Seasonal variation of free tropospheric aerosol particles at Mt. Tateyama, central Japan

K. Osada,[1] M. Kido,[2,3] H. Iida,[4] K. Matsunaga,[1] Y. Iwasaka,[1]
M. Nagatani,[5] and H. Nakada[5]

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[1] Number-size distribution of atmospheric aerosol particles and O_3 concentration were measured at Murododaira (36.6N, 137.6E, 2450 m above sea level (asl)) on the western flank of Mt. Tateyama in central Japan from January 1999 to November 2002. This study used nighttime data from 2400 to 0500 hours (local time) on the basis of analysis of their diurnal variation to characterize free tropospheric aerosols and O_3 over Japan. The O_3 concentration shows small variability (standard deviation of 4 ppbv) with the mean value of 40 ppbv in winter (October to February), large variability (8 ppbv) with the higher mean value of 51 ppbv in spring (March to May), and large variability (14 ppbv) with the lower mean value of 32 ppbv in summer (June to September). Highest monthly mean volume concentration (2.7 $\mu m^3/cm^3$) of accumulation particles (0.3 $\mu m < D <$ 1.0 μm) was observed in June, while the mean value in winter (October to February) was 0.7 $\mu m^3/cm^3$. On the basis statistics of backward air trajectory analyses, a stagnant airflow in summer over the coastal areas of the Yellow Sea and near Japan is inferred to be a suitable meteorological condition to form enhanced volume concentration of accumulation particles during transport. Associating with the seasonal changes in the dominant air trajectories, SO_2 emission from Miyakejima volcano since August 2000 is also an important source of the summer enhancement of accumulation particles. Highest monthly mean volume concentration (11.2 $\mu m^3/cm^3$) of coarse particles ($D > 1.0$ μm) was found in April, which was about 10 times higher than the mean value of 1.2 $\mu m^3/cm^3$ from summer to winter. Variability of daily nighttime volume concentrations of the coarse particles was high (standard deviation of 13.6 $\mu m^3/cm^3$) in spring and low (about 2 $\mu m^3/cm^3$) in the rest of the year. High volume concentration with large variability of the coarse particles in spring is caused by frequent arrival of Kosa (yellow dust) particles from the Asian continent. Rapid enhancement of coarse volume concentration was often observed to increase as much as 30 times within 3 hours during Kosa phenomena. The year 2001 had particularly strong Kosa activity with a prolonged season starting early January and ending early July. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* Asian dust, free troposphere, seasonal variation

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1. Introduction

[2] Atmospheric aerosol particles in the free troposphere play an important role in direct and indirect radiation effects in Earth's atmosphere [*Yamamoto and Tanaka*, 1972; *Charlson and Heintzenberg*, 1995; *Andreae*, 1995]. Their number-size distribution and temporal variation of concentration are fundamental parameters to evaluate and predict effects of aerosols on climatic changes. These aerosol data should be seasonally characterized to provide parameters for climate models because of their large variability with seasons. Seasonal variations in free tropospheric aerosols

[1]Graduate School of Environmental Sciences, Nagoya University, Nagoya, Japan.
[2]Niikawa Water Museum, Uozu, Japan.
[3]Now at Toyama Prefectural Environmental Science Research Center, Toyama, Japan.
[4]Tateyama Caldera SABO Museum, Ashikuraji, Japan.
[5]Solar-Terrestrial Environment Laboratory, Nagoya University, Toyokawa, Japan.

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have been observed by lidar [*Sakai et al.*, 2000] and by a scanning spectral radiometer [*Shiobara et al.*, 1991] over Japan. Scanning spectral radiometer measurement indicated that aerosols in spring and summer seasons had different features: coarse particle mode aerosols were predominant in spring, while accumulation mode aerosols were predominant in summer [*Shiobara et al.*, 1991]. However, their method of atmospheric measurements provides only column-integrated properties of aerosols. In contrast, lidar observation [*Sakai et al.*, 2000] offers the advantage of obtaining vertical distributions of values related to volume concentration and nonsphericity of aerosols. Their report addressed the relation of seasonal and altitudinal characteristics of aerosol optical properties over Japan to the ambient relative humidity and transport pathways from source areas. However, those data are limited to clear sky conditions.

[3] In situ measurements at a high elevation site may provide valuable data to obtain year-round, all weather basic information regarding free tropospheric aerosols [e.g., *Nyeki et al.*, 1998a, 1998b; *Huebert et al.*, 2001]. Our previous studies at Mts. Tateyama and Norikura, both in central Japan, provided primary results of aerosol chemistry and water soluble gases from fall to early spring [*Kido et al.*, 2001a, 2001b; *Osada et al.*, 2002]. This paper reports seasonal variations of aerosols and O_3 concentrations during almost four consecutive years at Murododaira (36.57°N, 137.60°E, 2450 m asl at Mt. Tateyama. First, diurnal variations of aerosols and O_3 concentrations are evaluated for screening the free tropospheric data. Second, daily and the monthly values of multiyear composite are presented to characterize temporal variations of aerosols and O_3. Then seasonal features of aerosols and O_3 are discussed with statistics of backward air trajectories. Short-term events are also discussed as examples of rapid aerosol variations.

2. Instrumentation and Data Screening

[4] Number-size distributions of atmospheric aerosol particles were measured with a laser particle counter (KC-01C; Rion Co., Ltd.) from late January 1999 at the Hotel Tateyama in Murododaira. The laser particle counter (LPC) measures the number of aerosol particles for five size ranges: larger than 0.3, 0.5, 1.0, 2.0, and 5.0 μm in diameter. It is calibrated by the manufacturer using standard polystyrene latex particles. Sample air humidity was always below 40% because the room temperature was higher than outside temperature. In this paper, aerosol concentrations are reported as the values of standard temperature (25°C) and pressure (1 atm). Although the size range covered by the LPC is rather limited, the terms "accumulation" and "coarse" particles in this paper refer to size ranges from 0.3 to 1.0 μm, and larger than 1.0 μm, respectively. In addition to the LPC, an UV absorption O_3 monitor (1006-AHJ and later DY115, Dylec) was placed in a room. A 3-m long electroconductive tube (8 mm i.d. × 12 mm o.d.) was connected to the system to introduce outside air. An auxiliary air pump was used at a flow rate of about 6 l/min to introduce air from the inlet to the LPC. Sampling losses caused by the long pipe from the inlet might be large particularly for supermicron particles. For our air sampling system, the flow Reynolds number is about 1000; hence the air flow through the inlet tube was laminar. According to

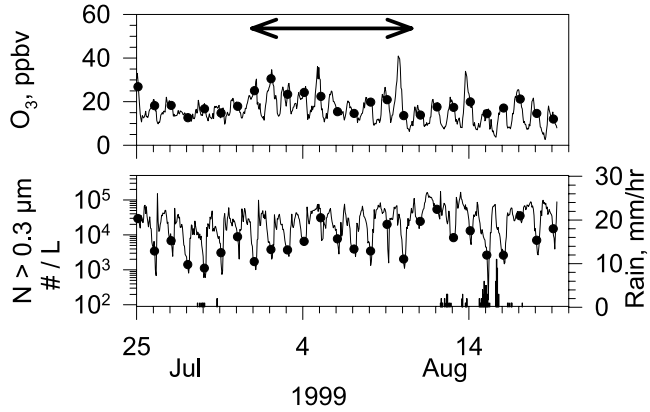

Figure 1. Concentrations of O_3, aerosol number (D > 0.3 μm) and precipitation amount at Tateyama in summer 1999. Filled circles represent mean values between 2400 and 0500 hours. The horizontal arrow in the upper panel indicates the period of detailed analysis (Figure 2) for diurnal variations without rain.

Cheng and Wang [1981] and *Pui et al.* [1987], the particle loss caused by the bend (in our case, the curvature ratio was 20) was estimated to be about 4% for particles of 5 μm in diameter. Furthermore, gravitational settling within the inlet tube was estimated as 6% for 2 μm and 38% for 5 μm particles using particle density of 2 g/cm^3 and assuming perfect stickiness of particles to the tube wall [*Hinds*, 1999; *Nichols*, 1998]. Thus the particle loss may be significant for coarse particles larger than 5 μm, but number-size data were not corrected in this study. During the winter monsoon period (November to April), strong northwesterly winds prevailed with frequent snowfalls with rime ice. A snow-clogging preventer similar to the "Frisbee sampler" in the work of *Heidam et al.* [1993] was installed at the tip of the inlet tube.

[5] Upslope valley winds and downslope mountain winds occur on the slope of Mt. Tateyama, as reported for other high elevation sites [*Mendonca*, 1969; *Parrington and Zoller*, 1984; *Nyeki et al.*, 1998a]. Upslope valley winds are caused by surface heating of the mountain slope by solar radiation during the day. Downslope mountain winds are caused by radiative cooling of the mountain surface during the night [*Whiteman*, 2000]. Dense cooler air flows down the mountain slope, flushing the mountain surface with clean air from the free troposphere. To select free tropospheric data at Mt. Tateyama, hourly data were analyzed for the result of August, the most suitable month to test upslope winds because of persistent fine weather. Figure 1 shows variations of O_3 concentrations, the number concentrations of aerosols larger than 0.3 μm and the precipitation amount near the site. Aerosol number concentrations decreased during precipitation in some cases. The O_3 concentrations were high (~20 to 40 ppbv) at night and low (~5 to 20 ppbv) during the day, whereas aerosol concentrations were low (10^3 to 10^4 L^{-1}) at night and high (>10^4 L^{-1}) in the daytime. Regarding O_3 variation, evening enhancements (from 6 to 10 p.m.) were also seen sporadically on 4, 9, and 13 August. As reported for Mt. Fuji (3776 m asl, 170 km southeast of Mt. Tateyama), enhanced O_3 in the

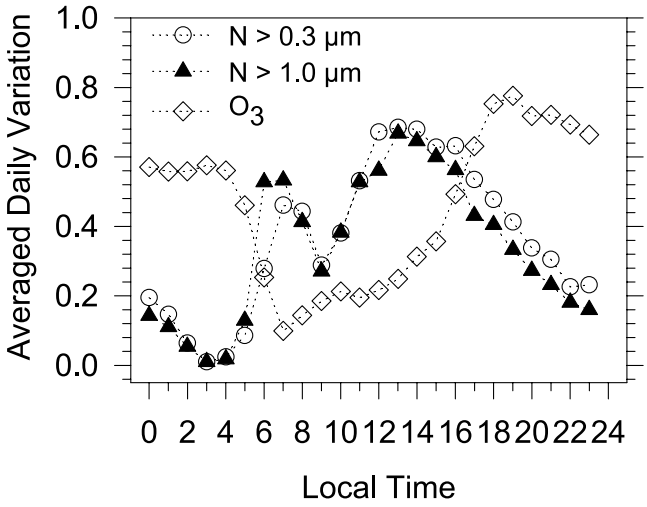

Figure 2. Averaged diurnal variations during 12 days without precipitation in Figure 1. Hourly values were normalized to the amplitude between respective daily maxima and minima.

convective boundary layer developed over industrial areas during the daytime were transported to the observation site at about dusk [*Tsutsumi and Matsueda*, 2000].

[6] Figure 2 summarizes the averaged daily variations for days without precipitation from 30 July to 10 August 1999. The averaged period shown by the horizontal arrow in Figure 1 was selected to avoid changes in aerosol concentrations caused by precipitation scavenging. Hourly data were normalized by daily amplitude: 0 and 1 correspond to the daily minima and maxima, respectively. Then normalized hourly data were averaged for 12 days. Diurnal variations of aerosol number concentrations and O_3 were both evident in Figure 2. Increased concentration of aerosols during daytime is associated with vertical upward transportation of pollutants from the lowland area near the mountain. A second peak in aerosol concentrations during morning hours is probably caused by activity associated with visitors and mountain hikers. Lower concentrations at nighttime from 2400 to 0500 hours are attributed to the subsidence of clean air from the free troposphere aloft. Similarly, high and stable O_3 concentrations from 2400 to 0500 hours result from subsidence of the O_3-rich free troposphere. Thus to collect the free tropospheric data for O_3 and aerosols, averaged data from 2400 to 0500 hours were used in this study as indicated by the filled circles in Figure 1.

3. Results and Discussion

3.1. Seasonal Variation

[7] Figure 3 shows temporal variations of the nighttime data from January 1999 to November 2002. Instrument malfunction and logistical problems caused several interruptions of continuous measurements. Variation of O_3 concentrations shows remarkable seasonal features. In spring, O_3 concentration gradually increased from 40 ppbv in February to 53 ppbv in April and May. During June to September, O_3 concentrations frequently oscillated between higher (40 ppbv) and lower (<0 ppbv) values. A shift of O_3 concentration from a lower (<0 ppbv) value in summer to a higher (40 ppbv) value was observed in middle to late September. Then O_3 concentrations of 40 ppbv with less variation remained until the following spring. During the study period, maximum nighttime average O_3 concentration of 79 ppbv was observed on April 20, 2001. Sporadic enhanced O_3 events (>100 ppbv) influenced by the upper tropospheric air were frequently observed at Mt. Fuji [*Tsutsumi et al.*, 1998], but such events were rarely observed at Mt. Tateyama.

[8] Number concentrations of aerosols (>0.3 μm and >1.0 μm) show large variability due partly to the precipitation scavenging. Maximum number concentrations were observed on 26 May 2001 for N > 0.3 μm (3.4×10^5 L^{-1}) and 17 March 1999 for N > 1.0 μm (9.1×10^3 L^{-1}). Volume concentration was calculated as particles to be spherical. For volume concentration of accumulation ($0.3 < D < 1.0$ μm) and coarse ($1.0 < D$ μm) particles, maximum values were observed on 26 May (15 $\mu m^3/cm^3$) and 11 April (86 $\mu m^3/cm^3$) in 2001, respectively. High (>20 $\mu m^3/cm^3$) volume concentration of the coarse particles was frequently found in spring. Variability of adjacent data in the coarse particles was also remarkable in spring.

[9] Figure 4 summarizes monthly box plots for seasonal variations. Table 1 lists statistical summary of seasonal variations of O_3 and aerosol concentrations. Variation of O_3 concentrations showed three regimes: small variability (standard deviation of 4 ppbv) with the mean value of 40 ppbv in winter (October to February), large variability (8 ppbv) with the higher mean value of 51 ppbv in spring

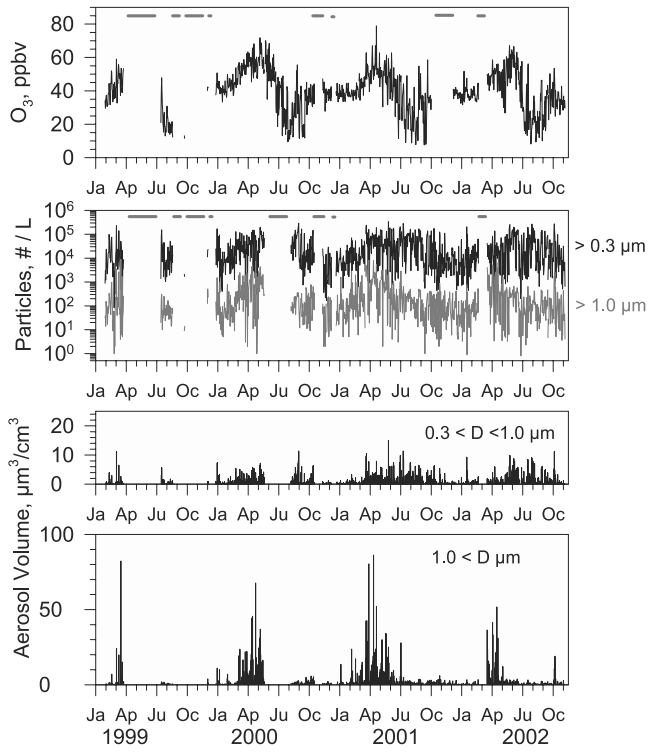

Figure 3. Nighttime (2400 to 0500 hours) daily mean values of O_3 and number and volume concentrations of aerosol particles at Mt. Tateyama. Horizontal bars in O_3 and number concentrations represent periods of missing data.

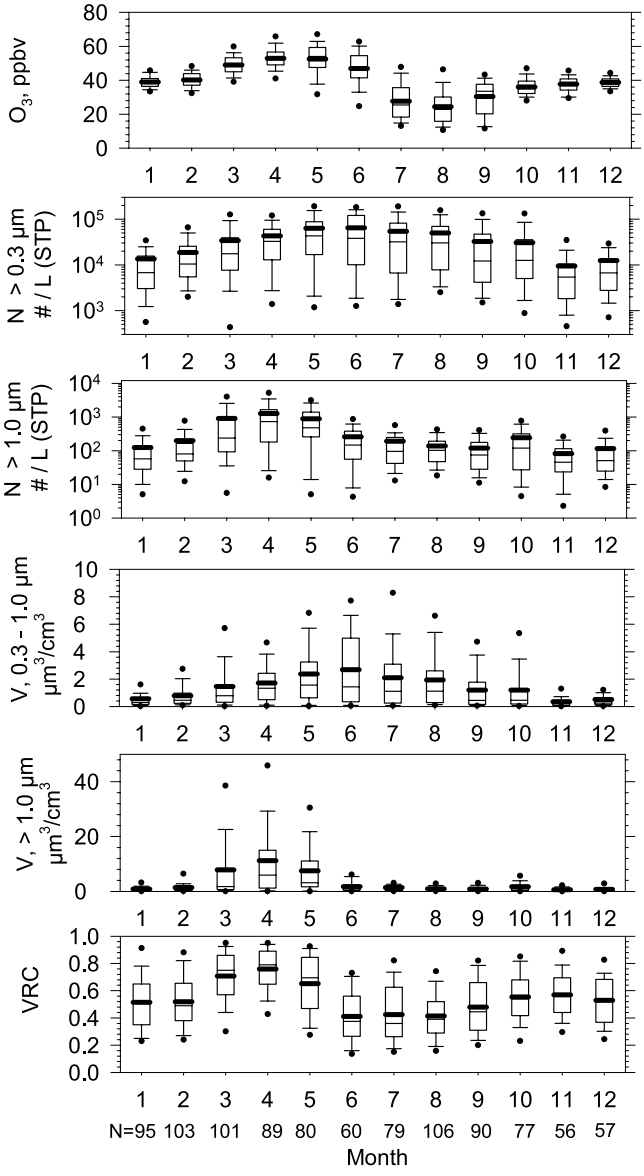

Figure 4. Monthly box plots for O_3 and aerosol concentrations at Mt. Tateyama. The lower boundary of the box indicates the 25th percentile, the line within the box marks the median, and the upper boundary of the box indicates the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentiles. The mean is also drawn as a thick line; 5th and 95th percentiles are plotted as filled circles. VRC in the lowest panel represents the volume ratio of the coarse fraction ($V > 1.0$ μm) to the total volume ($V > 0.3$ μm). Numbers (N=) under the month indicate data points used in monthly data for aerosols.

(March to May), and large variability (14 ppbv) with the lower mean value of 32 ppbv in summer (June to September). A similar seasonal variation of the free tropospheric O_3 concentration over Japan was observed by ozonesonde soundings [*Ogawa and Miyata*, 1985] and at Mt. Fuji [*Tsutsumi et al.*, 1994]. Appearance of the spring maximum in O_3 concentration remains an intriguing problem [e.g., *Harris et al.*, 1998; *Monks*, 2000]. Correlation between O_3 and CO concentrations at Mt. Happo (1850 m asl, 25 km north of Mt. Tateyama) has suggested contribution of photochemical O_3 production in the spring troposphere [*Kajii et al.*, 1998]. Recent model study [*Mauzerall et al.*, 2000] has also indicated photochemically produced O_3 export from east Asia in spring. On the other hand, it has been considered that the summer minimum of O_3 concentration is caused by typical air flow pattern from southern marine latitudes bringing clean low-O_3 air [*Ogawa and Miyata*, 1985]. Details of seasonal O_3 variation are discussed later with the backward air trajectory analysis.

[10] Number and the volume concentrations of the accumulation mode particles ($0.3\ \mu m < D < 1.0\ \mu m$) were enhanced in warmer months from March to October. The maximum monthly volume concentration of accumulation particles was observed in June (2.7 $\mu m^3/cm^3$). The monthly maximum value was approximately four times higher than the winter mean of 0.7 $\mu m^3/cm^3$. The number and the volume concentration of the coarse particles (>1.0 μm) show distinct high concentrations in spring. The maximum monthly volume concentration of the coarse particles was observed in April (11.2 $\mu m^3/cm^3$); it was about 10 times higher than the mean value of 1.2 $\mu m^3/cm^3$ from summer to winter. In October, a small peak of coarse volume concentration was seen with the monthly mean of 1.7 $\mu m^3/cm^3$. Variability of daily nighttime values was high (standard deviation of 13.6 $\mu m^3/cm^3$) in spring and low (about 2 $\mu m^3/cm^3$) during the rest of the year. The volume ratio (VRC) of the coarse volume ($V > 1.0$ μm) to the total volume concentration ($V > 0.3$ μm) indicates that the coarse fraction dominates in spring, and that the accumulation particle is the main fraction in summer.

[11] In east Asia, spring is the period of high aerosol loading caused by frequent Asian dust outbreaks [e.g., *Koizumi*, 1932; *Iwasaka et al.*, 1988; *Arao et al.*, 2003]. Such Asian dust events are called "Kosa" in Japan. They are characterized by significant haze that consists of yellow or brown windborne dust particles. The Kosa particles are predominantly coarse particle size [*Ishizaka and Ono*, 1982]. Aerosol weight concentrations during Kosa events were several times higher than monthly average values and showed significant sporadic variations [*Hao et al.*, 1995]. Thus high volume concentrations with large variability of the coarse aerosol in spring suggest frequent arrival of Kosa dust at the site.

[12] Figure 5 shows composite plots of 5-day backward trajectories from Mt. Tateyama for 2 years: Figure 5a for February 2001 and 2002, Figure 5b for April 2001 and 2002, Figure 5c for June 2001 and 2002, Figure 5d for August 2001 and 2002, and Figure 5e for November 2000 and 2001. The start height of the trajectories was set at 3000 m above sea level. Trajectories were calculated from the HYSPLIT 4 (Hybrid Single-Particle Lagrangian Integrated Trajectory) model, 1997 (Web address: http://www.arl.noaa.gov/ready/hysplit4.html, NOAA Air Resources Laboratory, Silver Spring, MD). Figures 5a to 5e indicate that major source areas and the transport distance during 5 days vary with the season. We divided trajectories according to their three regions of origin: the coastal area, the Pacific ocean, and west of 100°E, as shown in Figure 5f because the major source areas of anthropogenic sulfate and its precursor (SO_2) are located near the coastal area of the

Table 1. Statistical Summary of Ozone and Aerosol Data at Mt. Tateyama, 1999–2002

	December–February	March–May	June–August	September–November	Annual
		Ozone, ppbv			
Mean	40	51	32	33	40
Median	39	51	29	35	40
Standard deviation	4	8	5	9	13
Count	252	283	312	154	1001
		$N > 0.3$ μm, *Number per L*			
Mean	1.5 E4	4.6 E4	5.5 E4	2.7 E4	3.6 E4
Median	7.6 E3	3.0 E4	3.3 E4	9.4 E3	1.6 E4
Standard deviation	2.4 E4	5.2 E4	6.0 E4	4.0 E4	4.9 E4
Count	255	270	245	223	993
		$N > 1.0$ μm, *Number per L*			
Mean	1.5 E2	1.0 E3	1.9 E2	1.6 E2	4.0 E2
Median	69	4.3 E2	1.1 E2	84	1.1 E2
Standard deviation	2.9 E2	1.5 E3	2.5 E2	2.5 E2	8.8 E2
		V 0.3–1.0 μm, $\mu m^3/cm^3$			
Mean	0.7	1.8	2.2	1.0	1.4
Median	0.3	1.2	1.2	0.4	0.6
Standard deviation	1.0	2.1	2.4	1.5	2.0
		$V > 1.0$ μm, $\mu m^3/cm^3$			
Mean	1.1	8.9	1.3	1.1	3.2
Median	0.4	3.1	0.7	0.5	0.7
Standard deviation	2.5	13.6	2.2	1.8	8.1
		VRC			
Mean	0.52	0.71	0.42	0.53	0.55
Median	0.50	0.75	0.38	0.53	0.54
Standard deviation	0.19	0.20	0.19	0.19	0.22

Yellow Sea and the rim of west Pacific including Japan. In Figure 5f, trajectory endpoints north of 60°N are also included in the category of the west for simplicity because of long duration of transport. The statistical summary of the trajectory analysis is listed in Table 2.

[13] In February, most (86%) trajectories were transported from the west of 100°E. Trajectories ending near the coastal area doubled in April; then the coastal area became the dominant (78%) source region in June. Air masses arriving in August originated from the Pacific (56%), the coastal (37%), and the west (7%). In November, the dominant (75%) origin of air masses was reverted to the west. Using these data for trajectories, seasonal variations of O_3 and aerosols are discussed with synoptic meteorological features.

[14] The summer monsoon between June and July is characterized by frequent and heavy rain in Japan; it is the so-called Bai-u season, resulting from frontal activity between cold midlatitude air and warm Pacific subtropical moist air [*Barry and Chorley*, 1987]. According to studies on seasonal variation of O_3 concentration at Hahajima, of the Ogasawara Islands [*Matsumoto et al.*, 1998; *Nagao et al.*, 1999], marine air mass derived from the central Pacific contains lower (<5 ppbv) O_3 concentration. Figure 5c shows that frequency of the Pacific air mass increases in June and August. This leads to the oscillation of O_3 concentrations during the Bai-u season and lower monthly mean value of O_3 concentration in summer. Another shift of O_3 concentration from lower to higher values during September corresponds with the Shurine period: vicissitude of the Pacific subtropical air masses and midlatitude air masses under the westerly winds condition [*Barry and Chorley*, 1987]. After the shift, predominance of westerly flow patterns from October to February lead to less variation of O_3 concentrations.

[15] Enhanced volume concentration of the accumulation particles was observed in spring and summer, especially from May to July. The enhancement of fine particles associated with the Kosa phenomenon was reported previously [*Uematsu et al.*, 2002]. However, the cause of the high volume concentration of accumulation particles in summer remains unclear. Factors contributing to the volume enhancement of accumulation particles may include: (1) suitable conditions in summer for conversion from SO_2 to particulate SO_4^{2-} and (2) influence of volcanic SO_2 in Japan. Both factors are related to seasonal variation of the dominant trajectory pattern. On the basis of the analysis of non-sea-salt SO_4^{2-} per SO_2 ratio and backward air trajectories in November, *Kido et al.* [2001a] suggested that oxidation of anthropogenic SO_2 derived from the coastal areas of the Yellow Sea was a significant source of fine SO_4^{2-} particles at Mt. Tateyama. The coastal area of the Yellow Sea is the dominant source region of anthropogenic SO_2 [*Akimoto and Narita*, 1994]. According to the trajectory statistics in Table 2, the frequency of air masses remaining at the coastal areas during the preceding 5 days increases from April to June. In the trajectory of June (Figure 5c), transport from the coastal area of the Yellow Sea to Japan took about 3 to 4 days, which is nearly double that of conditions of November and February. In addition to slowing trajectory speed from the SO_2 source areas, conversion from gaseous SO_2 to SO_4^{2-} is particularly prevalent during transport because incoming solar radiation is strong from May to July. Assuming a conversion of SO_2 to SO_4^{2-} in the troposphere at a rate of 1% per hour [*Warneck*, 1999],

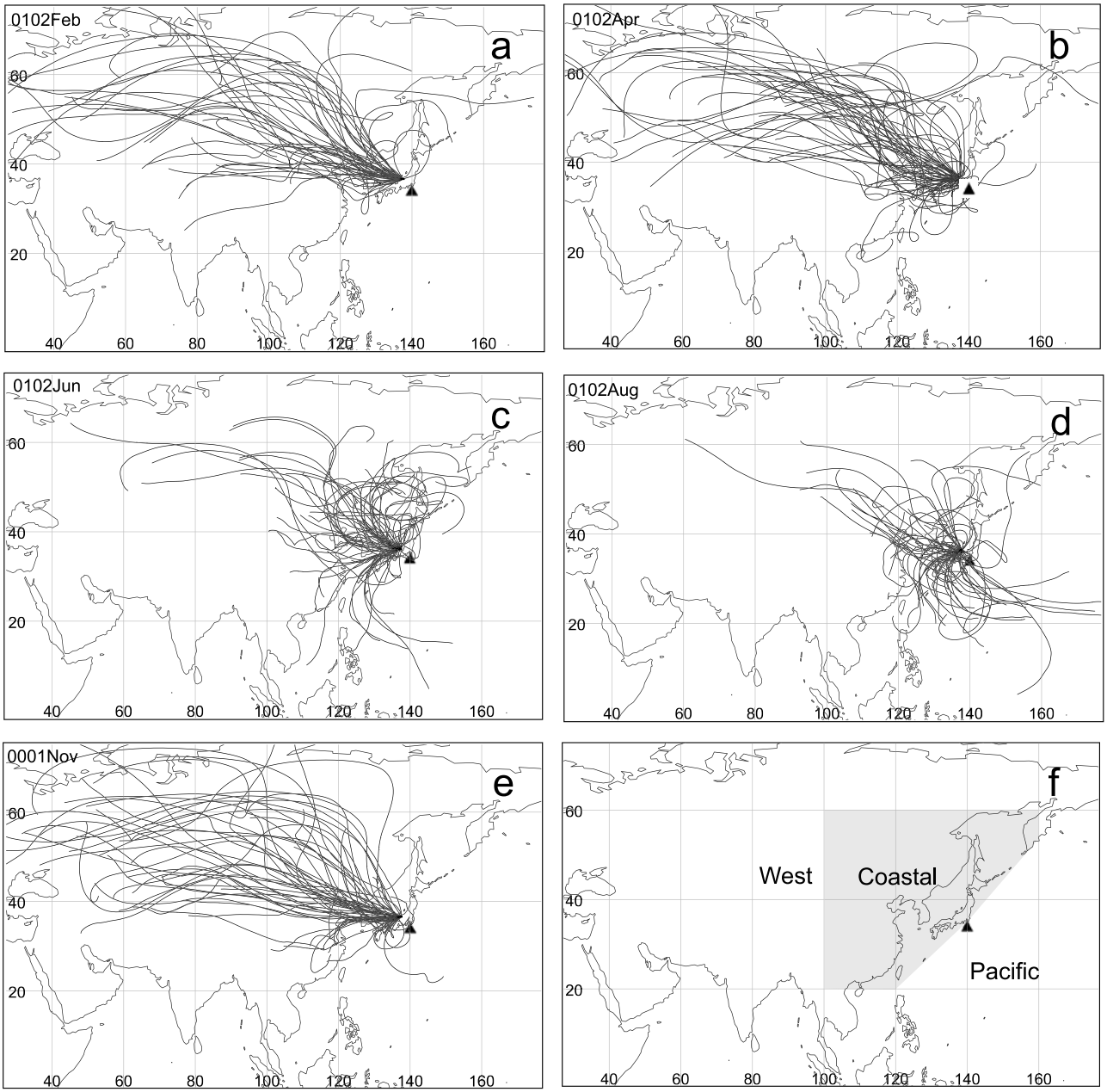

Figure 5. Five-day backward air trajectories from the observation site by the HYSPLIT 4 model, 1997. Figures 5a–5e are composites of trajectories. (a) January 2001 and 2002, (b) April 2001 and 2002, (c) June 2001 and 2002, (d) August 2001 and 2002, (e) November 2000 and 2001. Figure 5f represents three regions characterized in the text. Starting height of the trajectories was set at 3000 m asl. The triangle in the panels represents the location of Miyakejima volcano.

most SO_2 emitted at the coastal area of the Yellow Sea is converted to SO_4^{2-} during transportation at about 4 days. For these reasons, in summer and especially in June, meteorological conditions during transport are suitable to form fine SO_4^{2-} particles through conversion of anthropogenic SO_2. Stagnant airflow over western Japan and the Yellow Sea during the Bai-u season has been suggested as a suitable meteorological condition to form the high sulfate aerosols observed in the northern Kyushu area [*Uno et al.*, 1998].

[16] Furthermore, strong volcanic SO_2 sources exist in Japan. The amount of volcanic SO_2 emission in Japan before the year 2000 was estimated to be 1.1 Tg SO_2 per year mostly (~75%) from Kyusyu such as Sakurajima volcano (31.58°N, 130.63°E) [*Fujita et al.*, 1992]. The annual emission rate of volcanic SO_2 before the year 2000

Table 2. Statistical Summary of Backward Air Trajectories

Month	Case Day	West, %	Pacific, %	Coastal, %
February	56	86	0	14
April	60	70	2	28
June	60	14	8	78
August	62	7	56	37
November	60	75	2	23

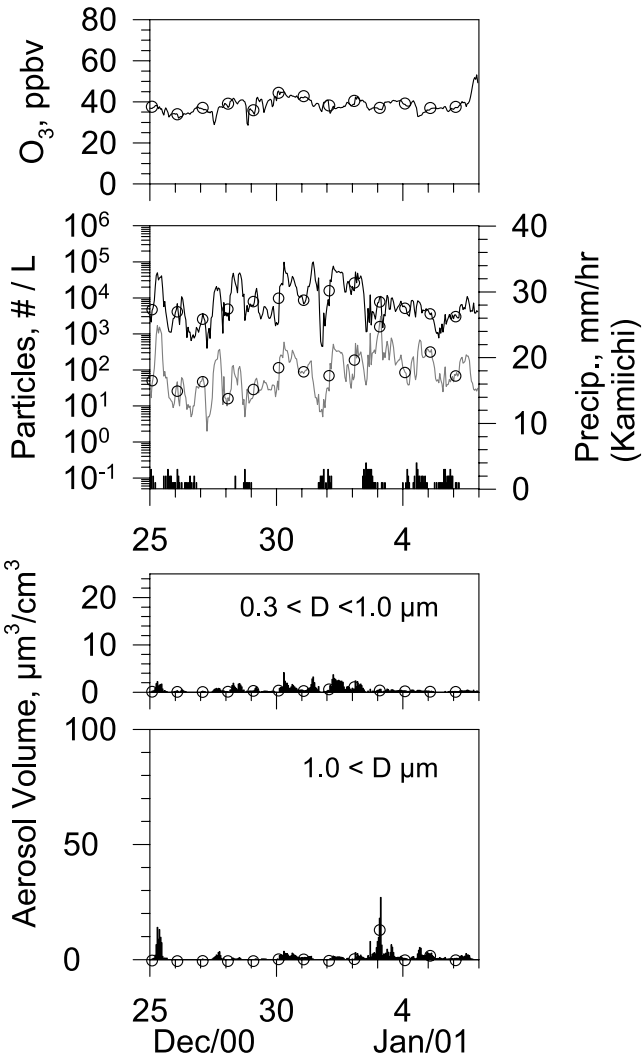

Figure 6. O_3 concentrations and the number and volume of aerosols at Mt. Tateyama with the precipitation amount (vertical bar) at Kamiichi from 25 December 2000 to 6 January 2001. Upper and lower lines in the panel of the number concentration represent values of >0.3 μm and >1.0 μm in diameter, respectively. All data are hourly mean values. Open circles represent mean values from 2400 to 0500 hours.

is comparable to that of anthropogenic SO_2 in Japan [*Akimoto and Narita*, 1994]. Since August 2000, Miyakejima volcano (34.07°N, 139.55°E) has been emitting huge amounts of SO_2. The approximate SO_2 emission rate from Miyakejima volcano was estimated to be as high as 14.6 Tg (SO_2) per year for the first several months, 8.4 Tg (SO_2) per year as the average of 2001, and 1 to 3.7 Tg (SO_2) per year for the winter of 2002 (*Kazahaya et al.* [2000], *Shinohara et al.* [2003] and the Japan Meteorological Agency). Elevated SO_2 and particulate SO_4^{2-} concentrations caused by volcanic gas from Sakurajima and Miyakejima volcanoes have been observed at Mt. Happo [*Satsumabayashi et al.*, 1999; *Katsuno et al.*, 2002] and Mt. Norikura [*Osada et al.*, 2002]. Figure 5d shows that the dominant air trajectories in August are from the Pacific Ocean, implying that the frequency of volcanic impact from Miyakejima volcano increases during summer. Thus slow advection suitable for conversion of anthropogenic SO_2 to SO_4^{2-} and influence of volcanic emission are suggested as important factors contributing to enhanced accumulation particles in summer.

3.2. Kosa Events in 2001

[17] The year 2001 showed a particularly strong Kosa phenomenon over a prolonged season, starting in early January and ending early July. Figures 6 to 8 show the earliest (January), the highest volume concentration, and the latest (July) events in the year 2001. Figures 6 to 8 also include hourly precipitation amounts at Mt. Tateyama. Unfortunately, the amount of precipitation at Mt. Tateyama is obtained only for summer; for winter, we used precipitation data at Kamiichi, located 25 km northwest of Mt. Tateyama.

[18] Figure 6 shows the earliest event observed on 2 to 3 January 2001. Unlike temporal variations in summer seen in Figure 1, diurnal variation is not significant for O_3 and aerosol concentrations. Precipitation (mostly snow) was observed frequently because of the winter monsoon. During the period shown in Figure 6, maximum volume concen-

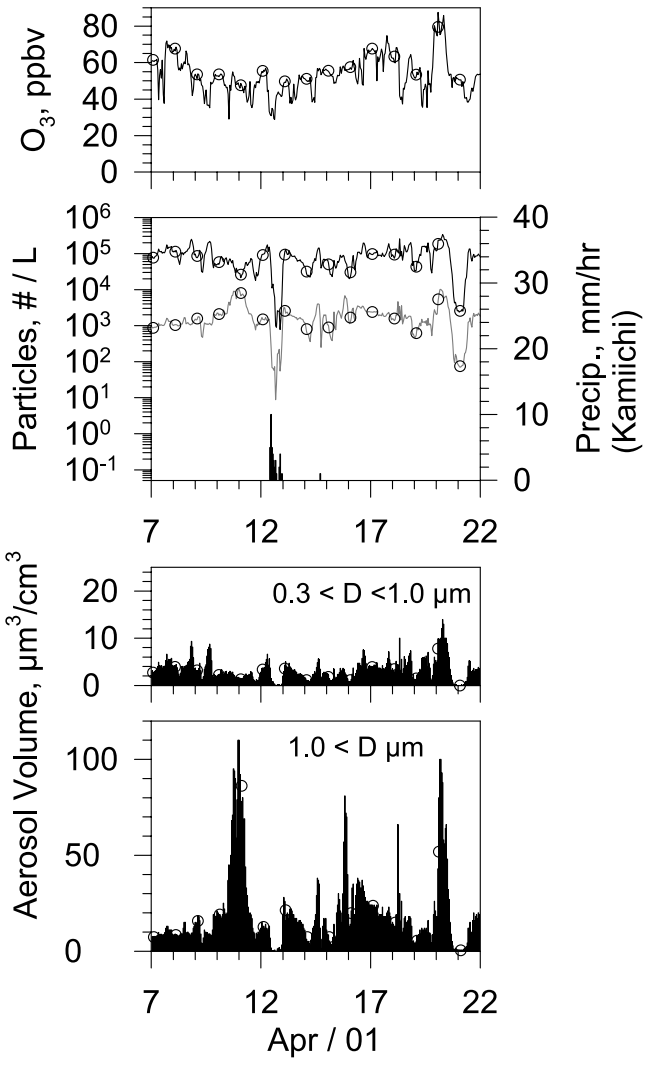

Figure 7. As Figure 6, but from 7 to 22 April 2001.

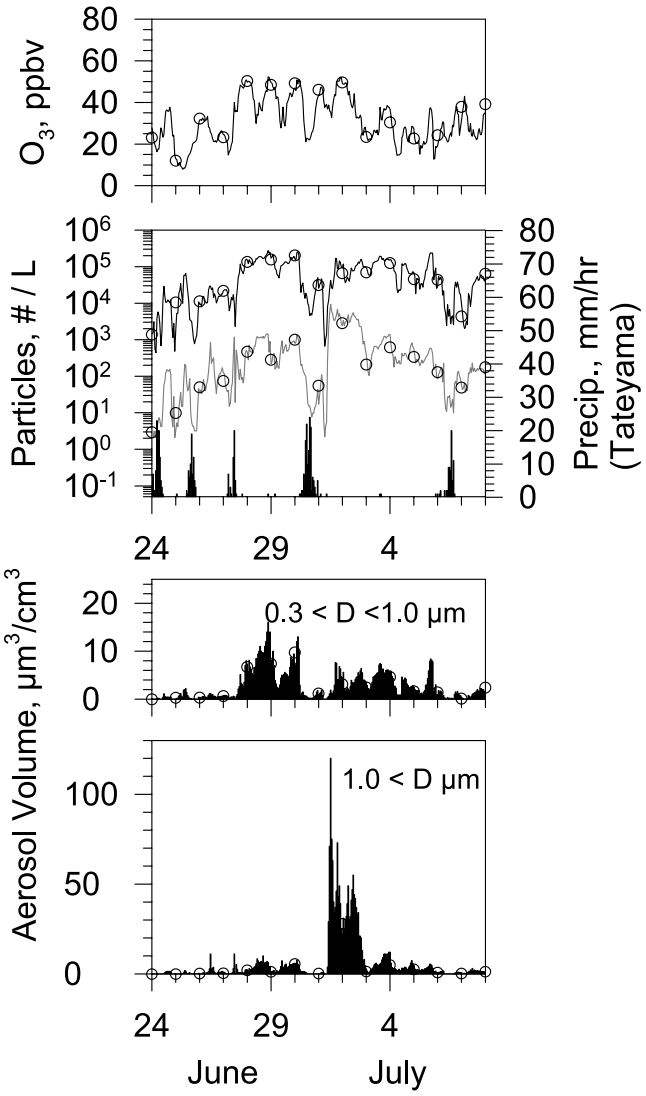

Figure 8. As Figure 6, but from 24 June to 7 July 2001. Precipitation data are available at Mt. Tateyama in summer.

trations of accumulation particles were 3 $\mu m^3/cm^3$ or lower. From 21 on 2 January to 03 on 3 January, volume concentration in the coarse particles increased nine-fold from 3 to 27 $\mu m^3/cm^3$. According to the Japan Meteorological Agency (JMA), Kosa phenomena were reported for western Japan from the afternoon of 2 January. Weather conditions at Mt. Tateyama and the nearby Hokuriku district were rain or snow at that time. On 3 January, dirty snow caused by the mixing of Kosa particles was found at a town near Mt. Tateyama (H. I., unpublished data) and at Takada Meteorological Observatory, located some 80 km to the northeast. Observation of Kosa in early January is rare in Korea and Japan [*Kim and Park*, 2001; *Arao et al.*, 2003].

[19] Figure 7 shows the Kosa event of the highest volume concentration of coarse particles in 2001. From March to May, volume concentrations of the coarse particles were continuously high (generally >5 $\mu m^3/cm^3$) except for precipitation periods as seen on 12 April. Among these major Kosa events, the maximum volume concentration was obtained on 11 April. From 7 to 9 April, volume concentration of coarse particles was about 10 $\mu m^3/cm^3$ and then increased to about 20 $\mu m^3/cm^3$ on 10 April. From 11 to 23 on 10 April, the coarse volume concentration increased 5.6 times from 20 to 111 $\mu m^3/cm^3$, but the volume concentration of accumulation particles decreased during this time. Other peaks in coarse particles found in the night of 15 April and early morning of 20 April showed rapid increase of about five times within 3 to 4 hours. Interestingly, not only coarse particles, but also concentrations of accumulation particles and O_3 showed high values on 20 April. Although O_3 destruction during transport of air mass containing Saharan dusts has been reported [*Prospero et al.*, 1995; *de Reus et al.*, 2000], it is difficult to see apparent depletions of O_3 concentration during Kosa phenomena.

[20] Figure 8 shows the latest event, from 1 to 2 July 2001. After rain stopped at 08 on 1 July, a 30-fold increase of volume concentration of coarse particles was observed from 09 (4 $\mu m^3/cm^3$) to 12 (119 $\mu m^3/cm^3$). High concentration of non-sea-salt Ca^{2+}, an indication of Kosa particles, was observed on the same day at Mt. Norikura (see *Osada et al.* [2002] and manuscript in preparation with new data). Observation of Kosa in July is very rare [*Koizumi*, 1932; *Arao et al.*, 2003]. Regarding the relationship between the coarse aerosol volume and O_3 concentration, it is difficult to recognize a clear depletion of O_3 concentration on 2 July. Figure 8 also shows an example of high (up to 16.1 $\mu m^3/cm^3$) volume concentrations of accumulation particles (28 to 30 June).

4. Summary and Conclusions

[21] Number-size distribution of atmospheric aerosol particles and O_3 concentrations were measured from January 1999 to November 2002 at Mt. Tateyama, central Japan. Free tropospheric concentration of aerosols and O_3 showed seasonal variations related to dominant air trajectories. In spring, increased concentrations of the coarse (>1 μm) particles and O_3 were observed at the site. Asian dust outbreaks and photochemical production are inferred to be dominant factors of these spring enhancements. In summer, increased accumulation particles ($0.3 < D < 1.0$ μm) and oscillation of O_3 concentration are also related to features of air trajectories because of slow movement and vicissitude of air masses between midlatitudes and the subtropical Pacific. Stagnant airflow around the coastal area of the Yellow Sea and around Japan provides suitable conditions for conversion from SO_2 to fine sulfate during transport. Anthropogenic and volcanic emissions were suggested for the major source of summer enhancement of the accumulation particles. Reduced concentration of aerosol particles and less variable O_3 concentrations were maintained from fall to winter under strong westerly winds engendered by the winter monsoon.

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H. Iida, Tateyama Caldera SABO Museum, Ashikuraji 930-1450, Japan. (iidah@nsknet.or.jp)

Y. Iwasaka, K. Matsunaga, and K. Osada, Graduate School of Environmental Sciences, Nagoya University, Nagoya 464-8601, Japan. (iwasaka@stelab.nagoya-u.ac.jp; kmatsu@stelab.nagoya-u.ac.jp; osada@stelab.nagoya-u.ac.jp)

M. Kido, Niikawa Water Museum, Uozu 937-0066, Japan. (kidomizu@nifty.com)

M. Nagatani and H. Nakada, Solar-Terrestrial Environment Laboratory, Nagoya University, Toyokawa 442-8507, Japan. (nagatani@stelab.nagoya-u.ac.jp; nakada@stelab.nagoya-u.ac.jp)