qquad (2)$$

where

 $p_{i0}$ : the known saturation vapor pressure at temperature  $T_0$ ,

 $T_s$ : the frost point temperature.

In Fig. 1, we show the vertical profile of the frost point temperature calculated from eq. (2) for water vapor mixing ratios of 1, 5, 10 and 20 ppmV. The dotted curves in the figure are atmospheric temperatures. Obviously, supersaturation can occur with reasonable values of water vapor mixing ratio over a region of a few kilometers in depth near the mesopause or lower stratosphere.



Fig. 1. Frost point temperatures for the water vapor mixing ratio = 1, 5, 10 and 20 ppmV. Dotted curve  $T_a$  is the atmospheric temperature profile measured by THEON et al. (1967), and curve  $T_b$  is the temperature profile of the Antarctic winter stratosphere estimated from various meteorological observations.

Although the occurrence of supersaturation is necessary for the growth of ice crystal particles, it is not a sufficient condition. The spontaneous growth of particles in the absence of any condensation or sublimation nuclei is known as a homogeneous nucleation, and it generally requires a high dgree of supersaturation. The nucleation

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process is, however, greatly helped by the presence of small particles on whose surfaces the water molecules can collect in a protected environment. And it is reasonable to consider that a homogeneous nucleation is very unlikely to occur and that small-size aerosol particles exist in sufficient concentrations to act as nuclei.

The rate at which molecules strike particles per unit area of the surface is given by,

$$\alpha p / \sqrt{2\pi m_w kT} , \qquad (3)$$

where

p: the partial pressure of the water vapor,

 $\alpha$ : the probability of a water molecule adhering to the surface.

According to the definition of the saturation point, the rate at which water molecules are lost by evaporation is equal to the rate at which they are added under a saturation condition. And the net rate of gain of water molecules under all conditions is given by,

$$P_w = \alpha (p - p_i) / \sqrt{2\pi m_w kT}$$
 (4)

Thus, the growth rate is given by the following form assuming that the particles are spherical in shape,

$$dR/dt = (\alpha/\rho) \sqrt{m_w(2\pi kT)(p-p_i)}, \qquad (5)$$

where

R: the particle radius,

 $\rho$ : the density of particle.

The terminal speed of particle is given by,

$$v = dZ/dt$$
  
=  $\rho g R \sqrt{\pi/(2m_a kT)/2N_a}$ , (6)

where

g: the acceleration due to gravity,

 $N_a$ : the concentration of air molecules,

 $m_a$ : the mass of air molecules.

From eqs. (5) and (6), we can solve R directly as a function of Z in the following manner,

$$dR/dZ = [dR/dt]/[dZ/dt].$$
(7)

In Fig. 2, examples of the calculation of growing rate are shown for the temperature profile of T(z)=213-(4/3)Z for 8 km < Z < 35 km. The value of dR/dt becomes negative (evaporation of particle) under the low mixing ratio (1 ppmV), but the positive growth rate is found for the mixing ratio of 5 ppmV. In Fig. 3, particle trajectory determined by eq. (7) is presented. Curves suggest that the particle having the initial size of 0.1  $\mu$ m radius can grow to the large-size particle which can conduct "freezout" from the stratosphere to the troposphere under the condition of water vapor

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Fig. 2. Growth rate of ice particles for the condition of water vapor mixing ratio=1, 5 and 10 ppmV. Plus and minus signs mean condensation of water vapor molecules onto the ice crystal particle surface and evaporation from the particle, respectively. Calculation is made for the temperature distribution T(Z)=213-(4/3)Z, Z in unit of km.

mixing ratio=5 ppmV. For a colder condition, precipitation of ice crystals can take place even in the drier atmosphere.

### 3. Source and Sink of Stratospheric Water Vapor

The mechanism which is controlling the stratospheric water vapor budget has not been made clear, and various possibilities have been presented, such as 1) oxidation of solar proton, 2) stratospheric oxidation of  $CH_4$  transported from the troposphere, 3) thunderstorm penetrations into the tropopause, 4) meridional circulation about the westerly jet streams, upward diffusion, so-called "freez-out" in the Antarctic stratosphere, and so on. No data have been produced to unequivocally support any of these sources as a significant mechanism which controlls the water vapor



Fig. 3. Trajectory of one particle having initial size of 0.1 µm on R-Z plane.

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budget of the stratosphere on a global scale. Some investigators suggest that the most important sink and source of the stratospheric water vapor are due to the Hadley cell type transportation through the tropical tropopause (*e.g.* NEWELL *et al.*, 1969; ELLSAESSER, 1974). Typical values due to the Hadley cell transportation are listed in Table 1. The oxidation of CH<sub>4</sub> transported from the troposphere has been also emphasized as an important source of the stratospheric water vapor (*e.g.* NICOLET, 1972, 1975). Some estimations are compared with the Hadley cell type source or sink in Table 1. The calculations presented here suggest that "freez-out" process can occur under the condition of water vapor mixing ratio=5 ppmV (if temperature can decrease more than the values used here, "freez-out" can occur under the drier atmospheric condition), and that the particle size is the order of 1  $\mu$ m. The downward flux can be roughly estimated by using reasonable values, namely the density of particle is 5/cm<sup>3</sup> and average size 2  $\mu$ m,

flux of water =  $5.02 \times 10^{-11} \text{ g/cm}^2/\text{s}$ 

and this corresponding to the flux having the unit of  $(H_2O)$  molecules/cm<sup>2</sup>/s,

flux of water molecules =  $1.68 \times 10^{12}$  (H<sub>2</sub>O)molecules/cm<sup>2</sup>/s.

Considering the horizontal scale= $1-2 \times 10^7$  km<sup>2</sup>, and the duration time=1-3 months of the "freez-out" phenomenon, the global average flux comes to be

flux of water due to "freez-out" =  $1.3 \times 10^7$  tons/year -  $7.8 \times 10^7$  tons/year.

Most of the values listed in Table 1 have large uncertainty, but, the "freez-out" in

Oxidation of CH₄		
Water vapor influx estimated by the CH <sub>4</sub> influx	CH₄ influx	
$0.48 \times 10^8$ tons/year	$5 \times 10^9$ molec/cm <sup>2</sup> /s	NICOLET and PEETERMANS, 1973
$0.58 - 1.8 \times 10^8$ tons/year	$6-19 \times 19^9$ molec/cm <sup>2</sup> /s	Ehhalt, 1978
Hadley cell injection		
$3.28 \times 10^8$ tons/year		Ellsaesser's estimation using the date by Newell et al. (1969)
5.76 $\times 10^8$ tons/year		Ellsaeseer's estimation using the by SISSEWINE et al. (1972)
11.3 $\times 10^8$ tons/year		Ellsaesser (1974)
Hadley cell return out from strato- sphere to troposphere		
8.3 $\times 19^8$ tons/year		Ellsaesser (1974)
Freez-out in Antarctic stratosphere		
$0.17-0.78 \times 10^8$ tons/year		Present estimation

Table 1. Source and sink of stratospheric water vapor.

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the Antarctic stratosphere possibly affect the glogal budget of the stratospheric water vapor.

### 4. Summary

The careful analysis of cloud observations by STANFORD (1973, 1977) reveals that the "Stratospheric Cist" was most probably ice clouds lying in the lower stratosphere. The numerical results presented here and other investigations suggest the possibility that the "Stratospheric Cist" affects the stratospheric water budget. Recently satellite measurements of SAGE (Stratospheric Aerosol and Gas Experiment) caught the region of highly concentrated particulate matter in the high-latitude stratosphere which was speculated to be due to "Stratospheric Cist" (private communications, FUJIWARA, 1980).

The polar stratosphere has been considered to be an active sink of stratospheric aerosols, and some investigators pointed out the fact that the stratospheric aerosols were possibly transported into the troposphere on the basis of the chemical analysis of the aerosol particles sampled in the south polar region (CADLE *et al.*, 1968; HOGAN, 1975; MAENHAUT *et al.*, 1979). Thus, the effect of the stratospheric aerosol particles on the behavior of "Stratospheric Cist" presents interesting problems.

Unfortunately, available data on "Stratospheric Cist" are very limited, and it is impossible to reach a definite conclusion about "Stratospheric Cist". More detailed observations are required in order to make clear the behavior of "Stratospheric Cist" and related problems.

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