Studies on relationship between Asian dust outbreak and the stratosphere-to-troposphere transport in spring with coupled ice-core-meteorology

アイスコア及び気象学的解析を用いた春季黄砂発生と成層圏対流圏輸送の関係についての研究

安成　哲平

Teppei J. Yasunari

Ph.D. Dissertation

Division of Earth System Science

Graduate School of Environmental Science

Hokkaido University

2008

Abstract

The ability to predict climate is becoming increasingly important. The ice core is one of the most useful tools for reconstructing previous atmospheric data to assess past climatic changes. The core data provides direct access to past atmospheric information, as the snow surface is in direct contact with the atmosphere. Therefore, ice cores drilled in mountain glaciers and ice sheets are suitable for reconstructing previous atmospheric data. However, an important limitation of ice core research is that ice-core data is a one-point data, making it difficult to discuss spatial variation of past atmospheric information. In general, the spatial representation of ice cores is unknown. However, we should extract past spatial data of atmospheric materials from ice cores for discussing spatial material circulation. Hence, a detailed comparison of the results of meteorological analysis with ice core data is a very important first step for reading past atmospheric circulation from ice-core data.

This study consists of two main chapters. Chapter 2 covers the North Pacific region from East Asia to the North Pacific Ocean. This region is subject to Asian dust outbreaks and stratosphere-to-troposphere transport (STT) associated with cyclonic activities, which may be important for assessing climate change. Therefore, investigating climatic variations in this region from the past to the present is very important for future climate prediction. Intra-annual ice-core data is useful for an assessment of those variations from the past to the present. However, there have been no ice core data with full seasonal dating and no study focusing on the interaction between the stratosphere and the troposphere by means of ice core research in this region thus far. Hence, the tropospheric and stratospheric materials (dust and tritium, respectively) have been analyzed in an Alaskan ice core and the data obtained are discussed in terms of ice core research. In Chapter 3, in terms of meteorological

analyses, investigations on the interaction between the troposphere and the stratosphere are carried out as case studies of severe dust storms in East Asia in 2001 and 2002 with simultaneous examination of their impacts on the ice core site. Additional investigations on the seasonal march and interannual variation of atmospheric circulation in spring–early-summer (March–June) were carried out to better understand atmospheric circulation patterns, which may have impact on the Alaskan ice-core data and the interaction between the tropospheric and the stratospheric circulation of material in the North Pacific region.

For research (1), to assess past variation of materials of tropospheric and stratospheric origin, a 50-m ice core was drilled at the summit of the Mount Wrangell Volcano, Alaska (62°N; 144°W; 4100 m) and the dust number, tritium concentrations, and stable hydrogen isotope were analyzed. The period covered was from 1992 to 2002. We found that the concentrations of both fine dust (0.52–1.00 μm), an indicator of long-range transport, and coarse dust (1.00–8.00 μm) increased simultaneously every spring. Moreover, their concentrations increased drastically after 2000, corresponding to the recent increase in Asian dust outbreaks in spring. In addition, an increase in the spring of 2001 corresponded to the largest dust storm recorded in East Asia since 1979. Therefore, our findings imply that Asian dust strongly pollutes Mount Wrangell every spring. The stratospheric tracer, tritium, had late spring maxima almost every year and we found it to be useful for ice-core dating for identifying late spring in the North Pacific region. We also found that a high positive interannual correlation existed between the calculated tritium and fine dust fluxes from late spring to summer. We propose that an interannual relationship between the STT and Asian dust storms is most closely connected in late spring because their activities are weak in summer. Therefore, the Mount Wrangell ice core is important and useful for assessing dust and tritium circulation in the distant past around the North Pacific with probable intra-annual time scale information.

For research (2), the investigations on both the Asian dust and the STT and the impacts on the Alaskan ice core site are described in terms of meteorological analyses. Previous results (1) suggested the hypothesis of an interannual connection between Asian dust outbreaks and STT in spring from the Mount Wrangell ice core, Alaska. However, no direct meteorological evidence explaining the results was shown. Hence, we investigated five severe trans-Pacific dust storms in the spring of 2001 and 2002 in East Asia using forward trajectory and meteorological analyses. We also examined the seasonal march of atmospheric circulation in the North Pacific region in spring and its effects on Mount Wrangell. Results showed that severe Asian dust storms with STT (ADSTT) exactly affected the ice-core site in three of five cases. We also found that snowfall at Mount Wrangell is important for explaining the tritium data in the ice core because tritium mainly exists as tritiated water vapor in the atmosphere. When tritium due to ADSTT in the two cases in 2002 was deposited onto the ice-core site, the tritium concentration drastically increased in the ice-core data. Hence, ADSTT may considerably increase tritium concentration in the troposphere. However, if it were not snowing at the ice-core site when the ADSTT tritium was transported to the site, the tritium information would have been lost, as was the case in 2001. When it was snowing at the site, an eddy developing near Alaska was often seen. It implies that the developing eddy caused the snowfall at the core site and Other STT (OTSTT) near Alaska. All ADSTT, OTSTT near Alaska, and snowfall conditions at Mount Wrangell are closely connected to each other because of the atmospheric circulation in the North Pacific region. In fact, eddies in Siberia, the Aleutians, and the Gulf of

Alaska have a high interannual correlation in spring (March–May). As a result, the high interannual connection between fine dust and tritium variations in spring in the ice core can be well explained by this connection among Asian dust storms with STT, other STT in the North Pacific Ocean, and snowfall conditions at the ice core site. In normal years, tritium concentration maxima in the ice core are well explained by the seasonal cycle of Brewer–Dobson circulation in the stratosphere with normal eddy-forced STT. In abnormal years, such as the strong ADSTT outbreaks in 2002, exceptional tritium intrusion into the troposphere in early spring because of strong eddy-forced ADSTT were added to the general tritium cycle where it shifted the tritium maximum from late spring to early spring. Our results of dust and tritium analyses of the ice core are useful for reconstructing the past circulation of the atmospheric material associated with Asian dust, STTs in East Asia and the North Pacific Ocean, and storm track activities.

This Ph.D. study contributes to basic research on future climate and environmental prediction. To predict global warming, it is essential to assess the relationship between Asian dust storms and ADSTT in spring because both the atmospheric dust and one of the stratospheric tracers, ozone, have the capacity to change the global radiation budget in terms of radiative forcing. Therefore, we should actively focus on this area of research involving both fields of study, and more of such kinds of studies need to be conducted in the near future. This will be a very useful research for predicting climate.

Contents

Chapter 1

Introduction

Predicting climate is becoming increasingly important these days, because of some anthropogenic emissions such as CO_2, CH_4, ozone-forming chemicals, which contribute to tropospheric ozone production, sulfate, organic carbon, black carbon, nitrate, and dust, have been drastically affecting the global heat balance since the Industrial Revolution, circa 1750 in *IPCC* [2007]. Atmospheric aerosol and a stratospheric material, ozone, are important materials affecting the atmospheric radiation balance (Fig. 1.1). Therefore, studies that can assess these variations in atmospheric materials are essential for predicting climate.

This study focuses on variation of atmospheric dust and stratospheric-origin-material in the troposphere in the North Pacific region. In the North Pacific region from East Asia to the North Pacific Ocean, the activities of Asian dust storms in spring and storm tracks in late fall and early spring [e.g., *Sun et al.*, 2001; *Nakamura*, 1992] are caused by cyclonic activities. Cyclonic activities are well associated with the stratosphere–troposphere exchange (STE) [e.g., *Holton et al.*,

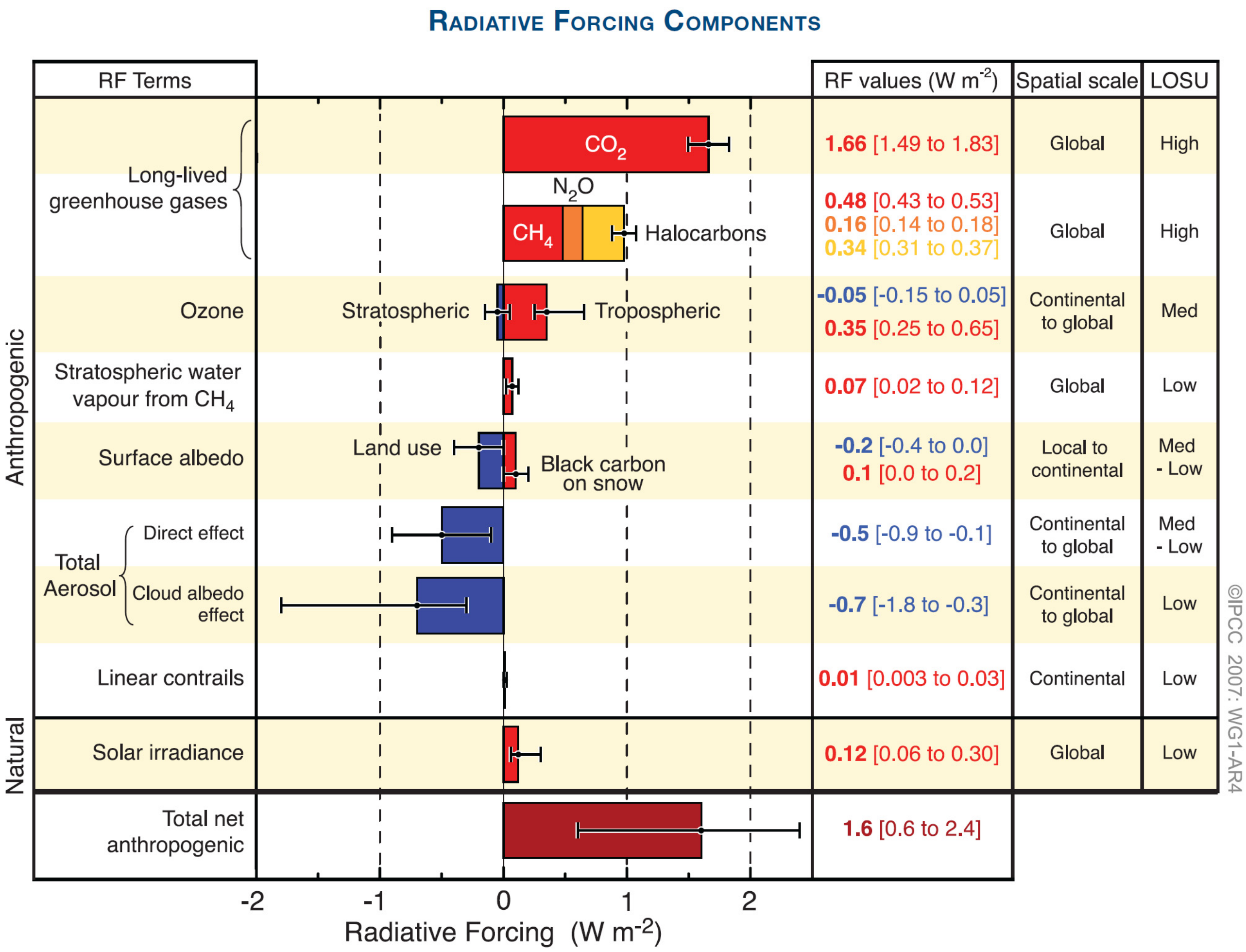


Figure1.1. Global average radiative forcing (RF) estimates and ranges in 2005 for anthropogenic carbon dioxide (CO2), methane (CH4), nitrous oxide (N_2O), and other important agents and mechanisms, together with the typical geographical extent (spatial scale) of the forcing and the assessed level of scientific understanding (LOSU). The net anthropogenic radiative forcing and its range are also shown. These require summing asymmetric uncertainty estimates from the component terms, and cannot be obtained by simple addition. Additional forcing factors not included here are considered to have a very low LOSU. Volcanic aerosols contribute an additional natural forcing but are not included in this figure due to their episodic nature. The range for linear contrails does not include other possible effects of aviation on cloudiness. (2.9, Figure 2.20 in *IPCC* [2007])

1995; *Stohl et al.*, 2003a, 2003b]. In STE, the intrusion of stratospheric materials into the troposphere is called stratosphere-to-troposphere transport (STT) [*Stohl et al.*, 2003a, 2003b]. This intrusion is often seen in cases of severe weather [*Browning and Reynolds*, 1994; *Goering et al.*, 2001] such as cyclonic activities, and is considered important for both dust outbreaks and STT. Dust is a kind of solid aerosol and affects the atmospheric radiation budget. The amount of advection of the stratospheric materials into the troposphere is important for material circulation and, in addition, affects the radiation balance in the atmosphere if ozone is considered. As a result, variations in dust and stratospheric materials in the troposphere are likely to affect the material circulation and the radiation balance in the atmosphere as suggested in Figure 1.1.

In this study, dust (as a tropospheric tracer) and tritium (as a stratospheric tracer) concentrations were analyzed using an ice core. The ice core is one of the most useful tools for reconstructing atmospheric information from the past to the present, thus helping understand and assess past climatic changes. The ice core data is capable of providing information on atmospheric materials and circulations in the past because the snow surface comes in direct contact with the atmosphere. Ice core sites are generally located at higher altitudes and are likely to reflect atmospheric circulation. Many such ice cores have been drilled in mountain glaciers [e.g., *Schwikowski et al.*, 1999; *Shiraiwa et al.*, 2003; *Shiraiwa et al.*, 2004; *Thompson et al.*, 1998; *Thompson et al.*, 2000; *Thompson et al.*, 2002] and ice sheets in Greenland and Antarctica [e.g., *Petit et al.*, 1990; *Watanabe et al.*, 2003; *EPICA community members*, 2004; *North Greenland Ice*

Core Project members, 2004], and are now available for reconstructing past atmospheric information based on material circulations.

An important problem for reconstructing past atmospheric information is that ice core data is a one-point data making it difficult for discussing spatial variations in the past atmospheric information. In general, the degree to which ice core data reflects spatial information is unknown. However, we should extract past spatial data of atmospheric materials from ice cores for discussing spatial material circulation. Therefore, it is essential and important to understand the spatial information from ice core data. Meteorology is helpful in understanding the circulation of atmospheric material circulation in the horizontal and vertical scale, which can support the validity of ice core data. Comparing detailed results of meteorological analyses with ice core data at a higher time-resolution is a very important first step for interpreting the information of past atmospheric circulation from ice core data.

In general, many meteorological studies on ice cores, often using annual ice core data, have been mainly associated with atmospheric circulation in the troposphere and in the horizontal scale [e.g., *Moore et al.*, 2002; *Kang et al.*, 2003]. Because seasonal dating is very difficult for ice core research, an annual data set has often been used. However, in order to extract detailed atmospheric information from ice core data, higher time-resolution ice core analysis with detailed meteorological analyses are necessary as mentioned above. In this study, the material data from the troposphere to the stratosphere in horizontal and vertical scales using coupled ice-core-meteorology is discussed. The ice core in this study was analyzed at higher time-resolution and seasonal dating was performed. This will help understand better the past material variations in the atmosphere as detailed spatial information. This is an original concept and is discussed in this.

Information about tropospheric material circulation is known in the distant past to some extent, in contrast to the stratospheric information, which remains almost entirely unknown. Some ice core studies analyzed stratospheric tracers such as isotopes of beryllium and tritium in the ice core [e.g., *Beer et al.*, 1988; *Fujii et al.*, 1990; *Schwikowski et al.*, 1999]. Unfortunately, most of them were used for assessing solar activity and ice core dating. Thus, there have been no ice core studies focused on the direct interaction between the stratosphere and the troposphere, especially in the North Pacific region.

Assessing the impact of circulating stratospheric material associated with the circulating tropospheric material is very important for understanding climatic changes in the present and the past. For example, one of the stratospheric tracers, ozone, has unique characteristics such as greenhouse effect and high oxidation. If the ozone balance in the troposphere and stratosphere changes, it may considerably affect the environment leading to a global environmental change. However, direct stratospheric ozone measurement from ice cores is difficult. In addition, ozone is produced by a photochemical process in the atmosphere [e.g., *Monks*, 2000]. Separating the ozone into tropospheric ozone and stratospheric ozone is, in general, difficult except for STT studies. Hence, ozone may not be considered the best stratospheric tracer for ice core studies. On the other hand, another stratospheric tracer, tritium, has often been used in ice core research and is relatively easy to measure [*Fujii et al.*, 1990; *Kamiyama et al.*, 1997]. The tritium background level in the atmosphere increased drastically after the nuclear tests during the1950s and 1960s [*Gat et al.*, 2001]. However, except for the nuclear tests, tritium is mainly produced by cosmic rays in the stratosphere. It primarily exists as tritiated water vapor and a clear spring peak of tritium in rain samples in the troposphere has been well explained as advection from the stratosphere into the troposphere [*Gat et al.*, 2001]. Thus, tritium is considered the best stratospheric tracer involving STT information in ice core research. Another stratospheric tracer, an isotope of beryllium (^{10}Be), has also been measured in ice cores [e.g., *Beer et al.*, 1988], but an accelerator is necessary to measure this tracer, which is more difficult than tritium measurement. Therefore, tritium was considered and was analyzed as the stratospheric tracer in this study.

Simultaneous investigation of both stratospheric and tropospheric materials in specific ice cores can be useful for interpreting the interaction between the troposphere and stratosphere in the past. Until date, there have been no ice core studies where both tropospheric and stratospheric origin materials are measured at the same time to focus on the interaction between the troposphere and the stratosphere followed by a detailed comparison of the data to corresponding specific atmospheric circulation patterns or weather events in each year in terms of meteorological analyses. Therefore, this study provides an important progress in reconstructing past information of the interaction between the troposphere and the stratosphere. Based on the result presented here, a more detailed spatial information on the past troposphere and stratosphere from ice core studies can be discussed. This doctoral research contributes to the progress in research associated with material circulating in the troposphere and the stratosphere in the North Pacific region, and will be useful for improving the estimation of Figure 1.1 by *IPCC* [2007] in near future.

The main parts of this study are comprised of two chapters (Chapters 2 and 3). Chapter two focuses on the North Pacific region from East Asia to the North Pacific Ocean, a region which is subject to Asian dust outbreaks and STT associated with cyclonic activities which may be important for assessing climate change. Therefore, investigations on variation in this region from the past to the present are very important for future climate prediction. Thus, the tropospheric and stratospheric materials (dust and tritium in this study) were analyzed in an Alaskan ice core and the data are discussed in terms of ice core research in Chapter 2. In Chapter 3, the investigations on the interaction between the troposphere and the stratosphere were carried out as case studies on severe dust storms in East Asia in 2001 and 2002 and, simultaneously, their impact on the ice core site were examined. In addition, more investigations on the seasonal march and interannual variation of atmospheric circulation in spring–early-summer (March–June) were carried out for understanding atmospheric circulation patterns, which may affect the Alaskan ice-core data and the interaction between the tropospheric and the stratospheric material circulation of the North Pacific region.

Chapter 2

Intra-annual variations in atmospheric dust and tritium in the North Pacific region detected from an ice core from Mount Wrangell, Alaska

2.1 Background

The North Pacific region is known to exhibit various climate phenomena such as the Pacific Decadal Oscillation (PDO) [*Mantua et al.*, 1997], El Niño-Southern Oscillation (ENSO) [*Bjerknes*, 1969], and Arctic Oscillation (AO) [*Thompson and Wallace*, 1998]. Therefore, a lot of geoscientists focus on this region and carry out research on subjects associated with these climate phenomena.

Ice-core study is one of the important fields for the reconstruction of historical climate changes in the North Pacific region. In the northern hemisphere, various ice cores were drilled mainly in the Atlantic section at locations such as Greenland and mountain glaciers [e.g., *Thompson et al.*, 2000; *North Greenland Ice Core Project members*, 2004]. In the North Pacific section, however, only few ice cores have been drilled so far. The locations are Mount Logan [*Holdsworth et al.*, 1992; *Shiraiwa et al.*, 2003], Eclipse ice field [*Wake et al.*, 2002] in the Yukon Territory, Canada, Mount

Bona Churchill by L. G. Thompson, Mount Ushkovsky in Kamchatka, Russia [*Shiraiwa et al.*, 2001], and Mount Wrangell in the Saint Elias Range, Alaska [*Shiraiwa et al.*, 2004].

We particularly focus on the dust concentration in the Mount Wrangell ice core because it is known to have been associated with a large dust outbreak in the Asian continent in the North Pacific section. The atmospheric dust outbreaks in the northern hemisphere are mainly associated with dust storms [*Iwasaka et al.*, 1983], particularly storms in the Gobi and Taklamakan deserts of East Asia. The outbreak frequency is highest in March, April, and May, and the dust outbreak frequency has particularly increased after the year 2000 [*Japan Meteorological Agency* (*JMA*), 2006; *Chun and Lim*, 2004]. The transport of Asian dust to Alaska was first identified in 1977 from aircraft samples [*Rahn et al.*, 1977] and it was also observed recently by elemental analysis using trajectory analysis [*Cahill*, 2003] and satellite observations [*Darmenova et al.*, 2005]. Hence, Asian dust has had impacts on Alaska for a long time.

In June 2003, a 50-m ice core was recovered from the summit of Mount Wrangell—a volcano located at 62°N, 144°W and 4100 m above sea level (a.s.l.) in Alaska [*Shiraiwa et al.*, 2004]. Mount Wrangell is an ideal site for ice-core study because of its proximity to the North Pacific, its high snow accumulation rate (1.3 m yr^{-1} in water equivalent near the North Crater [*Benson and Motyka*, 1978]), and the relatively flat glacier surface (Figure 2.1). Consequently, an ice core with few melt-frozen layers was obtained. The summit is in the free troposphere and its surface is strongly affected by a westerly jet. Since insoluble microparticles (dust) are transported from East Asia to the

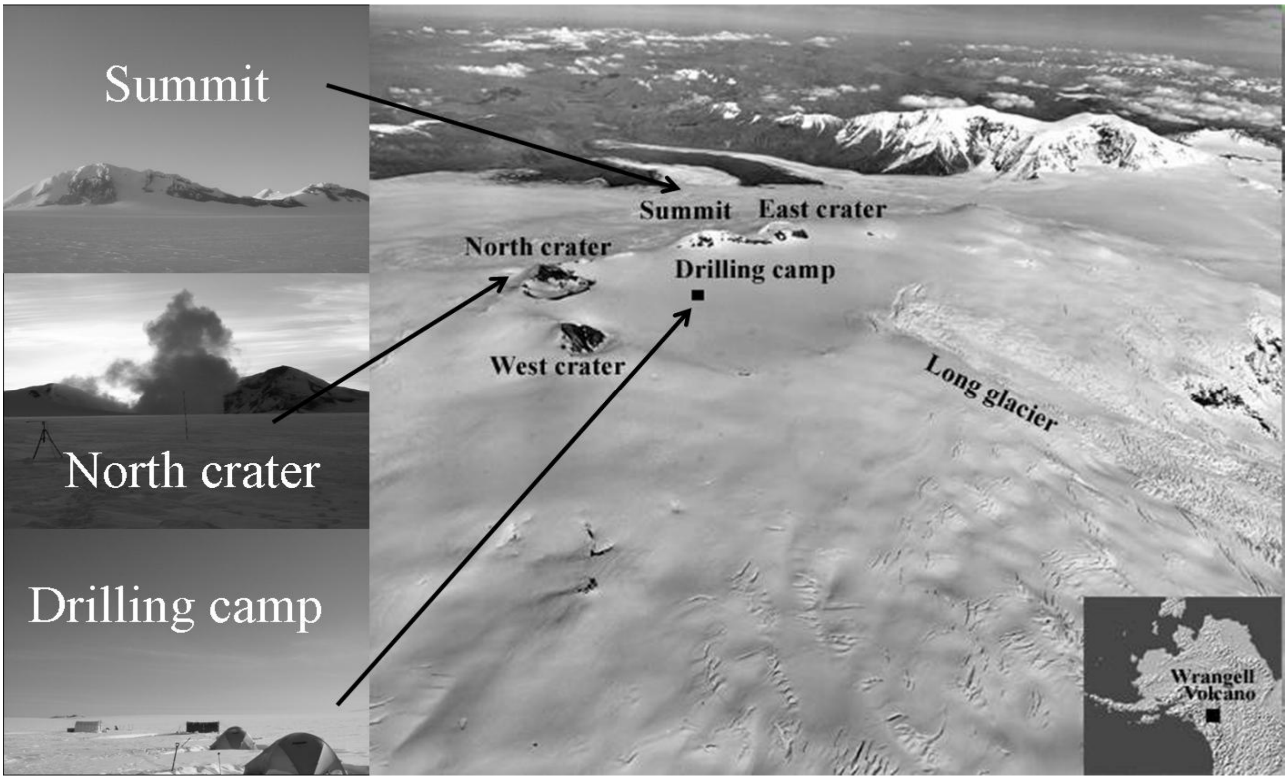


Figure 2.1. Schematic depiction of the summit of Mount Wrangell (elevation: 4100 m). The large picture is an aerial photograph at the summit taken by AeroMap, Anchorage, US. The summit caldera shapes a 4 × 6 km oblong crown and has 1 km deep [*Shiraiwa et al.*, 2004].

summit of Mount Wrangell by this jet, the summit is probably the best site to detect the Asian dust. Therefore, the ice core drilled at the summit of Mount Wrangell can be ideal for investigating the past seasonal variations of dust from East Eurasia to Alaska around the North Pacific region.

In this work, we have analyzed the ice core drilled by *Shiraiwa et al.* [2004] and have successfully obtained various datasets of Mount Wrangell through seasonal dating. The other ice cores drilled in the region were drilled at sites with considerably lesser snow accumulations, and it was unfortunately difficult to discuss the seasonal events. The goal of an ice-core study is generally to reconstruct long-term information of the past atmospheric behavior extending beyond contemporary meteorological observations. However, climate phenomena may vary on intra-annual time-scales. Therefore, a seasonal dataset of ice cores and their examination for seasonal variations of climate are necessary. If we could accurately discuss the details of seasonal climate variations from ice cores using many meteorological datasets that are available, then we could understand the distant past. It will also be helpful to simulate the past climate by General Circulation Model (GCM) studies in the future. In this study, we discuss important seasonal ice-core data in the North Pacific region.

2.2 Method

2.2.1 Ice core and snow sample analyses

The ice core was cut at intervals of 5–10 cm (some sections were slightly less than 5 cm) for a total of 643 samples for dust analysis, which corresponded to 31–143 samples per year and to roughly 3–12 days resolution. After cutting, we removed the surface of the samples to an extent of approximately 5 mm with a ceramic knife to remove the external contamination; this was done with clean-room clothes and poly gloves on a clean bench in a low-temperature clean room in a manner similar to that of *Fujii et al.* [2003]. The 50-m ice core was analyzed over its entire length for the dust particle number concentration in each range (0.52–0.71, 0.71–1.00, 1.00–1.42, 1.42–2.00, 2.00–2.82, 2.82–4.00, 4.00–5.70, 5.70–8.00, 8.00–11.15, and 11.15–16.00 μm) as well as the tritium concentration and hydrogen isotopic ratio (δD). As the size of the dust influences its atmospheric lifetime, we divided the dust sizes into fine dust (FD) with diameters 0.52–1.00 μm, coarse dust

(CD) with diameters 1.00–8.00 μm, and huge dust (HD) with diameters 8.00–16.00 μm. Their characteristics and the reason why we defined these three ranges are discussed later. All the analyzing processes for the dust were identical to those in the previous study [*Fujii et al.*, 2003] except for the rinsing process for the samples using ultra pure water because the ice core was firn, and the estimated error in the dust analysis in this study was within 10%. We measured the tritium concentration for every 40 cm (total 124 samples) by using a liquid scintillation counter [*Kamiyama et al.*, 1997]. The analytical error was $\pm 5\%$ in a 50 g and 7 TU sample, where the tritium unit (TU) is given by 1 TU = 10^{-18} [$^3H/H$] = 0.1181 Bq/L. The ratio δD was analyzed using the IsoPrime apparatus from Micromass [UK]. Measurements were made for every 10 cm for a total of 495 samples. The estimated measurement error was less than 0.1‰.

We also carried out a field observation in May 2004 at the summit of Mount Wrangell. The snow samples were obtained approximately 3 times a day. The dust concentrations in these were analyzed with the same method as that for the ice core.

2.2.2 Trajectory analysis

A 10-day backward trajectory analysis was performed for the understanding of air mass transport routes to Mount Wrangell for the duration of the field observation at the summit in May 2004. A trajectory model was produced by *Yamazaki* [1986]. This trajectory model was constructed based on the Lagrangian tracking method. The horizontal and vertical wind data (3D wind data) from the Grid Point Values (GPV) data for May provided by the JMA were used for the calculations. The wind data were complemented by a linear interpolation into the horizontal wind data and a cubic spline interpolation into the vertical wind data. Air parcels were scattered in the space comprising a latitude line, longitude line, and vertical layer. The initial space was divided into 25 bins (5×5) horizontally and 5 layers vertically ($5 \times 5 \times 5$, total 125 bins). Abundant air parcel scatterings make reappearance of air mass pathways to Mount Wrangell better. Subsequently, one air parcel was scattered in each bin (total: 125 parcels). The calculation time step was 2 h. The initial space centered on the summit of Mount Wrangell was set at 145.02˚W–143.02˚W in longitude, 60.59˚N–62.59˚N in latitude, and 700–500 hPa in altitude.

2.3 Results

2.3.1 Ice core dating

We use two important calibration layers for the ice-core dating. One is the drastic increase in the dust concentration in all the ranges at the depth from 26.824 to 26.873 m in water equivalent (hereafter denoted as m w.e.) due to Mt. Spurr Volcano's eruption in the fall of 1992. The other is the top of the ice core because we know that the drilling date was June 20, 2003. The ice core was drilled from a level of 0.85 m below the surface and the details will be discussed later.

The former was observed at the points labeled Y in Figure 2.2. The only visible sediment in the

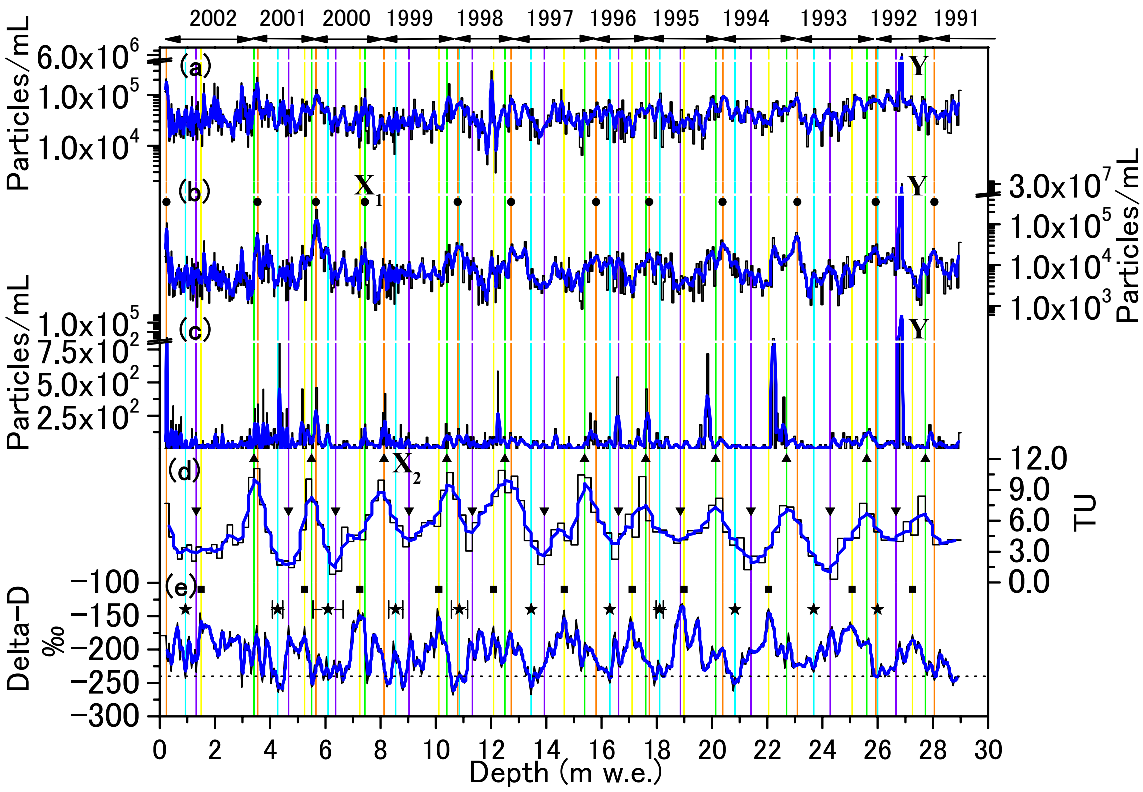


Figure 2.2. Dust, tritium, and δD variations in the Mount Wrangell ice core. Concentrations in the **(a)** fine dust (FD) (0.52–1.00 μm); **(b)** coarse dust (CD) (1.00–8.00 μm); and **(c)** huge dust (HD) (8.00–16.00 μm). **(d)** Tritium concentration. **(e)** Ratio of stable hydrogen isotope (δD). Depth was converted to water equivalent depth by using the density profiles [*Kanamori*, 2004]. Circles, triangles, squares, reverse-triangles, and stars denote the approximate positions of early spring, late spring, summer, fall, and winter, respectively, and each symbol is placed next to the curve that was used in their determination. The symbols X_1 (circle) and X_2 (triangle) mark late spring and early spring in 2000, respectively. The vertical solid lines of orange, yellowish green, yellow, violet, and sky-blue also denote early spring, late spring, summer, fall, and winter, respectively. The intervals between same colored vertical solid lines delineate annual increments. Symbol Y in **(a)**, **(b)**, and **(c)** corresponds to the clear dust layer from 26.824 to 26.873 m in water equivalent. The error bars in **(e)** denote broad minima which were used to determine the center depths in winter as mentioned in section 3.1. All the bold blue lines denote running means of 5 data points. The horizontal lines denote –240‰ level of δD.

50-m ice core was observed at this depth (Figure 2.3). The drastic increases in the dust concentrations were seen in the dust layer. The increases in FD, CD, and HD were larger by a factor of more than 100, 1000, and 10000 with regard to the mean values; the ash layer was excluded during the computation of the mean values in each range. These concentrations were abnormally higher than the background level in all ranges. In particular, the drastic HD increase suggests that a huge dust outbreak occurred very near the location. In 1992, the Crater Peak of Mount Spurr near Anchorage in Alaska erupted. The September 16-17 eruption ejected a large amount of volcanic ash and the trajectory analysis showed that the ash was transported to Mount Wrangell; the amount of dust deposition on the summit was presumed to be in the range from 50 to 100 g m^{-2} in Figure 16 in the work of *McGimsey et al.* [2002].

The preliminary results that were obtained using a petrographic microscope showed that the ash-layer samples were from the 1992 eruption of Crater Peak [*K. L. Wallace of USGS Anchorage*, personal communication, 2005]. A Comparison with the 1992 ash deposited in Anchorage showed petrographic and glass morphologic similarity to the ash samples in the ice core. The distances from Mount Spurr to Mount Wrangell and Anchorage are approximately 444 and 127 km, respectively.

Additionally, we calculated the mass of the ash deposition in the dust layer. The representative radius for each dust range, which was determined by the geometric mean between the minimum and maximum radius in the dust range, was used to calculate the single-particle volume. The single-particle mass at each range was then calculated with a typical mineral density of 2600 kg m^{-3}

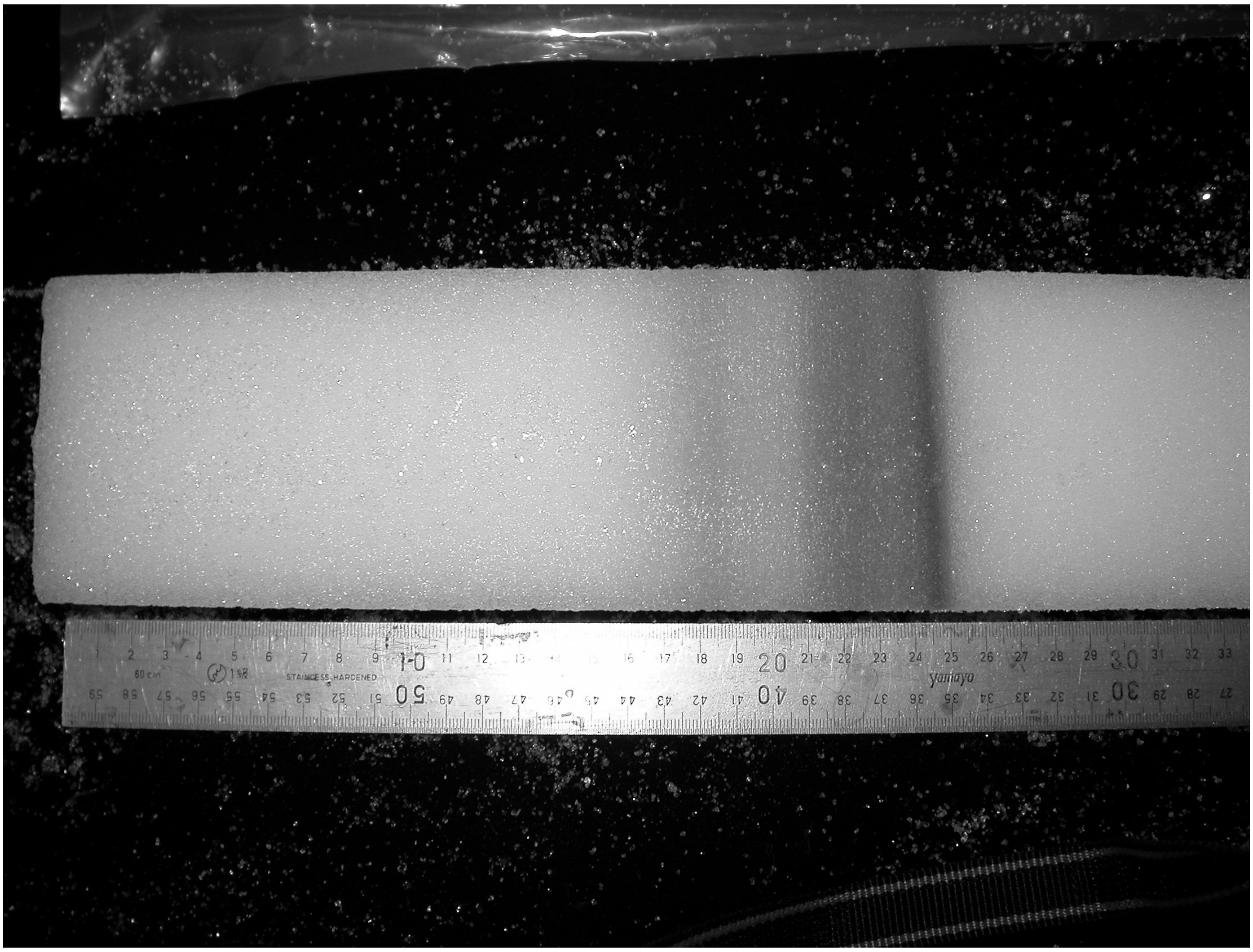


Figure 2.3. The picture of the ash layer corresponds to Mount Spurr eruption on 16-17 September in 1992 at a core depth of 26.824 to 26.873 m in water equivalent. The clear ash layer was found only at this depth in the ice core.

for each range. Subsequently, we estimated the amount of snow accumulation from the top of the dust layer to the bottom and the dust concentration in the dust layer. Finally, we obtained the total amount of ash deposition in all the dust ranges (0.52–16.00 μm) as 77.34 g m^{-2} by using the abovementioned data. This value was in the range from 50 to 100 g m^{-2} that was presumed by *McGimsey et al.* [2002]. In conclusion, the dust in the dust layer definitely corresponded to the Mount Spurr eruption. Taking into account all the abovementioned facts, we were convinced that the dust from the Mount Spurr eruption on 16–17 September 1992 was deposited onto the summit of Mount Wrangell.

If we take a look at the relationship of the ice-core data with the ash layer of Mount Spurr, we can determine the seasons corresponding to certain depths. We used a running mean of five data points because the raw data greatly fluctuated over short distances, making it difficult to determine each peak depth. The δD minima sometimes had broad depths in Figure 2.2e; similar minimum values continued. Therefore, if more than two δD minima less than –240‰ were found around the minima, we determined the minimum position in a certain year as the mean depth among the minimum peak depths.

Now, we know that the dust layer was deposited in September (fall). If we take a look around the ash layer, we find that just before the δD minimum peak depth the tritium concentration was decreasing from the maximum to the minimum. Therefore, the tritium minima are regarded as corresponding to the fall season, which occurred after 16-17 September (Mount Spurr eruption).

The dust peak of Mount Spurr eruption also was seen between the maximum and minimum of δD, and the δD minima occurred after the tritium minima. Therefore, the δD maxima and minima are regarded as corresponding to summer and winter, respectively. This δD seasonal cycle was identical to that of $\delta^{18}O$ at the nearest ice-core site, Eclipse Icefield (60.51˚N, 139.47˚W; 3017 m asl) in the Saint Elias Range [*Wake et al.*, 2002]. Therefore, the δD data in our ice core also reflect past seasonal cycles and are similar to those of past ice-core studies [*Holdsworth et al.*, 1992; *Thompson et al.*, 2000; *North Greenland Ice Core Project members*, 2004].

The other dating point comprises the dust, tritium, and δD at the top layer of the ice core. The ice core was drilled in June 2003 from a depth of 0.85 m from the surface (0.207 m w.e.). If we calculate the annual layer thickness by using δD, tritium, and FD and CD peaks, we obtain the annual mean snow accumulations from 2.43 to 2.58 m w.e. A snow accumulation of 0.207 m w.e. roughly corresponds to one month. Therefore, the ice-core data at the top roughly correspond to May. At the time, the concentration of FD and CD had just crossed their annual maxima, while that of tritium was located after the δD minimum and positioned near the maximum. We cannot determine whether the tritium concentration in the top layer was the maximum. The δD values in the top layer were almost the middle values between the minimum and the maximum in the annual cycles. The tritium concentrations and δD values before and after the Spurr eruption were consistent. All the tritium maxima occurred between the δD minima and maxima. Therefore, the tritium maxima are regarded as corresponding to springtime.

Moreover, we found that the CD maxima in Figure 2.2 appeared between the δD minima and the tritium maxima except for the year 2000 (X_1 in Figure 2.2). Two possible origins of the CD are generally speculated. One is dust supply from a local source, while the other is dust transport from significant dust events in a remote place. Therefore, a seasonal pattern in the CD is not generally useful for intra-annual dating. However, it is useful to use the CD maxima for dating in this study because of clear seasonal positions between the δD minima and tritium maxima. Additionally, the correlation analysis between the FD and CD and field observations support the usefulness of dust seasonality as discussed later.

Among the three types of dust (FD, CD, and HD), the FD dominated the dust from distant sources, presumably due to its long residence time in the atmosphere. When long-range transport from East Asia occurred, we observed that the FD increased in the field observation in May 2004 (Figure 2.4). This was verified by a 10-day backward trajectory model in which all 125 air parcels were scattered simultaneously [*Yamazaki*, 1986] (Figure 2.5). It indicates that the dust originated around the Gobi and Taklamakan deserts (Figure 2.5). In our data, we were generally able to identify a major CD or HD peak with a local dust-generating event, including volcanic eruptions; however, the CD levels also seasonally increased with the FD levels. For example, Figures 2.2a and 2.2b show that the FD and CD have similarly shaped broad peaks and minima, whereas the HD profile in Figure 2.2c has different characteristics.

These HD peaks are probably due to dust from volcanic plumes from the summit craters of

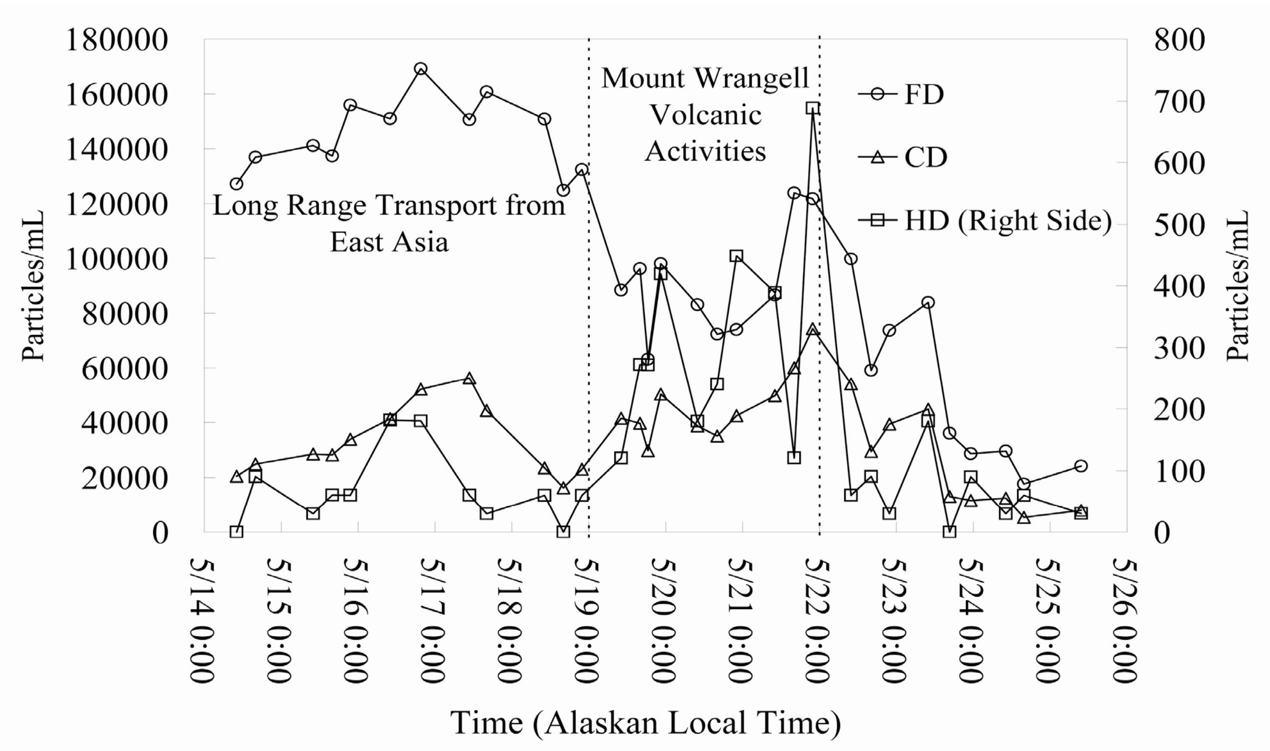
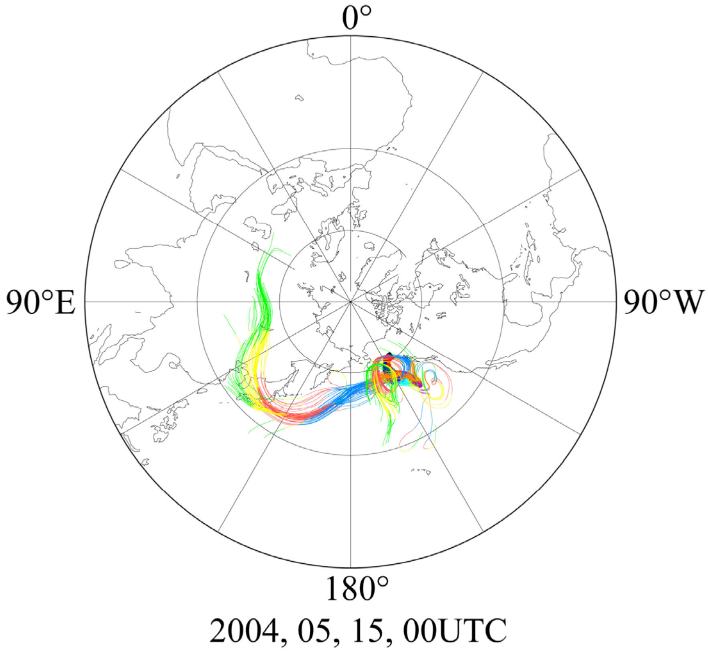


Figure 2.4. Dust content in the uppermost 2 cm of surface snow at the summit. Variations of FD, CD, and HD were measured about 3 times per day.

(a)



(b)

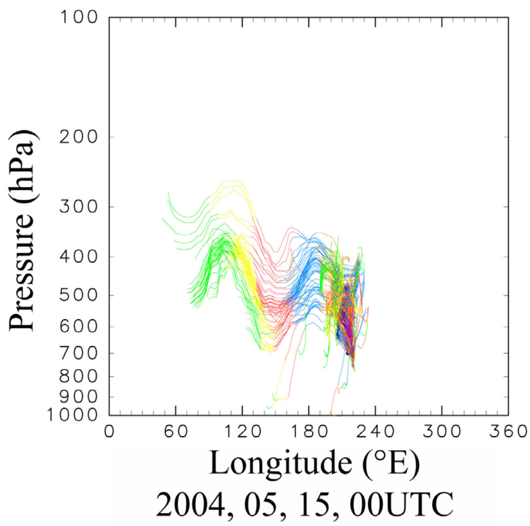


Figure 2.5. Air mass trajectories to Mount Wrangell in May 2004. The 10-day backward trajectories were analyzed using the trajectory model [*Yamazaki*, 1986] with 3D wind data from the GPV obtained from the JMA for May 2004. Each color denotes the air mass trajectory for a day. The denoted time is the start time of trajectory calculation in Coordinated Universal Time (UTC).

Mount Wrangell when the wind was blowing from the north crater (Figure 2.4) because we observed a drastic increase in the HD characteristics at the summit in 2004 from 2-cm surface snow samples. Therefore, in the case of Mount Wrangell, the HD contributes the most to the dust variations in the ice core as its volcanic activities and the CD do not contribute significantly. After removing the peak from the Mount Spurr ash layers (symbol "Y" in Figure 2.2), the correlation coefficient between the FD and CD was calculated to be 0.62 (99.9% significance level; two-tailed t-test; $n = 638$; $t = 19.928$). It also indicates that the FD and CD outbreak events were mostly generated in the same place. Hence, the seasonal cycle of the CD is related with the FD variation.

We find that the depth positions for tritium and CD in 2000 in Figure 2.2 are inversely related, which is an exceptional feature. The tritium and CD peaks for this year occurred only in early spring and late spring, respectively, because the tritium peaks were located before the CD peaks. With minor exceptions, we determined that the CD peaks mainly occur in early spring due to their clear seasonal positions. In conclusion, we will consider the CD maxima in Figure 2.2 as corresponding to early spring because almost all the CD peaks are located between the δD minima and tritium maxima. Moreover, we observe a 11-year maxima in the FD, CD, tritium, and δD and these maxima also fully explain the timing of the Y peaks in Figure 2.2 as being due to the dust event in 1992. Finally, we obtained the ice-core data, which spanned the years 1992–2002, that were divided into five seasons: early spring, late spring, summer, fall, and winter.

2.3.2 Dust and tritium fluxes in each season

To compare the annual trends in each season, we calculated the dust and tritium fluxes (Figure 2.6). The fluxes reflect all the dust and tritium increase events in each distinct season. The fluxes were calculated by multiplying the concentration of each element by the snow accumulation rate in each season. For the tritium fluxes, we considered the tritium half-life of 12.32 years [*Lucas and Unterweger*, 2000]. We used a representative month for each season in the calculation of tritium flux—April for early spring to late spring, June for late spring to summer, August for summer to fall, November for fall to winter, and February for winter to early spring. The HD was excluded from all further calculations because it is affected by the level of Mount Wrangell's volcanic activities. Correlation analyses among the FD, CD, and tritium were done with the two-tailed t-test. The results between dust and tritium with a significance level greater than 90% are shown in Figure 2.6. Because tritium mainly exists as tritiated water vapor (HTO) in the atmosphere, snow deposition is more important than dry deposition. However, dust deposits occur either by dry deposition or with snow. For an ideal comparison between tritium and dust fluxes, we need to exclude data during abnormally large and small snow accumulations. We thus used the flux data in each divided season when the snow accumulations were within 1σ of the mean values. Because we focused on the Asian dust contribution, we excluded the data from summer through winter of 1992 when the Mount Spurr eruption [*McGimsey et al.*, 2002] occurred. We excluded the data from the summer to the fall of 1998 too because large boreal forest fires occurred both in Siberia and Alaska in this year

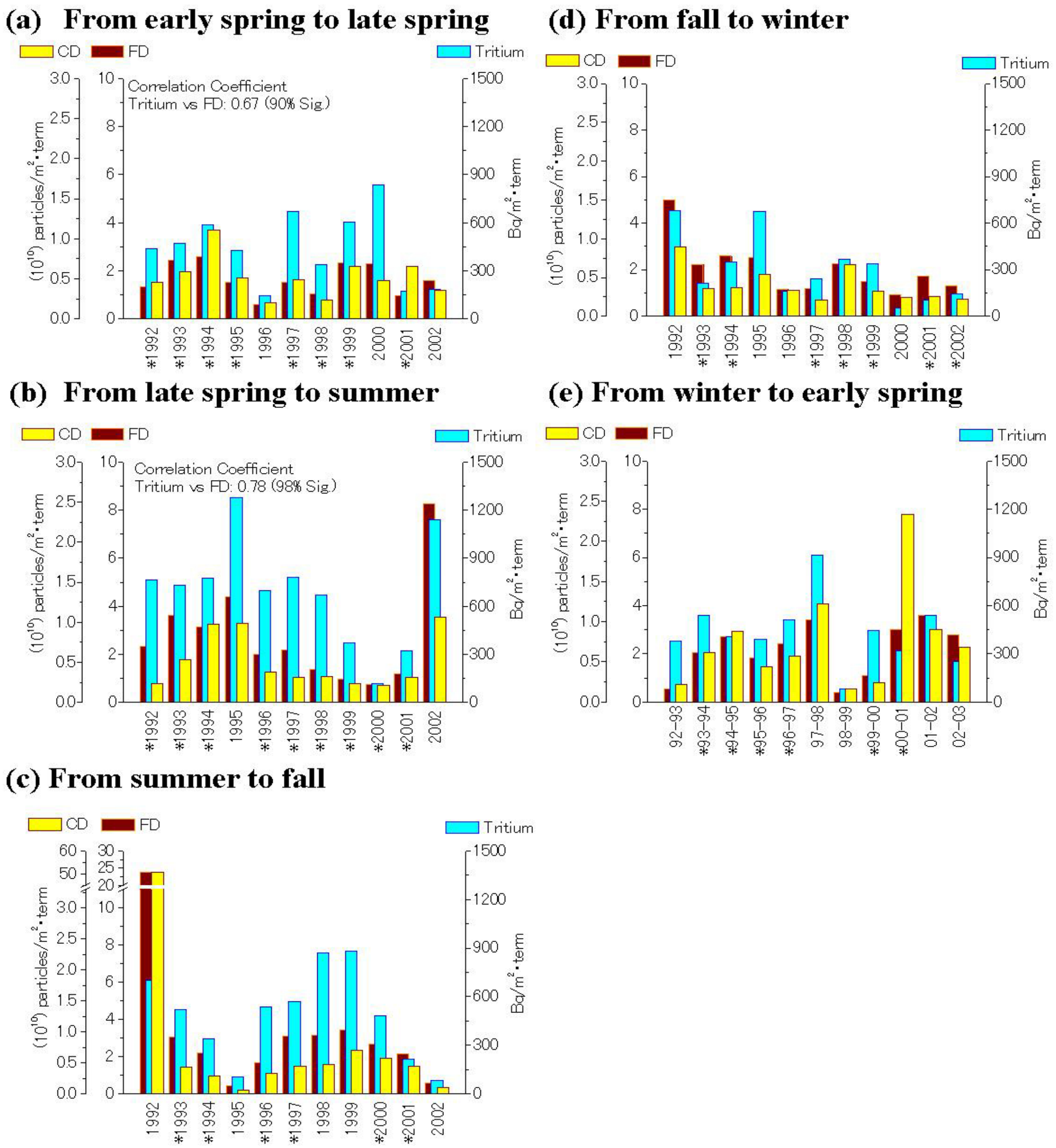


Figure 2.6. FD, CD, and tritium fluxes in each seasonal period. **(a)**: Early spring to late spring. **(b)** Late spring to summer. **(c)** Summer to fall. **(d)** Fall to winter. **(e)** Winter to early spring. Asterisks denote the data used for the correlation calculations (sample number: n). Also shown are the correlation coefficients between dust and tritium that had a significance level exceeding 90%.

[*Kasischke and Bruhwiler*, 2002; *M. Fukuda of ILTS, Hokkaido Univ., Japan*, personal communication, 2005] and black carbon aerosol (soot) due to forest fires can be transported from central Alaska to southern Alaska glaciers [*Kim et al.*, 2005], probably corresponding to the drastic increase of FD from the summer to the fall of 1998 (Figure 2.2a). All the correlation matrixes among the FD, CD, and tritium fluxes and concentrations and snow accumulations in each season are also shown in Table 2.1.

By dividing the annual ice-core signals into signals corresponding to five distinct seasons, we obtained important data on the past atmospheric annual flux variations for each season (Figure 2.6). The amounts of dust and tritium in the atmosphere varied for each year and season. In springtime, abnormally high FD and CD peaks were found after 2001 (Figures 2.2a, 2.2b, 2.6b, and 2.6e). It has been known that the outbreak frequency of Asian dust events is largest in March, April, and May. In particular, this has increased after the year 2000 [*Chun and Lim*, 2004; *JMA*, 2006]. Our results nicely support these two features and imply that Asian dust was definitely transported and deposited onto the summit of Mount Wrangell. Our backward trajectory analyses suggest that there are a lot of transport pathways from East Asia to Alaska and these vary with time. However, the background levels of dust concentration were particularly higher in springtime after 2001 (Figures 2.2a and 2.2b), implying that the Asian dust contribution to Mount Wrangell was significant. These flux increases in both the FD and CD were observed from the winter of 2000 to early spring of 2001 and from late spring to the summer of 2002 (Figures 2.2a, 2.2b, 2.6b, and 2.6e). This shows that severe

Table 2.1. Correlation Coefficients Among the FD, CD, Tritium Fluxes and Concentrations, and Snow Accumulation in Each Season

Significance level	90%	95%	98%	99%	99.9%

(a) From Early Spring to Late Spring (degree of freedom: n = 6)

	FD Flux	CD Flux	Tritium Flux	FD Con.	CD Con.	Tritium Con.	Snow Accum.
FD Flux	1.00	0.69	0.67	0.61	−0.03	0.39	0.69
CD Flux	0.69	1.00	0.30	0.58	0.58	−0.05	0.35
Tritium Flux	0.67	0.30	1.00	−0.14	−0.52	0.70	0.98
FD Con.	0.61	0.58	−0.14	1.00	0.54	−0.09	−0.14
CD Con.	−0.03	0.58	−0.52	0.54	1.00	−0.63	−0.48
Tritium Con.	0.39	−0.05	0.70	−0.09	−0.63	1.00	0.55
Snow Accum.	0.69	0.35	0.98	−0.14	−0.48	0.55	1.00

(b) From Late Spring to Summer (degree of freedom: n = 7)

	FD Flux	CD Flux	Tritium Flux	FD Con.	CD Con.	Tritium Con.	Snow Accum.
FD Flux	1.00	0.70	0.78	0.69	0.08	0.29	0.77
CD Flux	0.70	1.00	0.47	0.41	0.64	0.04	0.55
Tritium Flux	0.78	0.47	1.00	0.19	−0.35	0.62	0.90
FD Con.	0.69	0.41	0.19	1.00	0.37	0.14	0.08
CD Con.	0.08	0.64	−0.35	0.37	1.00	−0.35	−0.26
Tritium Con.	0.29	0.04	0.62	0.14	−0.35	1.00	0.23
Snow Accum.	0.77	0.55	0.90	0.08	−0.26	0.23	1.00

(c) From Summer to Fall (degree of freedom: n = 4)

	FD Flux	CD Flux	Tritium Flux	FD Con.	CD Con.	Tritium Con.	Snow Accum.
FD Flux	1.00	0.56	0.41	0.54	−0.24	−0.21	0.81
CD Flux	0.56	1.00	0.17	−0.05	0.47	−0.31	0.73
Tritium Flux	0.41	0.17	1.00	0.19	−0.19	0.76	0.33
FD Con.	0.54	−0.05	0.19	1.00	0.02	0.08	−0.04
CD Con.	−0.24	0.47	−0.19	0.02	1.00	0.00	−0.25
Tritium Con.	−0.21	−0.31	0.76	0.08	0.00	1.00	−0.34
Snow Accum.	0.81	0.73	0.33	−0.04	−0.25	−0.34	1.00

(d) From Fall to Winter (degree of freedom: n = 5)

	FD Flux	CD Flux	Tritium Flux	FD Con.	CD Con.	Tritium Con.	Snow Accum.
FD Flux	1.00	0.65	0.46	0.81	0.45	0.27	0.65
CD Flux	0.65	1.00	0.66	0.69	0.95	0.69	0.23
Tritium Flux	0.46	0.66	1.00	0.19	0.51	0.95	0.52
FD Con.	0.81	0.69	0.19	1.00	0.67	0.15	0.09
CD Con.	0.45	0.95	0.51	0.67	1.00	0.63	−0.08
Tritium Con.	0.27	0.69	0.95	0.15	0.63	1.00	0.25
Snow Accum.	0.65	0.23	0.52	0.09	−0.08	0.25	1.00

(e) From Winter to Early Spring (degree of freedom: n = 4)

	FD Flux	CD Flux	Tritium Flux	FD Con.	CD Con.	Tritium Con.	Snow Accum.
FD Flux	1.00	0.77	−0.36	0.91	0.75	−0.77	0.15
CD Flux	0.77	1.00	−0.67	0.78	0.99	−0.94	−0.05
Tritium Flux	−0.36	−0.67	1.00	−0.68	−0.74	0.53	0.74
FD Con.	0.91	0.78	−0.68	1.00	0.80	−0.69	−0.26
CD Con.	0.75	0.99	−0.74	0.80	1.00	−0.91	−0.15
Tritium Con.	−0.77	−0.94	0.53	−0.69	−0.91	1.00	−0.17
Snow Accum.	0.15	−0.05	0.74	−0.26	−0.15	−0.17	1.00

dust events occurred in the springtime of 2001-2002 compared to those in the springtime of 1992–2000. The CD concentration in early spring of 2001 was the highest since 1992 except for the Mount Spurr eruption in 1992 (Figure 2.2b). Further, the FD concentration also increased together with the CD concentration (Figures 2.2a and 2.2b). This implies that the severest dust storm occurred in this year since 1992. Therefore, we can state that the atmosphere in early spring of 2001 was the most contaminated (both by the FD and CD) during the period 1992–2002.

The tritium concentration was high from early spring to summer and the highest in late spring in the raw data (Figure 2.2d). Tritium mainly exists as tritiated water vapor in the atmosphere, and prominent tritium peaks are detectable from spring to summer at the ground surface in the Northern Hemisphere [*Gat et al.*, 2001]. Therefore, our results correspond to those of previous tritium studies. Most of the tritium that reaches the ground is produced in the stratosphere by cosmic rays and has a half-life of 12.32 years [*Lucas and Unterweger*, 2000]. Airborne tritium is also produced by nuclear tests. It has been used for determining the year 1962/1963 in ice-core researches [e.g., *Fujii et al.*, 1990] because of the massive tritium injections by the nuclear tests from 1961-1962 and the one- or two-year time lags of these effects. The tritium background level in the atmosphere has now returned to the level that existed before the nuclear tests [*Gat et al.*, 2001]. Therefore, the recent seasonal tritium variations in precipitation are mainly produced by cosmic rays in the stratosphere and intrusions from the stratosphere to the troposphere.

Tritium is exchanged between the troposphere and stratosphere by the stratosphere-troposphere exchange (STE). This process plays an important role in the material exchange (e.g., tritium and ozone) between the stratosphere and troposphere [*Holton et al.*, 1995; *Monks*, 2000; *Gat et al.*, 2001; *Stohl et al.*, 2003]. In this study, we use the term STE to refer to the one-way material intrusion from the stratosphere to the troposphere. The STE is actively generated through the tropopause folding by cutoff low, blocking high, or cyclonic activities [*Holton et al.*, 1995; *Monks*, 2000; *Gat et al.*, 2001; *Stohl et al.*, 2003a]. The seasonal maximum of the STE is predominantly seen in springtime in the northern hemisphere [*Holton et al.*, 1995; *Monks*, 2000; *Gat et al.*, 2001; *Stohl et al.*, 2003]. The maximum concentration of stratospheric tracers such as tritium and ozone is also predominantly observed at the ground surface in springtime [*Monks*, 2000; *Gat et al.*, 2001]. Tritium has the highest concentrations north of 30° latitude [*Gat et al.*, 2001].

Two modifying processes delay the arrival of tritium at the ground surface by 26 days [*Ehhalt*, 1971]. One is the vertical transport process between high and low altitudes. The other is the re-evaporation of the winter and spring precipitation. The latter effect is dominant at altitudes below 3.05 km [*Ehhalt*, 1971]. Both effects contribute to the delay in the arrival of tritium at the ground surface. Due to the study location being at 4100 m, the delay caused by the vertical transport process may be the most important for tritium transport to Mount Wrangell. Therefore, tritium may be transported from the stratosphere to the summit of Mount Wrangell within one month.

We compared the tritium concentration at Mount Wrangell with the seasonal characteristics of tritium concentration in the precipitation at Anchorage, Alaska, by using the Global Network of

Isotopes in Precipitation (GNIP) from the International Atomic Energy Agency (IAEA) and the World Meteorological Organization (WMO), Austria [*IAEA/WMO*, 2004] (Figure 2.7). Seasonal tritium peaks appeared from late spring to early summer (May–July) at Anchorage. Even if we consider a one-month delay at the ground surface as mentioned above, the tritium concentrations at the surface have maxima from early spring to summer (Figure 2.7). The tritium peaks in late spring in our ice-core results nicely correspond to those in the precipitation data; therefore, our ice-core results also reflect the seasonal cycle of tritium in the atmosphere. The tritium concentration and flux are the highest in late spring in Figures 2.2d and 2.6b, supporting the fact that tritium intrusions into the troposphere are the most active in late spring.

All the correlation coefficients with the two-tailed t-test among the fluxes and concentrations of FD, CD, and tritium and snow accumulations in each season were calculated within the limited period as mentioned earlier and are shown in Table 2.1. In the season from early spring to late spring (Table 2.1a), the FD and tritium fluxes were strongly correlated. The FD flux correlated with both the snow accumulation and the FD concentration, although the correlation between the FD concentration and flux was less than the 90% significance level. However, the correlation coefficient between the FD flux and concentration was similar in degree with the other high-significance correlations such as that between the dust and tritium fluxes or the dust flux and snow accumulation. It indicates that an annual relationship between the dust and tritium should exist between early and late spring. However, variations in the snow accumulation also strongly

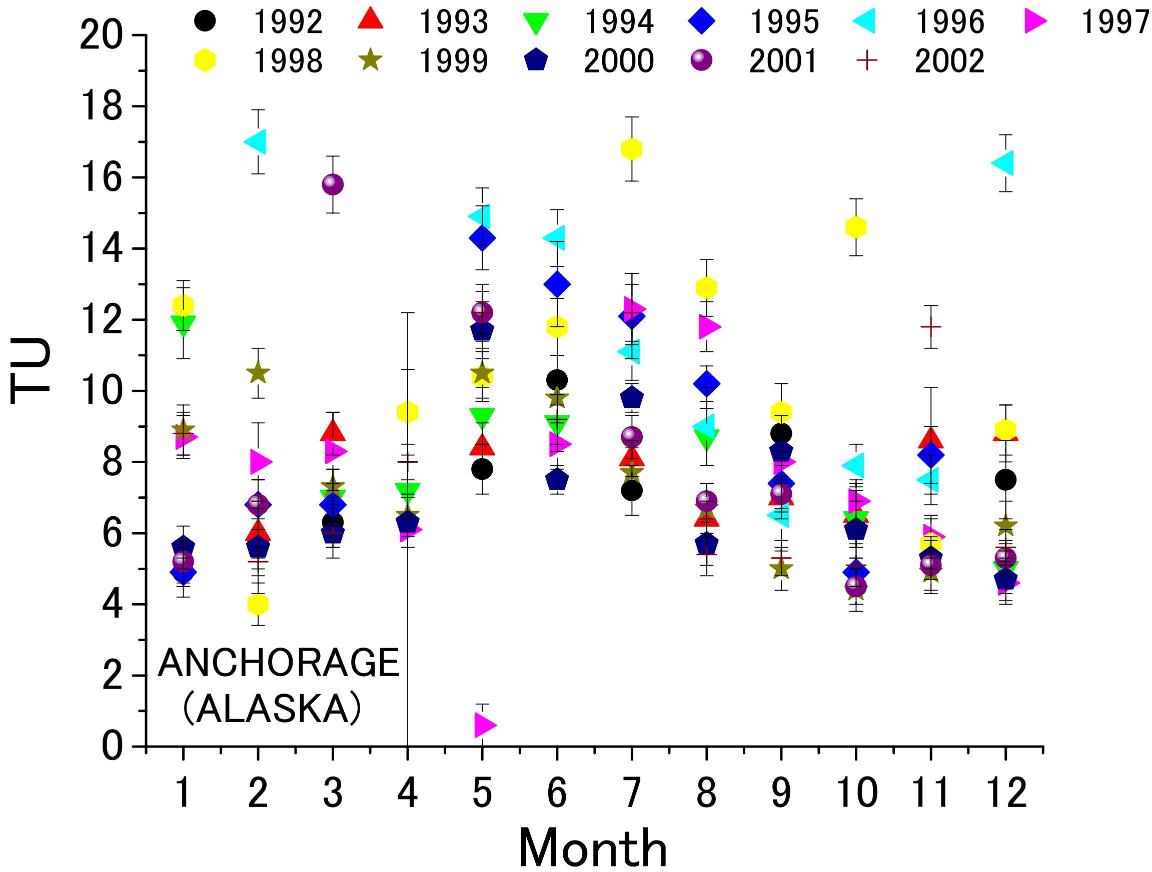


Figure 2.7. Seasonal variations of tritium concentration from 1992–2002 in precipitation data at Anchorage station, which was obtained from IAEA-GNIP data [*IAEA/WMO*, 2004]. Error bars denote measurement errors.

control this relationship. Hence, the annual relationship between the dust and tritium fluxes was observed due to the variations of dust and tritium concentrations and those of snow accumulation.

The tritium concentration and flux from early spring to late spring were highly correlated, indicating that the annual variation of tritium flux was due to variations in the concentration (Table 2.1a). The tritium flux was also correlated with snow accumulation, indicating that the snow effectively decreased the tritium background level in the atmosphere in this season. This is reasonable because tritium existed as tritiated water vapor in the atmosphere as mentioned above and therefore may have an increased affinity to snow or wet depositions rather than dry depositions.

In the season from late spring to summer, the fluxes of FD and tritium had the highest correlation (Table 2.1b). These also had high correlations with the concentrations of FD and tritium, CD flux, and the snow accumulation, indicating that the strong annual variations of the FD and tritium fluxes were due to both the variations in the concentration and the snow accumulation. However, the CD flux was not highly correlated with the snow accumulation. Therefore, the annual variations of both the FD and tritium fluxes were associated with snow deposition and that of the CD was associated with the dry deposition in this season (late spring to summer). It indicates that FD from a remote place may contribute to the annual relationship between dust and tritium. This is an interesting feature. In conclusion, the FD had the strongest annual relationship with tritium in this season.

In the season from summer to fall, the FD and CD were correlated only with snow accumulation (Table 2.1c) because the increases in dust and tritium were small and this season is the summer rainy season in Alaska [*Polissar et al.*, 1998]. Hence, the mean atmospheric dust levels were almost constant in this season from 1993 to 2002 and the amount of snowfall controlled the amount of dust deposition.

In the season from fall to winter, snow accumulation did not contribute to the flux variations significantly and variations in the concentrations of FD, CD, and tritium contributed the most to the fluxes (Table 2.1d). Therefore, the variations in the concentrations strongly controlled the annual flux variations.

In the season from winter to early spring, the FD and CD fluxes were correlated with their concentrations (Table 2.1e). However the correlations for tritium were weak, indicating that the annual variations of the dust fluxes depended on variations in their concentrations and that of tritium depended on the snow accumulation in this season.

2.3.3 Comparison of dust deposition amount in Alaska with those in other areas

We calculated the annual mean and seasonal mean of the dust particle number density, dust mass, and fluxes with the same method that was used in sections 2.3.1 and 2.3.2, as shown in Table 2.2. The abbreviations for ES, LS, SU, FA, and WI in Table 2.2 denote early spring, late spring, summer, fall, and winter, respectively. The dust size range 8–16 μm was excluded because Mount Wrangell's volcanic activities contributed the most in this range as mentioned above. Table 2.2 was originally produced by *Zdanowicz et al.* [1998] and we added our results to it. The Mount Spurr eruption strongly contributed to the mass deposition. However, if we exclude it, the values were less than those observed in China [*Wake et al.*, 1994]. The annual mean mass in this study was the same as that for the Alaskan range near Mount Wrangell [*Hinkley*, 1994]. The range of the dust flux for Mount Wrangell was the same as those of the Canadian Basin and Saint Elias Range (USA) [*Windom*, 1969; *Mullen et al.*, 1972; *Darby et al.*, 1974]. Greenland and Canadian Arctic were cleaner than our site [*Murozumi et al.*, 1969; *Kumai*, 1977; *Hammer*, 1977; *Fisher and Koerner*, 1981; *Koerner and Fisher*, 1982; *Hammer et al.*, 1985; *Steffensen*, 1988, 1997;

Table 2.2. Atmospheric Dust Concentration and Flux in Snow and Ice at Various Northern Hemisphere Sites[a]

	Site[b]				Dust concentration and flux					
	lat	long	elevation (m asl)	Period[c] (years AD)	analytical method[d]	size range (μm)	number (10^3 mL^{-1})	mass (μg kg^{-1})	flux[e] (μg cm^{-2} yr^{-1})	Reference
Mount Wrangell (Including Spurr)	62°N	144°W	4100	1992–2002	LLS	0.52–8	80.2	1511	84.7	This work
Mount Wrangell (Excluding Spurr)							58.2	303	12.6	
Mount Wrangell WI-ES							70.7	498	20.3	
Mount Wrangell FA-WI							48.4	207	9.6	
Mount Wrangell SU-FA							151.6	6162	368.3	
Mount Wrangell SU-FA (Excluding Spurr)							41.7	122	8.0	
Mount Wrangell LS-SU							51.7	228	13.1	
Mount Wrangell ES-LS							78.7	458	11.9	
					Canadian Arctic					
Penny Ice Cap	67°N	65°W	1980	1988–1994	CC	0.65–12	31.6	143	4.8	*Zdanowicz et al.* [1998]
					CC	1-12	13.7	129	4.4	
Agassiz Ice Cap	81°N	73°W	1600	1950–1977	CC	>1	18.3	na	na	*Koerner and Fisher* [1982]
				last 5000 years	CC	>1	18.2	na	na	
Devon Ice Cap	77°N	82°W	1800	last 7000 years	CC	>1	8.3	235	4.2	*Fisher and Koerner* [1981]
					Arctic Ocean					
Canada Basin	75°N	150°W	sea level	recent snow	FLTR	na	na	na	1.4–13.3	*Darby et al.* [1974]
Canada Basin	84°N	72°W	sea level	recent snow	FLTR	na	na	na	8.5–21	*Mullen et al.* [1972]
Arctic Ocean (Average)			sea level	recent snow	CHEM	na	na	na	3.1	*Rahn et al.* [1979]
					Greenland					
Summit	72°N	38°W	3207	recent snow	CC	0.5–12	na	46	1	*Steffensen* [1997]
Dye3	65°N	43°W	2479	last 10,000 years	LLS	0.2–4	na	50	2.5	*Hammer et al.* [1985]
				1978–1983	LLS	0.2–4	9.4	na	na	
				1780–1951	LLS	0.2–4	20.0	na	na	
Camp Century	77°N	61°W	1885	recent snow	CHEM	na	na	35	1.4	*Murozumi et al.* [1969]
				1753–1965	EM	0.02–8	na	35	1.4	*Kumai* [1977]
Sites A and D (Average)	70°N	39°W	3070	1891–1910	LLS	0.2–4	na	74	2.5	*Steffensen* [1988]
Inland Sites (Average)			>2400	1940–1950	LLS	>1	14.0	na	na	*Hammer* [1977]
					Other sites					
Alaska Range (USA)	63°N	151°W	2500	recent snow	CHEM	na	na	300	na	*Hinkley* [1994]
Saint Elias Range (USA)	60°N	139°W	2600	recent snow	FLTR	na	na	na	16	*Windom* [1969]
Mount Olympus (USA)	47°N	123°W	2000	recent snow	FLTR	na	na	na	32	*Windom* [1969]
Mont Blanc (France)	45°N	6°E	4270	1955–1985	CHEM	na	na	na	21–35	*De Angelis and Gaudichet* [1991]
Colle Gnifetti (Switzerland)	46°N	7°E	4450	1936–1982	CC	0.63–16	37.0	na	na	*Wagenbach and Geiss* [1989]
					CHEM	na	na	na	20	
Mustagh Ata (China)	38°N	75°W	5910	1990–1992	CC	1–22	276.4	6780	247	*Wake et al.* [1994]
Ngozumpa Glacier (Nepal)	28°N	86°W	5700	1989–1990	CC	1–13	18.2	379	27	*Wake et al.* [1994]
Chongce Ice Cap (China)	35°N	81°W	6327	1980–1987	CC	1–22	616	8220	607	*Wake et al.* [1994]

[a]Modified from Zdanowicz et al. [1998] with permission from Zdanowicz et al. and Blackwell Publishing.
[b]Lat, latitude; Long, longitude.
[c]Period of snow accumulation represented by the data.
[d]Analytical methods: CC, Coulter counter or equivalent method; CHEM, calculated from chemical data (typically [Al], [Ca], or [Si]); EM, electron microscope; FLTR, based on dry filter weights; LLS, laser light scattering.
[e]Flux values were taken from the literature or calculated from mass concentrations using published estimates of snow accumulation rates. The unit of seasonal Mount Wrangell Fluxes is μg cm^{-2} season^{-1}.

Zdanowicz et al., 1998]. However, the seasonal means of the mass from late spring to fall for Mount Wrangell were similar to the annual means in Canadian Arctic and larger by a factor of 2–3 as compared to Greenland. The contamination in the atmosphere around the summit of Mount Wrangell was higher in spring than in the season from summer to fall by a factor of 4. With regard to the history of dust studies in the Northern Hemisphere, the annual mean value in this study corresponded to those of the Younger Dryas event and the post-Last Glacial Maximum in Greenland, and the seasonal mean value in springtime (WI-ES and ES-LS) in the present ice core agreed with that of the Eemian event 2 [*Steffensen*, 1997].

2.4 Discussion

2.4.1 Dust characteristics for each defined range and their origins

Dust concentration in the FD and the CD ranges showed clear seasonal variations (Figure 2.2), and these two ranges were closely correlated with each other as mentioned in section 2.3.1. In the Mount Wrangell ice core, we determined that the dust maxima in Figure 2.2 corresponded to early spring. Previous studies have mentioned that soil aerosols in Alaska increase the most in April [*Malm et al.*, 1994; *Malm et al.*, 2004]. The Si variation also has spring maxima in Alaska [*Polissar et al.*, 1998]. These previous results were obtained from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network (http://vista.cira.colostate.edu/IMPROVE/); this network was established by an interagency consortium of federal land management agencies and

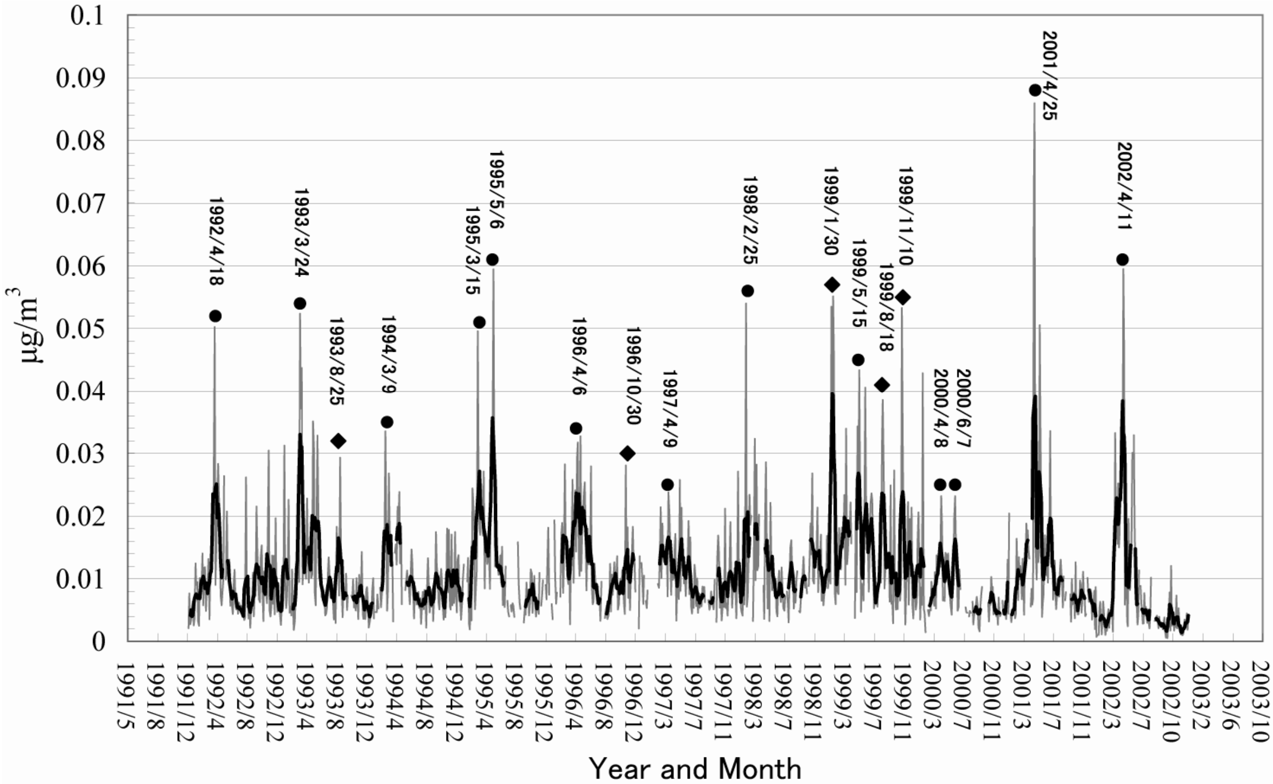


Figure 2.8. Mass of calcium particles less than 2.5 μm in diameter observed at Denali National Park; the data was supplied by the IMPROVE network. Bold line denotes 5-point running mean. Circles and diamonds with date denote the maxima of 5-point running mean corresponding to spring and other seasons, respectively.

the Environmental Protection Agency in the spring of 1985 to assess visibility and aerosol monitoring for the purpose of tracking spatial and temporal trends of visibility and visibility-impairing particles in rural areas. A sample of the IMPROVE data in Alaska at Denali National Park (63.72˚N, 148.97˚W, 658 m a.s.l.; highest site in Alaska) is shown in Figure 2.8, which is calcium data for particles less than 2.5 μm. The Ca data mainly have distinctive spring maxima; however, they sometimes have additional peaks corresponding to another season. Our result for the early spring maxima in the FD and CD corresponds to the seasonal variations in Figure 2.8. Therefore, our results for the dust at the summit of the mountain may also represent the typical seasonal variation of the atmospheric soil dust in Alaska.

We discuss the origins of dust in springtime in the following. First, we consider two typical origins. One is from a local area in Alaska. The other is due to transport from a remote area in the world. *Polissar et al.* [1998] mentioned that Alaskan time periods had been divided according to different synoptic conditions in the Arctic as follows. Spring from March to June is associated with a period of intensive long-range transport and effective photochemical transformation of aerosols in the Arctic troposphere during polar sunrise. Summer from July to September is related to a period of low long-range transport and more effective scavenging processes by precipitation and clouds. Winter from October to February is represented by a transitional synoptic situation. Therefore, it is known in Alaska that long-range transport is dominant in springtime and not dominant in summer because of the summer rainy season. In fact, in our results, the FD concentration increases the most in springtime due to long-range transport, as shown in Figure 2.2, and both the FD and CD flux are correlated with the snow accumulation only from summer to fall in Table 2.1, indicating that wet deposition is the most dominant in this season. Our drilling site is located at a high altitude and synoptic atmospheric circulations may influence the summit directly. The long-range transport of FD from East Asia was actually observed at the summit in the field observation (Figures 2.4 and 2.5). The case of long-range transport showed one example of a typical springtime pattern for material transport from East Asia. It is presumed that the seasonal FD and CD cycles are due to dust outbreak events in remote places. Of course, dust from local areas near Mount Wrangell may contribute to the variation of the CD in the ice core to some extent. However, we assumed that the clear seasonal cycles of dust mainly depend on contributions from remote areas because the FD generally has a long residence time and the FD and CD were highly correlated with regard to their seasonal cycle.

For long-range transport of the CD, a stronger wind velocity as compared to the FD is required to cover the required distance in the atmosphere. Additionally, the CD needs to reach a higher altitude because long-range transport by the westerly jet occurs in the free troposphere. It was pointed out by *Sun et al.* [2001] that cold fronts with a strong wind velocity were important in springtime for the long-range transport of dust and the dust of the Taklamakan Desert contributed more to the long-range transport as compared to that of the Gobi Desert. The maxima of the FD and CD were observed in early spring with a high positive correlation in our dating (Figures 2.2a and 2.2b). In this season, the main sources of dust from East Eurasia to the North Pacific are the Gobi and Taklamakan deserts and arid regions in East Asia. These deserts are known to have frequent dust storms in springtime [*Sun et al.*, 2001]. A strong vertical convection and high wind speeds at the ground surface are needed to lift dust to the altitude of the strong westerly jet.

Long-range transport of Asian dust over the Pacific Ocean has been observed [*Rahn et al.*, 1977; *McKendry et al.*, 2001; *VanCuren and Cahill*, 2002; *Cahill*, 2003; *Darmenova et al.*, 2005]. In the springtime of 2001, the CD dust concentrations and fluxes increased the most in the ice core (Figures 2.2b and 2.6e); The FD also increased together (Figure 2.2a). In fact, severe dust storms occurred in East Asian deserts and large amounts of dust were transported to the North Pacific and

Alaska [*Yu et al.*, 2003; *Zhang et al.*, 2003; *Darmenova et al.*, 2005]. The Sea-viewing Wide Field-of-view Sensor (SeaWiFS) images also showed abnormally large and long-lasting dust clouds in April 2001 (NASA science news, 17 May 2001 at http://science.nasa.gov/headlines/). In the article, Herman, the principal investigator of NASA's Total Ozone Mapping Spectrometer (TOMS) experiment, mentioned that in terms of the area covered, this was the largest dust storm observed in the Northern Hemisphere since 1979. The largest Asian dust storm broke out on April 6-7, 2001 and its large dust cloud reached Alaska on April 13, 2001 (see the web site of Visible Earth: http://visibleearth.nasa.gov/view_rec.php?id=14051). Although our ice core covers the years from 1992 to 2002, the CD maximum in the spring of 2001 in Figure 2.2b is undoubtedly the largest after 1992, except for the eruption of Mount Spurr and probably corresponds to the dust storm from East Asia. In the case of northern Chinese deserts during this spring, the peak mass loading occurred for the range of 4 to 8 μm in most dust-storm cases and these dust storms were commonly associated with dry and windy conditions [*Zhang et al.*, 2003]. The dominant dust range corresponds to our CD range. Of course, generally, CD has a lower residence time in the atmosphere as compared to FD. However, the contribution of the westerly jet to the long-range transport of dust to Mount Wrangell is large as mentioned above. Therefore, we cannot exclude the contribution of the Asian dust; it is also not possible to explain the clear seasonal variations in the CD that are associated with those of the FD at altitudes greater than 4000 m in the free troposphere without the Asian dust contribution. Therefore, we propose that a site at a high altitude, like the summit of Mount Wrangell, is largely affected by the Asian dust in the springtime every year; this is also true for other places in Alaska and regions around the northeast Pacific Basin and Canada [*Rahn et al.*, 1977; *McKendry et al.*, 2001; *VanCuren and Cahill*, 2002; *Cahill*, 2003].

In the North Pacific region, ice-core results are still sparse. However, dust loading is large in this area and various transport models have calculated the dust distribution and transport pattern [e.g., *Takemura et al.*, 2002; *Uno et al.*, 2003; *Tanaka et al.*, 2005]; however, information on the actual dust deposition is desirable. In particular, this importance is larger with regard to the past for which there is no meteorological data. Therefore, our intraseasonal dust data in the ice core may contribute to these demands, enabling an assessment of the past seasonal climate phenomena.

2.4.2 Seasonal variation of tritium in the troposphere

We obtained the maximum tritium concentration in late spring, except for the year 2000 (early spring), for the period from 1992 to 2002 (Figure 2.2d). The observed precipitation data near the ground surface depicted in Figure 2.7 also showed similar seasonal variations. Therefore, the seasonal variation of tritium with the maximum in late spring may be a typical phenomenon in this region. The origin of tritium in recent years is mainly from the stratosphere where it is produced by cosmic rays; tritium that resulted from the effect of thermonuclear tests has disappeared [*Gat et al.*, 2001]. Although the half-life of tritium is short (12.32 years [*Lucas and Unterweger*, 2000]), we can detect the seasonal cycle till the year 1963, which is one year after the year of a thermonuclear test.

This anthropogenic effect may help to reconstruct the seasonal variation as a visible variation in the concentration and compensate the half-life decay. After around 1980, tritium concentration levels have been returning to pre-1963 values [*Gat et al.*, 2001]. Hence, the effects of nuclear tests have almost disappeared since 1980 and the seasonal variation of tritium after 1980 may mainly be due to their production by cosmic rays in the stratosphere and their intrusion into the troposphere in springtime. In conclusion, in the ice-core study at Mount Wrangell, tritium is very useful to determine the late spring period if a time resolution is sufficient for about one month.

2.4.3 Comparison among dust and tritium fluxes

We examined the strength of the annual relationship between tritium and the dust fluxes. The highest positive correlation (98% significance level; two-tailed t-test) occurred in late spring through summer in the case of tritium and FD fluxes (Figure 2.6b). This suggests that the long-range transport of FD and the amount of STE may be related on an annual basis and strongest in late spring because the STE and strong frontal storms with Asian dust outbreaks are the most active in springtime and the weakest in summer [*Holton et al.*, 1995; *Monks*, 2000; *Gat et al.*, 2001; *Sun et al.*, 2001; *Stohl et al.*, 2003; *Chun and Lim*, 2004; *JMA*, 2006]. The drastic increases in the FD and CD fluxes after 2000 (Figures 2.6b and 2.6e) coincide with the recent increase in Asian dust outbreaks [*Chun and Lim*, 2004; *JMA*, 2006]. As discussed in section 2.4.1, the contribution of Asian dust in springtime may be large in this region. For such a long-range transport of dust, the

Asian dust must be lifted up to the altitude of the strong westerly jet by severe weather at the ground surface and a strong vertical convection. The severe weather such as strong cyclonic activities also contributes to the STE [*Browning and Reynolds*, 1994]. Further, the air from the stratosphere into the troposphere may tend to increase in strength, and an increase in stratospheric tracers such as tritium and ozone might be observed near the ground surface; an example is the case of ozone increase in East Asia with the Asian dust storm [*Kim et al.*, 2002]. The annual relationship between the STE and strong frontal storms with Asian dust outbreaks is weak, except for the seasons from early spring to late spring in Figures 2.6a and 2.6b; this indicates that the contribution of strong frontal storms with Asian dust outbreak to the STE is the largest in late spring. Hence, we propose that the annual relationship between the STE and strong cyclonic activities with Asian dust outbreaks may be the strongest in late spring (Figure 2.9).

Although there have been many studies on the Asian dust and the STE independently, there have been no studies on the annual relationship between the strong frontal storms with Asian dust outbreaks and STE because it has been difficult to obtain observations at a single location for several years. Because ice cores continuously preserve the past atmospheric information, it is possible to detect the relationship for several years. Unfortunately, our results are still not sufficient to fully discuss the annual relationship between the STE and strong cyclonic activities with Asian dust outbreaks because the STE mostly occurs on a few days; however, our timely high-resolution data for the dust and tritium roughly correspond to one week and one month, respectively. Of course,

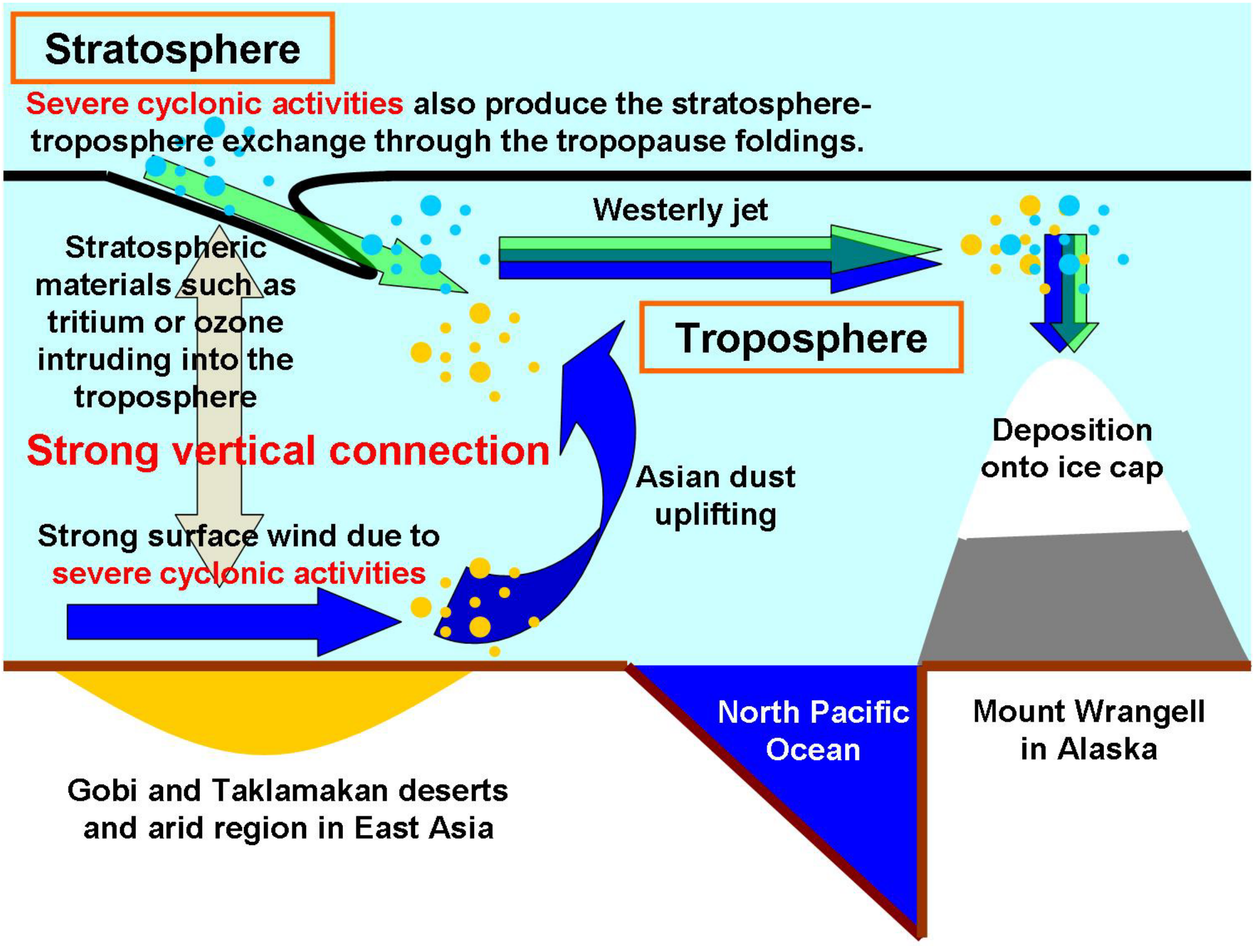


Figure 2.9. Schematic of the annual relationship between Asian dust storm and the STE in late spring according to our proposal. If the Asian dust annually increases, the amount of STE will also increase.

we also cannot know the locations where the STE occurred from only the ice-core results. However, the flux data reflect all the seasonal atmospheric dust and tritium events and our results suggest that Asian dust effect to Mount Wrangell in springtime is evident. Therefore, our speculation is probable and we suggest that this study is the first step to assess the annual relationship between the STE and strong frontal storms with Asian dust outbreaks; henceforth, we should monitor this relationship over a longer time because it leads to various climate changes due to intrusions of the stratospheric tracer into the troposphere and also causes changes in the radiative balance due to an increase in the dust in the troposphere. More studies involving field observations and model studies are necessary and in the near future they will elucidate the annual relationship between the STE and strong cyclonic activities with Asian dust outbreaks and assess their influence on climate change.

2.5 Conclusions

By analyzing an ice core from the summit of Mount Wrangell with an intra-annual dating resolution, we obtained data for clear seasonal variations in δD, tritium, and dust from 1992 to 2002. We succeeded in dating the intra-annual layers and divided each year into five distinct seasons to show the clear annual variations of tritium and dust for each season. The parameter δD has summer maxima and winter minima. The FD and CD concentrations were strongly correlated and had early spring maxima, and Asian dust storms in springtime may strongly contribute to the seasonal variations of the dust. The annual mean dust deposition flux (Table 2.2) at the summit of Mount Wrangell, which is in the free troposphere, was larger than that for the Canadian Arctic and Greenland; less than that for China, Nepal, France, and USA mainland; and similar to that for the Arctic Ocean. The tritium in the ice core had a late spring maximum and fall minimum and its fluxes had the highest annual correlation with the FD fluxes in the late spring season. Subsequently, the strongest annual link between Asian dust storms and the STE may occur in late spring. However, the time resolution of our ice-core data and the analysis for transport pathways in springtime are still not sufficient for a full discussion, and more studies are necessary in the future. Our result is the first step in the study on the annual relationship between Asian dust storms and the STE. The number of days on which Asian dust storms have been observed in East Asia has been increasing recently [*Chun and Lim*, 2004; *JMA*, 2006]; this may not only increase the dust in the atmosphere but also the amount of STE depending on our result that FD and tritium fluxes were annually correlated in late spring the most (Figure 2.9). This can lead to an increase in tritium and ozone in the troposphere. More studies in the future on both the STE and Asian dust will explain this.

In the North Pacific region, a seasonal variation in the climate is evident. Therefore, seasonal datasets of ice cores are necessary to reconstruct the past climate here. However, such datasets in the North Pacific region are still sparse. We fortunately succeeded in intra-annual dating with 5 seasons in this region. If we analyze longer ice cores drilled at the summit of Mount Wrangell and covering several hundred years, we can present detailed discussions on intra-annual climate changes in the past for which there is no meteorological data. The Mount Wrangell ice core will also contribute considerably to paleoclimate studies and model researches on dust circulation around the North Pacific involving probable features on intra-annual time scales. Our preliminary results in this study have covered only the recent 11 years, but the results will contribute to further studies.

Chapter 3

Case studies, monthly characteristics, and interannual connections between Asian dust storms and stratosphere-to-troposphere transport and their impacts on the Mount Wrangell ice core site, Alaska

3.1 Background

Nowadays, we have a better understanding of the impact of atmospheric dust on the global radiation budget [*Intergovernmental Panel on Climate Change* (*IPCC*), 2007] than in the past. Atmospheric dust directly scatters solar radiation [*Zhou et al.*, 1994], is involved in cloud processes [*Wurzler et al.*, 2000], and acts as ice nuclei [*Sassen et al.*, 2003; *Sassen*, 2005]. Thus it indirectly affects the global radiative balance through cloud physics [*Sassen et al.*, 2003; *Sassen*, 2005]. It is deposited onto the ocean and also affects the oceanic biogeochemistry [*Jickells et al.*, 2005]. Therefore, atmospheric dust is an important factor in the assessment of the present and future climate.

The Gobi and Taklamakan deserts and other arid regions in East Asia are important natural dust sources. These dusts are called Asian dust or "Kosa" in Japanese. Spring is the preferred season for dust outbreaks in East Asia. Asian dust storms occur because of cyclonic activities with strong surface winds in spring [*Kurosaki and Mikami*, 2003, 2005; *Yamamoto et al.*, 2007], and they are often associated with cold fronts [*Sun et al.*, 2001; *Hayasaki et al.*, 2006]. Transpacific long-range transport of these Asian dusts has been known; occasionally they reach Taiwan, Alaska, Canada, North America, and Greenland [*Rahn et al.*, 1977; *Biscaye et al.*, 1997; *McKendry et al.*, 2001; *Bory et al.*, 2002; *Takemura et al.*, 2002; *VanCuren and Cahill*, 2002; *Cahill*, 2003; *Darmenova et al.*, 2005; *Zdanowicz et al.*, 2006; *Yasunari et al.*, 2007 (hereafter, YS2007); *Wang*, 2007]. Widespread Asian dust in spring is one of the important factors affecting climate and material circulation.

On the other hand, the stratospheric air mass intrusion into the troposphere (also called stratosphere-troposphere exchange, STE) is an important meteorological phenomenon for material exchange between the troposphere and the stratosphere [*Holton et al.*, 1995; *Stohl et al.*, 2003a, 2003b]. *Stohl et al.* [2003a, 2003b] proposed that the STE should be considered for exchanges in both directions, i.e., stratosphere-to-troposphere transport (STT) and troposphere-to-stratosphere transport (TST). We focus on STT in this study. Stratospheric tracers such as tritium, ozone, beryllium isotopes, etc. move into the troposphere by STT [*Gat et al.*, 2001; *Monks*, 2000; *Zanis et al.*, 2003]. The STT accompanied by tropopause foldings is induced by strong cyclonic activities, cut-off lows, etc. Those stratospheric tracers are mainly observed with spring maxima in the troposphere. Although ozone is produced by photochemical production, STT also contributes to the ozone budget in the troposphere [*Monks*, 2000]. Stratospheric materials such as ozone may affect the radiation balance and oxidation process in the troposphere. Hence studies on STT with stratospheric tracers are essential for the assessment of material circulation and climate change.

Although it is well known that STT and Asian dust outbreaks are dominant in spring, sufficient information about those interannual is not available. For the first time, the possible interannual relationship between them was shown in YS2007 as mentioned in the next paragraph. In 2003, a 50-m ice core was drilled at the summit of Mount Wrangell, Alaska (62°N; 144°W; 4100 m asl) [*Shiraiwa et al.*, 2004]. The site is close to the Gulf of Alaska. YS2007 mentioned that the spring peaks of dust in fine and coarse sizes and tritium concentrations in the ice core may mainly occur as a result of Asian dust storms and STT, respectively. Recent studies have indicated that the effect of STT is lower at lower altitudes but greater at higher altitudes [*Stohl et al.*, 2003a, 2003b; *Zanis et al.*, 2003]. Therefore the Mount Wrangell ice core may be ideal for assessing the spring STT and Asian dust.

YS2007 presented a correlation analysis between the dust and tritium fluxes in each season and showed the highest interannual correlation between fine dust and tritium fluxes, particularly in late spring (a strong correlation from early spring to late spring was also observed). This suggests that the Asian dust outbreaks and stratospheric tritium intrusion into the troposphere are interannually connected by frontal cyclonic activities in spring. There have been no studies on the interannual relationship between the Asian dust outbreaks and stratospheric tritium intrusion at one observatory because it was difficult to measure those tracers at one location over a long period. The

STTs are accompanied by strong surface winds [*Browning and Reynolds*, 1994; *Goering et al.*, 2001]. Asian dust outbreaks require strong surface wind. Hence the hypothesis presented in YS2007 is the most probable. In fact, Asian dust storms associated with increased ozone in the troposphere have also been observed [*Kim et al.*, 2002]. Asian dust outbreaks accompanied by deep tropopause folding, which indicates STT, have also been observed [*Zhao and Zhao*, 2006]. These results support the hypothesis in YS2007. However, these studies did not focus on the Mount Wrangell ice core and therefore do not provide crucial support for the hypothesis. Unfortunately, the problem of ice core study lies in spatial representation. With ice core analysis, it is difficult to obtain spatial information and we cannot determine whether Asian dust storms and STT occur simultaneously and whether the air mass from the stratosphere and the troposphere due to the dust storm event is transported to the Wrangell ice core site.

To explain dust and tritium variations in spring in the Mount Wrangell ice core and develop the hypothesis in YS2007 for the interannual relationship between STT and Asian dust, the following three points should be investigated in terms of atmospheric circulation. (1) Simultaneous Asian dust outbreak and STT due to cyclonic activity. (2) Transport and deposition of Asian dust and stratospheric materials onto the Mount Wrangell ice core site due to the simultaneous outbreak event. (3) Atmospheric circulation patterns in the North Pacific region and snowfall conditions at the ice core site in each month from early spring to early summer. As a first step, investigation of the first two points is necessary to explain each dust and tritium variation in the Mount Wrangell ice core associated with the hypothesis in YS2007. After these case studies, we can investigate whether other contributions to dust and tritium variations exist in the ice core. This point is essential in discussing the mechanism of detailed dust and tritium variations and interannual correlation between Asian dust and STT in spring in the Mount Wrangell ice core in terms of the seasonal change in pressure patterns.

In order to explain and develop the hypothesis in YS2007, we investigated five important Asian dust storms that occurred in April 2001 and March and April 2002 in terms of meteorological analyses with forward trajectory analyses. We then investigated the spring–early-summer meteorological characteristics at the Mount Wrangell ice core site from typical atmospheric circulation patterns in the North Pacific region in 2001 and 2002.

In this paper, we discuss the following topics in section 3.3. In section 3.3.1, we summarize the variations of dust and tritium in the Mount Wrangell ice core in YS2007; we discuss the relationship between Asian dust storms and STT in East Asia and examine the air-mass impacts on the summit of the Mount Wrangell ice core site from five case studies in the spring of 2001 and 2002 in sections 3.3.2 and 3.3.3, respectively; we discuss the dust and tritium contributions to the Mount Wrangell ice core site due to Asian dust storms in the spring of 2001 and 2002 in section 3.3.4; in section 3.3.5, we examine the snowfall frequency at the ice core site in the climatology from 1992 to 2002, corresponding to the period corresponding to the YS2007 ice core, because tritiated water vapor in the atmosphere prefers snowy conditions [YS2007], and the conditions may be important for tritium deposition onto the ice core site; in section 3.3.6, we investigate the seasonal march of pressure patterns, potential vorticity fields, storm track activities, and meridional heat flux in the atmosphere from March to June. These investigations on spatial information may clarify the connections among Asian dust outbreaks, STT, and snowfall at Mount Wrangell. Finally, in section 3.3.7, we discuss a possible explanation for the mechanism of dust and tritium variations in spring in the Mount Wrangell ice core and the interannual connection between fine dust and tritium fluxes in spring suggested in YS2007. YS2007 mentioned that this relationship was due to Asian dust storms and STT in East Asia. We investigate whether this interannual correlation can be explained by only the contributions of Asian cyclonic activity.

Discussions on the interannual relationship between Asian dust outbreaks and STT in terms of other climate forcings such as the Pacific Decadal Oscillation (PDO) [*Mantua et al.*, 1997], El Niño-Southern Oscillation (ENSO) [*Bjerknes*, 1969], and the Arctic Oscillation (AO) [*Thompson and Wallace*, 1998] are the next steps after this study, and we will not discuss those details in this study. We hope that the hypothesis in YS2007 with meteorological support in this study will open a new research field involving STT, Asian dust, ice core, climate, etc. Our studies will be useful not only for the interpretation of ice cores in the North Pacific region, but also in other research fields associated with climate and material circulation changes.

3.2 Methods

The dynamical tropopause in the extratropics is usually defined as having a potential vorticity (PV) of 2 potential vorticity units (PVU; 1 PVU = 10^{-6} m^2 K s^{-1} kg^{-1}) [e.g., *Holton et al.*, 1995; *Goering et al.*, 2001; *Stohl et al.*, 2003a, 2003b]; we followed this definition in this study. The data pertaining to temperature, ozone mixing ratio, relative humidity, vertical and horizontal wind, and PV at each pressure level and wind at the 10-m surface were used from the European Centre for Medium-Range Weather Forecasts (ECMWF) 40-year reanalysis (ERA-40) [*Uppala* et al., 2005]. The 6-h analysis and monthly mean data are available in ERA-40. The potential temperature was calculated using the temperature and pressure data. Surface pressure data were calculated from sea level pressure, altitude, and geographical data. In this study, we focused on one of the stratospheric tracers, tritium, impacting on the Mount Wrangell ice core site; therefore, spatial information on stratospheric tracers was necessary. Unfortunately, tritium spatial data were not available; therefore, we used spatial ozone data from ERA-40 to understand the transport process of stratospheric tracers instead of tritium. Ozone data in ERA-40 are relatively well reproduced over a large part of the stratosphere [*Dethof and Holm*, 2002]. Although some problems partly exist in the troposphere with the vertical structure of the analyzed ozone in absolute values, we will discuss the transport pattern of ozone from the upper troposphere to the lower stratosphere, and those problems are not particularly serious in this study. In addition, when STT occurs, we also interpret the potential vorticity (PV) values of 2 PVU as the tropopause and air in the upper troposphere with increased ozone along 2 PVU line as dry stratospheric air. Each data from the other stratospheric information with ozone data can be used to determine whether the ozone data are proper as a double-check. Relative humidity data from the grid point value (GPV) data for May 2004 provided by the Japan Meteorological Agency (JMA) were also used as a reference because the data for 2004 are not available in ERA-40. The 12 h-analysis data are available in JMA-GPV. Monthly mean sea surface temperature (SST) data from the UK Meteorological Office [*Rayner et al.*, 2006; UKMO – GISST/MOHMATN4/MOHSST6 – Global Ice coverage and SST (1856–2006); available from http://badc.nerc.ac.uk/data/gisst/] were used for comparison with atmospheric data.

In order to understand the air mass transport to the summit of the ice core site of Mount Wrangell, forward trajectory analyses were performed on five severe dust-storm cases in East Asia in the spring of 2001 and 2002. Those dust storms broke out on 6–7 April 2001, 8–9 April 2001, 18(19)–22 March 2002, 24–25 March 2002, and 5(6)–9 April 2002 [*Liu et al.*, 2003; *Shao and Wang*, 2003; *Shao et al.*, 2003; *Sugimoto et al.*, 2003; *Zhou and Zhang*, 2003; *Darmenova et al.*, 2005; *Sun et al.*, 2006]. These dust storms were also detected as transpacific dust storms by Total Ozone Mapping Spectrometer (TOMS) (see the web site: http://toms.gsfc.nasa.gov/index_v8.html; hereafter called, Website 3.1). The trajectory model was constructed based on the Lagrangian tracking method [*Yamazak*i, 1986]. The horizontal and vertical wind data (3D wind data) from the ERA-40 data for 2001 and 2002 were used for the calculations. The wind data were interpolated linearly in the horizontal direction and with a cubic spline interpolation in the vertical direction. The time step for the trajectory calculation was 20 min, and the output time interval was 2 h. Initial air parcels were set in boxes defined initially (Table 3.1). The initial regions correspond to the central area of a cyclone and a STT in each dust storm case. The initial area was divided horizontally into 900 bins (30 × 30) and vertically into 30 levels. At that time, these vertical levels extended from the lowermost stratosphere to the lower free troposphere. A total of 27000 air particles were set in the initial box. We defined the Wrangell Area as the box of 57–67°N in latitude, 139–149°W in

Table 3.1. Initial boxes for forward trajectory calculations

Time set	Longitude	Latitude	Pressure (hPa)	Calculation term (days)
12:00 UTC on 6 April 2001	100–120ºE	40–50ºN	700–250	8
6:00 UTC on 9 April 2001	100–120ºE	35–50ºN	700–250	6
6:00 UTC on 20 March 2002	105–120ºE	35–48ºN	700–250	8
6:00 UTC on 24 March 2002	100–120ºE	42.5–52.5ºN	700–250	8
0:00 UTC on 6 April 2002	105–115ºE	42–48ºN	700–250	9

longitude, and 500–650 hPa in pressure level. Mount Wrangell is centered on the Wrangell Area. Only the trajectories that pass through the Wrangell Area are drawn in the next section.

We investigated the monthly march of subweekly fluctuations associated with migratory synoptic scale eddies as in *Nakamura and Wallace* [1990] and *Nakamura* [1992]. In those studies, local instantaneous storm track activity as the baroclinic wave amplitude at the upper troposphere was measured by the "envelope function" (Ze). The 9-day high-pass-filtered daily mean geopotential height at 250 hPa was used in this study. The quantity Ze was defined locally as doubling the squared high-pass-filtered time-series-data. It was then smoothed using a 9-day low-pass filter once again and its square root was taken. This quantity represents the local instantaneous amplitude of geopotential height fluctuations with periods shorter than 9 days. Regions with larger values of Ze show frequent cyclone passages and are regarded as a storm track. Finally, Ze was multiplied by a factor of sin(45°N)/sin(lat.) to mimic eddy amplitude in the geostrophic streamfunction. In the figures, we will plot monthly mean Ze values obtained from daily Ze data.

A monthly mean poleward (meridional) heat flux due to subweekly fluctuations ($\overline{v'T'}$) was also calculated with the 9-day high-pass-filtered meridional wind velocity and temperature at 850 hPa in daily mean data. To extract the long-wave component in the subweekly fluctuations, the flux data were smoothed by a 9-day low-pass filter once again. The method was the same as the calculation of the envelope function. This quantity represents the instantaneous storm track activity in terms of the baroclinic nature of migratory synoptic scale disturbances in the lower troposphere.

In the figures, we will plot the monthly mean of the poleward (meridional) heat flux values.

3.3 Results and discussion

3.3.1 Variations of dust and tritium in the Mount Wrangell ice core

YS2007 presented analysis of the dust amount and tritium concentrations in a 50-m ice core drilled at the summit of Mount Wrangell, Alaska (Figure 3.1). In Figure 3.1, clear dust and tritium peaks were seen in the spring of 2001 and 2002. The largest dust peaks were seen in fine and coarse dust in the early spring of 2002 and 2001, respectively (Figure 3.1). The variations of fine and coarse dust in the ice core had a high-positive correlation in the raw data [YS2007]. Hence fine and coarse dust vary together in annual cycles and their spring concentrations were higher. The 2001 dust peak in spring was discussed in YS2007 and may be mainly due to a perfect transpacific severe dust storm on April 6–7, 2001 [*Darmenova et al.*, 2005]. The 2002 dust peaks in spring have not been discussed before. Three larger coarse dust peaks with fine dust increasing from early spring to late spring in 2002 are seen in Figure 3.1. In 2002, four severe dust storms were observed in East Asia on 18(19)–22 March, 24–25 March, 5(6)–9 April, and 21–24 April [*Shao and Wang*, 2003; *Shao et al.*, 2003; *Zhou and Zhang*, 2003; *Sun et al.*, 2006]. No severe dust storms were recorded in May 2002 [*Sun et al.*, 2006; *Zhou and Zhang*, 2003]. *Sun et al.* [2006] mentioned that the dust storms on 18(19)–22 March and 5(6)–9 April were severe and that the area and intensity of dust emissions in the former case were wider and larger than those in the latter case. *Sugimoto et al.* [2003] also focused on dust storms on 20 March and 6 April 2002 and mentioned that the dust

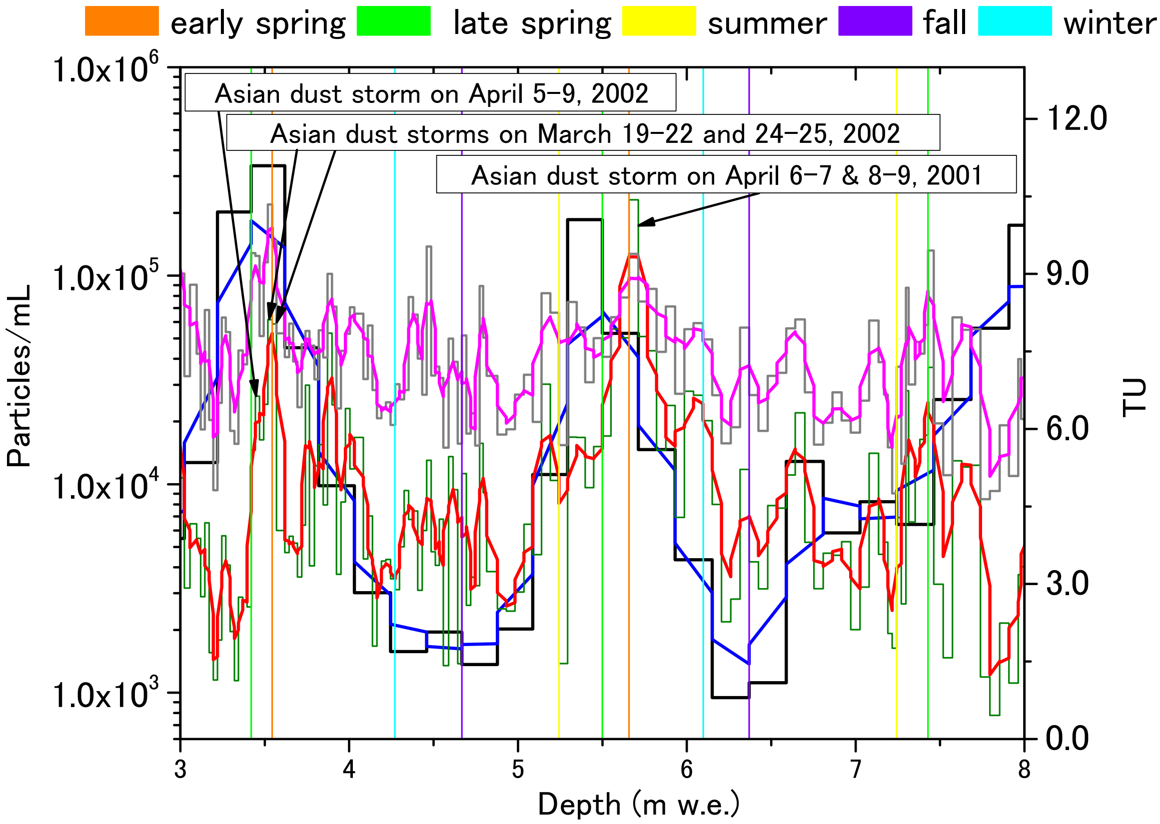


Figure 3.1. Recorded ice core data from 2000 to 2002 of fine dust (gray solid line; left side scale; size range of 0.52–1.00 μm), coarse dust (green solid line; left side scale; size range of 1.00–8.00 μm), and tritium concentrations (black solid line; right side scale) from Figure 2 of YS2007 (Figure 2.2). Solid lines in pink, red, and blue denote five data-point running means of fine dust, coarse dust, and tritium concentrations, respectively. The vertical orange, yellowish green, yellow, violet, and sky-blue solid lines denote the relative center positions of early spring, late spring, summer, fall, and winter, respectively. The written dates denote Asian dust contributions as discussed in section 3.3.

storm on 20 March was the largest in the last 10 years in Beijing; i.e., the 18(19)–22 March dust storm was the largest dust storm in 2002. If we consider four severe dust storms in the TOMS data, all of the Aerosol Index intensity data due to those dust storms showed transpacific transport and strong intensities (see Website 3.1). Except for the dust storm on 21–24 April, all of the dust was transported to higher latitudes, such as the Alaskan region. Hence the dust storms on 18(19)–22 March, 24–25 March, and 5(6)–9 April may be important and impact on the Alaskan ice core site. Therefore we will examine the detailed parts of those three dust storms in section 3.3.3.

In the ice core data from Mount Wrangell, dust peaks generally show spring maxima earlier than those of tritium in the raw data in Figure 2 of YS2007 (Figure 2.2). Time resolutions in dust and tritium analyses in the ice core were different because tritium analysis requires a large amount of ice core sample as melt water. Tritium time resolution was roughly 1 month and that of dust was roughly 3–12 days. Hence we cannot compare dust maxima with tritium maxima at the same time resolution. However, occasionally spring dust maxima are positioned within the range of tritium maxima in the raw ice core data in Figure 2 of YS2007 (Figure 2.2) (e.g., the dust peaks in the spring of 1995 and 2002; the 2002 case is shown in Figure 3.1). In addition, occasionally a spring dust peak was located within the second tritium maximum (e.g., the dust peaks in the spring of 1998). The spring maxima of dust and tritium in 2001 were positioned at different depths, and the spring maximum of dust in 2002 was within the tritium maximum in the spring of 2002. Hereafter, in the raw observed tritium data in Figure 3.1, TF2001 is the tritium first maximum in 2001, TS2001 is the tritium second maximum in 2001, TF2002 is the tritium first maximum in 2002, and

TS2002 is the tritium second maximum in 2002. Hence in sections 3.3.2–3.3.7, we will compare and discuss those differences in terms of meteorological analyses including case studies and the seasonal march of atmospheric circulation in the spring of 2001 and 2002.

Although spring dust peaks in the ice core are mainly due to Asian dust contributions as discussed in YS2007, tritium spring peaks are due to STT events, which are possibly not only STT due to Asian dust storms (hereafter, ADSTT) but also other STT events including STT due to eddy development and slow downward motion of meridional transport of the stratospheric air, called the Brewer–Dobson circulation [*Brewer*, 1949; *Dobson*, 1956] (hereafter, OTSTT). The OTSTT contribution to the ice core site cannot be excluded because STT also occurs in the North Pacific Ocean [*Sprenger and Wernli*, 2003]. It is worth noting that if there are no connections between ADSTT and OTSTT, the highest interannual correlation between fine dust and tritium fluxes in spring in YS2007 cannot be explained. The independent contribution of tritium due to OTSTT may disturb the high correlation. This is due to the high interannual correlation between fine dust and tritium fluxes in spring. For explaining the mechanism of dust and tritium variations in spring in the Mount Wrangell ice core and developing the hypothesis on the interannual connection between Asian dust and STT in Figure 9 of YS2007 (Figure 2.9), we investigate the details of ADSTT and OTSTT and discuss these in sections 3.3.2–3.3.7.

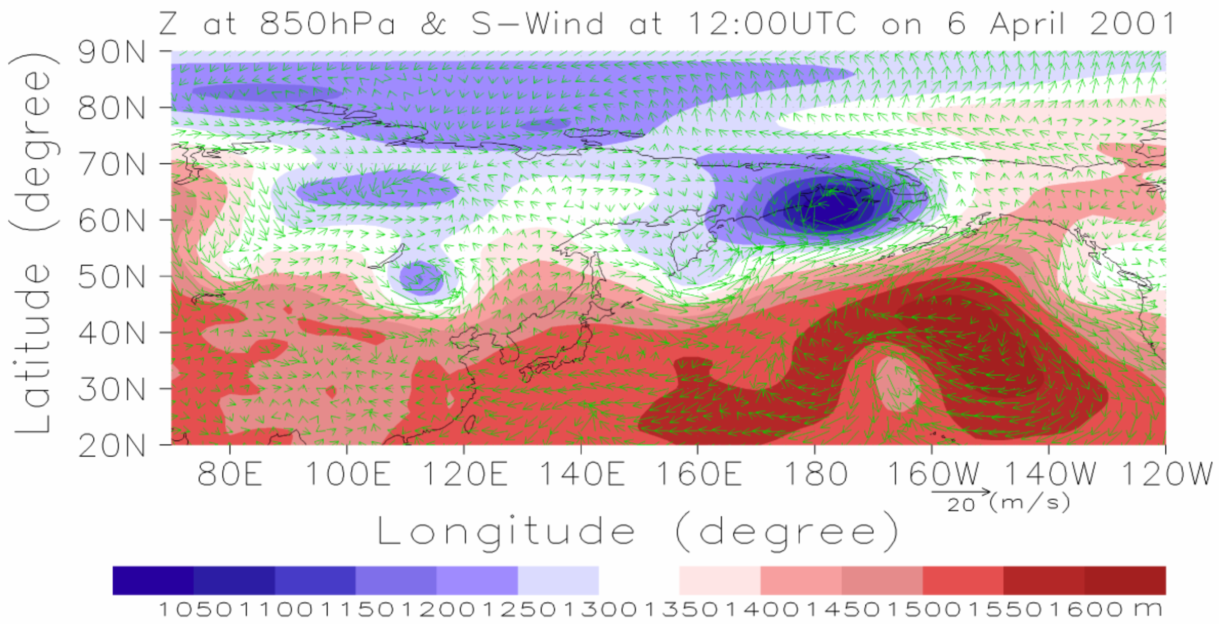
3.3.2 Simultaneous Asian dust storm and STT in April 2001

On 6–7 April 2001, a severe dust storm (a perfect dust storm) occurred in East Asia

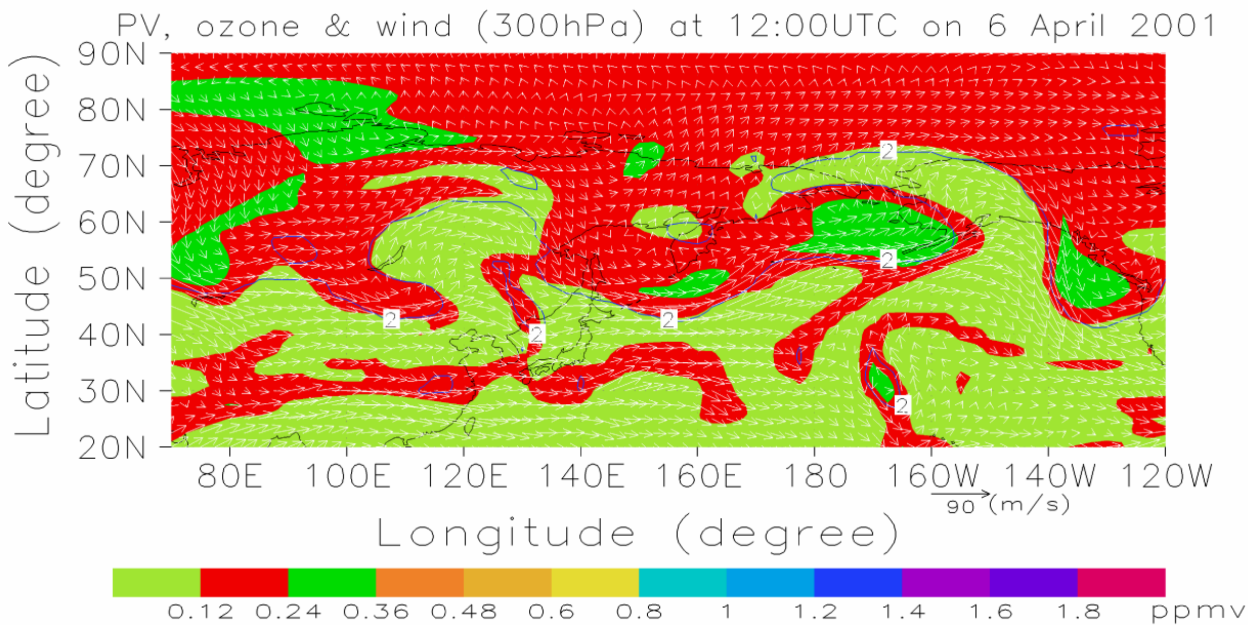
[*Darmenova et al.*, 2005]. The presence of the dust cloud over the North Pacific was verified by several satellite sensors such as the Moderate Resolution Imaging Spectroradiometer (MODIS), Total Ozone Mapping Spectrometer (TOMS) (Scientific Visualization Studio at NASA: http://svs.gsfc.nasa.gov/vis/a000000/a002800/a002860/; hereafter, Website 3.2), and the dust storm was mentioned in the NASA science news on 17 May 2001 (see the web site at: http://science.nasa.gov/headlines/). The dust cloud over the Gulf of Alaska on 12–13 April was detected based on TOMS data [*Darmenova et al.*, 2005]. *Cahill* [2003] mentioned that the dust from East Asia was transported in the lower troposphere and reached Adak Island, Alaska, which is one of the southernmost Aleutian Islands, on 13 April 2001. The dominant dust was coarse with a size of more than 1.15 μm. *Zdanowicz et al.* [2006] also detected the Asian dust at higher altitudes in the St. Elias Mountains, Yukon Territory, Canada, and its size was also coarser. YS2007 also showed a drastic increase in the dust concentration in coarser sizes (1.00–8.00 μm) in the spring of 2001 in the ice core from Mount Wrangell, Alaska, which may correspond to this dust storm.

At 12:00 UTC on 6 April 2001, a cyclone was developing in the area extending from approximately 40–50°N to 100–120ºE (Figure 3.2a). Stratospheric ozone advection into the troposphere along the 2 PVU tropopause line was seen from northwest to southeast in this area (Figure 3.2b, 3.2c, and 3.2d). We found a tropopause folding at the south-southwest side of this cyclone with strong downward wind (Figure 3.2c and 3.2d). Surface wind was very strong in this area (Figure 3.2a), and the dust storm broke out in this area on 6 April [*Darmenova et al.*, 2005]. The stratospheric air moved into the troposphere along the 310 K isentropic surface with strong

(a)


Z at 850hPa & S-Wind at 12:00UTC on 6 April 2001
Latitude (degree)
90N
80N
70N
60N
50N
40N
30N
20N
80E 100E 120E 140E 160E 180 160W 140W 120W
20 (m/s)
Longitude (degree)
1050 1100 1150 1200 1250 1300 1350 1400 1450 1500 1550 1600 m

(b)


PV, ozone & wind (300hPa) at 12:00UTC on 6 April 2001
Latitude (degree)
90N
80N
70N
60N
50N
40N
30N
20N
80E 100E 120E 140E 160E 180 160W 140W 120W
90 (m/s)
Longitude (degree)
0.12 0.24 0.36 0.48 0.6 0.8 1 1.2 1.4 1.6 1.8 ppmv

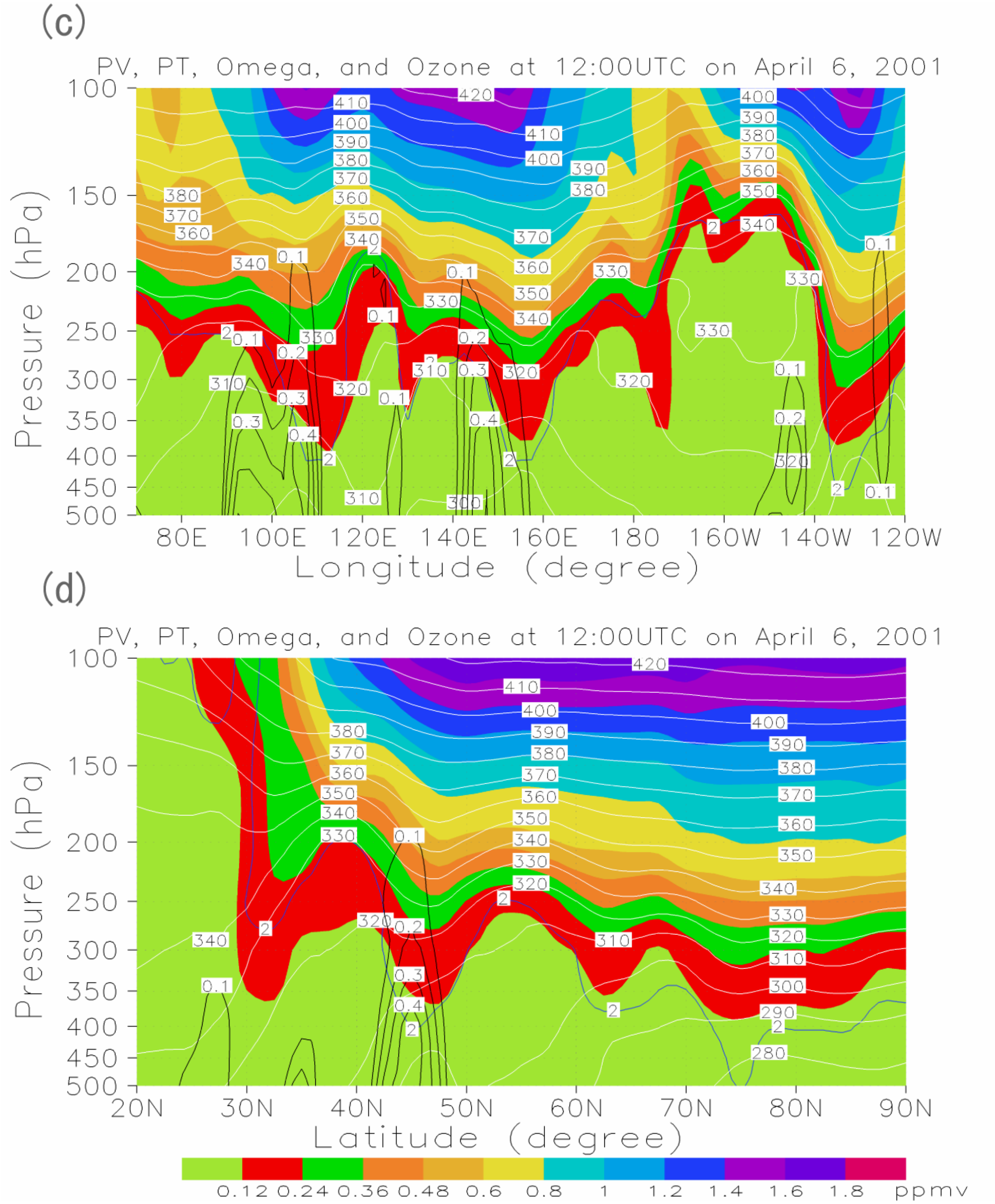


Figure 3.2. Meteorological conditions for the Asian dust storm at 12:00 UTC on 6 April 2001. (a) Geopotential height at 850 hPa (shaded contour; unit: m) and surface wind at 10 m (green vector; unit: m s^{-1}). (b) Ozone volume mixing ratio (color shaded contour; unit: ppmv), 2 PVU tropopause line (blue solid line), and wind (white vector; unit: m s^{-1}) at 300-hPa level. (c) Ozone volume mixing ratio (color shaded contour; unit: ppmv), potential temperature (PT; white solid line; unit: K), and vertical p-velocity, omega (black contour; only downward components are plotted; unit: Pa s^{-1}), at the 45°N line for the cross section between longitude and pressure level (<500 hPa). (d) Same as (c), but for the 107.5°E line for the cross section between latitude and pressure level (<500 hPa). The data grid is interpolated to a finer grid using cubic interpolation.

downward wind (Figure 3.2c and 3.2d). Then the large dust cloud due to this Asian dust storm reached the Gulf of Alaska on 12–13 April 2001 (see Website 3.2).

Figure 3.3 shows the 8-day forward trajectories starting from the dust storm area at 250–700 hPa in East Asia, which finally reached the Wrangell Area. The trajectories in the lower free troposphere passed near the boundary layer over the east side of the Eurasian continent during the first 2 days (Figure 3.3-a2 and 3.3-b2), and these corresponded to the pathway of the dust cloud from East Asia to the North Pacific region in shown Websites 3.1 and 3.2. The air mass from the lower troposphere reached near the summit of Mount Wrangell on 14 April (red and deep blue colored trajectories in Figure 3.3-a1 and 3.3-b1, respectively). Thus we have verified the Asian dust transport due to the 6–7 April dust storm in 2001 in terms of trajectory analysis. Hence we conclude that the 2001 coarse dust peak with fine dust increase shown in Figure 3.1 is mainly due to this severe dust storm in East Asia. The results of *Zdanowicz et al.* [2006], YS2007, and this study imply that long-range transport of coarse Asian dust in spring may be regarded as an ordinary phenomenon at higher altitude in the North Pacific region.

On 6 April, part of the stratospheric air parcels in the initial area rapidly moved into the lower free troposphere (Figure 3.2c and 3.2d) within 1 day because of the developing cyclone, which corresponds to the yellow colored trajectories in Figure 3.3-a2 and the southward branch of red colored trajectories in Figure 3.3-b1 and 3.3-b2. Most of the other branches shown as red trajectories traveling toward northward in Figure 3.3-b1 (near 60ºN) remained mainly in the upper troposphere for a few days after the Asian dust storm (Figure 3.3-a2). From 12:00 UTC on 9 April

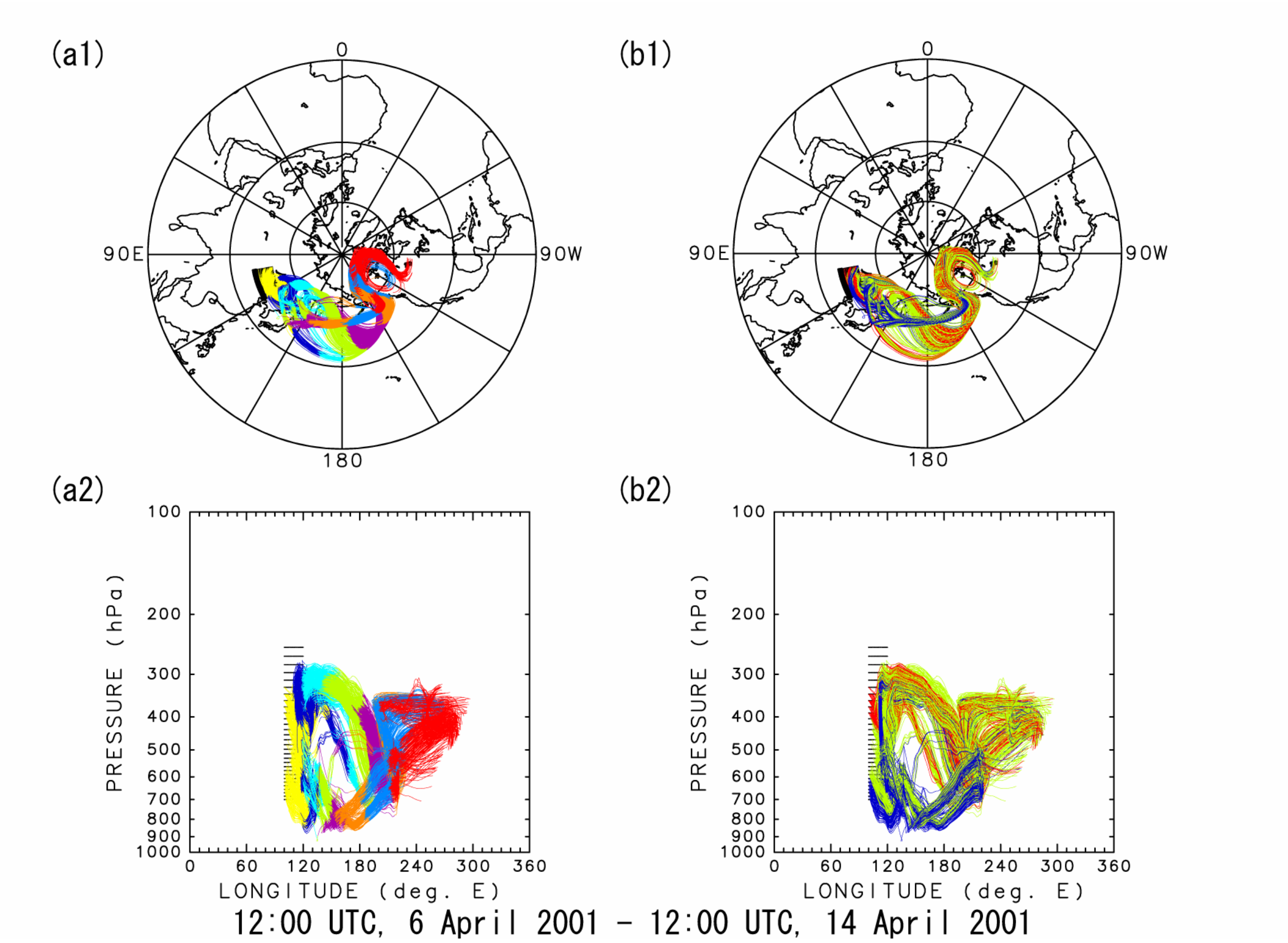


Figure 3.3. Recorded trajectories from the initial area of the Asian dust storm at 12:00 UTC on 6 April 2001 to the Wrangell Area. Left side figures (a1) and (a2) denote the trajectories in each day (each color shows 1 day transport length). Right side figures (b1) and (b2) denote the trajectories in each initial pressure level defined as follows: the initial air mass positions in the 700–600 hPa layer, 600–400 hPa layer, and 400–250 hPa layer were indicated in deep blue, yellowish green, and red, respectively. Only the air parcels that reached the Mount Wrangell Area are drawn.

to 12:00 UTC on 11 April, the air mass in the upper troposphere was transported rapidly to the middle troposphere corresponding to the yellowish green and purple trajectories downstream in Figure 3.3-a2. That was due to a cyclone developing around the Alaska Peninsula intensifying the downward motion of the air mass at the southwest end of the trough (Figure 3.4). This cyclone functioned effectively as an escalator transporting the air mass from the upper troposphere to the middle troposphere. In the process of downward motion, the advection of other stratospheric-origin materials into this air mass due to this developing cyclone (OTSTT) was also considered. The stratospheric-origin air masses due to both the Asian dust storm (ADSTT) and this developing cyclone (OTSTT) in the North Pacific met here near 180ºE. Then, both the stratospheric origin air masses mixed and the major parts of the air passed the Mount Wrangell Area on 12 April. Some parts returned above the summit of Mount Wrangell from the Arctic region on 14 April (the upper red trajectories in Figure 3.3-a2 and the thin parts of red trajectories in Figure 3.3-a1 and 3.3-b1). The dust cloud reached the Wrangell Area on 14 April 2001. The most important point here is that the Asian dust storm and the STT broke out simultaneously, but the major parts of the dust cloud and the stratospheric air masses were transported along different pathways and on different dates (two-day time lag).

Zdanowicz et al. [2006] mentioned that the Asian dust fell in the St. Elias Mountains due to snowfall and the assumed deposition dates were between 11 April and 19 April 2001. Mount Wrangell and Mount Logan in the St. Elias Mountains are very close, and both sites are located at higher altitudes. The distance between them is approximately 240 km. Hence synoptic-scale

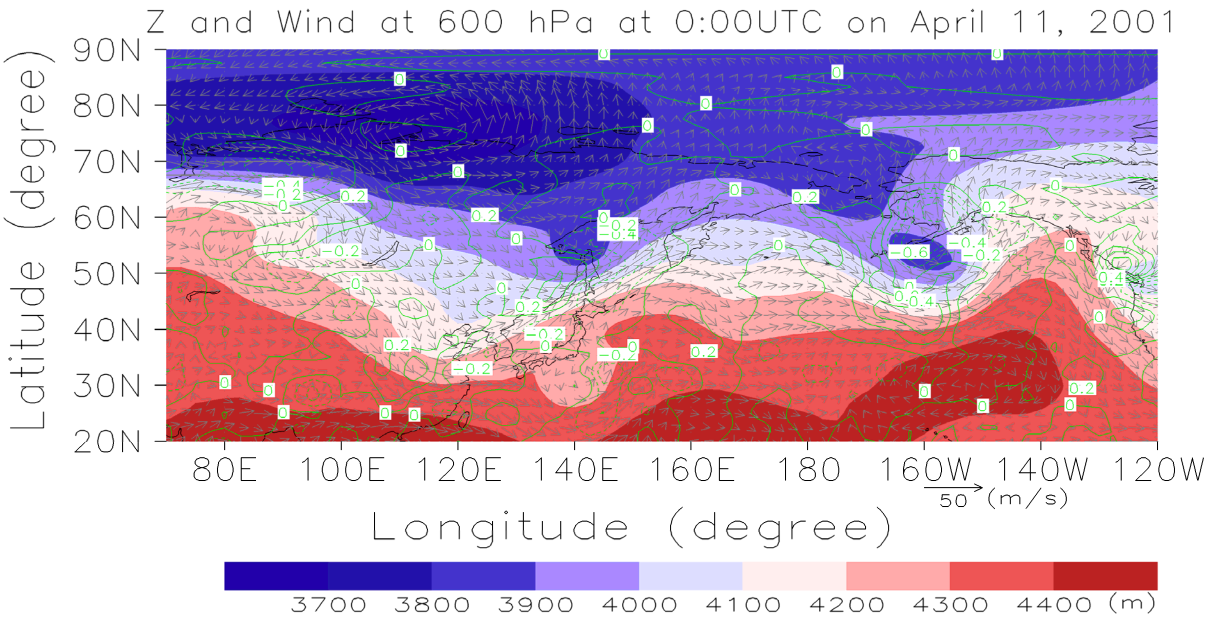


Figure 3.4. Geopotential height (color shaded contour; unit: m), horizontal wind (gray vector; unit: m s^{-1}), and vertical p-velocity, omega (green contour; unit: Pa s^{-1}), at the 600-hPa pressure level at 0:00 UTC on 11 April 2001. The data grid is interpolated to a finer grid using cubic interpolation.

atmospheric circulation around Mount Wrangell and Mount Logan may affect both sites simultaneously.

We also checked the relative humidity and ozone of ERA-40 at and above the Mount Wrangell ice core site when dust and the stratospheric tracers were transported (Figure 3.5). Our ice core site at Mount Wrangell (4100 m asl) roughly corresponds to a 600-hPa surface. If we consider the trajectories in the upper transport pathways (red and yellowish green trajectories in Figure 3.3-b2), those including the stratospheric air mass reached the Wrangell Area on 12 April 2001 (the boundary between orange and blue trajectories in Figure 3.3-a1). On 12 April, a relatively dry air zone with increased ozone was seen above Mount Wrangell (Figure 3.5). These ozone increments were due to the air mass transport of stratospheric origin mixed with the ADSTT on 6–7 April and the OTSTT on 9–11 April near 180º. After 12 April, the air of stratospheric origin turned around one turn clockwise (Figure 3.3-a1 and 3.3-b1), then the dust cloud with relatively higher moist air was transported to Mount Wrangell on 14 April because the dust cloud passed through the lower free troposphere above the North Pacific Ocean. Most of the upper trajectories reached Canada through the Arctic Ocean. Then, that part of the air trajectories from the upper troposphere returned above the Mount Wrangell ice core site. However, dry stratospheric origin air stayed above about 400 hPa (Figures 3.3-a2 and 3.5). Air with high humidity was present below about 400 hPa on 13–15 April. It was not efficient for tritium deposition due to snowfall because tritium mainly exists as tritiated water vapor in the atmosphere [*Gat et al.*, 2001] and may preferentially undergo wet deposition [YS2007]. Next, we will compare the relative humidity variation at Mount Wrangell in

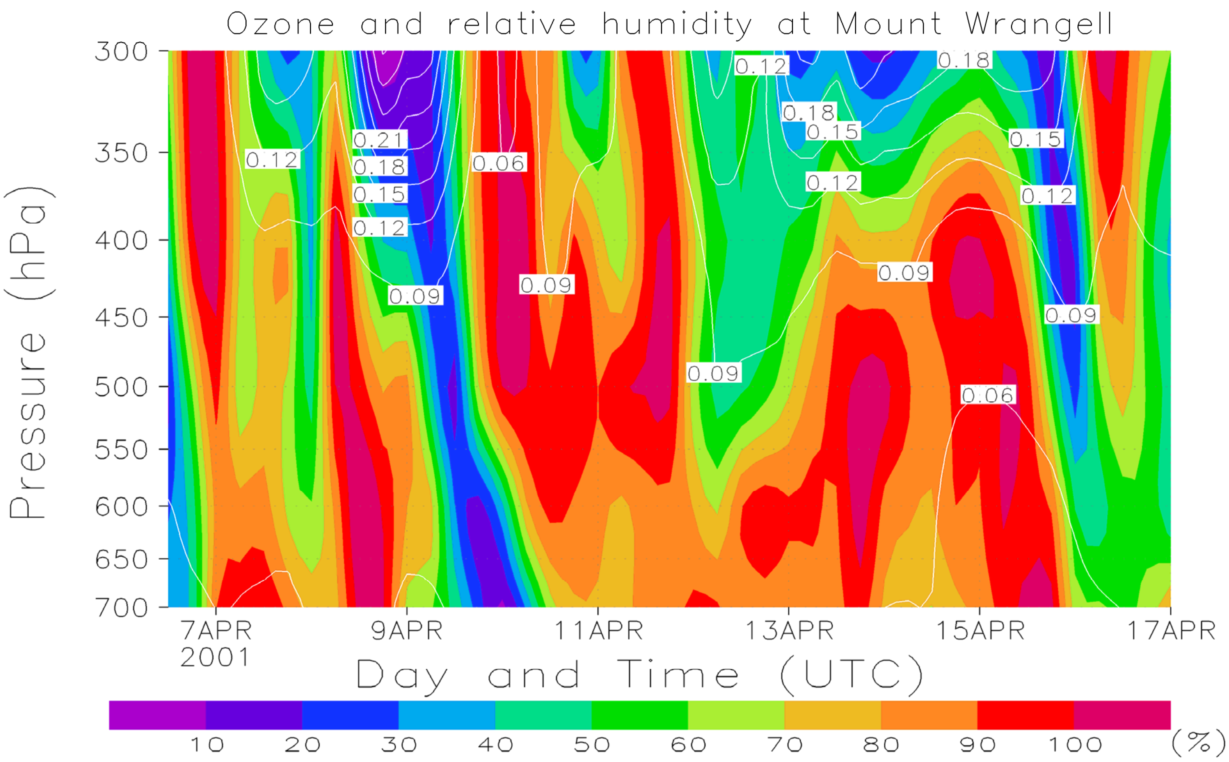


Figure 3.5. Ozone volume mixing ratio (white contour; unit: ppmv) and relative humidity (shaded contour; unit: %) at Mount Wrangell (nearest grid point in ERA40 data is 62.5°N, 145°W) from 12:00 UTC on 6 April 2001 to 0:00 UTC on 17 April 2001. The summit of Mount Wrangell is roughly at the 600-hPa level. The data grid is interpolated to a finer grid using cubic interpolation.

April 2001 with those in May 2004 when it was snowing at the ice core site.

From noon, 13 April 2001 to 14 April 2001, the relative humidity exceeded 80% below 400 hPa at Mount Wrangell (Figure 3.5). We compared this higher relative humidity with those in the field campaign in May 2004 at the summit of Mount Wrangell. When we stayed at the summit of Mount Wrangell in May 2004 (see Figure 4 of YS2007; Figure 2.4), two snowfall events occurred (Figure 3.6a, 3.6b-2, and 3.6b-4). The first snow event was from 16 May to 18 May (17 May to 18 May) in Alaskan local time (UTC), and the second snow event was from 22 May to 26 May (Alaskan local time and UTC). When snow was falling, the relative humidity exceeded 80%, which was verified in the GPV data from JMA (Figure 3.6). These relative humidity data reproduced the time series of weather changes at the summit of Mount Wrangell relatively well. The summit of Mount Wrangell is very flat (Figure 1 of YS2007; Figure 2.1), and synoptic scale atmospheric circulation impacts strongly on the flat ice core site. In conclusion, relative humidity greater than 80% in grid point atmospheric data may become a good snowfall index at the summit of the Mount Wrangell ice core site. Hence we will adopt this relative humidity greater than 80% in grid point data sets such as GPV or ERA-40 as a snowfall index at the summit of Mount Wrangell from now on.

When the relative humidity was greater than 80% on 13–15 April 2001, the substances below about 400 hPa at Mount Wrangell might have been deposited onto the summit due to snowfall (Figure 3.5). The dust cloud existed below about 400 hPa (blue trajectories in Figure 3.3-b2) and might have been deposited due to snowfall. However, the stratospheric air on 13–14 April mainly

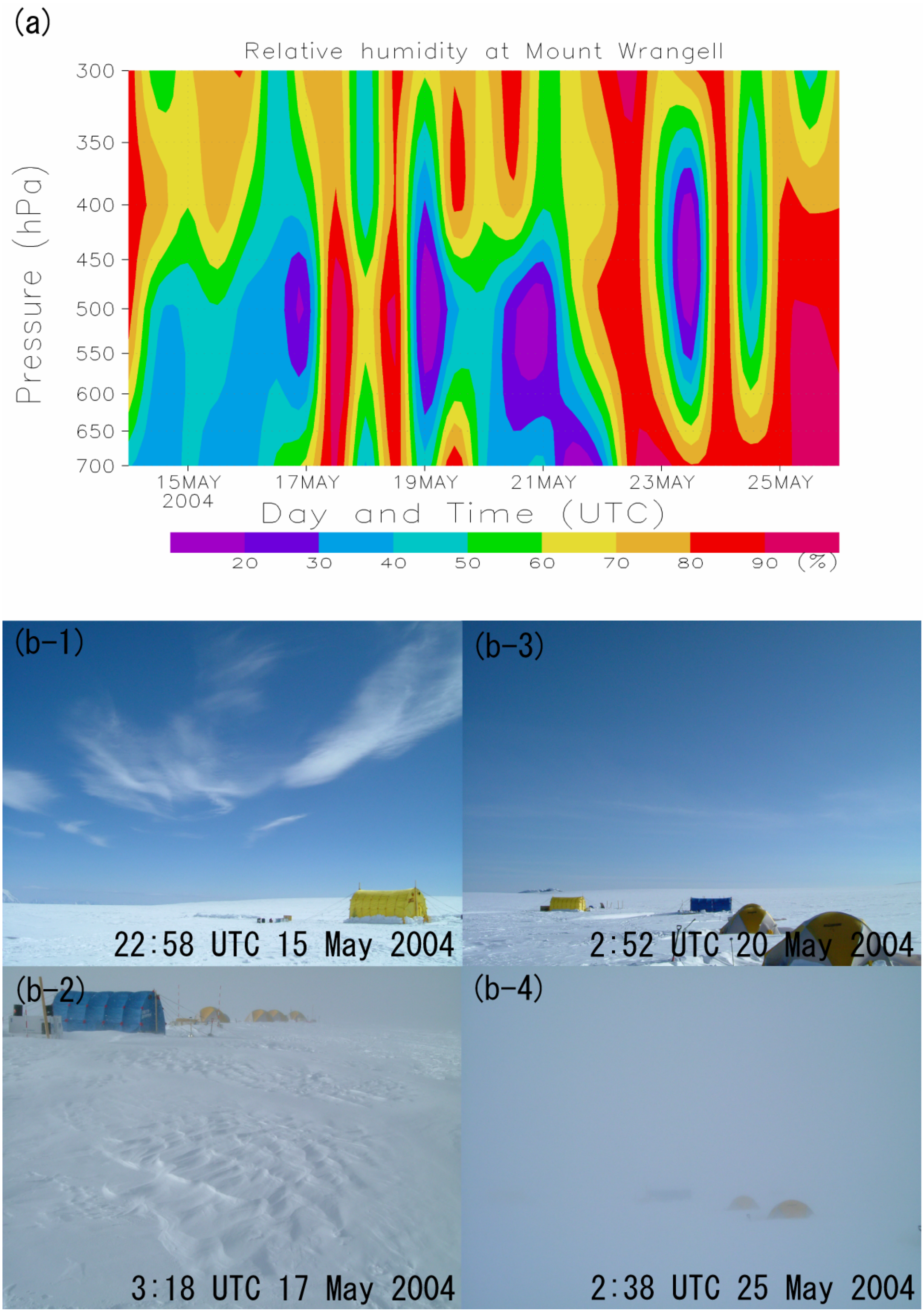
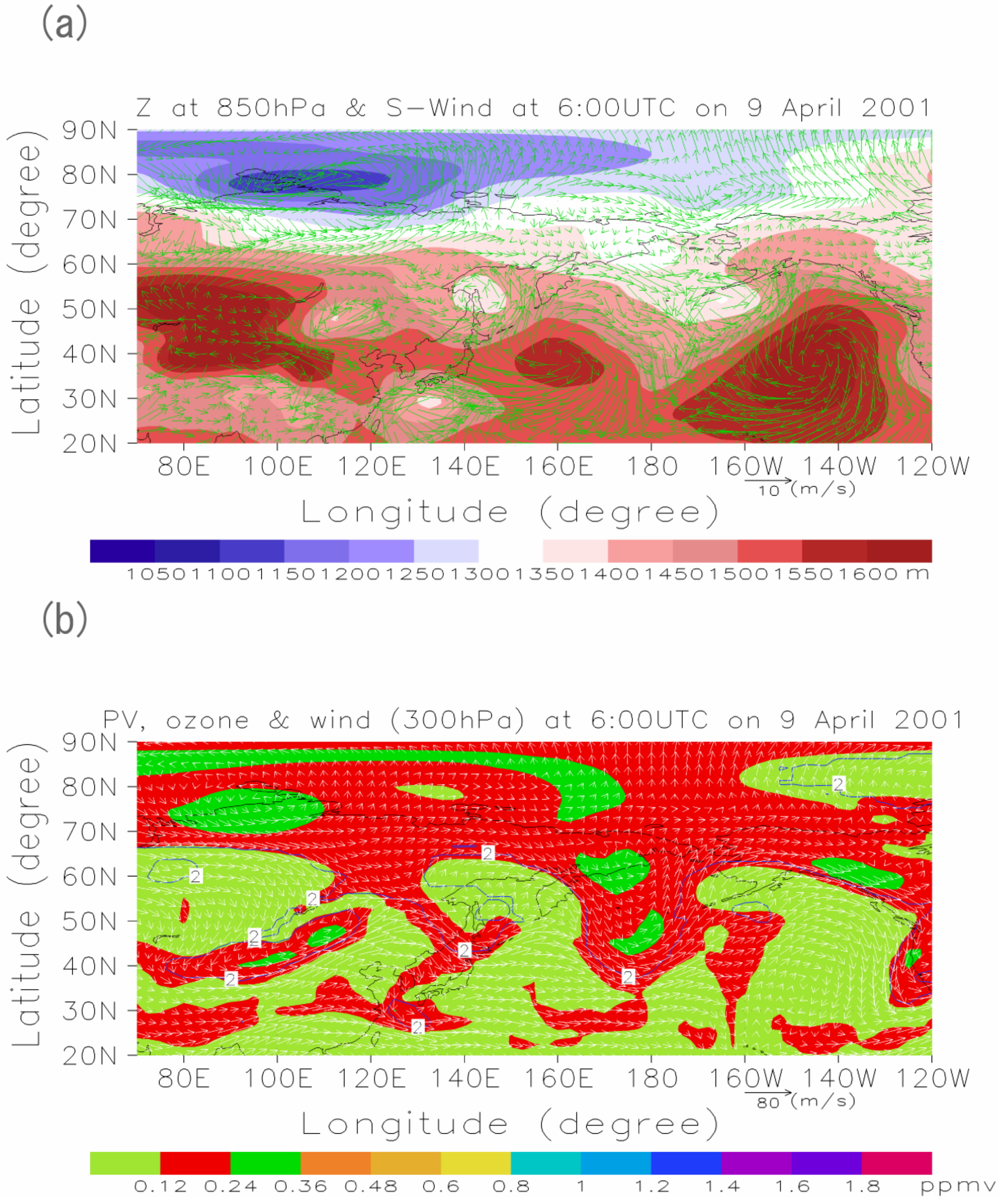


Figure 3.6. Weather conditions at Mount Wrangell (nearest grid point in GPV data is 62.5°N, 143.75°W) from 0:00 UTC on 14 May 2004 to 0:00 UTC on 26 May 2004. (a) Relative humidity (unit: %) at Mount Wrangell. The term approximately corresponds to the field observations at the summit of the Mount Wrangell ice core site in Figure 4 of YS2007 (Figure 2.4). The data grid is interpolated to a finer grid using cubic interpolation. (b) A few snapshots corresponding to Figure 3.6a. Figure 3.6b-2 and 3.6b-4 correspond to fine and snowy conditions, respectively. Note that Alaskan local time in Figure 4 of YS2007 (Figure 2.4) corresponds to UTC time minus 8 h.

stayed above 400 hPa and was dry (red trajectories above 400 hPa in Figure 3.3-a2). Hence snow deposition may have mainly occurred below 400 hPa. As mentioned above, tritium prefers wet (snow) deposition. The contribution of stratospheric air to the site was strongest on 12 April because of the major trajectories passed in the Mount Wrangell Area on that day. On 12 April, however, relatively dry air with increased ozone was dominant above the ice core site; therefore, strong snowfall might not have occurred (Figure 3.5). As a result, tritium deposition in this event may be considerably lower than in clear snowfall cases. Hence the tritium concentration corresponding to TS2001 might not have increased too much and was not the highest in 2001. However, the TS2001 peak was considerably higher than that of the background tritium level (Figure 3.1). Then, the OTSTT contribution to TS2001 is also considered for raising tritium background level as mentioned later.

On 8–9 April 2001, the second dust storm broke out in the same region as it followed the dust storm on 6–7 April (Figure 3.7). Then we also found that tropopause folding and STT occurred in the same region as for the case of the 6–7 April dust storm (Figure 3.7b, 3.7c, and 3.7d). We calculated the 6-day forward trajectories from 6:00 UTC on 9 April 2001 to 6:00 UTC on 14 April 2001. However, only a small number of trajectories from the dust storm transported to the Wrangell Area were present together with air mass contributions from the lower troposphere (Figure 3.8). The trajectory pathway is the same as that of the 6–7 April dust storm. These results imply that dust contribution to the Wrangell Area in this case is expected, whereas the stratospheric air contribution is expected to be less.


(a)
Z at 850hPa & S-Wind at 6:00UTC on 9 April 2001
Latitude (degree)
90N
80N
70N
60N
50N
40N
30N
20N
80E 100E 120E 140E 160E 180 160W 140W 120W
10 (m/s)
Longitude (degree)
1050 1100 1150 1200 1250 1300 1350 1400 1450 1500 1550 1600 m
(b)
PV, ozone & wind (300hPa) at 6:00UTC on 9 April 2001
Latitude (degree)
90N
80N
70N
60N
50N
40N
30N
20N
80E 100E 120E 140E 160E 180 160W 140W 120W
80 (m/s)
Longitude (degree)
0.12 0.24 0.36 0.48 0.6 0.8 1 1.2 1.4 1.6 1.8 ppmv

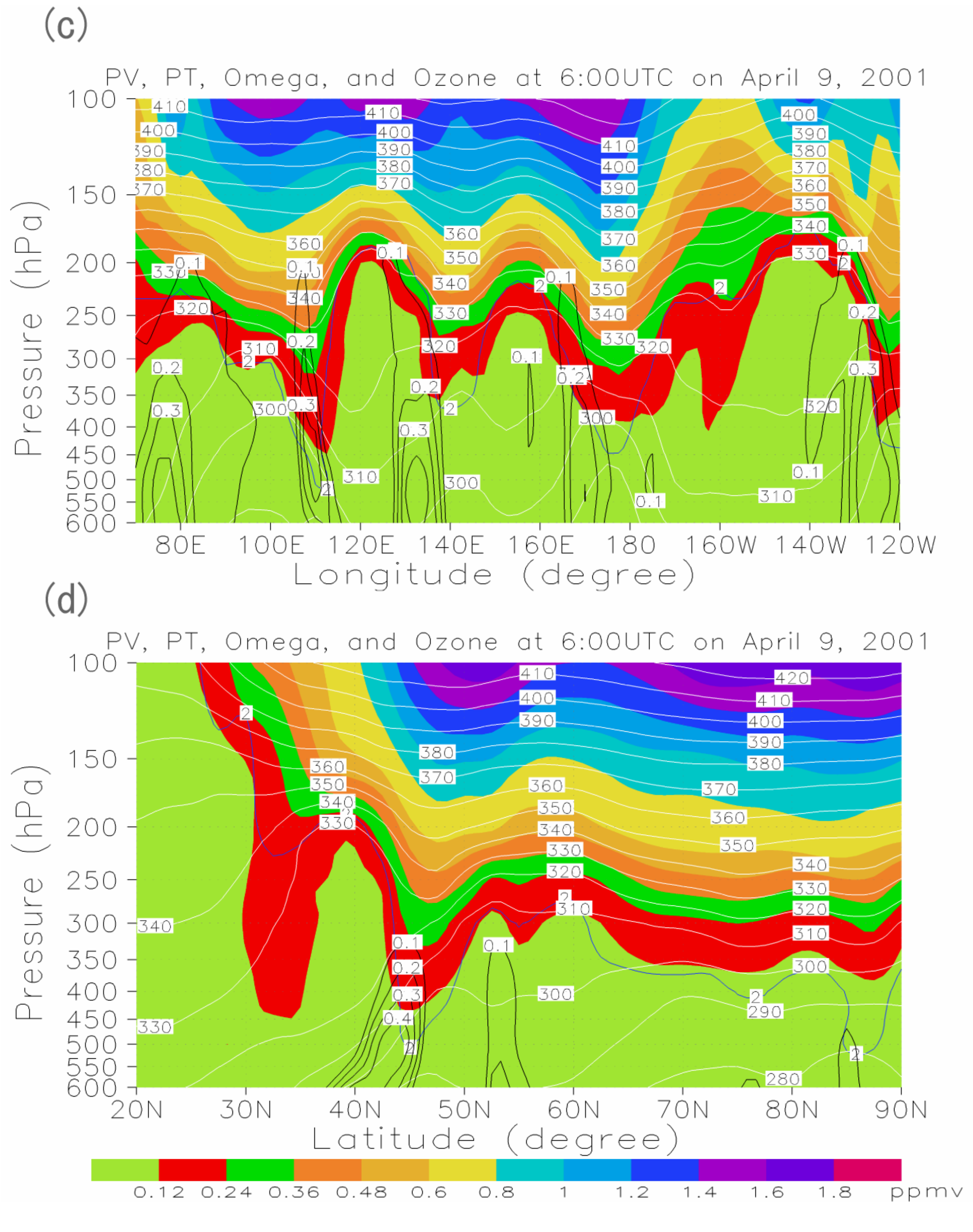


Figure 3.7. Similar to Figure 3.2 but for the Asian dust storm at 6:00 UTC on 9 April 2001. (c) is at the 45°N line for the cross section between longitude and pressure level (<600 hPa). (d) Same as (c), but for at the 110°E line for the cross section between latitude and pressure level (<600 hPa).

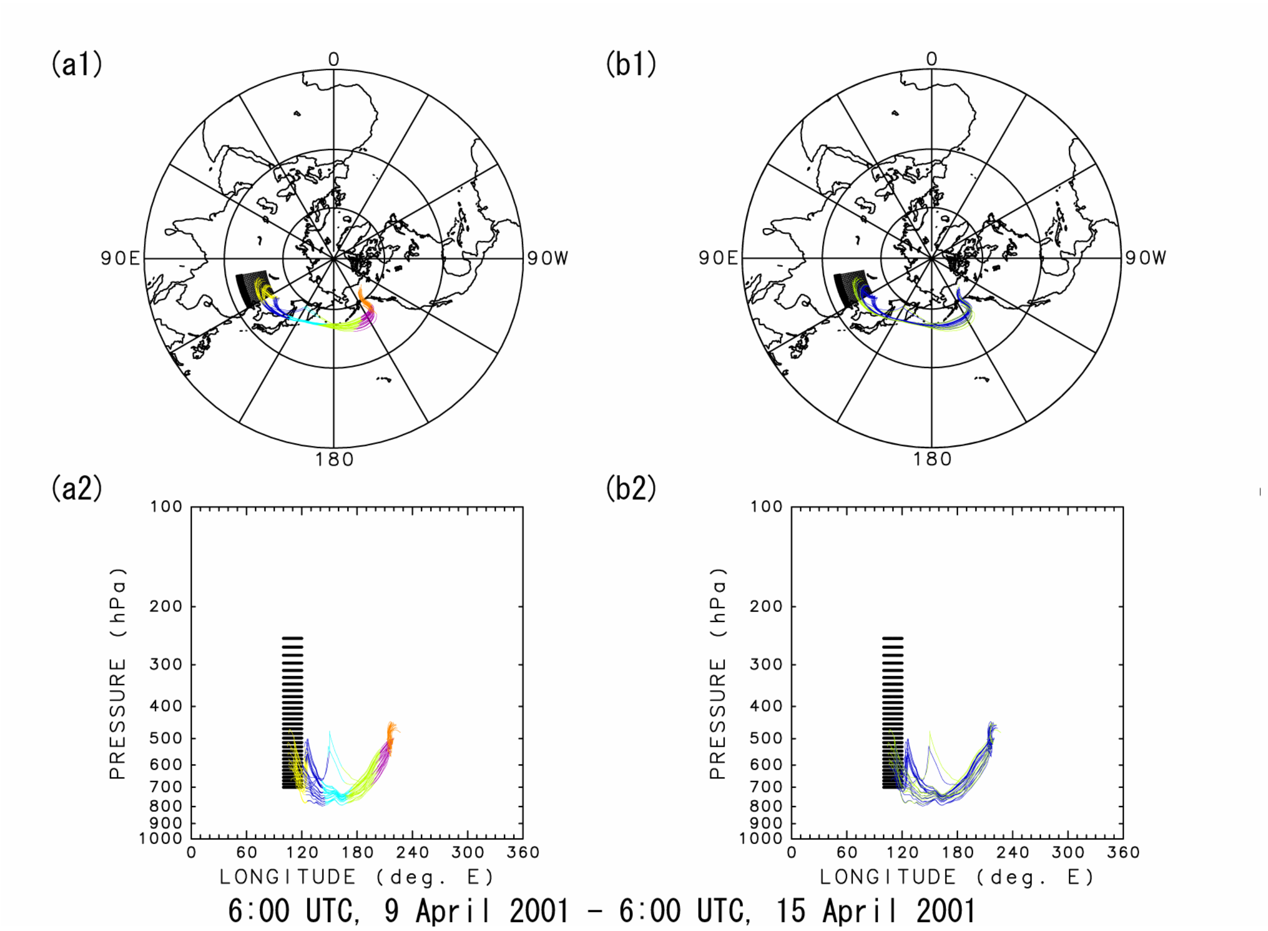


Figure 3.8. Similar to Figure 3.3 but for the Asian dust storm at 6:00 UTC on 9 April 2001.

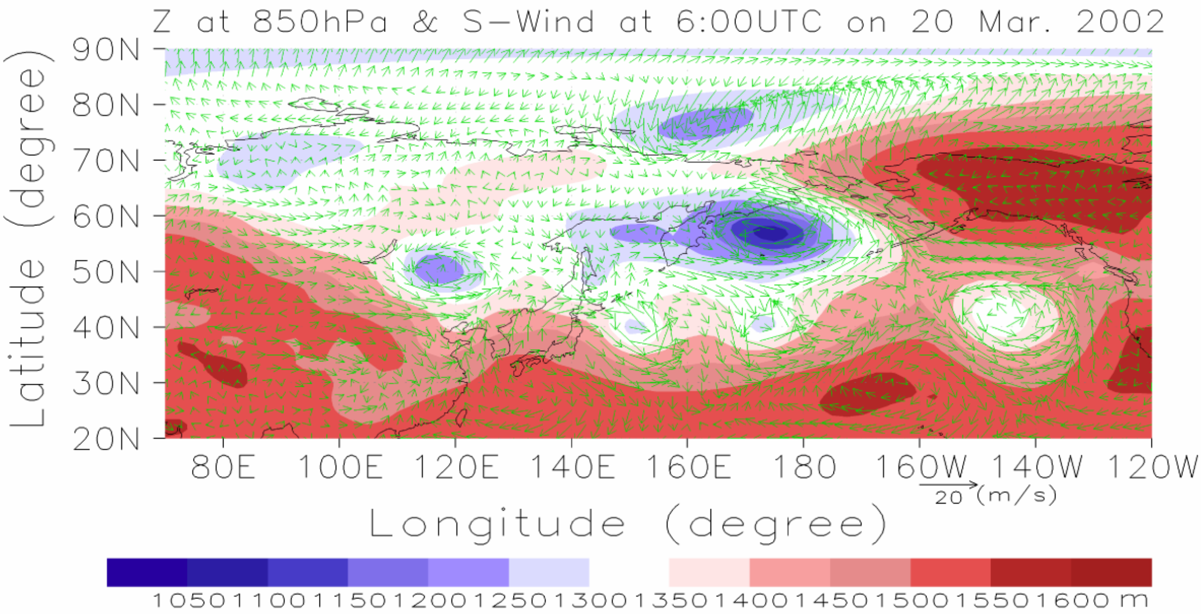
We summarize the impact of Asian dust and ADSTT onto the ice core site in the spring of 2001 as follows: (1) Asian dust was transported to the ice core site by the Asian dust storm on 6–7 and 8–9 April; (2) the ADSTT on 6–7 April impacted on the TS2001 peak in Figure 3.1 to some extent but was not expected to be perfect because of the absence of snowfall above the ice core site; and (3) the impact of ADSTT to the ice core site on 8–9 April was not expected. The lower humidity above 500 hPa on 12–13 April and above 400 hPa at Mount Wrangell on 13–14 April 2001 did not result in efficient tritium deposition on the Mount Wrangell ice core site, as mentioned in the previous paragraph. As a result, the tritium concentration in the ice core might not be the highest in 2001. Next, we discuss a case of simultaneous Asian dust outbreak and STT in April 2002.

3.3.3 Simultaneous Asian dust storm and STT in March and April 2002

On 20 March 2002, a cyclone was deepening on the southeast side of Lake Baikal (Figure 3.9a) and severe dust broke out from the Gobi desert [*Sugimoto et al.*, 2003; *Sun et al.*, 2006]. Then a deep STT occurred with strong downward wind on the south side of the cyclone (Figure 3.9b, 3.9c, and 3.9d). The ozone intrusion from the stratosphere and vertical strong wind contours corresponded well (Figure 3.9c and 3.9d). One of the stratospheric tracers, tritium, might also have intruded into the troposphere in this region at this time. The STT of this case was the deepest in terms of downward wind velocity and ozone descending to the 750 hPa layer.

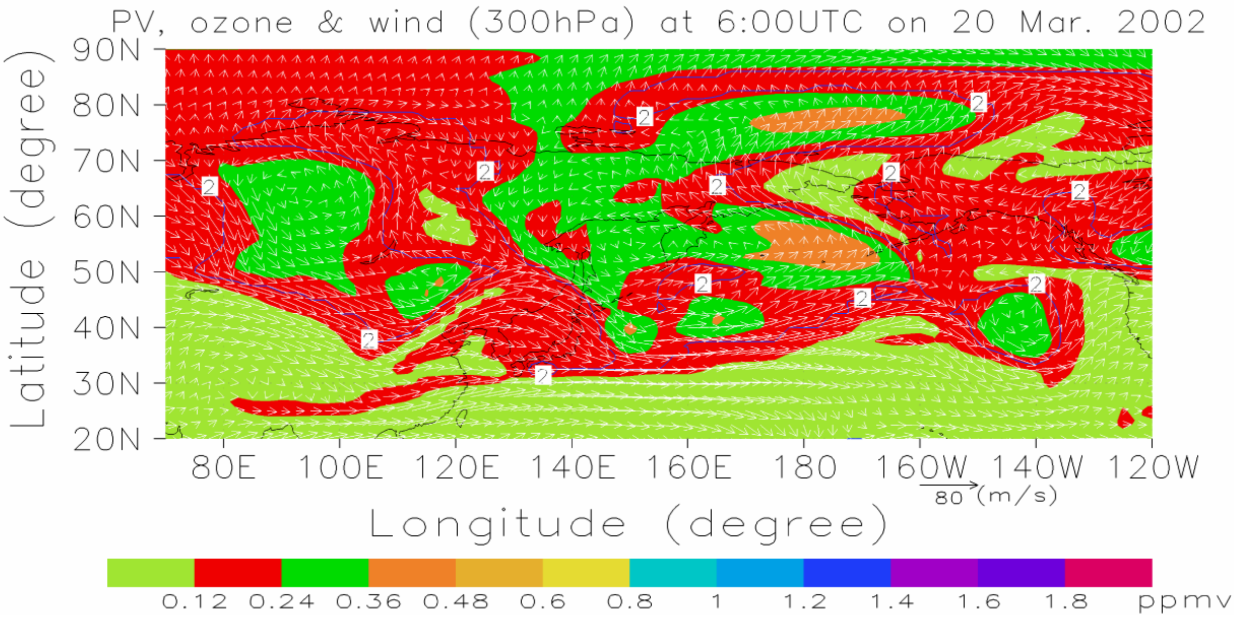
The trajectory was calculated from the initial region of the dust storm (Table 3.1). On the

(a)



Z at 850hPa & S-Wind at 6:00UTC on 20 Mar. 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E
100E
120E
140E
160E
180
160W
140W
120W
20 (m/s)
Longitude (degree)
1050 1100 1150 1200 1250 1300 1350 1400 1450 1500 1550 1600 m

(b)



PV, ozone & wind (300hPa) at 6:00UTC on 20 Mar. 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E
100E
120E
140E
160E
180
160W
140W
120W
80 (m/s)
Longitude (degree)
0.12 0.24 0.36 0.48 0.6 0.8 1 1.2 1.4 1.6 1.8 ppmv

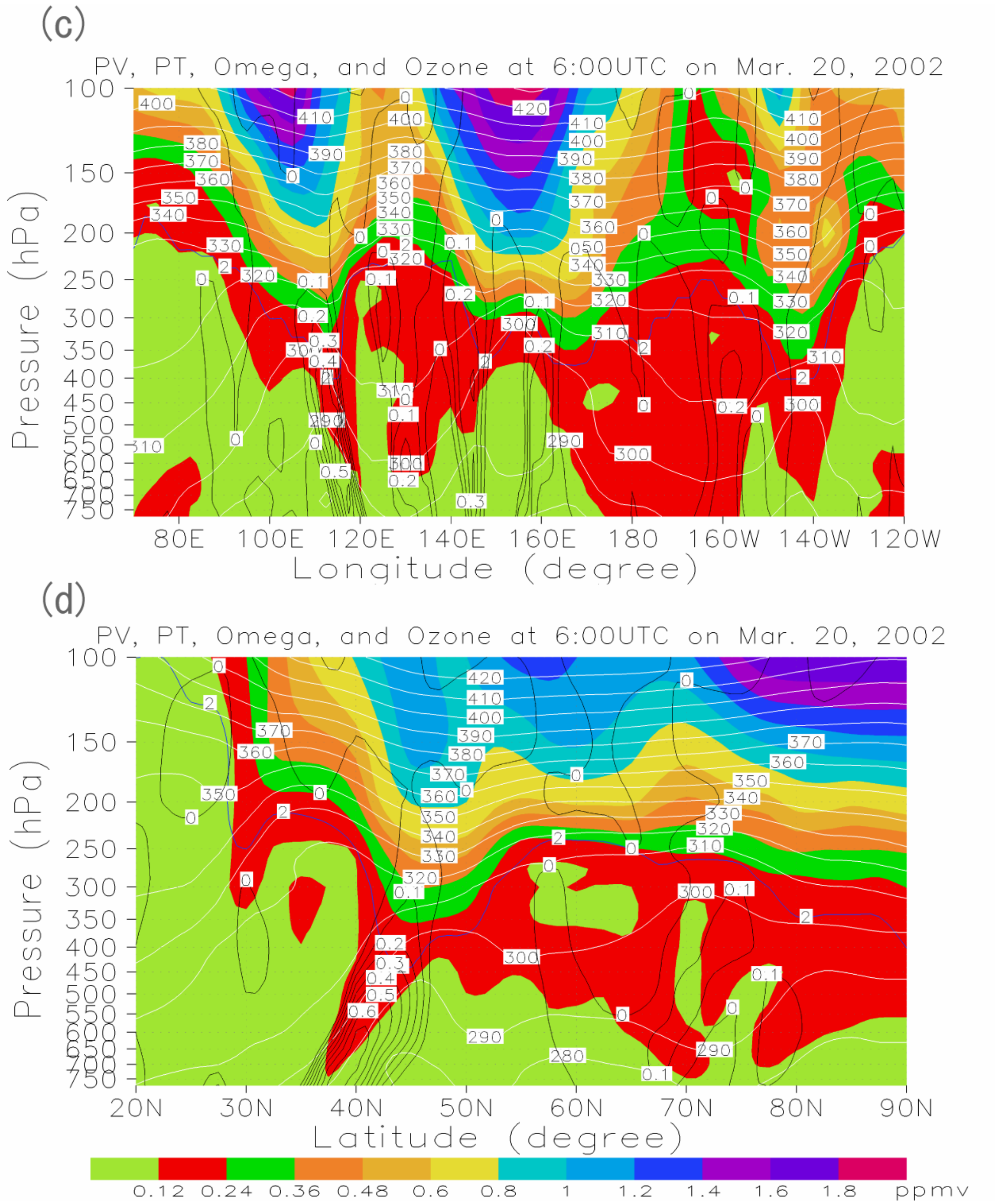


Figure 3.9. Similar to Figure 3.2 but for the Asian dust storm at 6:00 UTC on 20 March 2002. (c) is at the 42.5°N line for the cross section between longitude and pressure level (<775 hPa). (d) Same as (c), but for the 115°E line for the cross section between latitude and pressure level (<775 hPa).

transport pathways to Mount Wrangell, the air masses starting from the lower troposphere and the lower stratosphere were well mixed vertically. The trajectories did not branch off, which is an important difference from the case of the 6–7 April 2001 dust storm (Figures 3.3 and 3.10).

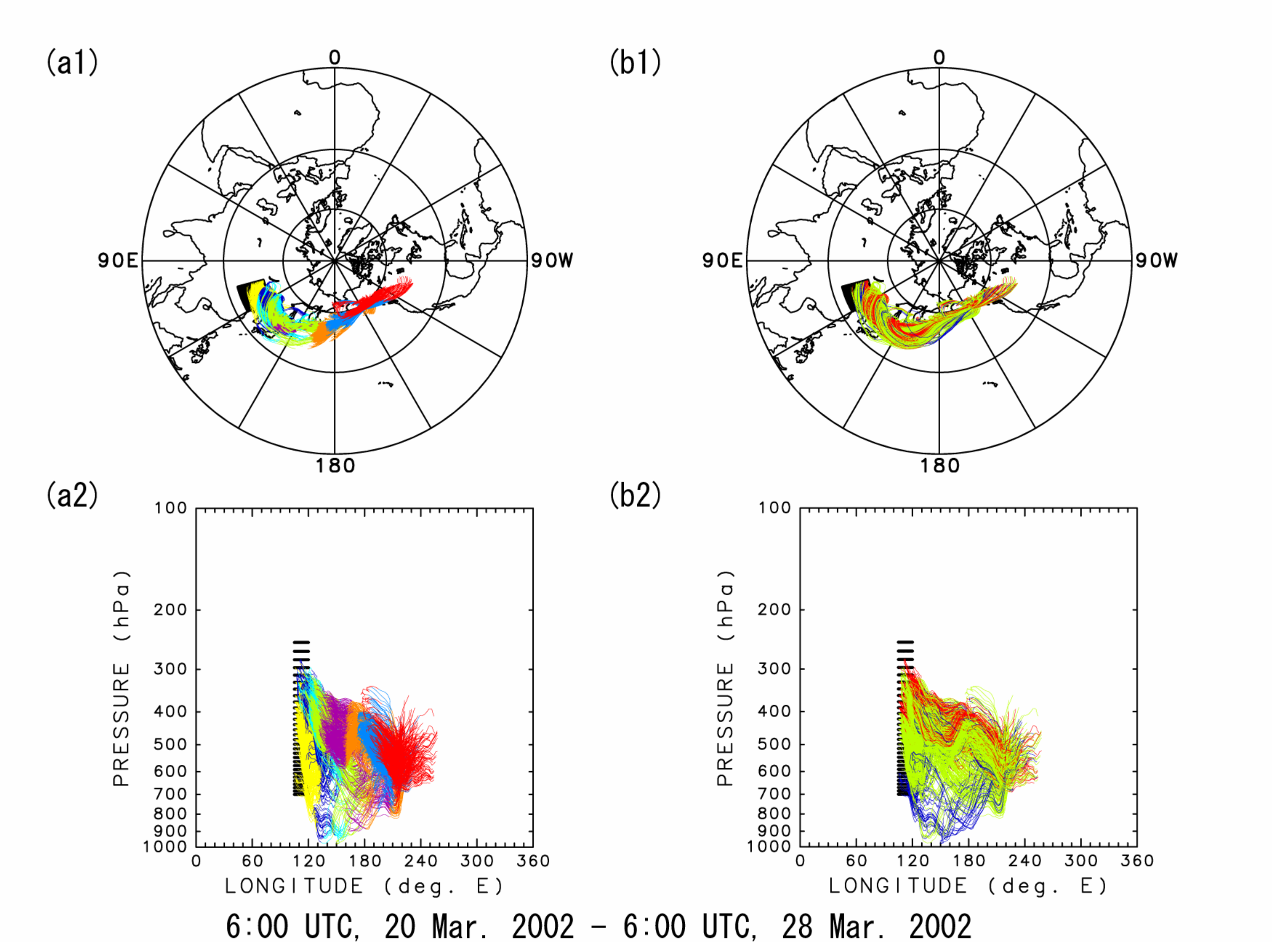


Figure 3.10. Similar to Figure 3.3 but for the Asian dust storm at 6:00 UTC on 20 March 2002.

The dust and stratospheric air reached the Mount Wrangell Area after 26 March (Figure 3.10a). Relative humidity on 26–28 March exceeded 80%, and snowfall was expected at that time at the summit of Mount Wrangell (Figure 3.11a). Because the air mass from the lower troposphere may efficiently involve highly moist air when it passed over the North Pacific Ocean (blue trajectories in

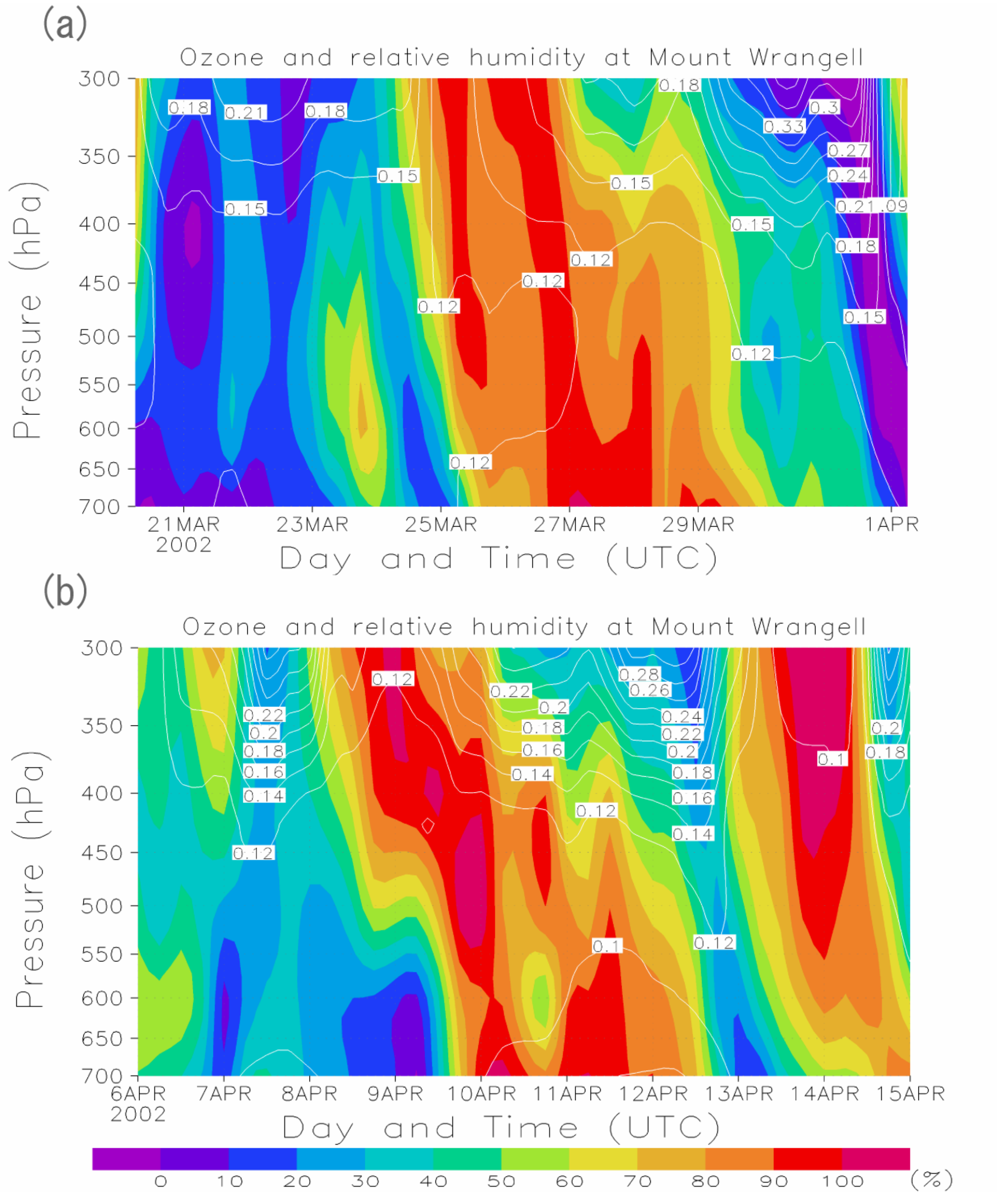
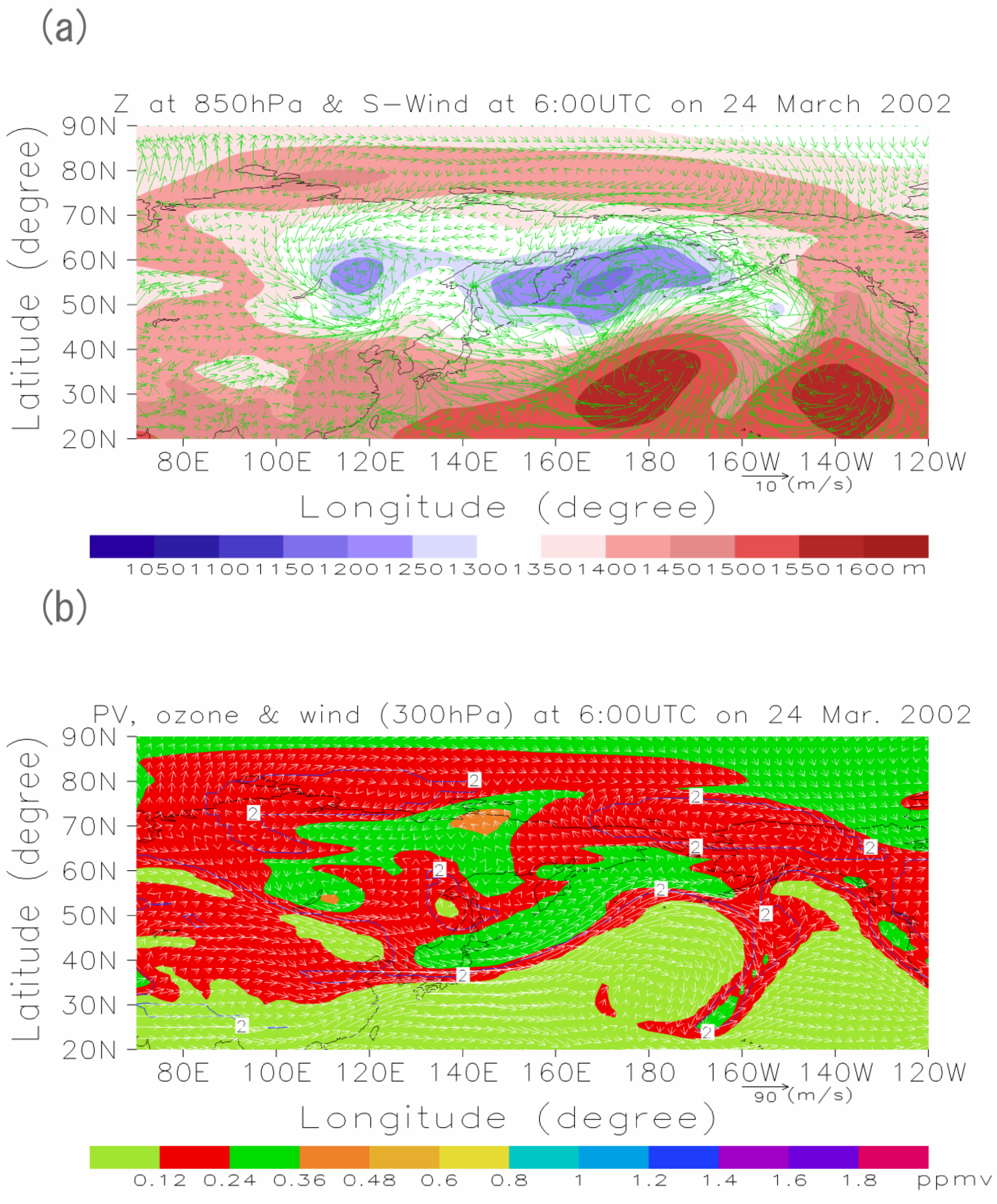


Figure 3.11. Ozone volume mixing ratio (white contour; unit: ppmv) and relative humidity (shaded contour; unit: %) at Mount Wrangell (nearest grid point in ERA-40 data is 62.5°N, 145°W) (a) from 6:00 UTC on 20 March 2002 to 6:00 UTC on 1 April 2002 and (b) from 0:00 UTC on 6 April 2002 to 0:00 UTC on 15 April 2002. The summit of Mount Wrangell is roughly at the 600-hPa level. The data grid is interpolated to a finer grid using cubic interpolation.

Figure 3.10-b), the arrival timings of the relative humidity increase and air mass reaching Mount Wrangell were almost identical (Figures 3.10 and 3.11a). The ozone volume mixing ratio at the ice core site on 26 March was higher than 0.12 ppmv, and stratospheric air contribution from East Asia is considered. Ozone transport from the Asian dust storm to Mount Wrangell was also observed in the horizontal scale motion of each 6-h animation starting from Figure 3.9b (not shown). Hence one of the stratospheric tracers, tritium, must have also been deposited efficiently by snowfall at this time.

A dust storm occurred on 24–25 March following the dust storm on 18(19)–22 March (Figure 3.12). Ozone intrusions were seen with tropopause folding at about 52.5ºN, but it was not a strong intrusion (Figure 3.12b, 3.12c, and 3.12d). Clear descending air is seen at about 42.5ºN and 100ºE as red trajectories in Figure 3.13b, but the origin was the upper troposphere and not the stratosphere because the tropopause was at about 250 hPa and very flat in this region (not shown). In fact, the descending area was from the tropopause line of 2 PVU in the horizontal view, namely, in the troposphere (Figure 3.12b). The trajectories from the lower troposphere contributed to the Wrangell Area in this case. The total number of trajectories reaching the Wrangell Area was much less than that of the dust storm on 18(19)–22 March 2002 (Figures 3.10 and 3.13). The starting area of those trajectories from the lower troposphere was not the dust outbreak area, but the trajectories on 26 March passed through the dust cloud around Hokkaido, Japan (see the sky blue and deep blue trajectories in Figures 3.13-a1 and 3.13-b1, respectively; see Website 3.1). Hence although the trajectories were fewer than those on 18(19)–22 March, the Asian dust on 24–25 March might have



(a)
Z at 850hPa & S−Wind at 6:00UTC on 24 March 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E 100E 120E 140E 160E 180 160W 140W 120W
10 (m/s)
Longitude (degree)
1050 1100 1150 1200 1250 1300 1350 1400 1450 1500 1550 1600 m
(b)
PV, ozone & wind (300hPa) at 6:00UTC on 24 Mar. 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E 100E 120E 140E 160E 180 160W 140W 120W
90 (m/s)
Longitude (degree)
0.12 0.24 0.36 0.48 0.6 0.8 1 1.2 1.4 1.6 1.8 ppmv

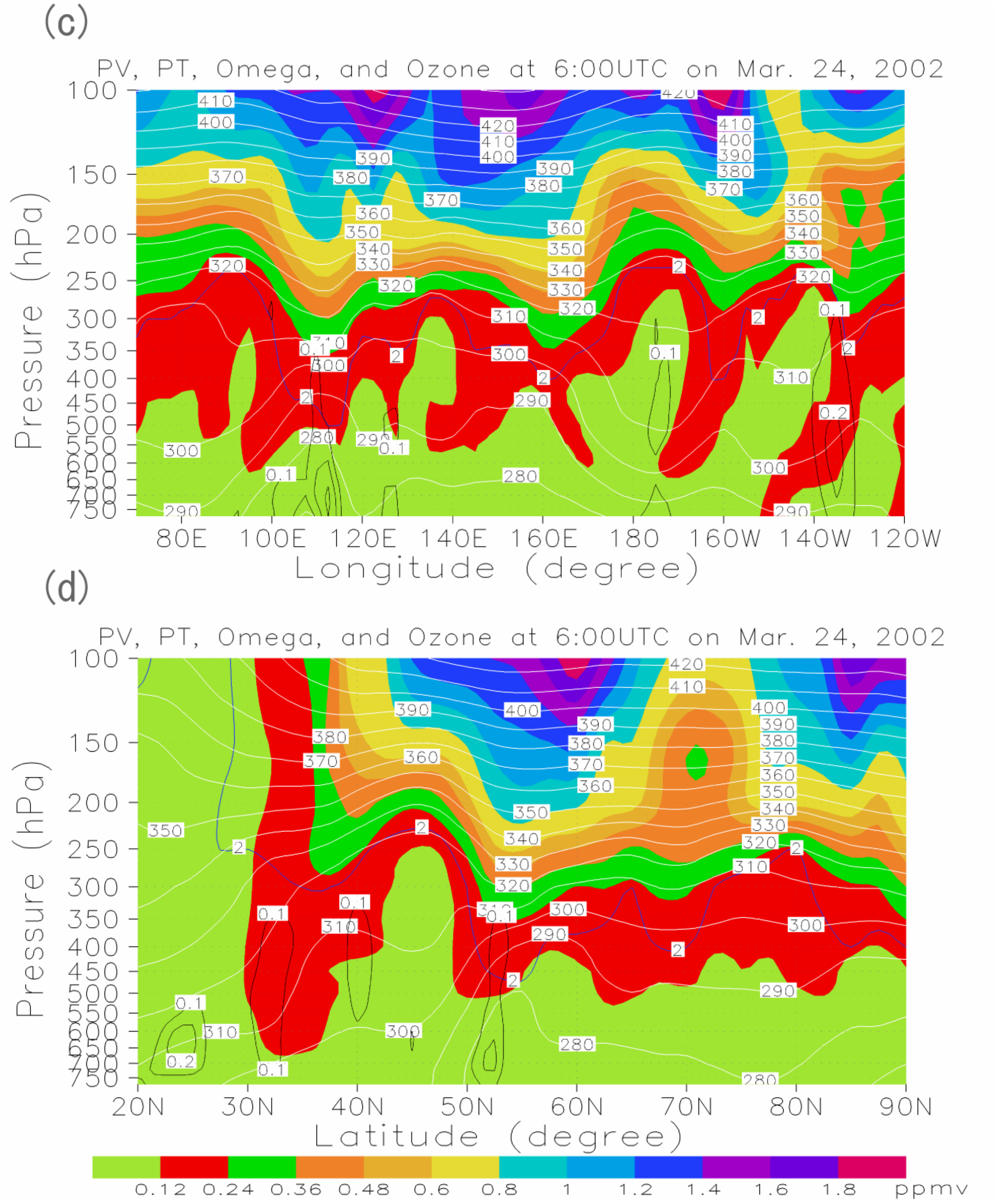


Figure 3.12. Similar to Figure 3.2 but for the Asian dust storm at 6:00 UTC on 24 March 2002. (c) is at the 52.5°N line for the cross section between longitude and pressure level (<775 hPa). (d) Same as (c), but for at the 110°E line for the cross section between latitude and pressure level (<775 hPa).

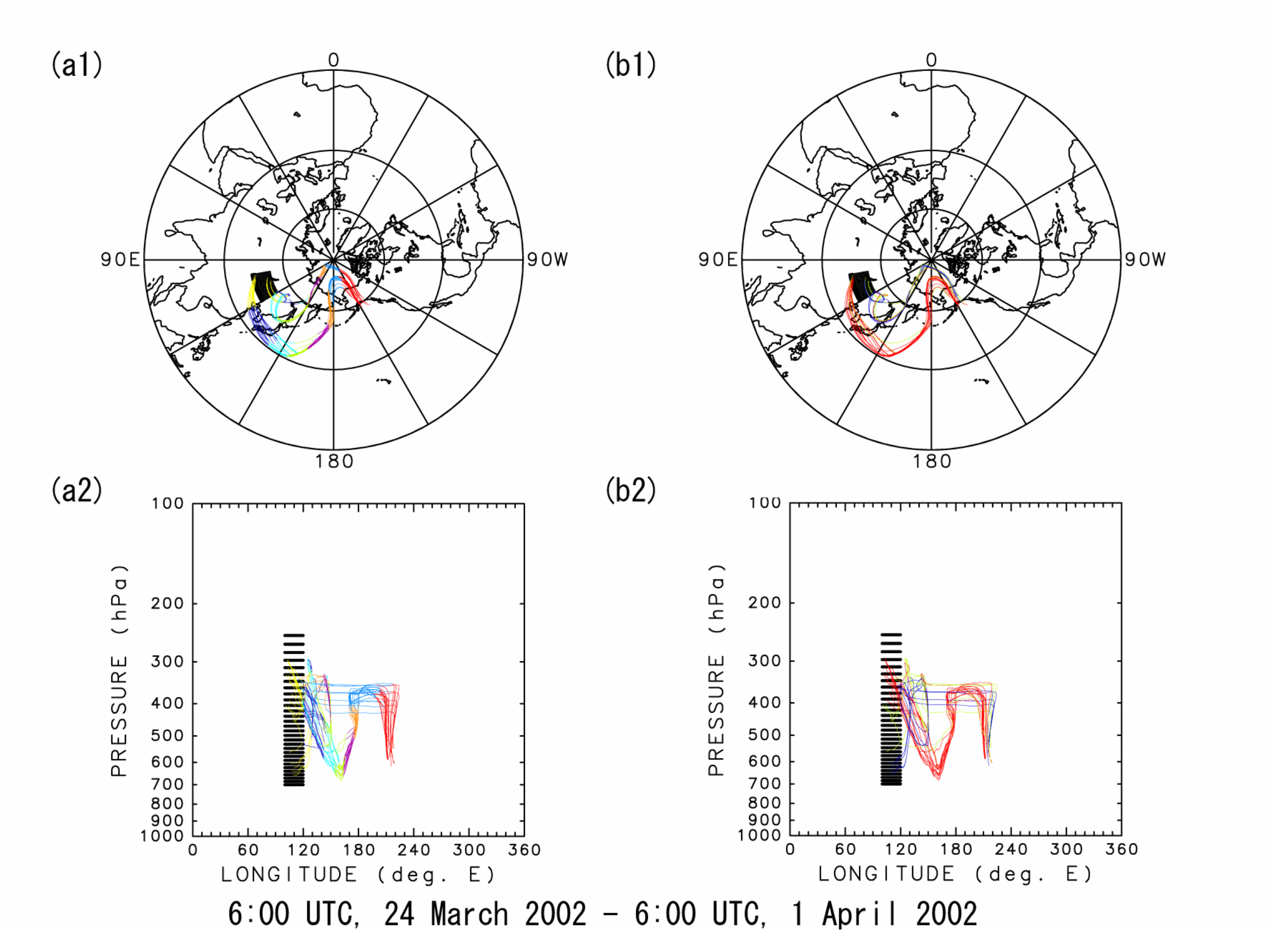


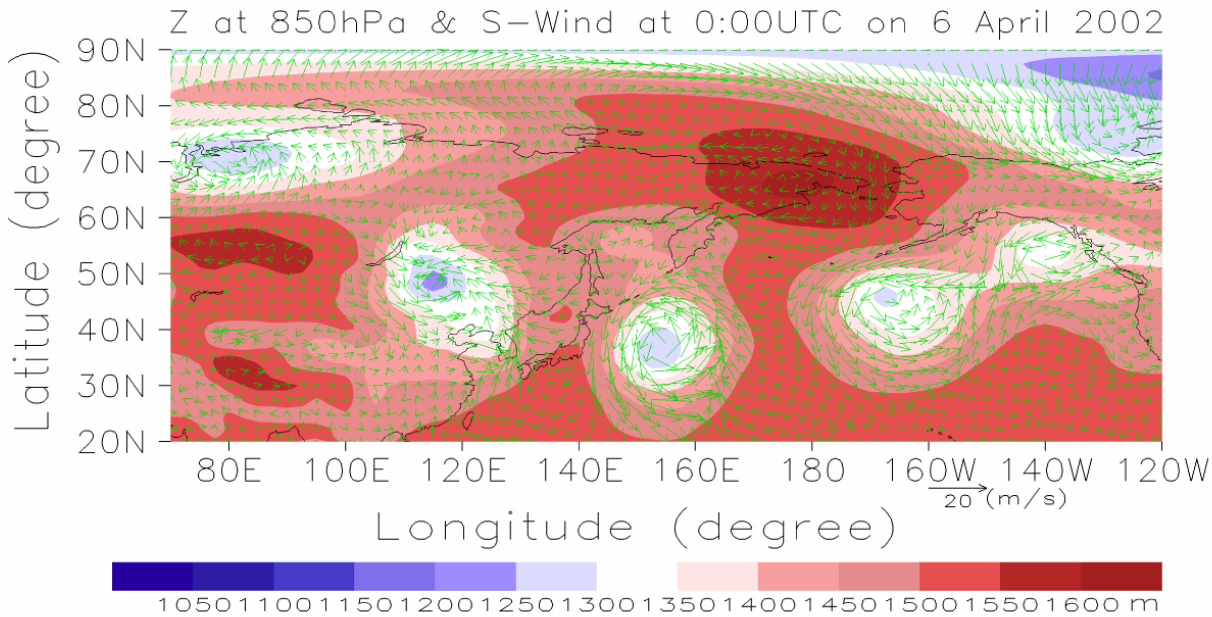
Figure 3.13. Similar to Figure 3.3 but for the Asian dust storm at 6:00 UTC on 24 March 2002.

contributed to the Wrangell Area. On the other hand, the stratospheric air (tritium) associated with the ADSTT might not have been transported to the Wrangell Area. Although ozone above 500 hPa from 30 March to 1 April increased with low relative humidity (Figure 3.11), this air mass was due to OTSTT from Arctic stratosphere origin (not shown). The relative humidity on those days was also very low at the ice core site, and efficient tritium deposition by snowfall was not expected in this case. Hence both the ADSTT and OTSTT contributions to the ice core site were expected to be low in this case.

Now we consider the other dust storm case. On 5(6)–9 April 2002, a severe dust storm occurred in East Asia. As mentioned in section 3.3.1, four severe dust storms broke out in the spring of 2002 [*Shao and Wang*, 2003; *Shao et al.*, 2003]. The TOMS data showed that the Aerosol Index intensity on the transpacific transport was stronger (see Website 3.1). Hence this dust storm may considerably impact on remote areas over transpacific transport in 2002, the same as the 18(19)–22 March dust storm.

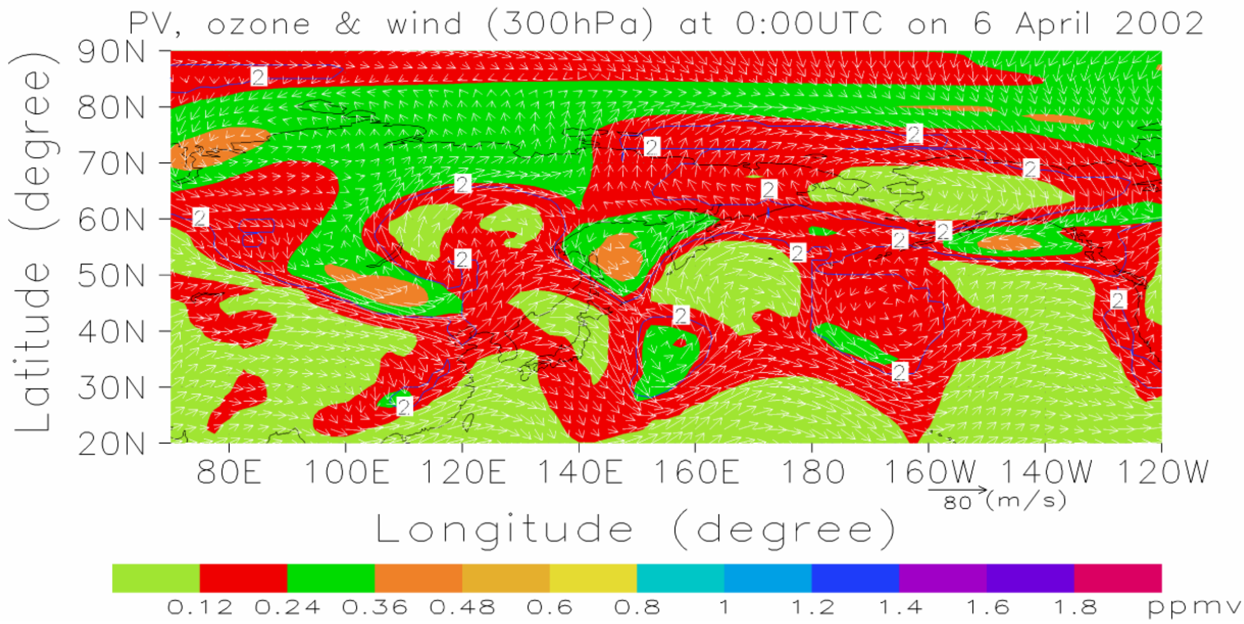
A cyclone was developing on the southeast side of Lake Baikal on 6 April (Figure 3.14). This developing position of the cyclone was very similar to the 6–7 April 2001 dust storm (Figures 3.2a and 3.14a). The ozone advection pattern from northwest to southeast was also almost the same as the 6–7 April 2001 dust storm (Figures 3.2b and 3.14b). However, if we consider the vertical structure, the air mass intrusion due to ADSTT was much deeper than that of the 6–7 April 2001 dust storm (Figures 3.2c, 3.2d, 3.14c, and 3.14d). The MODIS mapped the dust plume from this dust storm on 7 April 2002 (see http://visibleearth.nasa.gov/view_rec.php?id=3220). Trajectory

(a)



Z at 850hPa & S−Wind at 0:00UTC on 6 April 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E 100E 120E 140E 160E 180 160W 140W 120W
20 (m/s)
Longitude (degree)
1050 1100 1150 1200 1250 1300 1350 1400 1450 1500 1550 1600 m

(b)



PV, ozone & wind (300hPa) at 0:00UTC on 6 April 2002
90N
80N
70N
60N
50N
40N
30N
20N
Latitude (degree)
80E 100E 120E 140E 160E 180 160W 140W 120W
80 (m/s)
Longitude (degree)
0.12 0.24 0.36 0.48 0.6 0.8 1 1.2 1.4 1.6 1.8 ppmv

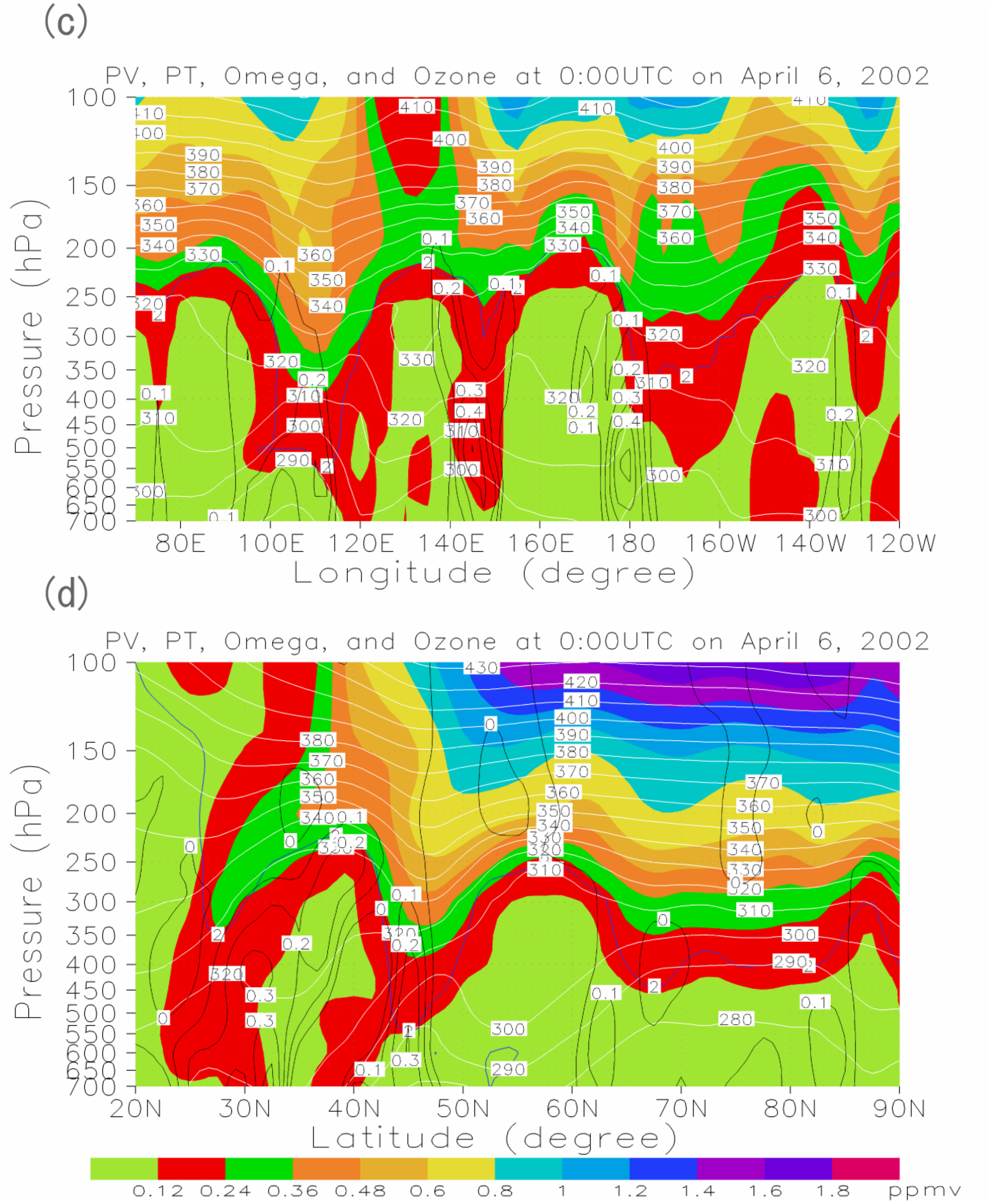


Figure 3.14. Similar to Figure 3.2 but for the Asian dust storm at 0:00 UTC on 6 April 2002. (c) is at the 45°N line for the cross section between longitude and pressure level (<700 hPa). (d) Same as (c), but for at the 110°E line for the cross section between latitude and pressure level (<700 hPa).

calculation was performed from 0:00 UTC on 6 April 2002 in the cyclone developing area (Table 3.1). The air masses reached the Wrangell Area on 14 April 2002 and were well mixed vertically in their transport (Figure 3.15-a2 and 3.15-b2). Relative humidity on 14 April was very high above the summit of Mount Wrangell, and highly saturated layers were developing toward the upper troposphere (Figure 3.11b). Then the Asian dust and the tritium due to the ADSTT were expected to be efficiently deposited by snowfall onto the ice core site.

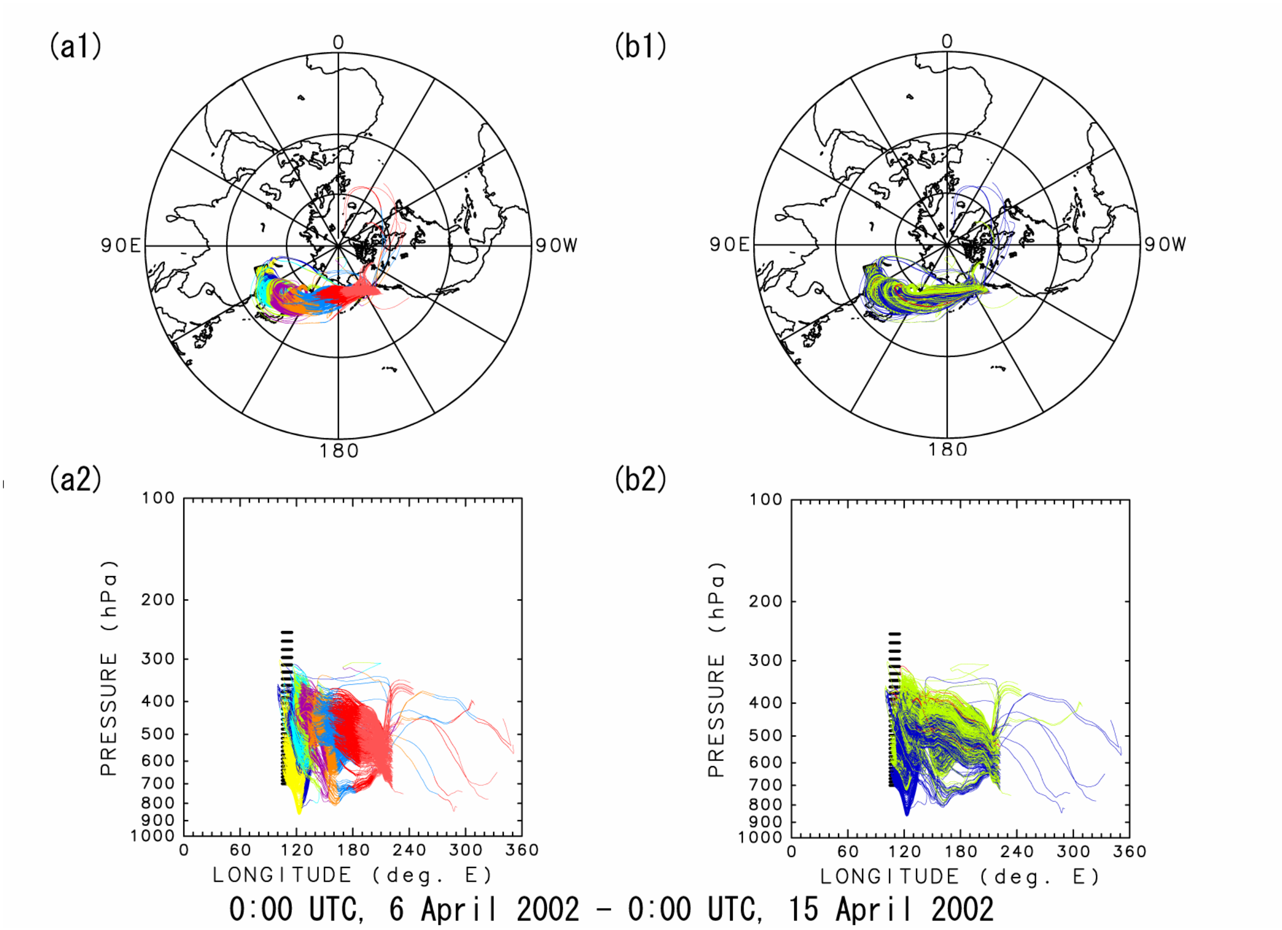


Figure 3.15. Similar to Figure 3.3 but for the Asian dust storm at 0:00 UTC on 6 April 2002.

We now summarize the characteristics of the Asian dust storms on 20 March 2002 and 6 April 2002. The transport and deposition patterns of these cases were very similar: (1) The air was vertically well mixed. (2) Trajectories from the lower troposphere to the lower stratosphere did not branch off in the horizontal direction. (3) The air from the lower troposphere and the lower stratosphere reached the Wrangell Area almost simultaneously. (4) A relatively high humidity layer was developing in the upper troposphere above Mount Wrangell when the air masses reached it. This led to efficient tritium deposition due to snowfall. These four conditions may contribute considerably to the simultaneous deposition of dust and tritium onto the Mount Wrangell ice core site. As a result, we conclude that these two dust storms with ADSTT in East Asia might have considerably contributed to the TF2002 peak.

3.3.4 Dust and tritium contributions due to Asian dust storms in the spring of 2001 and 2002

We found clear differences in transport and deposition patterns in the spring dust storms of 2001 and 2002 as discussed in sections 3.3.2 and 3.3.3. In all five cases of Asian dust storm, dust outbreak and STT occurred simultaneously in the dust storm area. However, the pathway patterns in the transpacific transport and snowfall conditions at Mount Wrangell were different between the cases of expected efficient tritium deposition and those of expected inefficient deposition. In the case of the 6–7 April 2001 dust storm, the dust cloud and the stratospheric air pathways branched off and reached the Wrangell Area with 2-day time lags (Figure 3.3). The dry stratospheric air reaching the Wrangell Area before the Asian dust cloud arrival lacked moist air from the North Pacific Ocean. Tritium mainly exists as tritiated water in the atmosphere [*Gat et al.*, 2001] and prefers wet deposition [YS2007]. In addition, minor parts of the stratospheric air returned to the ice core site on 14 April 2001, and stayed in the upper troposphere (Figures 3.3 and 3.5). A high humidity layer existed below 400 hPa and efficient tritium deposition was not expected because the stratospheric air existed above 400 hPa. Hence the impact on tritium deposition due to the dust storm on 6–7 April 2001 might have been weaker than deposition in the case of perfect snowfalls. As a result, the tritium concentration of TS2001 did not increase very much and might have been the second maximum.

However, TS2001 was considerably higher than the background level (Figure 3.1). If we consider the relative humidity at 600 hPa on 12 April 2001, it was 70–100% (Figure 3.5). Not-vertically-developed weak snowfall perhaps occurred at the summit of Mount Wrangell. Less developed relative humidity in the vertical scale in the upper troposphere is perhaps inefficient for tritium deposition onto the ice core site. As a result, tritium deposition onto the ice core site on 12 April is estimated to be less than that in the cases of vertically developed snowfall such as the cases of the 18(19)–22 March and 5(6)–9 April dust storms in 2002. Thus at the summit of Mount Wrangell, dry stratospheric air passages with weak snowfall may contribute to the tritium increase in the ice core to some extent, but the contribution is expected to be much less than in the cases of vertically developed snowfall events. In conclusion, the relatively higher tritium concentration of TS2001 cannot be explained by this ADSTT contribution alone. The OTSTT contribution to the

TS2001 may be important for explaining this relatively higher tritium concentration as mentioned in section 3.3.6.

On the other hand, in the spring of 2002, the 18(19)–22 March and 5(6)–9 April dust storm cases were perfect in terms of transport pathway patterns and snow deposition timings for both dust and tritium as mentioned in the last paragraph of section 3.3.3. The 24–25 March dust storm contributed to the ice core site in terms of only Asian dust deposition (i.e., no ADSTT contribution). The timings of dust and tritium depositions due to the 18(19)–22 March, the 24–25 March, and the 5(6)–7 April dust storms were estimated to be 26–28 March, April 1 (dust only deposition), and 14–15 April, respectively. The time lags between the 26–28 March and the 1 April depositions and between the 1 April and the 14–15 April depositions onto the ice core site are approximately 4–6 days and 13–14 days, respectively. In the ice core data of Figure 3.1, the time resolutions of tritium and dust are roughly 1 month and 3–12 days, respectively. From the late spring of 2001 to the late spring of 2002, the number of dust samples was 65 and the time resolution of dust was roughly 6 days. The second maximum of the coarse dust peak in the spring of 2002 and the first maximum of the coarse dust peak are aligned side by side. Hence the time lag is estimated to be within 6 days (i.e., one sample interval). Then, those maxima were considered to be associated with continuous Asian dust storms on 18(19)–22 March and 24–25 March. Those continuous dust peaks can be explained by a series of dust clouds due to these continuous Asian dust storms. Next, three sample intervals existed between the first maximum of the coarse dust peak and the third maximum of the coarse dust peak in the early spring of 2002. Hence the time lag is roughly estimated to be 12–18 days, well corresponding to the time lag of 13–14 days between 1 April and 14–15 April. In conclusion, the continuous second-first maxima and the third maximum of the coarse dust peak may be the Asian dust contributions due to the 18(19)–22 March, 24–25 March and 5(6)–9 April severe dust storms, respectively (Figure 3.1).

The dust and tritium depositions due to the 18–22 March and 5(6)–9 April dust storms in 2002 are considered to be mainly snow deposition, and those depositions due to the 24–25 March dust storm are expected to be dry deposition. As mentioned before, the latter case might have not contributed to the tritium deposition and might have contributed only to the dust deposition. The former two cases are expected to strongly impact the drastic increase in tritium concentration.

The half-life of tritium is 12.32 years [*Lucas and Unterweger*, 2000] and tritium decayed by about 5% from the spring of 2001 to the spring of 2002 in the Mount Wrangell ice core of YS2007. Even if the decay of tritium is taken into account, the tritium concentration of TF2002 is much larger than that of TS2001. The perfect transport of Asian dust and tritium and its deposition due to snowfall in the spring of 2002 may contribute considerably to produce the highest tritium concentration in 2002 as the TF2002 peak.

The additional importance of the ADSTTs on 20 March and 6 April 2002 is the much deeper ADSTT characteristics than in the other storms in the spring of 2001 and 2002 (Figures 3.2, 3.7, 3.9, 3.12, and 3.14). Deep STT is more effective for mixing of stratospheric material into the troposphere [*Stohl et al.*, 2003a, 2003b]. Our results are consistent with these descriptions. In addition, when the tritium from the ADSTTs was transported to Mount Wrangell, the weather conditions were better for tritium deposition because of the expected snowfall (Figure 3.11). As a result, the tritium concentration of TF2002 in the ice core could have increased considerably (Figure 3.1). On the other hand, in section 3.3.6, the TF2001 in late spring of 2001 will be mainly explained in terms of OTSTT contributions.

3.3.5 Estimated snow deposition contributions onto the ice core site in spring

Snow deposition onto Mount Wrangell is a favorable condition for simultaneous dust and tritium deposition as mentioned before. In particular, snowfall is more important for tritium deposition than dust, because tritium in the atmosphere mainly exists as tritiated water vapor [*Gat et al.*, 2001]. Snowy conditions at Mount Wrangell may contribute to the monthly and interannual variations of tritium in the ice core. Hence in this section, we discuss the characteristics of snowfall at Mount Wrangell from 1992 to 2002, which correspond to the ice core data in YS2007. In section 3.3.6, we discuss the effects of OTSTT contributions in terms of atmospheric circulation patterns. Finally, in section 3.3.7, we discuss the mechanism of dust and tritium variations in spring in the North Pacific region and the possible explanation and progress on the hypothesis of YS2007 that Asian dust outbreaks and STT are interannually correlated in spring due to cyclonic activities.

We assumed that snow deposition occurred at the ice core site when relative humidity exceeded 80% as seen in Figures 3.5, 3.6, and 3.11. Note that we focus on the snowfall frequency in 1 month and not on the amount of snow accumulation in 1 month. We used a daily mean relative humidity of more than 80% for the calculation of monthly snowy days to easily count the snowfall days. When the relative humidity in the raw data of JMA-GPV was greater than 80%, we observed snowfalls at the summit of Mount Wrangell in May 2004 (Figure 3.6). Hence a relative humidity of greater than 80% is considered to be an effective index for snowfall at the ice core site. In the calculations, one snowfall event was also estimated. If a daily mean relative humidity greater than 80% continued for more than 2 days, we regarded the total continuous snowfall days as one snow event. If one snow event continued beyond the final day in a certain month, the snow event was counted as one snow event in the next month.

The percentage of the number of snowy days defined by a relative humidity of greater than 80% and the number of single snow events in 1 month from January 1992 to August 2002 are shown in Figure 3.16a. The monthly composites of the percentage of the number of snowy days in the 11-year climatology are also shown in Figure 3.16b. It seems that there are no significant trends of these data in the recent 11 years (Figure 3.16a). The mean monthly values of the percentage of snowy days and number of snow events in all of the time series data are 42.05% and 5.48, respectively. This means that snowfall at the ice core site may occur about 12–13 days in 1 month, and one snow event often continues for more than 2 days. Hence we assume that one snowfall event at the ice core site occurs every few days in 1 month in terms of the mean monthly value. A few cases with these characteristics are actually seen in Figures 3.5, 3.6, and 3.11.

It is worth noting that the percentage of the number of snowy days with a daily mean relative humidity of more than 80% increases in April and May in the 11-year climatology (Figure 3.16b). This means that April and May have more snowy days than those in the other months. Snow

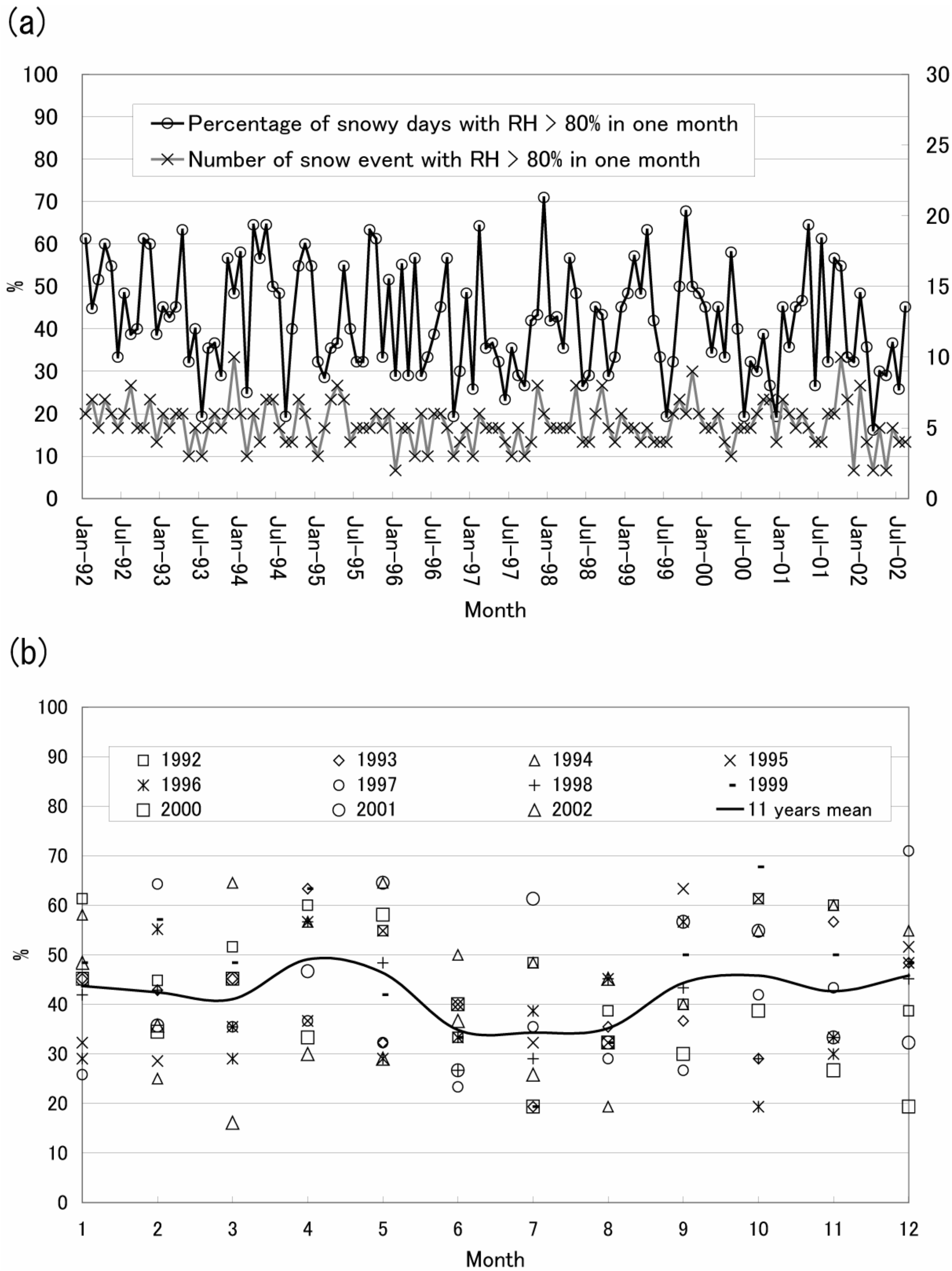


Figure 3.16. Percentage of snowy days (solid black line with circles) and number of snow events in 1 month (gray solid line with crosses) estimated by the condition of relative humidity greater than 80%. (a) Time series of those monthly data. (b) Monthly composite of the percentage of snowy days in each month. Bold line denotes monthly mean values in the 11-year climatology.

fall may easily occur at the summit in these months. The secondary maxima in September and October correspond to the rainy season in southeast Alaska [*Manley, W.F., and Daly, C.*, 2005, Alaska Geospatial Climate Animations of Monthly Temperature and Precipitation: INSTAAR, University of Colorado; see the website at http://instaar.colorado.edu/QGISL/AGCA]. The number of snowfall days in March is also higher than that in summer, but less than that in April and May (Figure 3.16b).

Monthly mean geopotential height, horizontal wind at 600 hPa, and PV at 300 hPa are shown in Figure 3.17. The composites of these in snowfall cases at the ice core site (relative humidity greater than 80% in the 6-h data) are also shown in Figure 3.18. The monthly changes in major monthly pressure patterns in 2001 and 2002 in Figure 3.17 are roughly similar to those in the climatology data (Figure 3.19a-d), but their intensities and positions are very different in 2001 and 2002. In addition, the rough shapes of Figures 3.17 and 3.18 are similar. This indicates that the monthly pressure pattern is important in controlling the snowfall conditions at the ice core site.

3.3.6 Monthly characteristics in atmospheric circulation for Asian dust outbreaks and STT

Although we have discussed the effect of ADSTT contributions to the ice core site in terms of case studies in the spring of 2001 and 2002, here we explain the dust and tritium variations in 2001 and 2002 in the ice core in spring in terms of monthly atmospheric circulation patterns. We investigate the characteristics of atmospheric circulation in each month in terms of ADSTT, OTSTT,

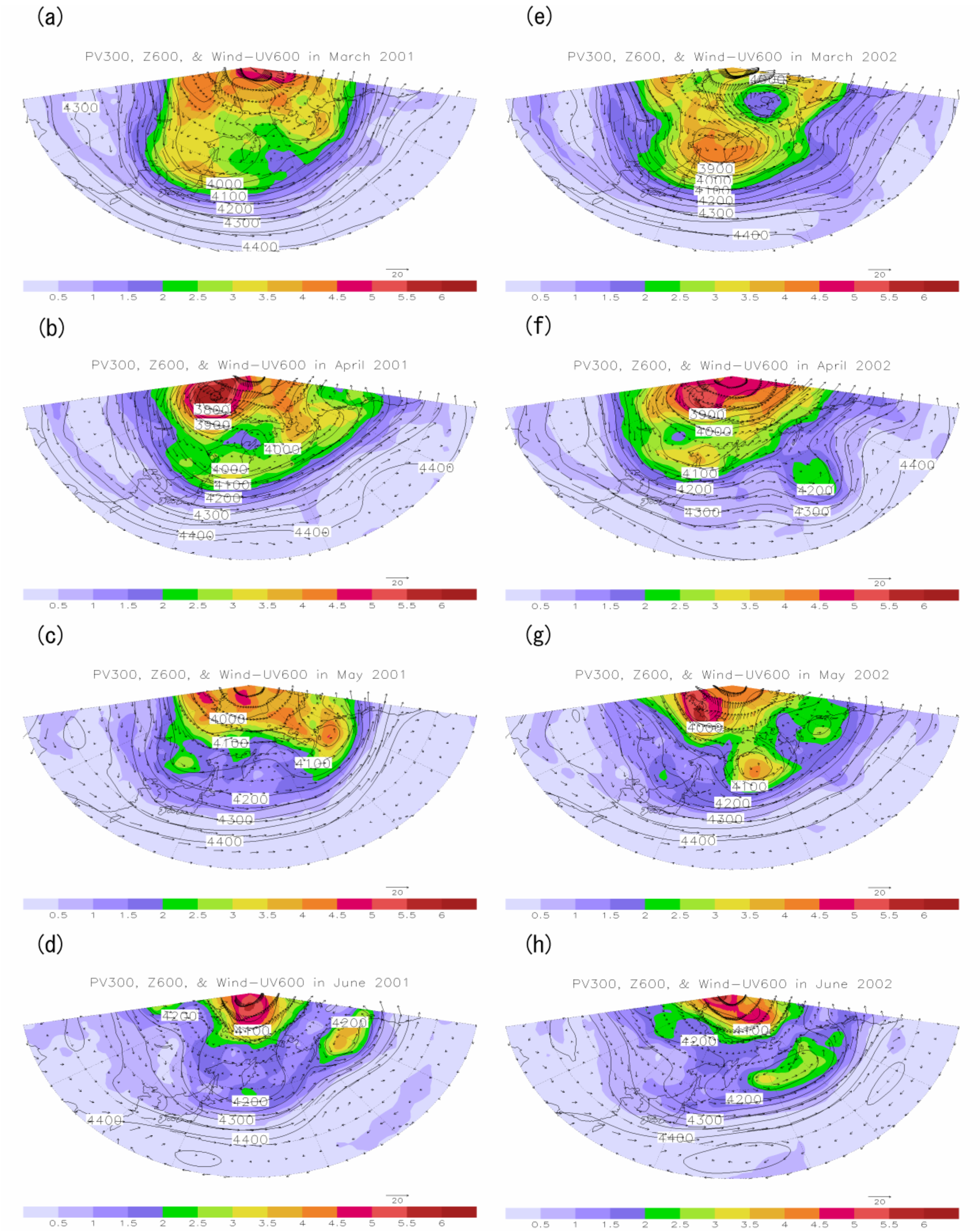


Figure 3.17. Monthly mean geopotential height (unit: m) and wind speed (unit: m s^{-1}) at 600 hPa and PV (unit: PVU) at 300 hPa from March to June in 2001 and 2002. (a)–(d) are March, April, May, and June in 2001, respectively. (e)–(h) are March, April, May, and June in 2002, respectively. The data grid is interpolated to a finer grid using cubic interpolation.

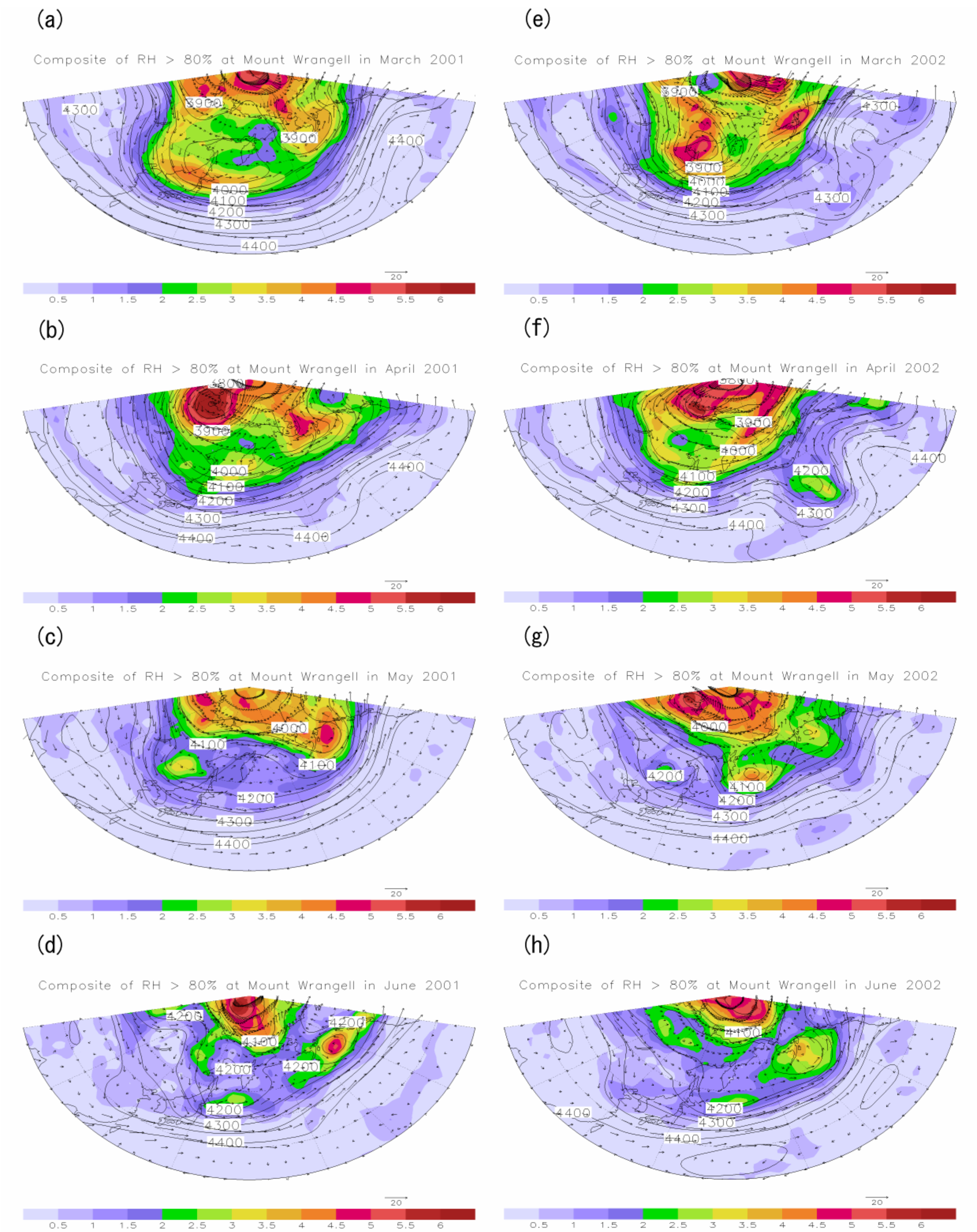


Figure 3.18. Monthly composites of geopotential height (unit: m) and wind speed (unit: m s^{-1}) at 600 hPa and PV (unit: PVU) at 300 hPa when it snowed at Mount Wrangell, which is estimated by relative humidity greater than 80% from March to June in 2001 and 2002. (a)–(d) are March, April, May, and June in 2001, respectively. (e)–(h) are March, April, May, and June in 2002, respectively. The data grid is interpolated to a finer grid using cubic interpolation.

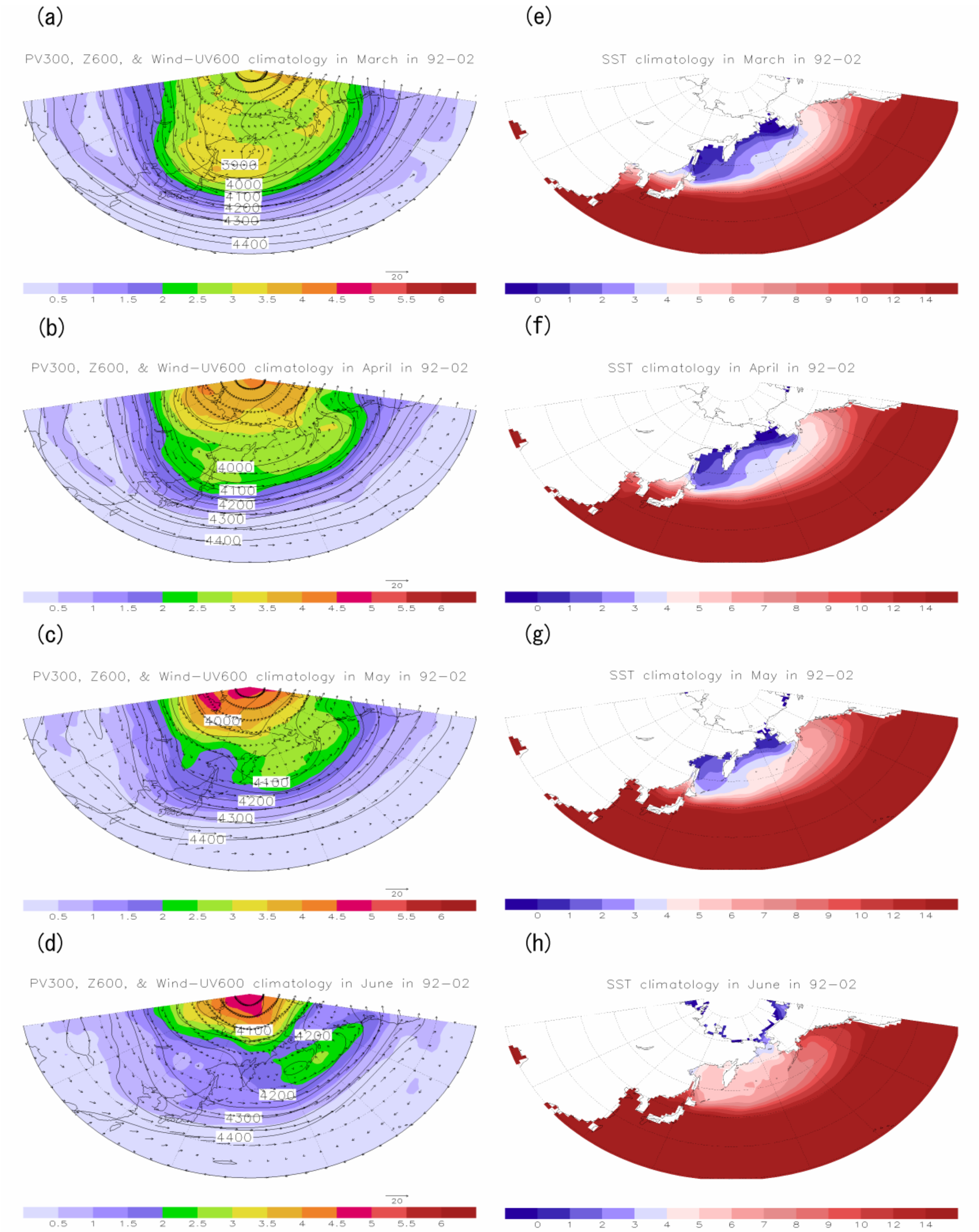


Figure 3.19. Monthly mean geopotential height (unit: m) and wind speed (unit: m s^{-1}) at 600 hPa, PV (unit: PVU) at 300 hPa, and SST (unit: ºC) from March to June in the 11-year climatology. (a)–(d) are March, April, May, and June in 92-02 for geopotential height and wind speed at 600 hPa, PV at 300 hPa, respectively. (e)–(h) are March, April, May, and June in 92-02 for SST, respectively. The data grid is interpolated to a finer grid using cubic interpolation.

and snowfall conditions at Mount Wrangell.

In March, the strong potential vorticities over Siberia are favorable for severe dust storms with STT in East Asia because of the easy production of a cold front from the Siberian and Mongolian regions (Figures 3.17a, 3.17e, and 3.19a). In fact, a cold front is very important for a dust outbreak [*Sun et al.*, 2001; *Hayasaki et al.*, 2006]. Cold front advection from Siberia may produce a large meridional temperature gradient and cause the near surface baroclinicity to intensify around the Gobi desert. Then, storm tracks are produced probably in the mid-latitude jet zone (Figure 3.20a and 3.20e) and surface winds also strengthened, such as in the severe dust storm cases in this study (Figures 3.2a, 3.7a, 3.9a, 3.12a, and 3.14a). In 2001 and 2002, Asian dust storms were observed more frequently than that in the 1990s [*Chun and Lim*, 2004] (see also *Japan Meteorological Agency* (*JMA*), Basic knowledge about Asian dust (Kosa) (in Japanese, 2006, http://www.data.kishou.go.jp/obs-env/kosahp/4-4kosa.html). Severe dust storms were actually observed in 2001 and 2002 as investigated before and in this study [*Liu et al.*, 2003; *Shao and Wang*, 2003; *Shao et al.*, 2003; *Sugimoto et al.*, 2003; *Zhou and Zhang*, 2003; *Darmenova et al.*, 2005; *Sun et al.*, 2006]. These are explained well by the stronger eddies over Siberia in March leading to cold air advection from north to south (Figure 3.17a and 3.17e). Atmospheric conditions in March are favorable for developing cyclones in East Asia, and these cyclones may also lead to dust outbreaks and STT.

Snowy days in March are fewer than those in April and May in the climatology data (Figure 3.16b). This characteristic is strongest in March 2002 (Figure 3.16b). This is probably due to

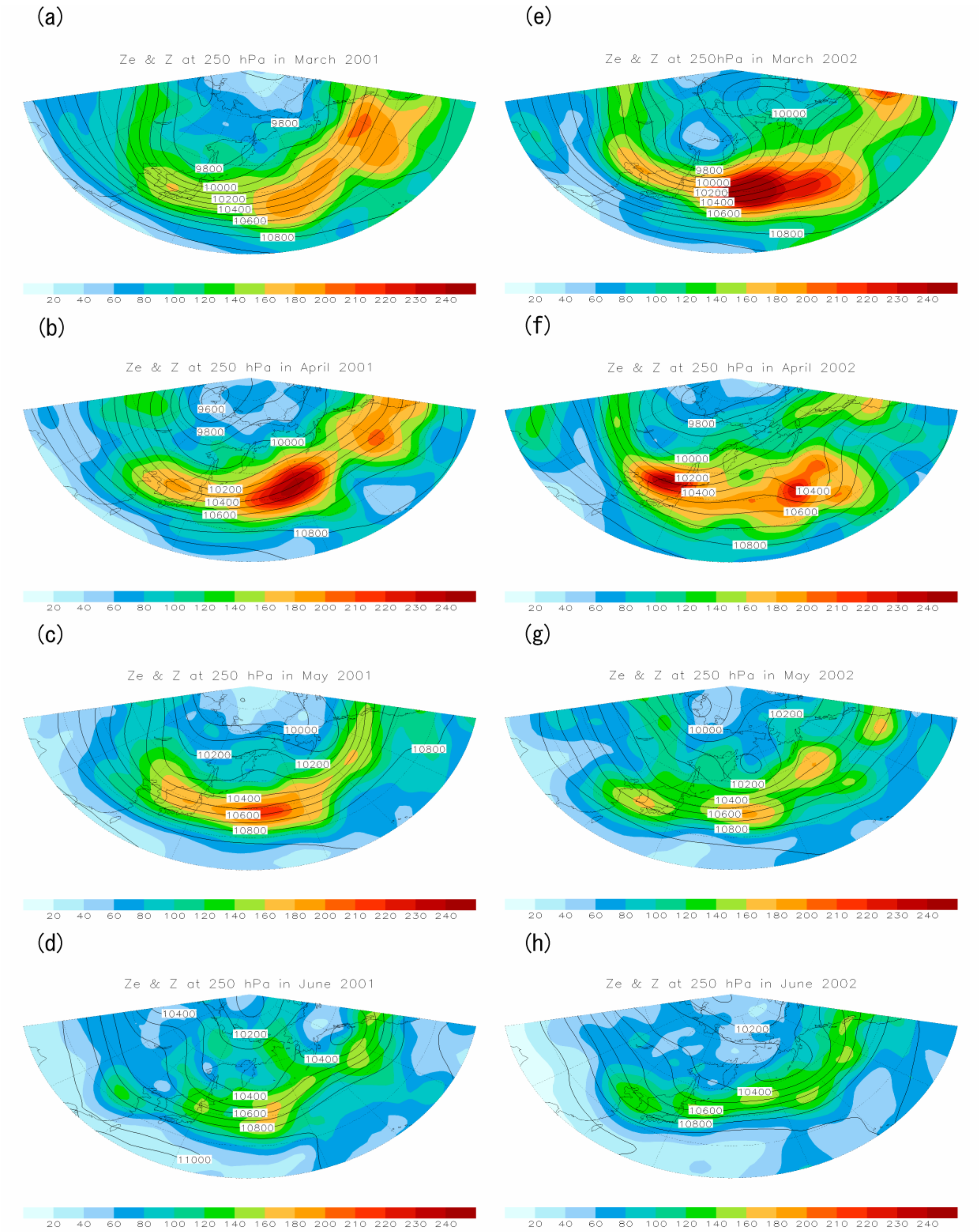


Figure 3.20. Monthly mean envelope function (Ze; unit: m) and geopotential height at 250 hPa from March to June in 2001 and 2002. (a)–(d) are March, April, May, and June in 2001, respectively. (e)–(h) are March, April, May, and June in 2002, respectively. The data grid is interpolated to a finer grid using cubic interpolation.

pressure patterns over Alaska and the Gulf of Alaska. It is worth noting that blocking-like high pressure areas with smaller PV values covered Alaska in March 2002 (Figure 3.17e). Then, these probably produced the least snowy days at the ice core site from 1992 to 2002 (Figure 3.16b). However, when it snowed at the ice core site in March 2002, low pressures with higher PV values were seen in the Gulf of Alaska (Figure 3.18e). Hence if the timing of the snowfall at the ice core site and transport of Asian dust and tritium to the ice core site synchronize at each event scale, dust and tritium may be efficiently deposited onto the ice core site such as in the 5(6)–9 April and the 18(19)–22 March dust storms in 2002 (Figure 3.11), although March is less favorable for snow deposition than April and May in terms of snowfall probability.

Storm tracks in the North Pacific region are stronger than in East Asia, and the central positions of storm tracks in March 2001 and 2002 were the Gulf of Alaska and the center of the North Pacific, respectively (Figure 3.20a and 3.20e). The regions where strong meridional heat flux was seen corresponded very well to the storm track regions at the upper troposphere (Figures 3.20a, 3.20e, 3.21a and 3.21e). Strong PV-values at 300 hPa in Figure 3.17 and Figure 3.18 imply that the tropopause height is lower and frequent STT occurrence is expected in the high PV region. *Yamashita* [2003] and *Mukougawa et al.* [2004] mentioned that the tropopause rose in the center of the storm track region and fell to the northwest and southeast. Our relationship between high PV-value and strong storm track regions corresponded well with their results. The tropopause height to the south of the storm track region is generally much higher than to the north, and we cannot see the high PV-value in the southern region at the 300 hPa level (Figures 3.17 and 3.18). Hereafter, we

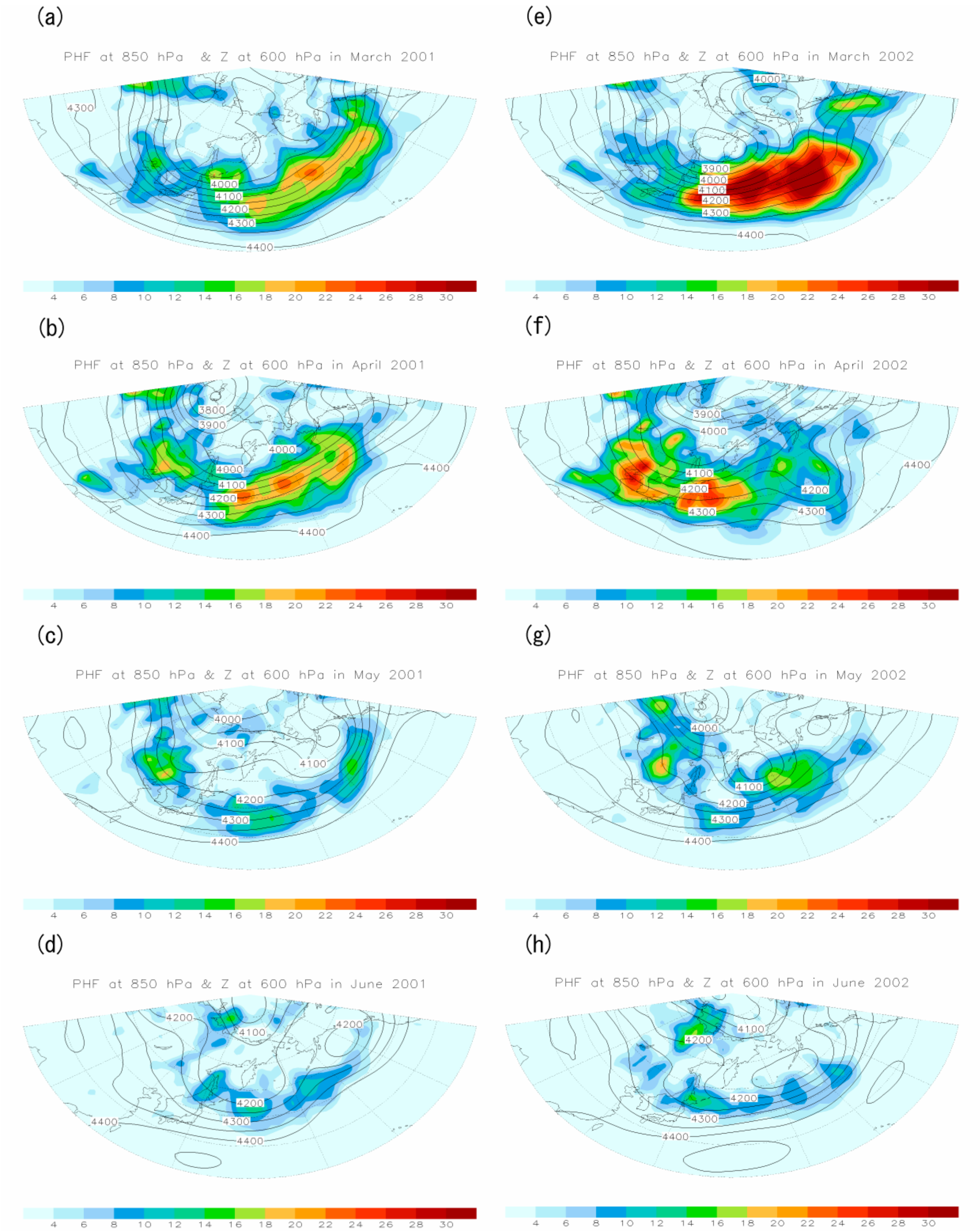


Figure 3.21. Monthly mean poleward (meridional) heat flux (unit: K m s^{-1}) at 850 hPa from March to June in 2001 and 2002. (a)–(d) are March, April, May, and June in 2001, respectively. (e)–(h) are March, April, May, and June in 2002, respectively. The data grid is interpolated to a finer grid using cubic interpolation.

call this relationship between the high PV-value (lower tropopause) and the strong storm track regions as the relationship between the storm track and the tropopause (RSTTP). In conclusion, cyclones developing near Alaska are important for both snowfall at the ice core site and the OTSTT contribution onto the ice core site.

In April, the PV developing with intensified low pressure over Siberia was the strongest in March–June 2001 and 2002 (Figure 3.17b and 3.17f). Intensified eddies over Siberia and Alaska in 2001 and 2002 showed a pair of high-PV structures (Figure 3.17b and 3.17f). These two structures were clearer when it snowed at the ice core site (Figure 3.18b and 3.18f). Clear extensions of the polar vortex to the Siberian region were seen (Figures 3.17b, 3.17f, and 3.19b). In particular, April is much better for producing a cold front from the Siberian region because of the more intensified polar vortices extending southward (Figure 3.17b and 3.17f). As a result, these cold-front regions prefer to cause a meridional temperature gradient and lead to Mongolian cyclone developing associated with Asian dust outbreaks. In fact, April is the main dust-outbreak month [*Sun et al.*, 2001]. In addition, Asian dust storms with deep tropopause folding were observed due to the intensified baroclinicity, which indicates STT [*Zhao and Zhao*, 2006]. The polar vortex over the Siberian region in April 2001 intensified more than that in April 2002 (Figure 3.17b and 3.17f). This implies that a much more severe dust storm could break out in 2001. In fact, the 6–7 April 2001 dust storm was the largest in terms of area covered since 1979 (SeaWiFS: NASA science news, 17 May 2001 at http://science.nasa.gov/headlines/).

The central positions of the cyclones developing in the North Pacific in April of 2001 and

2002 when it snowed at the ice core site were in the Aleutian-Alaskan region (Figure 3.18b and 3.18f). The higher PV-values in this region imply that STT occurs there. Namely, OTSTT contributions to the ice core site due to these cyclones are expected. The STT near Alaska with snowy conditions at Mount Wrangell is better for efficient tritium deposition. The background level of tritium concentration in the ice core in normal years may depend considerably on the degree of OTSTT near Alaska. If additional ADSTT contribution at the ice core site is expected with snowfall, the tritium concentration in the ice core may further increase due to both the ADSTT and the OTSTT contributions such as the TF2002 peak.

Although the snowfall frequency changes year by year, the probability of snowfall in April is more than in March (Figure 3.16b). If a large increase in tritium concentration is seen associated with the dust increase in the Mount Wrangell ice core in spring, the contributions of simultaneous dust and tritium transport and efficient tritium deposition due to snowfalls at the ice core site are expected with high probability. In the YS2007 ice core, the dust and tritium, which sometimes co-increased in spring in the observed raw data, may also be the simultaneous dust and tritium depositions identical with the TF2002 peak.

In April of 2001 and 2002, the Aleutian lows intensified when it snowed at the ice core site (Figure 3.18b and 3.18f). High PV-values were seen from the Bering Sea to Alaska in April of 2001 and 2002 (Figure 3.18b and 3.18f) and corresponded very well to the RSTTP (Figure 3.20b and 3.20f). The meridional heat flux was strong in the central positions of the storm track in the North Pacific Ocean (Figure 3.20b, 3.20f, 3.21b, and 3.21f). *Fan and Wang* [2007] mentioned that the deepening of the Aleutian low correlated with strong wind speeds at the Beijing station and caused dust storms in spring because the resulting cyclone induced cold air activity with strong winds. Intensified PV-values were also seen in the Siberian region (Figures 3.17b, 3.17f, 3.18b, and 3.18f). This may cause Asian dust storms due to cold fronts from Siberia. High-speed winds at the surface due to severe weather are also associated with STT [*Browning and Reynolds*, 1994; *Goering et al.*, 2001]. In fact, a case of increased ozone was reported in East Asia with the Asian dust storm [*Kim et al.*, 2002]. In addition, intensified PV-values in the Bering Sea, i.e. descending tropopause, were seen when it was snowing at the ice core site in April (Figure 3.18b and 3.18f). This suggests that the Aleutian low is probably connected with both Asian dust storms and snowfall at the ice core site. The deepening Aleutian low is probably accompanied by STT in its cyclone region. This will be mentioned in detail in the next section. Hence we consider that a developing Aleutian low in April is one of the important factors for the Asian dust outbreaks with ADSTT in East Asia, the OTSTT near Alaska, and the snowfall at Mount Wrangell.

In May, although a low pressure system still exists on the east side of the Eurasian Continent, cyclones were much stronger in the Aleutian region or the Gulf of Alaska (Figure 3.17c, 3.17g, and 3.19c). The baroclinic wave amplitude over the Eurasian Continent was weaker in May than that in March and April (Figure 3.20c and 3.20g). The number of severe dust storms in East Asia may be less than that in March and April, although dust storms still occur in May. In fact, Asian dust storms in East Asia in May are statistically less frequent than in April [*Sun et al.*, 2001], and no severe dust storms were observed in May 2002 [*Sun et al.*, 2006; *Zhou and Zhang*, 2003].

The probability of efficient tritium deposition onto the ice core site in May may be as high as in April because of the greater number of snowy days in May (Figure 3.16b). The pressure pattern in May is favorable for snowfall with OTSTT near Alaska because of low pressure developing in the Gulf of Alaska (Figures 3.17c, 3.17g, 3.18c, 3.18g, and 3.19c). This may lead to efficient water vapor transport to Mount Wrangell. The SST in the Gulf of Alaska is clearly different before and after May (Figure 3.19e–h). Evaporation from the Gulf of Alaska may increase after May. In addition, there are more snowy days at the ice core site in May as mentioned (Figure 3.16b). This may contribute sufficiently to snowfall as efficient tritium deposition onto the ice core site is also expected.

In the North Pacific region, storm track activities on intensity and meridional movement are very important for the variation of tropopause height [*Yamashita*, 2003; *Mukougawa et al.*, 2004]. The RSTTP was clearly seen in May 2001 and 2002 (Figures 3.18c, 3.18g, 3.20c, and 3.20g). The intensified storm track positions correspond very well to the strong heat flux regions (Figures 3.20c, 3.20g, 3.21c, and 3.21g). The OTSTT in the Gulf of Alaska was considered to be stronger in 2001 than in 2002 (Figures 3.17c, 3.17g, 3.18c, and 3.18g). This can be explained by the central positions of the storm track as the RSTTP. Stratospheric material such as tritium may have intruded considerably into the troposphere in May 2001 due to the stronger descending tropopause in the Gulf of Alaska and water vapor advection to the ice core site due to the cyclone may cause snowfall leading to efficient tritium deposition onto the ice core site. The frequency of snowy days at the ice core site was higher in 2001 than in 2002 and also the highest in the 11-year climatology (Figure

3.16b). Intensified OTSTT in the Gulf of Alaska with a developing cyclone may also increase both the probability of snowy days at the ice core site and efficient tritium deposition onto the site. In conclusion, the most important factor for the general tritium maxima in late spring in the ice core may be the OTSTT contribution in the Gulf of Alaska such as the 2001 tritium maximum in late spring (TF2001).

In June, no cyclone developing in East Asia was seen and the storm track position moved toward the North Pacific (Figures 3.17d, 3.17h, 3.19d, 3.20d and 3.20h). Moreover, the intensity of the storm track in the North Pacific region was weaker than that in March–May. This corresponds very well to the main months for Asian dust outbreak being March, April, and May. In addition, the position of Mount Wrangell is between the polar vortex and the North Pacific low (Figure 3.17d and 3.17h). This may weaken the advection of humid air from the North Pacific and increase the number of calm days at Mount Wrangell. The polar vortex and the North Pacific low are both weakened at Mount Wrangell. The weakening conditions may lead to weak OTSTT in the North Pacific region. It may also lead to the reduction in the number of snowy days at Mount Wrangell in June (Figure 3.16b). That condition is also not effective for tritium deposition onto the ice core site. Hence in general, the STT contributions to tritium concentration in the ice core in June are assumed to be weak. Therefore, the next tritium samples after the TF2001 and TS2002 peaks, in general, may correspond to the months after June.

3.3.7 Possible explanation for intraseasonal and interannual relationships between Asian dust and STT detected from the Mount Wrangell ice core

Finally, we discuss the relationship between dust and tritium variations in spring in the ice core and explain the mechanism of tritium variation in spring in the North Pacific region. Then we discuss the hypothesis presented in YS2007 that Asian dust and STT are interannually correlated due to cyclonic activity in East Asia. That suggestion is based on the positive correlation between fine dust and tritium fluxes in the spring season and the high contribution of Asian dust to Mount Wrangell in the spring season. The ADSTT is one of the important factors for explaining the interannual relationship between dust and tritium fluxes. However, it may not be sufficient for a detailed explanation, so we discuss the details next.

YS2007 presented detailed seasonal dating in the 50-m Mount Wrangell ice core. Dust and tritium were used for early spring and late spring dating points, respectively, by means of 5-data-point running mean data. The annual dust peaks in early spring are mainly produced by the Asian dust contribution. In general, tritium peaks in spring in the ice core tended to be located after the dust peaks along the ice core depth in terms of the running mean data as seasonal march. If we consider the measured raw tritium data, the tritium maxima were also located after the dust peaks at the early spring dating points in almost all of the years except for 1995, 2000, and 2002. The dust peak was located after the tritium maximum peak only in 2000. In 1995 and 2002, the periods of the maximal tritium peaks include the coarse dust maximal peaks in observed raw data. Hence we consider that the tritium maximal peak in the early spring of 1995 was also moved from late spring to early spring due to strong ADSTT in early spring as in the case of 2002. When we focus on the fine and coarse dust concentrations corresponding to the depths at the tritium maximal peaks in 2002 and 1995, considerably higher concentrations of fine and coarse dust were seen in the tritium peaks. Those higher concentrations of fine and coarse dust peaks were more than 85,000 and 25,000 particles mL^{-1}, respectively. The Asian dust maximal peak in 2001 also agreed with these concentrations (Figure 3.1). Hence these values can be used as a threshold value for severe Asian dust contribution to the Mount Wrangell ice core in spring. In 2002 and 1995, the ADSTT and efficient tritium deposition due to snowfall at the ice core site were expected, based on knowledge of the relationship between dust and tritium in the spring of 2002, as mentioned in section 3.3.3.

Now we consider the tritium raw data points in early spring, which are mainly included between the dating points from early spring to late spring but the edge of the tritium data is occasionally positioned outside the two dating points. If there are two tritium samples from early spring to late spring, we focus on the larger concentration tritium data here. In the period in which the early spring tritium data point is located, occasionally both the fine and coarse dust concentrations in 1993, 1995, 1998, 1999, 2001, and 2002 met the threshold value condition for severe Asian dust contribution. Hence ADSTTs due to severe dust storms may impact on the ice core site in these years. However, its effect may depend on the snowfall condition at Mount Wrangell.

We can mainly separate the relationship between dust and tritium in spring in the Mount Wrangell ice core into four types in terms of the relationships between dust threshold and tritium as mentioned above (Figure 3.22). In 1992, 1994, 1996, and 1997, the dust concentrations in the ice core were normal; Asian dust contributions in these seasons were considered to be normal, and tritium maximal peaks were also located in late spring (Figure 3.22a). Only the 1995, 1999, and 2002 seasons are considered to be perfect cases for simultaneous dust and tritium increases with tritium deposition information present; i.e., ADSTT information was nicely recorded in the Mount

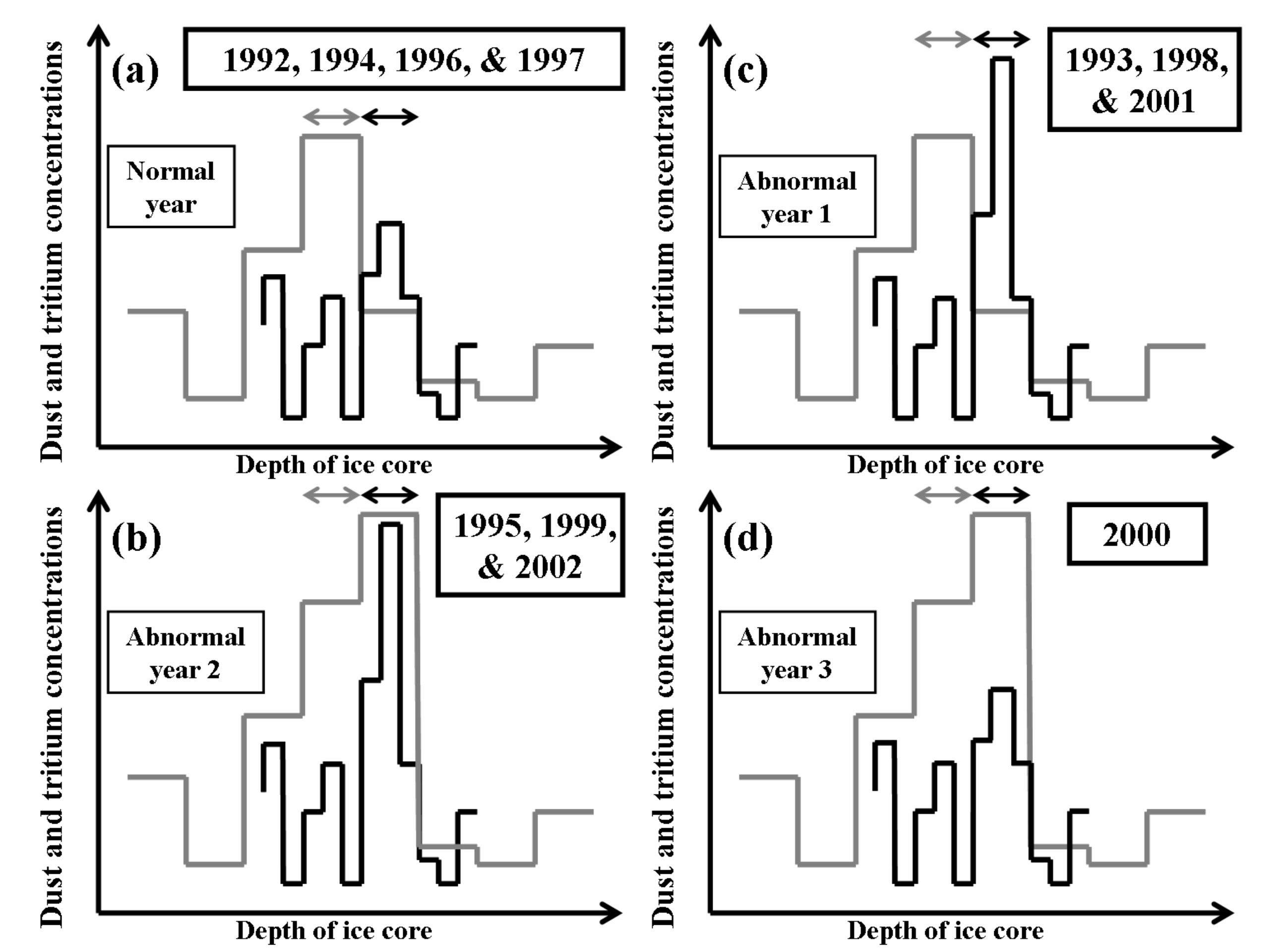


Figure 3.22. Schematic diagram of the relationships between dust and tritium concentrations in spring in the Mount Wrangell ice core. The relationships were divided into four types from (a) to (d). The gray and black solid lines denote tritium and dust concentrations. The gray and black double-headed arrows correspond to the relative positions in late spring and early spring, respectively. In types (b) and (c), there are contributions from severe Asian dust in early spring.

Wrangell ice core. In 1999, the dust increase in early spring was not at the early spring dating point, but met the threshold value for a severe Asian dust storm. In these 3 years, tritium maximal peaks are considered to be able to shift from late spring to early spring due to the ADSTT contribution to the ice core site (Figure 3.22b). Because the tritium annual cycle, in general, has a late spring maximum in normal years in the ice core, those years are considered to be abnormal years. The tritium concentrations from early spring to late spring in 1993, 1998, and 2001, which we focus on here, were not the maximal peaks. The tritium peaks from early spring to late spring in those years were the second, the third, and the second maxima, respectively (see Figure 2 in YS2007; Figure 2.2). In these 3 years, although the effect due to ADSTT probably reached Mount Wrangell, information about tritium deposition was perhaps lost to some extent because of the lack of snowfall at the ice core site, such as in the case of the 6–7 April 2001 dust storm (Figure 3.22c). In 2000, the tritium maximal peak was located from early spring to late spring (Figure 3.22d). However, fine and coarse dust concentrations were normal; therefore, we can consider two possibilities for the tritium peak in early spring. One is the lack of Asian dust information on the ice core site. The other is the strong OTSTT effect on the ice core site. The former is a better explanation because the number of Asian dust storms has increased since 2000 [*JMA*, 2006] and, in fact, deep tropopause folding occurred in the 5–7 April 2000 severe dust storm in East Asia [*Zhao and Zhao*, 2006]. Hence information about dust deposition on the ice core site for 2000 may be lost. In 1994 and 1997, normal tritium peaks, similar to those in Figure 3.22a, were observed. The tritium concentrations from early spring to late spring in those years were not maxima but were considerably higher with no expected ADSTT effect. Hence OTSTT effects on the ice core site were expected to some extent. In normal years, the tritium maximal peak in late spring can be explained by large meridional circulation in the stratosphere and normal eddy-forced STT, as mentioned below.

The late spring maximum of tritium in normal years can be explained well by the annual cycle of the Brewer–Dobson circulation in the stratosphere, which consists of upwelling in the Tropics and downward transport in the extratropics [*Brewer*, 1949; *Dobson*, 1956] as was also calculated in detail in *Appenzeller et al.* [1996]. Tritium is mainly produced by cosmic rays in the upper troposphere and the lower stratosphere [*Gat et al.*, 2001]. The concentration of tritium is much higher in the stratosphere [*Ehhalt et al.*, 2002], and advection of tritium into the troposphere is very important for the annual cycle of tritium concentration in the troposphere. The mass in the lower stratosphere has a maximum in mid-winter, and this corresponded well with the mass decrease in the overworld stratosphere in the same season [*Appenzeller et al.*, 1996]. The Brewer–Dobson circulation in the stratosphere intensifies in winter. This is well explained by the downward control theory as due to planetary-wave propagation from the troposphere and its breaking in the stratosphere [*Haynes et al.*, 1991]. In the results of *Appenzeller et al.* [1996], the stratospheric mass accumulates in the lower stratosphere from mid-winter to early spring and the outflow of the stratospheric mass into the troposphere is the largest in late spring (May and June). Tritium peaks in late spring in the ice core correspond well to this annual cycle. We also calculated the vertical velocity of residual mean meridional circulation ($\overline{w^*}$) at 100 hPa in the extratropics, which is near the boundary between the overworld and the lowermost stratosphere (Figure 3.23). This velocity means the intensity of the Brewer–Dobson circulation. $\overline{w^*}$ is defined as

$$\overline{w^*} \equiv \overline{w} + \frac{1}{a\cos\phi}\frac{\partial}{\partial\phi}\left(\cos\phi\frac{\overline{v'\theta'}}{\frac{\partial\overline{\theta}}{\partial z^*}}\right), \tag{3.1}$$

where a, ϕ, v, θ, and z^* are the radius of the earth, latitude, meridional wind velocity, potential temperature, and altitude in log-pressure coordinates, respectively [*Andrews et al.*, 1987]. The bar and prime denote zonal mean and eddy components, respectively. Figure 3.23 implies that the mass flux at 100 hPa is the highest in winter (December and January) every year, and it may take a few months for the dense stratospheric mass to reach the tropopause from the 100 hPa level. A large amount of stratospheric tracers in higher concentrations are transported from the middle-upper stratosphere to the lowermost stratosphere from winter to early spring due to intensified Brewer–Dobson circulation. The stratospheric mass in higher concentrations may reach the tropopause region in early spring (March and April). In late spring (May and June), mean transport due to the Brewer–Dobson circulation is minor and the stratospheric materials are discharged from the lowermost stratosphere because of eddy transport [*Miyazaki et al.*, 2005b]. Tritium maxima are observed in late spring in the troposphere in normal years. Some tritium peaks in raw observed tritium data in early spring in the ice core may be explained by the additional effect of strong eddy-forced STTs (ADSTT and OTSTT) on the seasonal tritium cycle by the Brewer–Dobson circulation. Hence the typical annual cycle of tritium variation in a normal year may be well explained by the Brewer–Dobson circulation with normal eddy-forced STT at the tropopause region.

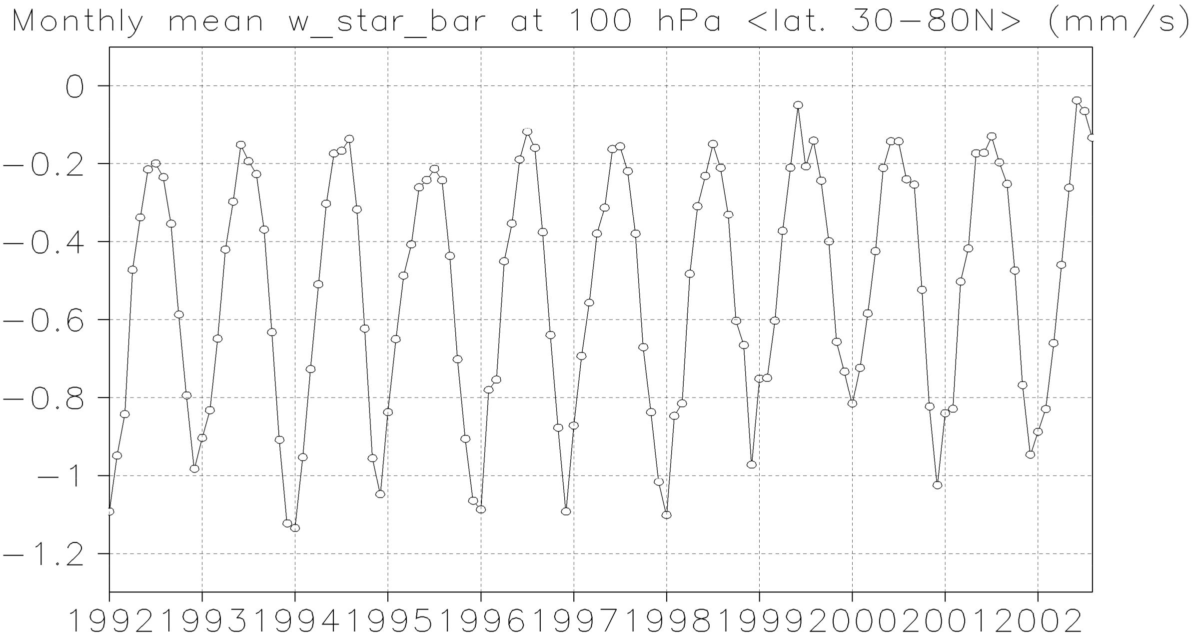


Figure 3.23. Vertical component of residual mean meridional circulation at the 100 hPa level. The unit is mm s^{-1}, and the values from 30ºN to 80ºN were averaged. Minus values denote downward velocity of the vertical component.

In addition, seasonal snowy conditions in April and May at the Mount Wrangell ice core site are perfect for detecting the annual tritium cycle. Tritium in the atmosphere mainly exists as tritiated water vapor and prefers snow (wet) deposition. In fact, a very high correlation was observed between tritium flux and snow accumulation in the ice core [YS2007]. In April and May, the number of snowy days is the highest at the ice core site among all the months (Figure 3.16b). When it snowed at the ice core site, high PV-values (tropopause descendings) were observed near Alaska from the Bering Sea to the Gulf of Alaska (Figure 3.18). The OTSTT may occur in the high-PV regions. These high PV regions are well explained by the RSTTP from the storm track positions (Figure 3.20). The intensified storm track may also efficiently bring water vapor to the ice core site and lead to snowy conditions at the summit.

If we consider the tritium concentration in the late spring of 2001 and 2002 (TF2001 and TS2002), the concentrations were 10.2 and 10.05 TU, respectively (Figure 3.1). Tritium decays by approximately 5% in 1 year from 2001 to 2002. TF2001 is just a few percent higher than that in TS2002, but there is not a great difference between the tritium concentrations in the late spring of 2001 and 2002. The tritium concentration from early spring to late spring in 2002 was much higher because of the abnormal ADSTT contribution. Considering the dates of the Asian dust peaks in 2001 and 2002 in Figure 3.1 and the approximate time resolution of the tritium samples, i.e. roughly 1 month resolution, TS2001, TF2001, TF2002, and TS2002 may mainly reflect the atmospheric information in April, May, March–April, and April–May, respectively (Figure 3.1). The PV values near Alaska in May 2001 intensified more than those in April–May 2002 (Figure 3.17c, 3.17f, and 3.17g). We consider that the differences in the intensity of OTSTT near Alaska may contribute a few percent differences of tritium concentrations in TF2001 and TS2002. The OTSTT near Alaska may produce efficient tritium deposition the ice core site by snowfall. Hence TF2001, which is a few percent higher than TS2002, is probably due to the difference in OTSTT near Alaska. However, quantitative discussions are still difficult at the present time. The abnormal ADSTT contribution due to a severe dust storm can induce a greater concentration increase in tritium, such as the 2002 case in Figure 3.1.

Eddy development in the Gulf of Alaska may be more favorable for efficient tritium deposition onto the ice core site than that in the Bering Sea or over Alaska, in terms of atmospheric water vapor. In April and May 2002, the number of snowy days were the lowest in 1992–2002 (Figure 3.16b) and PV values in the Gulf of Alaska were not as high (Figure 3.17f and 3.17g) as those in 2001 (Figure 3.17b and 3.17c). We think that this caused the least snowy days at Mount Wrangell in the spring of 2002 in the 11-year climatology. The probability of efficient tritium deposition onto the ice core site was possibly less in the spring of 2002. Fortunately, when the stratospheric air due to ADSTT was transported to Mount Wrangell, sufficient snowfall was expected in the two severe dust storms in 2002, as mentioned in section 3.3.3. The tritium concentration in TF2002 reached a maximum in 2002.

In general, the SST in the Gulf of Alaska is much higher than that in the Bering Sea (Figure 3.19e–h). Transport of a large amount of water vapor from the ocean to Mount Wrangell is favorable for snowy conditions at the ice core site. This leads to efficient tritium deposition by snowfall. On the other hand, cold SST in the Bering Sea is perhaps detrimental to a large amount of water vapor transport than warm STT. Hence OTSTT near Alaska including the Bering Sea, Alaska, and the Gulf of Alaska is important for tritium advection into the troposphere, but more efficient tritium deposition is possible when cyclones are developing in the Gulf of Alaska. In conclusion, storm track positions in the North Pacific Ocean may control efficient tritium deposition onto the Mount Wrangell ice core site.

On the other hand, the ADSTT contribution is also very important for tritium variations in the ice core as shown in sections 3.3.2 and 3.3.3. When both the ADSTT and snowfall at Mount

Wrangell occur in one dust storm event, the tritium of ADSTT origin can be efficiently deposited onto the ice core site such as in the cases of the 18(19)–22 March and 5(6)–9 April dust storms in 2002. It may have the ability to shift the annual tritium peak earlier than that in normal years, as shown by the TF2002 peak (Figure 3.1). Unfortunately, the ADSTT contribution due to the 6–7 April 2001 dust storm could not have contributed significantly to the increase in tritium concentration in the TS2001 peak because of lack of efficient tritium deposition due to snowfall.

If we consider the interannual relationship between dust and tritium fluxes in YS2007, a high positive correlation was only seen between fine dust and tritium fluxes. A relationship was not found between coarse dust and tritium fluxes. This can be interpreted as follows. The coarse dust in YS2007 showed a clear annual cycle with early spring maxima and its correlation with fine dust was high. In fact, severe Asian dust storms transported coarser dust with fine dust to Mount Wrangell as explained in sections 3.3.1–3.3.3. However, coarser dust is easy to deposit without snowfall by gravity settlement and dry deposition. The correlation between coarse dust flux and snow accumulation was lower in spring [YS2007]. In addition, a local dust contribution to coarse dust may exist to some extent, even though the most of the local dust, huge dust, was removed in their study. As a result, the correlation between coarse dust and tritium fluxes might have been poorer in their study.

Finally, a positive interannual correlation was found between fine dust and tritium fluxes in spring in YS2007. They suggested that Asian dust outbreaks and ADSTT are interannually connected due to cyclonic activities in East Asia. Taking our study into account, this is partly very likely but not certain, because we found that the tritium contribution to the Mount Wrangell ice core was mainly due to ADSTT and OTSTT in the spring season, as mentioned. As was discussed, snowy conditions at Mount Wrangell are also important for the increase in tritium concentration in the ice core. The correlation between fine dust and tritium from early spring to late spring in YS2007 was lower than that from late spring to summer. This may partly depend on the lower snowfall frequency in March than in April and May (Figure 3.16b). As a result, the correlation was higher in late spring corresponding to April and May, in which the snowfall frequency was higher.

A pair of vortex structures of eddies developing over Siberia and near Alaska is often seen in March–May (Figure 3.17a, 3.17b, 3.17c, 3.17f, and 3.17g). We consider that this is a most important factor in explaining the mechanism of interannual variations of dust and tritium and their connection in spring. This suggests that eddies of Siberian origin and the North Pacific storm tracks are closely connected. Hence if developed cyclones in East Asia, which are originally produced in the Siberian region, are able to reach areas near Alaska, such as the Bering Sea and the Gulf of Alaska, while maintaining their eddy intensity, the cyclones can also contribute to OTSTT near Alaska and snowfall at the Mount Wrangell ice core site. The 18(19)–22 March 2002 and 5(6)–9 April 2002 dust storms illustrate this case well. *Nakamura* [1992] mentioned that baroclinic waves from Siberia may play a roll in "seeding" baroclinic disturbances that grow over the western Pacific. Our results also support this description. The monthly mean atmospheric circulation in Figure 3.17 may also determine the snowy conditions at the ice core site because the circulation patterns in Figure 3.18 were similar to the monthly mean patterns.

To lend clear support to this discussion, we conducted correlation analysis for the interannual variations in each month (March–June) between one-point PV variation at 250 hPa, which was averaged in the Gobi Desert region in East Asia, and PV variations at 250 hPa (Figure 3.24). Here we focus on cyclones that are mainly produced in the Siberian region and pass the area of Asian dust outbreaks. The monthly mean data in March–June in ERA-40 was used from 1958 to 2002.

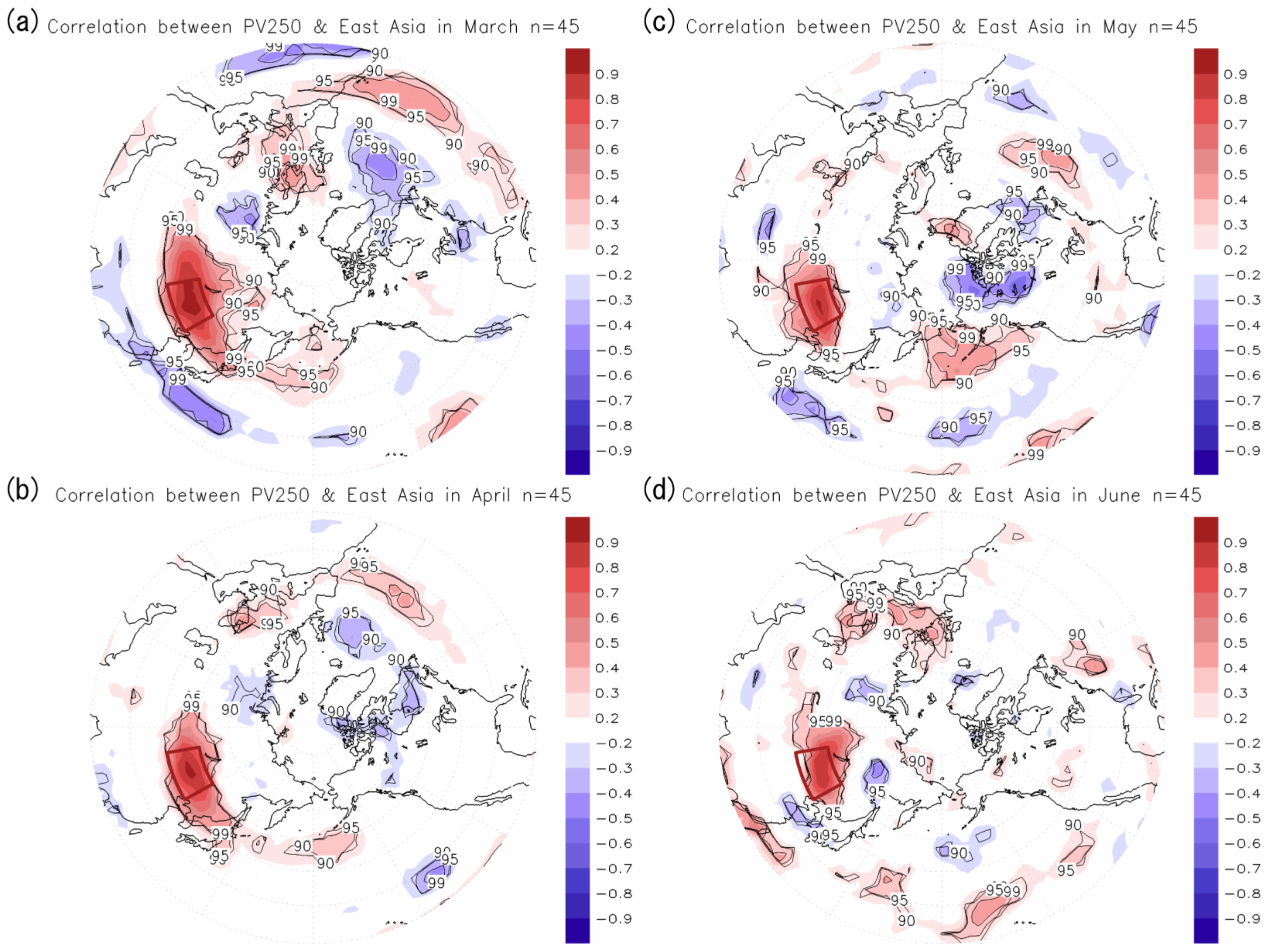


Figure 3.24. Correlation coefficients in each month (March–June) between one-point PV-variation at 250 hPa, which was averaged in the Gobi Desert region, and PV variations at the 250-hPa level. Color shaded and line contours denote correlation coefficients and 90%, 95%, and 99% confidence limits, respectively. Red fan-shaped area with bold lines is the Gobi Desert region where PV data were averaged.

The results clearly showed that if a vortex at the upper troposphere in Asian dust region is intensified, Aleutian low or cyclonic activities in the Gulf of Alaska are also simultaneously intensified. In March and April, a co-intensified eddy was positioned in the Aleutian region with a strong eddy in the Gobi Desert region. In May, its intensified position was extended to the Gulf of Alaska. In June, the pair of strong vortices was not seen. This indicates that an Asian dust outbreak with ADSTT, OTSTT in the North Pacific Ocean, and snowy conditions at the ice core site are closely connected to each other in March–May through the variation of a pair of strong PVs. Hence intensified PV values in the Gobi Desert region likely cause the Asian dust outbreak, ADSTT, OTSTT due to high PV values in the Aleutian and the Gulf of Alaska, and snowfall at Mount Wrangell in March–May. In conclusion, the high interannual correlation between dust and tritium flux in the Mount Wrangell ice core [YS2007] is well explained by the interannual variation of a pair of intensified vortices in the upper troposphere in spring in East Asia and the North Pacific.

In Figure 3.25, we summarize the mechanism of tritium variation in spring in the North Pacific region. This also leads to development of the hypothesis in YS2007 as follows. Three components may control the tritium variation in spring in the Mount Wrangell ice core. They are ADSTT due to Asian dust storms, OTSTT in the North Pacific region, and snowy conditions at the Mount Wrangell ice core site. A strong eddy in the Siberian region may cause an Asian dust storm with ADSTT. On the other hand, OTSTT is the result of slow Brewer–Dobson circulation and eddy-forced STT. Both the ADSTT and OTSTT increase the tritium concentration in the atmosphere. The Brewer–Dobson circulation in winter and eddy-forced STT in spring play important rolls in mass transport from the middle-upper stratosphere to the lowermost stratosphere including the tropopause region and from the tropopause region to the troposphere in the extratropics, respectively. This concept is consistent with previous studies by *Miyazaki et al.* [2005a; 2005b]. Both these processes increase the material of stratospheric origin in the troposphere.

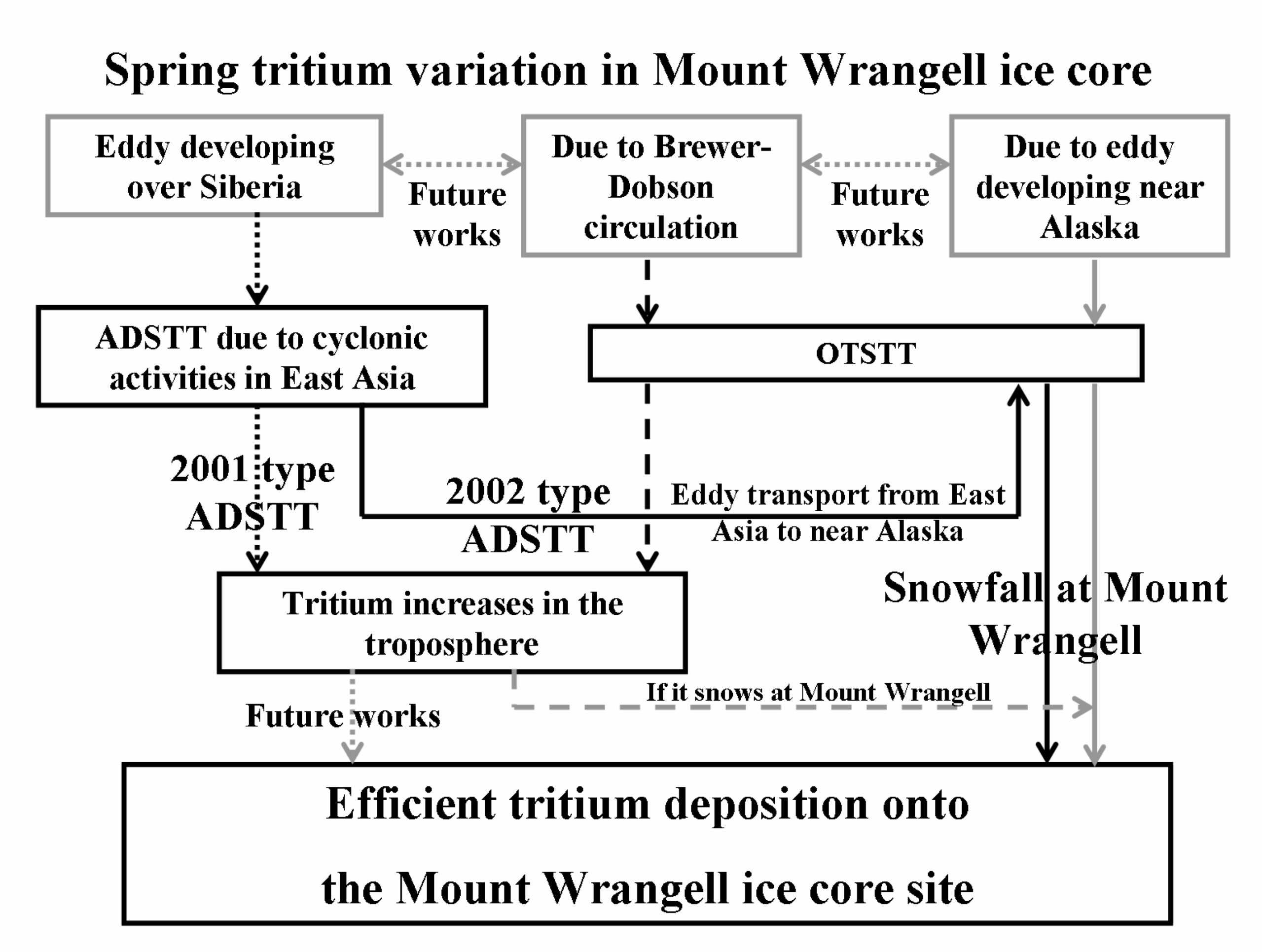


Figure 3.25. Schematic diagram of mechanism of tritium variation in spring in the North Pacific region detected from the Mount Wrangell ice core. The arrows in the same style and shade denote one flowchart. The dotted black arrow denotes the flowchart for the 6–7 April 2001 dust storm case. The solid black arrow denotes the flowchart for the 18(19)–22 March and 5(6)–9 April 2002 dust storms and is likely well explained by Figure 3.24. The gray dashed arrow denotes the flowchart when it snows at the ice core site due to a cyclone developing near Alaska such as in the Aleutians and the Gulf of Alaska. The process proceeds along this gray dashed arrow. The gray dotted arrows denote possible flow charts but the details have not been discussed in this study and will be considered in future studies.

However, the timing of snowfall at the ice core site is also very important for tritium deposition because tritium mainly exists as tritiated water vapor in the atmosphere [*Gat et al.*, 2001]. If snowfall does not occur when tritium from the stratosphere is transported to Mount Wrangell, the tritium increase will not be reflected sufficiently in the ice core, such as in the 6–7 April 2001 dust storm case (Figure 3.1). A pair of high-PV structures of eddies developing in the Siberian region, which may develop in East Asia, and near Alaska (Figure 3.17a, 3.17b, 3.17c, 3.17f, and 3.17g). Then, the interannual high positive correlation between PV in the Gobi Desert region and in the Aleutian region and the Gulf of Alaska was seen (Figure 3.24). It indicates that ADSTT and OTSTT may be dependently connected to each other. If the developed cyclones in East Asia reach areas near Alaska such as the Bering Sea and the Gulf of Alaska while maintaining eddy intensities, the cyclones can also contribute to OTSTT near Alaska and snowfall at the Mount Wrangell ice core site such as in the 18(19)–22 March 2002 and 5(6)–9 April 2002 dust storm cases. When the eddy-forced OTSTT developed near Alaska, snowfall occurred at the ice core site at the same time. This suggests that eddy-forced OTSTT near Alaska and efficient tritium deposition onto the ice core site by snowfall are closely associated. The eddy-forced OTSTT near Alaska corresponded well with the RSTTP. The contribution of the eddy-forced OTSTT in the Gulf of Alaska is expected to be very favorable for efficient tritium deposition due to snowfall at Mount Wrangell associated with much water vapor transport from warmer conditions in the Gulf of Alaska. The positions of eddy development in East Asia and the North Pacific Ocean are important in determining their relationships. This implies that if we compare the concentrations and peak positions of dust and tritium in spring in the Mount Wrangell ice core (Figure 3.22), we can anticipate the atmospheric circulation patterns and the central position of the storm track in the North Pacific region in the past. In conclusion, Asian dust outbreaks with ADSTT, OTSTT near Alaska, and snowfall at Mount Wrangell are closely connected, and they may be interannually connected. As a result, positive interannual correlations between fine dust and tritium were seen in the Mount Wrangell ice core.

3.4 Conclusions

We investigated Asian dust storms with ADSTT, OTSTT contributions to the Mount Wrangell ice core site, and snowfall conditions at the site to explain the dust and tritium variations in spring in the ice core and to develop the hypothesis in YS2007. We found the following in terms of meteorological analyses: (1) Severe Asian dust storms and STT occurred simultaneously in all five severe dust storm events in the spring of 2001 and 2002. (2) Both the Asian dust and the stratospheric air were transported exactly to the Wrangell Area in three of the five cases. (3) If snowfall occurs at the ice core site when tritium due to ADSTT is transported to Mount Wrangell, efficient tritium deposition is expected onto the ice core site. A strong ADSTT contribution may have the ability to shift the annual tritium peak earlier than in normal years. (4) The tritium spring maximum in the ice core can move from late spring to early spring if there is no loss of information about tritium deposition because of lack of snowfall at the ice core site. The tritium annual cycle with a late spring maximum in the ice core of YS2007 in normal years, in general, can be explained by the annual cycle of the Brewer–Dobson circulation in the stratosphere and normal eddy-forced

ADSTT and OTSTT. (5) The amount of stratospheric material intrusions into the troposphere in spring may largely depend on the intensities of eddy-forced ADSTT, eddy-forced OTSTT near Alaska, and OTSTT due to slow Brewer–Dobson circulation. (6) Furthermore, April and May are the most frequent months for snowfall in climatology with better conditions for tritium deposition onto the ice core site due to snowfall. (7) When it snows at the ice core site, eddies developing near Alaska, which imply eddy-forced OTSTT, are often seen and efficient tritium contributions to the Mount Wrangell ice core site are expected. This suggests that both the OTSTTs near Alaska and snowfall at Mount Wrangell are closely related. (8) A pair structure of eddies developing in East Asia, originally produced in the Siberian region, and near Alaska implies that ADSTT and OTSTT in the Aleutian and the Gulf of Alaska are associated with each other. In fact, they have a high interannual correlation. (9) Finally, the positive interannual correlation between fine dust and tritium fluxes in spring in the Mount Wrangell ice core in YS2007 is explained well by the close connection among Asian dust storms with ADSTT, the OTSTTs in the Aleutian region and the Gulf of Alaska, and snowfall conditions at the Mount Wrangell ice core site.

If dust and stratospheric tracers such as tritium are analyzed from the ice cores drilled in the North Pacific region, we can assess the historical relationships among Asian dust, ADSTT, and OTSTT information. In the case of the Mount Wrangell ice core, if we compare the concentrations and peak positions between dust and tritium in spring in the ice core data, such as in Figure 3.22, we can consider the atmospheric circulation patterns and the central position of the storm tracks in the North Pacific region in the past. For that purpose, as a first step, seasonal or intraseasonal ice core data, such as the YS2007 ice core data, are essential. Comparing the ice core data with the atmospheric circulation in detail in the past several decades is the second step. After that, we can discuss and reconstruct material and atmospheric circulation in the distant past. Then, ice cores in the North Pacific region can be used to reconstruct the information on these circulations from the distant past to the present. Finally, ice core research in the North Pacific region would greatly contribute to future climate projections in terms of material and atmospheric circulation from the stratosphere to the troposphere. Only a few ice cores have been drilled thus far in the North Pacific region. The locations are Mount Logan [*Holdsworth et al.*, 1992; *Shiraiwa et al.*, 2003], Eclipse ice field [*Wake et al.*, 2002] in the Yukon Territory, Canada; Mount Bona Churchill by L. G. Thompson; Mount Ushkovsky and Mount Ichinsky in Kamchatka, Russia [*Shiraiwa et al.*, 2001; *Matoba et al.*, 2007]; and Mount Wrangell in the Saint Elias Range, Alaska [*Shiraiwa et al.*, 2004]. Our results in this study will be very useful for these ice core studies.

The analysis of tritium in ice cores in the North Pacific region can be used to reconstruct tritium information of pure stratospheric origin, which is produced by cosmic rays in the stratosphere [YS2007], back to around 1980. However, the effects of past nuclear tests in the Northern Hemisphere might have persisted from 1954 to around 1980. We can discuss the increased timing of dust and tritium and the contributions of ADSTT and OTSTT in spring in each year before 1980, but a detailed year by year comparison between them may be difficult because of the disturbances by anthropogenic tritium. In either case, the half-life of tritium is 12.32 years [*Lucas and Unterweger*, 2000], and there were no drastic nuclear contributions to the atmospheric tritium before 1954 [*Gat et al.*, 2001]. Hence we cannot measure the tritium concentration in ice core before 1954 because it is probably below the detection limit for tritium measurement. Other methods are necessary to reconstruct pure STT information for the period before 1954. One possibility for the reconstruction of STT information in the distant past is the use of other stratospheric tracers such as a beryllium isotope with a longer half-life time (^{10}Be: 1.5×10^6 years) because it has been measured in ice cores [e.g., *Beer et al.*, 1988; *Smith et al.*, 2000]. However, if the interannual relationship between dust and tritium fluxes in YS2007 persisted before 1980, it is possible to reconstruct the STT information indirectly by means of dust analysis in ice cores in the North Pacific region. In all the cases, more comprehensive studies are required on STT, Asian dust storms, cyclonic activity, and ice cores in the North Pacific region.

Chapter 4

Summary

Studies on the relationship between atmospheric dust and tritium were carried out using ice cores. The Mount Wrangell ice core contained precise information on the materials from the past to the present suggesting clear seasonal cycles. As a result, the inter- and intra-annual relationships between atmospheric dust as a representative tracer in the troposphere and tritium in the stratosphere became clear in the North Pacific region for the first time. In spring, the contribution due to Asian dust storms in East Asia is large at Mount Wrangell. In addition, the higher altitude is favorable for the observation of stratospheric tracers. Ice cores can provide information on the stratospheric and tropospheric materials at a specific location from the past to the present. Therefore, these inter- and intra-annual relationships are important for material circulation with climate change in the North Pacific region.

The studies on the relationship between Asian dust outbreaks and the stratosphere-to-troposphere transport (STT) due to Asian dust storms (ADSTT) in East Asia and the effect of other STT (OTSTT) due to cyclonic activities in spring in the North Pacific region were examined by means of meteorological analyses. Ice cores only show point data at a certain drilling point and one of difficulties in ice core research is that the spatial representation is unknown. This difficulty can be corrected by providing spatial information in terms of meteorology. Accordingly, simultaneous Asian dust outbreaks and STTs were verified and the information exactly reached Mount Wrangell, providing spatial information. In addition, the relationships among Asian dust storms and ADSTT due to cyclones developing in East Asia, OTSTT due to eddies developing near Alaska, and snowfall conditions at Mount Wrangell have been clear. Such in-depth analysis could not have been possible without meteorological analyses. Coupling ice cores to meteorology is an essential approach to understand ice core data spatially and reconstruct atmospheric circulation in the past.

The main results obtained from these Ph.D. studies using coupled ice-core-meteorology are summarized as follows:

(1) Dust number and tritium concentrations were analyzed in the 50-m ice core drilled at the summit of Mount Wrangell. The ice-core data were successfully divided into 5 seasons in the North Pacific region for the first time (early spring, late spring, summer, fall, and winter). In general, the dust concentration in the ice core tended to increase in early spring and the tritium increased in late spring. In some years, drastic dust increases were seen in the corresponding tritium peaks in spring in the observed raw data. The dust increases in 2001 and 2002 were

dominant and in fact corresponded to recent increases of Asian dust outbreaks in East Asia. In addition, Asian dust transport was verified in the field campaign in May 2004 at Mount Wrangell. The data suggests that Asian dust loading to the Mount Wrangell ice core is large. It is worth noting that the inter- and intra-annual relationships between dust and tritium at higher altitude in the North Pacific region are now clearly understood. The high interannual correlation between fine dust and tritium fluxes was found only in the spring season. It suggests that Asian dust outbreaks and STT are closely connected due to the inter-annual activities of cyclones in East Asia. This assumption suggests that if Asian dust outbreaks increase, intrusion of stratospheric tracers such as tritium, ozone, and beryllium isotopes into the troposphere may also increase due to intensified cyclonic activities as interannual variation.

(2) Five severe dust storms in the spring of 2001 and 2002 were investigated in terms of Asian dust outbreaks with STT and its impact on the Mount Wrangell ice core site. Asian dust outbreaks and STT occurred simultaneously in all five cases. The transport of Asian dust and stratospheric material due to STT to the ice core site were seen in 3 of 5 cases, while Asian dusts were transported to the site in all cases. A clear difference was seen between the 2001 (a 6–7 April 2001 dust storm) and the 2002 (18(19)–22 March 2002 and 5(6)–9 April 2002 dust storms) cases when materials were transported to the ice core site. The difference was whether snowfall occurred or not when dust and stratospheric tracers were transported to Mount Wrangell. In the

2002 cases, snowfall did occur and the tritium concentration corresponding to the Asian dust peaks in the ice core showed a maximum in 2002. However, dominant snowfall was not seen in 2001 and the tritium concentration was the second highest. Thus, snowfall at Mount Wrangell is an important factor to determine tritium variation in the ice core. The atmospheric circulation patterns when it was snowing at Mount Wrangell were quite similar with the monthly mean atmospheric pressure pattern. This suggests that the monthly atmospheric circulation determines the snowfall conditions at Mount Wrangell. These atmospheric circulation patterns also showed possible tritium contributions to the ice core site due to OTSTT by an eddy developing near Alaska when it was snowing at the ice core site. Hence, snowfall and OTSTT due to eddies developing near Alaska are closely connected. In addition, the estimated number of snowy days at the ice core site was highest in April and May in an 11-year climatology. This leads to favorable conditions for efficient tritium deposition by snowfall at the site. In general, the annual cycle of tritium concentration with late spring maxima in the ice core can be well explained by previous studies such as *Appenzeller et al.* [1996] that the downward mass flux at tropopause takes maxima in late spring (May and June). Namely, the seasonal cycle of Brewer–Dobson circulation in the stratosphere may bring stratospheric materials in the middle-upper stratosphere to the lowermost stratosphere from winter to early spring and the discharge amount of those materials into the troposphere by eddy takes maxima in late spring as mentioned by *Miyazaki et al.* [2005b]. Then, tritium in the ice core has maximal peaks in late spring in normal years. Therefore, this may be determined by the total effect of clear Brewer–Dobson circulation from winter to spring and normal eddy-forced ADSTT and OTSTT near Alaska in spring. However, in abnormal years such as 2002, additional tritium contribution was dominant due to strong eddy-forced ADSTT in East Asia in early spring. Then, exceptional tritium discharge into the troposphere in early spring of 2002 may be added onto normal seasonal cycle of tritium concentration. As a result, the tritium maximal peak can shift from late spring to early spring, as in the 2002 case. In conclusion, past atmospheric circulation patterns associated with Asian dust and STT in spring in the North Pacific region can be determined by dust and tritium in the ice core based on peak positions and concentrations (Figures 3.25 and 4.1).

Still unsolved work remains for the future. Tritium may be difficult for reconstructing past stratospheric information before 1954 because the half-life of tritium is short and massive injections of tritium into the atmosphere by nuclear tests were carried out after 1954 [*Gat et al.*, 2001]. One of the alternative stratospheric tracers for reconstructing distant past changes before 1954 is beryllium 10 (^{10}Be). Its half-life is 1.5×10^6 yr and it has actually been measured in ice cores [*Beer et al.*, 1988; *Smith et al.*, 2000]. If we use ^{10}Be before 1954, we can reconstruct the stratospheric information of the distant past in the North Pacific region. Then, we can compare ^{10}Be with dust concentration of the distant past.

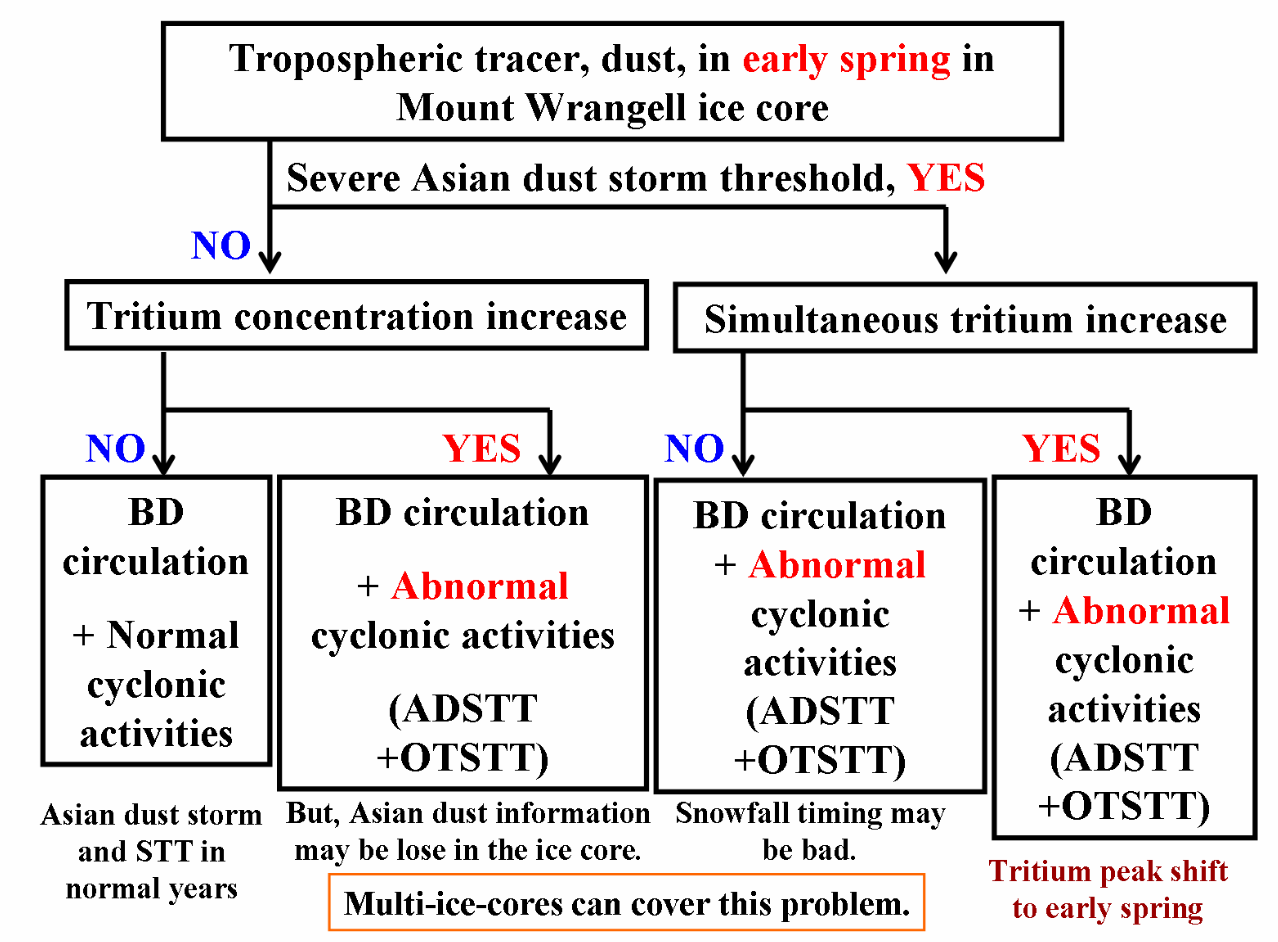


Figure 4.1. The method of extracting the information from the ice core regarding severe Asian dust storms, ADSTT, OTSTT near Alaska in the North Pacific Ocean, and snowfall at the Mount Wrangell ice core site. We compared dust and tritium concentrations in early spring. If the dust concentration was satisfied with the threshold of severe Asian dust storms as defined in chapter 3, the arrow goes to "YES." The following questions are the same. The timing of Brewer–Dobson circulation intensification is in winter (December and January) as mentioned in Figure 3.23. Then, higher concentrations of stratospheric tracers may reach around the tropopause in spring. Finally, the timing of its discharge into the troposphere may be because of the cyclonic activities in East Asia and near Alaska in the North Pacific Ocean. The final box at the left side and the right side are normal years and abnormal years, respectively. The center-boxes are involved in the case of the box at the right side, but dust or tritium information in the ice core is unfortunately lacking. This is a problem of using a single ice core. Using multi-ice-cores in the North Pacific region can eliminate this problem in the near future.

We need to investigate the following problems in order to understand the connection between the stratosphere and the troposphere and assess future climate change: (1) How much does ADSTT impact on total mass intrusion into the troposphere in spring in the Northern hemisphere? (2) What quantity of stratospheric tracers are transported to other ice-core sites in the North Pacific region such as Mount Logan [*Holdsworth et al.*, 1992; *Shiraiwa et al.*, 2003]; the Eclipse ice field [*Wake et al.*, 2002] in the Yukon Territory, Canada; Mount Bona Churchill by L. G. Thompson; and Mount Ushkovsky and Mount Ichinsky in Kamchatka, Russia [*Shiraiwa et al.*, 2001; *Matoba et al.*, 2007]? (3) How closely are ADSTT in East Asia and OTSTT in the North Pacific Ocean connected in intra-seasonal or decadal time-scales? (4) How strongly are the Brewer–Dobson circulation in the lowermost stratosphere and eddy-forced ADSTT and OTSTT near Alaska associated? (5) How widely does ADSTT affect the seasonal cycle of stratospheric tracers in the troposphere (i.e., the effects in the North Atlantic region, European region, North African region, etc.)? (6) How much does ozone intrusion into the troposphere due to ADSTT and OTSTT change the greenhouse effect in the troposphere and does Asian dust uplifting cancel ozone's greenhouse effect?

It is worth noting that reconstructing the connection between the stratosphere and the troposphere from the ice core from the near past to the present was shown in this study. In general, ice cores have often been used for reconstructing tropospheric climate information of the past. However, there have been no ice core studies focusing on the reconstruction of the direct interaction between the stratosphere and the troposphere in the past with any understanding of spatial information. The troposphere and the stratosphere are closely connected through the interactions between them. Thus, reconstructing the connection between them using ice cores is important for understanding atmospheric circulation from the troposphere to the stratosphere in the past. This Ph.D. study showed progress on reconstructing the past interaction between the stratosphere and the troposphere using ice core data. Ice cores are one of the most useful tools for reconstructing tropospheric and stratospheric information.

This Ph.D. study contributes to the basic research on future climate and environmental predictions. To predict global warming in the future, it is essential to estimate the amount of materials, which affect the radiative balance in the troposphere as mentioned in Figure 1.1. This study is useful for assessing the springtime relationship between Asian dust storms and STT because both atmospheric dust and ozone have the capacity to change the global radiation budget in terms of radiative forcing. Therefore, additional research focusing on both study fields of cyclonic activities in spring in the North Pacific region from East Asia to the North Pacific Ocean affecting Asian dust and STT should be encouraged. I hope more studies of this kind will develop in the near future, as they would prove to be very useful for predicting climate.

Acknowledgements

I would like to heartily thank Professor Takeo Hondoh, Institute of Low Temperature Science (ILTS), Hokkaido University, for giving me the opportunity to think about all kinds of studies thoroughly with logicalness, to take part in various symposiums, meetings and field observations for study, and for guiding me to right way in scientific field.

I express my cordial thanks to Dr. Takayuki Shiraiwa, Research Institute for Humanity and Nature (ILTS, Hokkaido University), for my participation to the field work at Mt. Wrangell drilling campaign in 2004, to support to observation and life in severe environment, and to give me useful comments for my study especially on ice core research.

Professor Koji Yamazaki, Graduate School of Environmental Earth Science, Hokkaido University, lets me use his trajectory model, atmospheric data in the data server, blade, of the Laboratory of Ocean and Atmospheric Dynamics, gave me a lot of comments on meteorological calculation techniques, and tell me a meteorological point of view with enough kindness.

Professor Yoshiyuki Fujii, National Institute of Polar Research (NIPR), kindly gave me great comments for dust analysis, for its data interpretation, and for sharing his interest of dust with me.

Dr. Makoto Igarashi, RIKEN Wako Institute, kindly told me ice core treatments and gave me useful comments for the details about ion chromatograph and its data analyses, and for dealing with some trouble for the instruments.

Professor Teruyuki Nakajima, Center for Climate System Research (CCSR), University of

Tokyo, and Dr. Yoshinori IIZUKA, ILTS, Hokkaido University, kindly gave me useful comments on my doctoral dissertation.

I would like to give my sincere appreciation to Dr. Carl S. Benson, Geophysical Institute, University of Alaska Fairbanks (UAF), for all the support in Mt. Wrangell 2004 campaign. I was very grad to collaborate with him, a great glaciologist. I appreciate he gave me a lot of useful comments on paper preparation to submit to research journals.

Dr. Yasunori Kurosaki, Georgia Institute of Technology, kindly gave me a lot of Asian dust information and useful comments. I was very glad to have had significant discussions associating with Asian dust.

Dr. Kumiko-Goto Azuma, NIPR, supported my research works and let me analyze my ice core samples at NIPR with kindness.

Dr. Masamitsu Hayasaki, National Institute for Environmental Studies, kindly gave me the information of Asian dust and cold surge, and discussed it with me with plenty of time.

Dr. Atsushi Matsuki, Laboratoire de Meteorologie Physique, Universite Blaise Pascal, kindly gave me a lot of information and comments on aerosol research and told me scanning electron microscope (SEM) techniques with sufficient kindness.

Dr. Yongwon Kim, International Arctic Research Center (IARC), UAF, gave me a lot of useful infromation on black carbon analysis and Alaskan aerosol information.

Dr. Atsushi Miyamoto, ILTS, Hokkaido University, always kindly check my manuscripts for the presentation in symposium, and gave me important comments on daily research life.

Dr. Teruo Aoki, Meteorological Research Institute, kindly gave me opportunities to participate in the Saroma observation for the work of the Global Imager Cryosphere group, to do frequent snow pit works at the Institute of Low temperature Science, let me use an albedometer for the Mt. Wrangell observation, and gave me useful comment for aerosol deposition by snowfall.

Professor Masahiko Hayashi, Fukuoka University, kindly gave me great comments for the aerosol measurement and its data analysis.

Professor Masami Fukuda, IARC, UAF, gave me useful comments and data about forest fire in Siberia and Alaska with enough kindness.

Dr. Yuji Kodama, ILTS, Hokkaido University, tell me a lot of knowledge on snow pit works and let me take part in the snow pit works at ILTS.

Dr. Konosuke Sugiura, Institute of Observational Research for Global Change (IORGC), Japan Agency for Marine-Earth Science and Technology (JAMSTEC), kindly let me use a snow particle counter in the Mt. Wrangell observation.

Dr. Keiko Konya, IORGC, JAMSTEC, kindly let me use an anemometer for the field observation at Mount Wrangell.

Dr. Keiichiro Hara, Fukuoka University, kindly gave me useful comments for aerosol measurement in cold region, and for aerosol study.

Dr. Yoshimitsu Chikamoto, CCSR, University of Tokyo, told me a lot of useful technical methods on numerical calculations in meteorological analyses.

Dr. Shinichiro Horikawa gave me useful comments on the program matters for programming and told me SEM tequniques.

Dr. Hiroaki Hatsushika, Central Research Institute of Electric Power Industry, Dr. Jun Inoue, IORGC, JAMSTEC, and Dr. Yoshihiro Tomikawa, NIPR, kindly gave me good comments about trajectory analysis. Dr. Yoshihiro Tomikawa also kindly gave me a lot of useful comments on middle-atmosphere dynamics with calculation techniques.

Dr. Sumito Matoba, ILTS, Hokkaido University, gave me useful comments on chemical analyses for ice core.

Mss. Tomoko Naka, Maki Nakada and Tomoko Kobayashi at NIPR, and Mika Watanabe at ILTS kindly supported me dust analysis, tritium analysis, ion analyses at NIPR and ILTS, and gave me useful comment about them. Dust and tritium in the snow samples and ice cores were analyzed at NIPR.

Tephra samples in the Mount Wrangell ice core were analyzed by Dr. Kristi Wallace of USGS Anchorage.

I would like to appreciate Ms. Kaori Kidahashi, ILTS, Hokkaido University, with great kindness for many procedures and supports to business trip and symposium.

I would like to greatly thank all the members of Mt. Wrangell Ice Core Drilling Campaign 2004. Mr. Syosaku Kanamori kindly gave me good advice for staying in mountain and had a lot of discussions on researches associating with Mount Wrangell. I'm also glad to have just finished doctoral dissertation at the same time in Hokkaido University.

I would like to appreciate all the members of Saroma Field Experiment 2004 and glad to have had nice snow field observations in Hokkaido.

I would like to appreciate all the members and staffs in our group and in NIPR for supporting my daily study life. Especially, Ms. Asako Sakamaki always arranged my stay at NIPR and supported my study at NIPR.

Professor Kunio Rikiishi, Dr. Yasumasa Kodama, and Dr. Sachinobu Ishida, Hirosaki University, Mr. Naohiko Hirasawa, NIPR and Dr. Masahiro Hori, Japan Aerospace Exploration Agency (JAXA), Earth Observation Research and application Center (EORC) gave me useful comments to determine the subject in graduate school and I'm glad to have had nice discussions.

The 21st COE Program in Graduate School of Environmental Science and Institute of Low Temperature Science, Hokkaido University, gave me opportunities to work for my research as a Research Assistant in doctoral course.

Finally, I would heartily thank my family for supporting my research life. Professor Tetsuzo Yasunari, my father, sometimes gave me great comments as a father and as a scientist.

References

Andrews, D. G., J. R. Holton, and C. B. Leovy (1987), Middle Atmosphere Dynamics, 489 pp., Elsevier, New York.

Appenzeller, C., J. R. Holton, and K. H. Rosenlof (1996), Seasonal variation of mass transport across the tropopause, *J. Geophys. Res.*, *101*, 15,071–15,078.

Beer, J., U. Siegenthaler, G. Bonani, R. C. Finkel, H. Oeschger, M. Suter, and W. Wolfli (1988), Information on past solar-activity and geomagnetism from Be-10 in the camp century ice core, *Nature*, *331*(6158), 675–679.

Benson, C. S., and R. J. Motyka (1978), Glacier-volcano interaction on Mt. Wrangell, Alaska, *Geophysical Institute Annual Report*, 1–25, Geophys. Inst., Univ. of Alaska Fairbanks, Fairbanks.

Biscaye, P. E., F. E. Grousset, M. Revel, S. VanderGaast, G. A. Zielinski, A. Vaars, and G. Kukla (1997), Asian provenance of glacial dust (stage 2) in the Greenland Ice Sheet Project 2 Ice Core, Summit, Greenland, *J. Geophys. Res.*, *102*(C12), 26,765–26,781.

Bjerknes, J. (1969), Atmospheric teleconnections from the Equatorial Pacific, *Mon. Wea. Rev.*, *97*, 163–172.

Bory, A. J. M., P. E. Biscaye, A. Svensson, and F. E. Grousset (2002), Seasonal variability in the origin of recent atmospheric mineral dust at NorthGRIP, Greenland, *Earth Planet. Sci. Lett.*, *196*(3–4), 123–134.

Brewer, A. M. (1949), Evidence for a world circulation provided by the measurements of helium and water vapor distribution in the stratosphere, *Q. J. R. Meteorol. Soc.*, *75*, 351–363.

Browning, K. A., and R. Reynolds (1994), Diagnostic study of a narrow cold-frontal rainband and severe winds associated with a stratospheric intrusion, *Q. J. R. Meteorol. Soc.*, *120*(516), 235–257.

Cahill, C. F. (2003), Asian aerosol transport to Alaska during ACE-Asia, *J. Geophys. Res.*, *108*(D23), 8664, doi:10.1029/2002JD003271.

Chun, Y., and J.-Y. Lim (2004), The recent characteristics of Asian dust and haze events in Seoul, *Korea, Meteorol. Atmos. Phys.*, 87, 143–152.

Darby, D. A., L. H. Burckle, and D. L. Clark (1974), Airborne dust on the Arctic ice pack, its composition and fallout rate, *Earth Planet. Sci. Lett.*, *24*, 166–172.

Dethof, A., and E. Holm (Eds.) (2002), Ozone in ERA40: 1991-1996, *Eur. Cent. for Medium-Range Weather Forecasts*, Reading, UK.

Darmenova, K., I. N. Sokolik, and A. Darmenov (2005), Characterization of east Asian dust outbreaks in the spring of 2001 using ground-based and satellite data, *J. Geophys. Res.*, *110*(D2), D02204, doi:10.1029/2004JD004842.

De Angelis, M., and A. Gaudichet (1991), Saharan dust deposition over Mont Blanc (French Alps) during the last 30 years, *Tellus*, *43B*, 61–75.

Dobson, G. M. B. (1956), Origin and distribution of the polyatmoic molecules in the atmosphere, *Proc. R. Soc. London*, *Ser. A*, *236*, 187–193.

Ehhalt, D. H. (1971), Vertical profiles and transport of HTO in the troposphere, *J. Geophys. Res.*, *76*, 7351–7367.

Ehhalt, D. H., F. Rohrer, S. Schauffler, and W. Pollock (2002), Tritiated water vapor in the stratosphere: Vertical profiles and residence time, *J. Geophys. Res.*, *107*(D24), 4757, doi:10.1029/2001JD001343.

EPICA community members (2004), Eight glacial cycles from an Antarctic ice core, *Nature*, *429*, 623–628.

Fan, K., and H. J. Wang (2007), Dust storms in North China in 2002: A case study of the low frequency oscillation, *Adv. Atmos. Sci.*, *24*(1), 15–23.

Fisher, D. A., and R. M. Koerner (1981), Some aspects of climatic change in the high Arctic during the Holocene as deduced from ice cores, in *Quaternary Paleoclimate*, edited by W. C. Mahaney, pp. 249–271, University of East Anglia Press, Norwich, U. K.

Fujii, Y., et al. (1990), 6000-Year climate records in an ice core from the Høghetta Ice Dome in northern Spitsbergen, *Ann. Glaciol.*, *14*, 85–89.

Fujii, Y., M. Kohno, S. Matoba, H. Motoyama, and O. Watanabe (2003), A 320 k-year record of microparticles in the Dome Fuji, Antarctica ice core measured by laser-light scattering, *Mem. Natl Inst. Polar Res., Spec. Issue*, *57*, 46–62.

Gat, J. R., W. G. Mook, and H. A. J. Meijer (2001), Tritium in the atmosphere, in *Atmospheric*

Water: Environmental Isotopes in the Hydrological Cycle Principles and Applications, vol. II, edited by W. G. Mook, pp. 63–74, UNESCO/IAEA Series, Paris. (Available at: http://www.iaea.org/programmes/ripc/ih/volumes/volumes.htm.)

Goering, M. A., W. A. Gallus, M. A. Olsen, and J. L. Stanford (2001), Role of stratospheric air in a severe weather event: Analysis of potential vorticity and total ozone, *J. Geophys. Res.*, *106*(D11), 11,813–11,823.

Hammer, C. U. (1977), Dust studies on Greenland ice cores, in *Isotopes and Impurities in Snow and Ice*, *Proceedings of the Grenoble Symposium*, *1975*, edited by H. Oeschger et al., *IAHS Publ.*, *118*, 365–370.

Hammer, C. U., H. B. Clausen, W. Dansgaard, A. Neftel, P. Kristinsdottir, and E. Johnson (1985), Continuous impurity analysis along the Dye 3 deep core, in *Greenland Ice Core: Geophysics, Geochemistry and the Environment*, *Geophys. Monogr.*, edited by C. C. Langway Jr., H. Oeschger and W. Dansgaard, pp. 90–94, AGU, Washington, D. C.

Hayasaki, M., S. Sugata, and H. L. Tanaka (2006), Interannual variation of cold frontal activity in spring in Mongolia, *J. Meteorol. Soc. Jpn.*, 84(3), 463–475.

Haynes, P., C. Marks, M. McIntyre, T. Shepherd, and K. Shine (1991), On the ''downward control'' of extratropical diabatic circulations by eddy-induced mean forces, *J. Atmos. Sci.*, 48, 651–678.

Hinkley, T. D. (1994), Composition and sources of atmospheric dust in snow at 3200 meters in the

St. Elias Range, southeastern Alaska, USA, *Geochim. Cosmochim. Acta*, *58*, 3245–3254.

Holdsworth, G., H. R. Krouse, and M. Nosal (1992), Ice core climate signals from Mount Logan, Yukon, A. D. 1700–1987, in Climate Since A. D. 1500, edited by R. S. Bradley and P. D. Jones, pp. 483–504, Routledge, Boca Raton, Fla.

Holton, J. R., P. H. Haynes, M. E. Mcintyre, A. R. Douglass, R. B. Rood, and L. Pfister (1995), Stratosphere-troposphere exchange, *Rev. Geophys.*, *33*(4), 403–439.

IAEA/WMO (2004), Isotope Hydrology Information System, *The ISOHIS Database*, International Atomic Energy Agency, Vienna, Austria. (Accessible at: http://isohis.iaea.org.)

IPCC, 2007: Summary for Policymakers. In: *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M.Tignor and H.L. Miller, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Iwasaka, Y., H. Minoura, and K. Nagaya (1983), The transport and spacial scale of Asian dust-storm clouds: a case study of the dust-storm event of April 1979, *Tellus*, *35B*, 189–196.

Japan Meteorological Agency (2006), Basic knowledge about Asian dust (Kosa), (Available at the website in Japanese: http://www.data.kishou.go.jp/obs-env/kosahp/4-4kosa.html).

Jickells, T. D., et al. (2005), Global iron connections between desert dust, ocean biogeochemistry, and climate, *Science*, *308*(5718), 67–71.

Kamiyama, K., W. Shimada, K. Kitaoka, K. Izumi, and S. Ezumi (1997), Determination of HTO content in polar ice/snow samples by low background liquid scintillation technique, *Nankyoku Shiryo (Antarctic Record)*, *41*, 631–642.

Kanamori, S. (2004), Seasonal variations in density profiles at cold mountain glaciers, M. S. thesis, 68 pp., Hokkaido University, Sapporo.

Kang, S. C., P. A. Mayewski, Y. P. Yan, D. H. Qin, T. D. Yao, and J. W. Ren (2003), Dust records from three ice cores: relationships to spring atmospheric circulation over the Northern Hemisphere, *Atmos. Environ.*, *37*(34), 4823–4835.

Kasischke, E. S., and L. P. Bruhwiler (2002), Emission of carbon dioxide, carbon monoxide, and methane from boreal forest fires in 1998, *J. Geophys. Res.*, *108*(D1), 8146, doi:10.1029/2001JD000461.

Kim, Y., H. Hatsushika, R. R. Muskett, and K. Yamazaki (2005), Possible effect of boreal wildfire soot on Arctic sea ice and Alaska glaciers, *Atmos. Environ.*, *39*, 3,513-3,520.

Kim, Y. K., H. W. Lee, J. K. Park, and Y. S. Moon (2002), The stratosphere-troposphere exchange of ozone and aerosols over Korea, *Atmos. Environ.*, *36*(3), 449–463.

Koerner, R. M., and D. A. Fisher (1982), Acid snow in the Canadian High Arctic, *Nature*, *295*, 137–140.

Kumai, M. (1977), Electron microscope analysis of aerosols in snow and deep ice cores from Greenland, in *Isotopes and Impurities in Snow and Ice*, *Proceedings of the Grenoble*

Symposium, *1975*, edited by H. Oeschger et al., *IAHS Publ.*, *118*, 341–350.

Kurosaki, Y., and M. Mikami (2003), Recent frequent dust events and their relation to surface wind in East Asia, *Geophys. Res. Lett.*, *30*(14), 1736, doi:10.1029/2003GL017261.

Kurosaki, Y., and M. Mikami (2005), Regional difference in the characteristic of dust event in East Asia: Relationship among dust outbreak, surface wind, and land surface condition, *J. Meteorol. Soc. Jpn.*, *83A*, 1–18.

Liu, M., D. L. Westphal, S. G. Wang, A. Shimizu, N. Sugimoto, J. Zhou, and Y. Chen (2003), A high-resolution numerical study of the Asian dust storms of April 2001, *J. Geophys. Res.*, *108*(D23), 8563, doi:10.1029/2002JD003178.

Lucas, L. L., and M. P. Unterweger (2000), Comprehensive review and critical evaluation of the half-life of tritium, *J. Res. Natl. Stand. Technol.*, *105*(4), 541–549.

Malm, W. C., B. A. Schichtel, M. L. Pitchford, L. L. Ashbaugh, and R. A. Eldred (2004), Spatial and monthly trends in speciated fine particle concentration in the United States, *J. Geophys. Res.*, *109*, D03306, doi:10.1029/2003JD003739.

Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill (1994), Spatial and seasonal trends in particle concentration and optical extinction in the United States, *J. Geophys. Res.*, *99*(D1), 1347–1370.

Mantua, N. J., S. R. Hare, Y. Zhang, J. M. Wallace, and R. C. Francis (1997), A Pacific interdecadal climate oscillation with impacts on salmon production, *Bull. Amer. Meteorol. Soc.*, *78*(6), 1069–1079.

Matoba, S., et al. (2007), The glaciological expedition to Mount Ichinsky, Kamchatka, Russia, *Bull. Glaciol. Res.*, *24*, 79–85.

McGimsey, R. G., C. A. Neal, and C. M. Riley (2002), Areal distribution, thickness, mass, volume, and grain size of tephra-fall deposits from the 1992 eruptions of Crater Peak Vent, Mt. Spurr Volcano, Alaska, *Open File Rep. 01–370*, 38 pp., U.S. Geol. Surv, Denver, CO, USA. (Available at : http://geopubs.wr.usgs.gov/open-file/of01-370/.)

McKendry, I. G., J. P. Hacker, R. Stull, S. Sakiyama, D. Mignacca, and K. Reid (2001), Long-range transport of Asian dust to the Lower Fraser Valley, British Columbia, Canada, *J. Geophys. Res.*, *106*(D16), 18,361–18,370.

Miyazaki, K., and T. Iwasaki (2005a), Diagnosis of meridional ozone transport based on mass weighted isentropic zonal means, *J. Atmos. Sci.*, 62, 1192–1208.

Miyazaki, K., T. Iwasaki, K. Shibata, and M. Deushi (2005b), Roles of transport in the seasonal variation of the total ozone amount, *J. Geophys. Res.*, *110,* D18309, doi:10.1029/2005JD005900.

Monks, P. S. (2000), A review of the observations and origins of the spring ozone maximum, *Atmos. Environ.*, *34*(21), 3545–3561.

Moore, G. W. K., G. Holdsworth, and K. Alverson (2002), Climate change in the North Pacific region over the past three centuries, *Nature*, *420*(6914), 401–403.

Mukougawa, H., K. Yamashita, and F. Hasebe (2004), Intraseasonal low-frequency variability of tropopause height associated with storm tracks, *Annuals of Disas. Prev. Res. Inst.*, *47B*, 349-358 (In Japanese with English abstract).

Mullen, R. E., D. A. Darby, and D. L. Clark (1972), Significance of atmospheric dust and ice rafting from Arctic Ocean sediment, *Geol. Soc. Amer. Bull.*, *83*, 205–212.

Murozumi, M., T. J. Chow, and C. Patterson (1969), Chemical concentrations of pollutant lead aerosols, terrestrial dusts and sea salts in Greenland and Antarctic snow strata, *Geochim. Cosmochim. Acta*, *33*, 1247–1294.

Nakamura, H., and J. M. Wallace (1990), Observed changes in baroclinic wave activity during the life cycles of low-frequency circulation anomalies, *J. Atmos. Sci.*, 47(9), 1100–1116.

Nakamura, H. (1992), Midwinter suppression of baroclinic wave activity in the Pacific, *J. Atmos. Sci.*, 49(17), 1629–1642.

North Greenland Ice Core Project members (2004), High-resolution record of Northern Hemisphere climate extending into the last interglacial period, *Nature*, *431*, 147–151.

Petit, J. R., L. Mounier, J. Jouzel, Y. S. Korotkevitch, V. I. Kotlyakov and C. Lorius (1990), Palaeoclimatological and Chronological Implications of the Vostok Core Dust Record, *Nature*, *343*, 56-58.

Polissar, A. V., P. K. Hopke, W. C. Malm, and J. F. Sisler (1998), Atmospheric aerosol over Alaska, 1. Spatial and seasonal variability, *J. Geophys. Res.*, *103*(D15), 19,035–19,044.

Rahn, K. A., R. D. Borys, and G. E. Shaw (1977), Asian source of Arctic haze bands, *Nature*, *268*, 713–715.

Rahn, K. A., R. D. Borys, G. E. Shaw. L. Schu¨tz, and R. Jaenicke (1979), Long-range impact of desert aerosol on atmospheric chemistry: Two examples, in *Saharan Dust: Mobilization, Transport, and Deposition*, edited by C. Morales, pp. 243–266, John Wiley, Hoboken, N. J.

Sassen, K., P. J. DeMott, J. M. Prospero, and M. R. Poellot (2003), Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results, *Geophys. Res. Lett.*, *30*(12), 1633, doi:1610.1029/2003GL017371.

Sassen, K. (2005), Dusty ice clouds over Alaska, *Nature*, 434(7032), 456–456.

Schwikowski, M., S. Brutsch, H. W. Gaggeler, and U. Schotterer (1999), A high-resolution air chemistry record from an Alpine ice core: Fiescherhorn glacier, Swiss Alps, *J. Geophys. Res.*, *104*(D11), 13,709-13,719.

Shao, Y. P., and J. J. Wang (2003), A climatology of Northeast Asian dust events, *Meteorol. Z.*, *12*(4), 187–196.

Shao, Y. P., *et al.* (2003), Northeast Asian dust storms: Real-time numerical prediction and validation, *J. Geophys. Res.*, *108*(D22), 4691, doi:10.1029/2003JD003667.

Shiraiwa, T., Y. D. Murav'yev, T. Kameda, F. Nishio, Y. Toyama, A. Takahashi, A. A. Ovsyannikov, A. N. Salamatin, and K. Yamagata (2001), Characteristics of a crater glacier at Ushkovsky volcano, Kamchatka, Russia, as revealed by the physical properties of ice cores and borehole thermometry, *J. Glaciol.*, *47*(158), 423–432.

Shiraiwa, T., K. Goto-Azuma, S. Matoba, T. Yamasaki, T. Segawa, S. Kanamori, K. Matsuoka, Y. Fujii (2003), Ice core drilling at King Col, Mount Logan 2002, *Bull. Glaciol. Res.*, *20*, 57–63.

Shiraiwa, T., S. Kanamori, C. S. Benson, D. Solie, and Y. D. Muravyev (2004), Shallow ice-core drilling at Mount Wrangell, Alaska, *Bull. Glaciol. Res.*, *21*, 71–77.

Smith, A. M., D. Fink, D. Child, V. A. Levchenko, V. I. Morgan, M. Curran, D. M. Etheridge, and G. Elliott (2000), Be-7 and Be-10 concentrations in recent firn and ice at Law Dome, Antarctica, *Nucl. Instrum. Meth. B*, *172*, 847–855.

Sprenger, M., and H. Wernli (2003), A northern hemispheric climatology of cross-tropopause exchange for the ERA15 time period (1979-1993), *J. Geophys. Res.*, *108*(D12), 8521, doi:8510.1029/2002JD002636.

Steffensen, J. P. (1988), Analysis of the seasonal variation in dust, Cl^-, NO_{3-}, and SO_{42-} in two central Greenland firn cores, *Ann. Glaciol.*, *10*, 171–177.

Steffensen, J. P. (1997), The size distribution of microparticles from selected segments of the Greenland Ice Core Project ice core representing different climatic periods, *J. Geophys. Res.*, *102*(C2), 26,755–26,763.

Stohl, A., et al. (2003a), Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO, *J. Geophys. Res.*, *108*(D12), 8516, doi:10.1029/2002JD002490.

Stohl, A., H. Wernli, P. James, M. Bourqui, C. Forster, M. A. Liniger, P. Seibert, and M. Sprenger

(2003b), A new perspective of stratosphere-troposphere exchange, *Bull. Amer. Meteorol. Soc.*, *84*(11), 1565–1573.

Sugimoto, N., I. Uno, M. Nishikawa, A. Shimizu, I. Matsui, X. H. Dong, Y. Chen, and H. Quan (2003), Record heavy Asian dust in Beijing in 2002: Observations and model analysis of recent events, *Geophys. Res. Lett.*, *30*(16), 1640, doi:10.1029/2002GL016349.

Sun, J. M., M. Y. Zhang, and T. S. Liu (2001), Spatial and temporal characteristics of dust storms in China and its surrounding regions, 1960-1999: Relations to source area and climate, *J. Geophys. Res.*, *106*(D10), 10,325–10,333.

Sun, J. H., et al. (2006), An integrated dust storm prediction system suitable for East Asia and its simulation results, *Glob. Planet. Change*, *52*(1-4), 71–87.

Takemura, T., I. Uno, T. Nakajima, A. Higurashi, and I. Sano (2002), Modeling study of long-range transport of Asian dust and anthropogenic aerosols from East Asia, *Geophys. Res. Lett.*, *29*(24), 2158, doi:2110.1029/2002GL016251.

Tanaka, T. Y., and M. Chiba (2005), Global simulation of dust aerosol with a chemical transport model, MASINGAR, *J. Meteorol. Soc. Jpn.*, *83A*, 255–278.

Thompson, D. W., and J. M. Wallace (1998), The Arctic Oscillation signature in the wintertime geopotential height and temperature fields, *Geophys. Res. Lett.*, *25*(9), 1297–1300.

Thompson, L. G., M. E. Davis, E. M. Thompson, T. A. Sowers, K. A. Henderson, V. S. Zagorodnov, P. N. Lin, V. N. Mikhalenko, R. K. Campen, J. F. Bolzan, J. Cole-Dai and B.

Francou (1998), A 25,000 year tropical climate history from Bolivian ice cores, *Science*, *282*, 1858-1864.

Thompson, L. G., T. Yao, E. Mosley-Thompson, M. E. Davis, K. A. Henderson, and P. N. Lin (2000), A high-resolution millennial record of the South Asian monsoon from Himalayan ice cores, *Science*, *289*, 1916–1919.

Thompson, L. G., E. Mosley-Thompson, M. E. Davis, K. A. Henderson, H. H. Brecher, V. S. Zagorodnov, T. A. Mashiotta, P. N. Lin, V. N. Mikhalenko, D. R. Hardy, and J. Beer (2002), Kilimanjaro ice core records: Evidence of Holocene climate change in tropical Africa. *Science*, *298*, 589-593.

Uno, I. et al. (2003), Regional chemical weather forecasting system CFORS: Model descriptions and analysis of surface observations at Japanese island stations during the ACE-Asia experiment, *J. Geophys. Res.*, *108*(D23), 8668, doi:10.1029/2002JD002845.

Uppala, S. M., et al. (2005), The ERA-40 re-analysis, *Q. J. R. Meteorol. Soc.*, *131*, 2961–3012.

VanCuren, R. A., and T. A. Cahill (2002), Asian aerosols in North America: Frequency and concentration of fine dust, *J. Geophys. Res.*, *107*(D24), 4804, doi:4810.1029/2002JD002204.

Wagenbach, D., and K. Geiss (1989), The mineral record in a high altitude alpine glacier (Colle Gnifetti, Swiss Alps), in *Paleoclimatology and Paleometeorology: Modern and Past Patterns of Global Atmospheric Transport*, edited by M. Leinin and M. Sarnthein, pp. 543–564, Springer, New York.

Wake, C. P., P. A. Mayewski, Z. Li, J. Han, and D. Qin (1994), Modern eolian dust deposition in Central Asia, *Tellus*, *46B*, 220–233.

Wake, C. P., K. Yalcin, and N. S. Gundestrup (2002), The climate signal recorded in the oxygen-isotope, accumulation and major-ion time series from the Eclipse ice core, Yukon Territory, Canada, Ann. Glaciol., 35, 416–422.

Wang, K. Y. (2007), Long-range transport of the April 2001 dust clouds over the subtropical East Asia and the North Pacific and its impacts on ground-level air pollution: A Lagrangian simulation, *J. Geophys. Res.*, *112*(D9), doi:10.1029/2006JD007789.

Watanabe, O., J. Jouzel, S. Johnsen, F. Parrenin, H. Shoji and N. Yoshida. (2003), Homogeneous climate variability across East Antarctica over the past three glacial cycles, *Nature*, *422*, 509-512.

Windom, H. L. (1969), Atmospheric dust records in permanent snowfields: implications to marine sedimentation, *Geol. Soc. Amer. Bull.*, *80*, 761–782.

Wurzler, S., T. G. Reisin, and Z. Levin (2000), Modification of mineral dust particles by cloud processing and subsequent effects on drop size distributions, *J. Geophys. Res.*, *105*(D4), 4501–4512.

Yamamoto, T., M. Yoshino, and J. Suzuki (2007), The relationship between occurrence of dust events and synoptic climatological condition, *J. Meteorol. Soc. Jpn.*, *85*, 81–99.

Yamashita, K. (2003), Atmospheric 3D-structures and -variations of the tropopausal region in the extratropics, *M.S. Thesis*, 67 pp., Hokkaido Univ., Sapporo (In Japanese).

Yamazaki, K. (1986), Preliminary calculation of trajectory analysis in the lower stratosphere of the southern-hemisphere, *Geophys. Res. Lett.*, *13*(12), 1312–1315.

Yasunari, T. J., T. Shiraiwa, S. Kanamori, Y. Fujii, M. Igarashi, K. Yamazaki, C. S. Benson, and T. Hondoh (2007), Intra-annual variations in atmospheric dust and tritium in the North Pacific region detected from an ice core from Mount Wrangell, Alaska, *J. Geophys. Res.*, *112*, D10208, doi:10.1029/2006JD008121.

Yu, H., R. E. Dickinson, M. Chin, Y. J. Kaufman, B. N. Holben, I. V. Geogdzhayev, and M. I. Mishchenko (2003), Annual cycle of global distributions of aerosol optical depth from integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, *108*(D3), 4128, doi:10.1029/2002JD002717.

Zanis, P., et al. (2003), An estimate of the impact of stratosphere-to-troposphere transport (STT) on the lower free tropospheric ozone over the Alps using Be-10 and Be-7 measurements, J. Geophys. Res., *108*(D12), 8520, doi:10.1029/2002JD002604.

Zdanowicz, C. M., G. A. Zielinski, and C. P. Wake (1998), Characteristics of modern atmospheric dust deposition in snow on the Penny Ice Cap, Baffin Island, Arctic Canada, *Tellus*, *50B*, 506–520.

Zdanowicz, C., G. Hall, J. Vaive, Y. Amelin, J. Percival, I. Girard, P. Biscaye, and A. Bory (2006), Asian dustfall in the St. Elias Mountains, Yukon, Canada, *Geochim. Cosmochim. Acta*, *70*(14), 3493–3507.

Zhang, X. Y., S. L. Gong, R. Arimoto, Z. X. Shen, F. M. Mei, D. Wang, and Y. Cheng (2003), Characterization and Temporal Variation of Asian Dust Aerosol from a Site in the Northern Chinese Deserts, *J. Atmos. Chem.*, *44*, 241–257.

Zhao, L. N., and S. X. Zhao (2006), Diagnosis and simulation of a rapidly developing cyclone related to a severe dust storm in East Asia, *Glob. Planet. Change*, *52*(1-4), 105–120.

Zhou, M. Y., Z. Chen, R. H. Huang, Q. M. Wang, R. Arimoto, F. Parungo, D. Lenschow, K. Okada, and P. M. Wu (1994), Effects of two dust storms on solar-radiation in the Beijing-Tianjin area, *Geophys. Res. Lett.*, *21*(24), 2697–2700.

Zhou, Z. J., and G. C. Zhang, 2003, Typical severe dust storms in northern China during 1954 - 2002, *Chin. Sci. Bull.*, 48(21): 2366–2370.