

氏名	當房 豊
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学位授与の題目	Microphysics of Aerosols and Clouds in the Upper Troposphere and Lower Stratosphere within the Tibetan Anticyclone (チベット高気圧内における上部対流圏・下部成層圏のエアロゾル・雲の微物理)
論文審査委員(主査)	岩坂 泰信(フロンティアサイエンス機構・特任教授)
論文審査委員(副査)	早川 和一(自然科学研究科・教授), 柏谷 建二(環日本海域環境研究センター・教授), 古内 正美(自然科学研究科・准教授), 張 代洲(熊本県立大学・准教授)

## Abstract

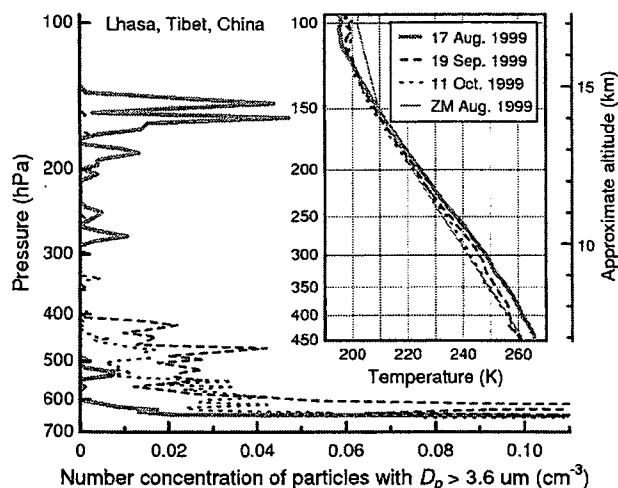
Vertical profiles of size-resolved particle concentrations were measured over the Tibetan Plateau by using a balloon-borne optical particle counter. During the observing period, an upper-tropospheric anticyclone (i.e., Tibetan anticyclone) linked to deep convection was formed over the Tibetan Plateau. The measurements on 17 August 1999 showed that there was a thin layer of particles with diameters of  $>3.6 \mu\text{m}$  at concentrations of  $>0.01 \text{ cm}^{-3}$  around 150 hPa. These particles are most likely composed of ice crystals. Within the layer, a large amount of submicron aerosols with diameters of  $0.3\text{-}1.2 \mu\text{m}$  still coexisted with the ice, indicating that not all of the background aerosols were removed by ice nucleation and scavenging. These results suggest an occurrence of selective ice nucleation involving a fraction of the background aerosols (i.e., effective ice nuclei) in the upper troposphere. Other features of the measurements were high number concentrations ( $0.7\text{-}0.8 \text{ cm}^{-3}$ ) of submicron aerosols between about 130 and 70 hPa. In regard to this result, direct convective transport of lower-tropospheric aerosols might affect atmospheric constituents near the tropopause. In addition, adiabatic cooling and hydration associated with deep convection might trigger enlargement of haze particles (e.g.,  $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  droplets) near the tropopause. These results should be considered in assessing the global/regional climate system and the geochemical cycle.

## 1. Introduction

The Tibetan Plateau, called “the roof of the world”, has an average elevation of  $>4000$  meters and occupies an area of about 2.5 million square kilometers in Asia. During the Asian summer monsoon period, thermal contrast between the Tibetan Plateau and its adjacent oceans becomes predominant due to elevated surface heating over the Tibetan Plateau, leading to deep convection. It has been suggested that the coupling of the Tibetan anticyclone and deep convection strongly influences on constituent behavior in the upper troposphere and lower stratosphere. Thus, deep convection is regarded as a major pathway for upward material transport from the

troposphere into the stratosphere.

Intensive measurements had been carried out in August-October 1999 at Lhasa (29.7°N, 91.1°E, 3650 m a.s.l.), located in the southern part of the Tibetan Plateau. This study addresses how aerosol and cloud particles in the upper troposphere/lower stratosphere are characterized under the influence of the Asian summer monsoon activities over the Tibetan Plateau.



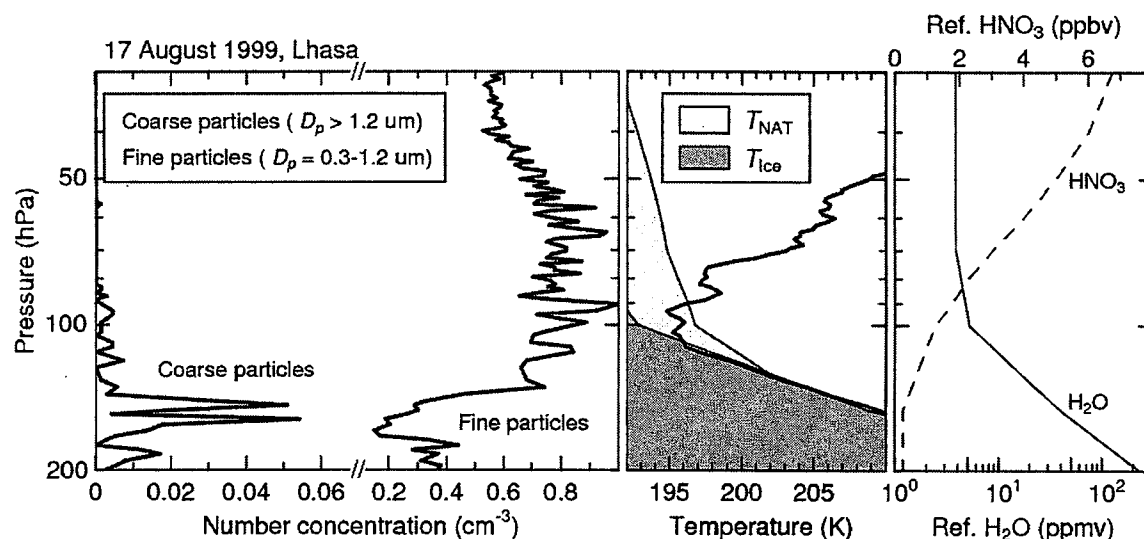
**Figure 1.** Vertical profiles of number concentrations of particles with  $D_p > 3.6 \mu\text{m}$  and air temperatures at Lhasa on 17 August, 19 September, and 11 October 1999. Also shown are the ECMWF analyzed zonal mean temperatures in August 1999.

## 2. Cirrus Clouds in the Upper Troposphere

Figure 1 shows the vertical profiles of number concentrations of particles with  $D_p > 3.6 \mu\text{m}$ , together with those of ambient temperatures, on 17 August, 19 September, and 11 October 1999. The 17 August flight (within the Tibetan anticyclone) showed the existence of some distinct layers between about 300 and 130 hPa, in contrast to the other two flights. In particular, relatively high concentrations ( $>0.01 \text{ cm}^{-3}$ ) of the particles were observed at  $\sim 150$  hPa. Such heights are typical of the top of cirrus clouds over the Tibetan Plateau during the Asian summer monsoon period. These particles are much larger than the background aerosols and most likely composed of ice crystals. Within the cirrus layer, a large amount of submicron aerosols with diameters of  $0.3\text{-}1.2 \mu\text{m}$  still coexisted with the ice (Figure 2), indicating that not all of the background aerosols were removed by ice nucleation and scavenging.

Number concentrations of ice crystals in cirrus clouds depend in part on the difference of the ice-nucleation mechanisms. Homogeneous ice nucleation in haze particles usually produces high number concentrations of ice crystals ( $1\text{-}100 \text{ cm}^{-3}$ ). On the other hand, heterogeneous ice nucleation on insoluble or non-resolved solute particles usually form cirrus clouds with small number concentrations of ice crystals ( $<0.1 \text{ cm}^{-3}$ ), corresponding approximately to the ice particle concentrations presented in this study. In addition, recent laboratory experiments with a cloud chamber have suggested that heterogeneous ice nucleation on mineral dust, soot, or solid-phased

ammonium sulfate (these particles are usually originated from lower troposphere) is efficient at temperatures about 210-220 K. These results suggest an occurrence of heterogeneous ice nucleation in the upper troposphere over the Tibetan Plateau.

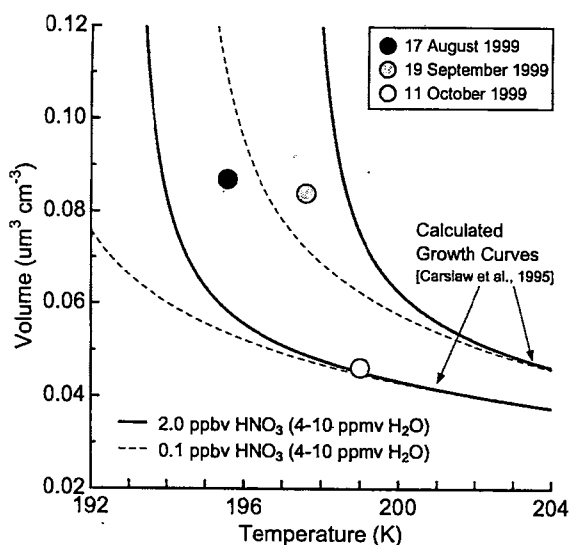


**Figure 2.** Vertical profiles of number concentrations of coarse ( $D_p > 1.2 \mu\text{m}$ ) and fine ( $D_p = 0.3-1.2 \mu\text{m}$ ), and those of temperatures at Lhasa on 17 August 1999. Also shown are the expected frost point of nitric acid trihydrate  $T_{\text{NAT}}$  and water ice  $T_{\text{ice}}$ .

The presence of heterogeneous ice nuclei in cloud-free air usually trigger ice nucleation below the threshold required for homogeneous ice nucleation in haze particles, resulting in cirrus clouds with low ice particle number concentrations, in which supersaturations might be sustained for relatively long periods. Hence, if heterogeneous freezing is true, this process affects strongly on the climate system.

### 3. Submicron Particles in the Tropopause and Lower Stratosphere

The present measurements showed that relatively high number concentrations ( $0.7-0.8 \text{ cm}^{-3}$ ) of aerosols with  $D_p = 0.3-1.2 \mu\text{m}$  were formed between about 130 and 70 hPa (Figure 2). Based on a comprehensive investigation, I found that these high aerosol events were associated with the Tibetan anticyclone; i.e., deep convection that is responsible for formation of the Tibetan anticyclone. There are two possible ways for the monsoon anticyclone to affect the enhancements of high aerosol concentrations. One way is the direct transport of natural and anthropogenic emissions from South Asia. The other way is the enlargement of aqueous solution droplets linked to adiabatic cooling and hydration of the air associated with deep convection. Model calculations (Figure 3) suggested that, if sufficient amounts of a variety of trace gases (e.g.,  $\text{H}_2\text{O}$ ,  $\text{H}_2\text{SO}_4$ , and  $\text{HNO}_3$ ) are present near the tropopause, the enlargement process will become an important process in explaining the present observations.



**Figure 3.** Aerosol volume concentrations from the measurements at Lhasa compared with model calculations for the growth of liquid particles. The dots are the aerosol volume concentrations near the tropopause ( $\sim 105$  hPa). The growth curves are for 0.1-2.0 ppbv total  $\text{HNO}_3$  and 4-10 ppmv total  $\text{H}_2\text{O}$  at 105 hPa altitude, assuming that the observed aerosols on 11 October (outside Tibetan anticyclone) are at equilibrium with their environment for 4.5 ppmv  $\text{H}_2\text{O}$ .

High aerosol concentrations near the tropopause may not only impact on the global/regional climate system and the geochemical cycle, but also lead to efficient heterogeneous chemistry on aerosols. It may be important to gain an understanding of the coupling of the enhanced aerosols to the ozone valley over the Tibetan Plateau. However, there are substantial uncertainties resulting from shortage of information about laboratory experiments, physico-chemical properties of aerosols (e.g., phases, sizes, shapes, chemical compositions, etc.), abundances of trace gases, and the magnitude of the monsoon circulation under the atmospheric conditions discussed here; therefore, these uncertainties merit further comprehensive investigations in future work.

## 学位論文審査結果の要旨

[審査経過] 打ち合わせた審査方針にもとづいて、専門分野の学力・学識を有することを確認し、面接し諮問を行った。2月8日に、口頭発表（最終試験）を行い、その後引き続いて行った<sup>最終</sup>採集審査委員会で審議の結果、以下のような結論を得た。

[審査結果] フロンの大量の放出によってオゾン層の破壊がすすみ、フロンの使用や製造が強く規制された今日においても、破壊された状態から回復せず、人の健康への影響をはじめ様々な生態影響に強い懸念が示されている。そのようなことからチベット上空に出現する低濃度オゾン層が、フロンなどの人為起源物質の影響を受けたものなのか否かについて強い関心が持たれてきた。

申請者は、1999年に行われたエアロゾルの気球観測結果を精査し、夏の期間に発生する強い対流運動によって形成する低温域で、活発な粒子形成が進行していることやそれらのプロセスがオゾン化学にかかわりを持ちうることを示した。これまでの極めて単純化された仮説「低濃度オゾンの大気が下部成層圏まで上昇するため成層圏オゾン濃度が低下する」だけではなく、エアロゾル等が関与した複雑な微物理過程が重なって起きていることを示唆した点で当該分野の発展に大きな貢献をした。

よって、審査委員会は博士（理学）を授与するにふさわしいものと判定した。