Atmos. Chem. Phys. Discuss., 9, 16527–16547, 2009
www.atmos-chem-phys-discuss.net/9/16527/2009/
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Atmospheric Chemistry and Physics Discussions

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Impact of Chinese SO_2 emissions on submicron aerosol concentration at Mt. Tateyama, Japan

K. Osada[1], T. Ohara[2], I. Uno[3], M. Kido[4], and H. Iida[5]

[1]GSES, Nagoya University, Nagoya, Japan
[2]NIES, Tsukuba, Japan
[3]RIAM, Kyusyu University, Fukuoka, Japan
[4]Toyama Prefectural Environmental Science Research Center, Imizu, Japan
[5]Tateyama Caldera SABO Museum, Tateyama, Japan

Received: 3 July 2009 – Accepted: 4 August 2009 – Published: 6 August 2009

Correspondence to: K. Osada (kosada@nagoya-u.jp)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Rapid Asian economic development might engender secondary impacts of atmospheric aerosol particles over the western Pacific after conversion of gaseous pollutants such as SO_2. To elucidate changes in aerosol concentrations in leeward areas undergoing remarkable industrialization, the number-size distributions of submicron (0.3–1.0 μm) aerosols were measured at Murododaira (36.6° N, 137.6° E, 2450 m a.s.l.) on the western flank of Mount Tateyama in central Japan during January 1999–February 2009. Nighttime data obtained from 24:00 to 05:00 (local time) were used to analyze free-tropospheric aerosol concentration. Monthly average volume concentrations were calculated for months with >50% daily data coverage. Volume concentrations of submicron aerosols were high in spring to early summer and low in winter. Significant increasing trends at 5% level were found for volume concentrations during December–January and March–April. Simulated monthly SO_4^{2-} concentrations at Mt. Tateyama from results of regional aerosol modeling with SO_2 emission inventory up to 2005 showed seasonal variation and winter-spring increasing trends similar to those of observed aerosol concentration. According to the model analyses, the contribution of Chinese SO_4^{2-} concentration was high in winter–spring (ca. 80% of total SO_4^{2-} at Mt. Tateyama). This accords with the increasing trend observed in winter–spring, suggesting that increasing SO_2 emission in China engender enhancement of submicron aerosols over Japan, especially during winter–spring.

1 Introduction

Free tropospheric aerosol particles play an important role in long-range transport of anthropogenic pollutants and in direct and indirect effects on the radiation balance on the earth. Rapid Asian economic development might affect atmospheric pollution, such as increasing SO_2 and NO_x (Streets et al., 2003; Akimoto, 2003; Ohara et al., 2007). These gaseous species might be converted to aerosols during atmospheric transport,

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engendering secondary impacts on aerosol concentrations in leeward regions such as the Western and Northern Pacific regions. Prospero et al. (2003) reported an increasing trend from 1981 to the mid-1990s for non-sea-salt (nss) SO_4^{2-} and NO_3^- in aerosols at Midway Island in the North Pacific. Their analysis ended at year 2000 with large variability and showed an even slightly decreasing tendency in the late 1990s. On the other hand, satellite data have shown a recent increase of an NO_2 column amount over China, which has been compared with results of numerical models (Richter et al., 2005; van der A et al., 2006; He et al., 2007; Uno et al., 2007a; van der A et al., 2008). Increased NO_x emissions in China might increase aerosol nitrate concentration and, consequently, wet and dry deposition flux of total nitrate in and around the Japan Islands (Uno et al., 2007b). The once flat SO_2 emission trend in China (27.1 and 27.6 Mt in 1995 and 2000, respectively) of the late 1990s has turned upward since 2000 (e.g., 36.6 Mt in 2003; Ohara et al., 2007). Although some reports (Huebert et al., 2001; Prospero et al., 2003) have described long-term anthropogenic aerosol impacts at far leeward areas of China, aerosol concentrations have not been described for the area since 2000.

In situ measurements at a high-elevation site might provide valuable data to elucidate year-round free tropospheric aerosols, including rainy days (e.g., Nyeki et al., 1998; Huebert et al., 2001; Osada et al., 2003). Our previous report on free tropospheric aerosols over Japan described a clear seasonal variation of submicron aerosols – high in summer and low in winter – based on nearly 3 yr observation from 1999 (Osada et al., 2003).

To elucidate interannual variations of submicron aerosols over Japan, this paper presents variations of submicron aerosols during 10 years at Murododaira (36.57° N, 137.60° E, 2450 m a.s.l.), on the western flank of Mt. Tateyama. We compare aerosol data with regionally simulated SO_4^{2-} concentration for Mt. Tateyama. We discuss seasonal variation of Chinese SO_4^{2-} contribution for this area to explain seasonally distinct increasing trend in aerosol data.

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2 Observation, data treatment, and numerical model

Number-size distributions of atmospheric aerosol particles were measured using a laser particle counter (KC 01C and KC 01D; Rion Co., Ltd.) from 27 January 1999 at the Hotel Tateyama in Murododaira (36.57° N, 137.60° E, 2450 m a.s.l.), on the western flank of Mount Tateyama in central Japan (Fig. 1). The laser particle counter (LPC) there measures the number of aerosol particles for five size ranges: >0.3, 0.5, 1.0, 2.0, and 5.0 μm diameter. It is calibrated every year by the manufacturer using standard polystyrene latex particles. The sample air humidity was mostly less than 40% because the room temperature was always higher than outside temperature. As described herein, aerosol concentrations are reported as the values of standard temperature (25°C) and pressure (1 atm). Loss of aerosol particles before entering the LPC from the outside was negligible for submicron (0.3–1.0 μm) particles. During the winter monsoon period (November–April), strong northwesterly winds prevailed with frequent snowfalls with rime ice. A snow-clogging preventer resembling the "Frisbee sampler" described by Heidam et al. (1993) was installed at the tip of the inlet tube.

Nighttime data from 2400 to 0500 (local time) were used to analyze free-tropospheric conditions (Osada et al., 2003). For the submicron (0.3–1.0 μm) size range, monthly average volume concentrations were calculated for the month with >50% coverage of daily nighttime data. In all, 90 monthly data were obtained for this period (122 months: January 1999–February 2009).

The three-dimensional regional-scale chemical transport model (Uno et al., 2005) used for this study was based on the Models-3 Community Multiscale Air Quality (CMAQ) version 4.4 modeling system released by the US Environmental Protection Agency (Byun and Schere, 2006). This model is driven by meteorological fields generated by the Regional Atmospheric Modeling System (RAMS) version 4.4 (Pielke et al., 1992). The horizontal model domain for the CMAQ simulation is 6240×5440 km on a rotated polar stereographic map projection centered at 25° N, 115° E, with a grid resolution of 80×80 km. For vertical resolution, we used 14 layers up to 23 km a.s.l in the

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sigma-z coordinate system. We adopted the Statewide Air Pollution Research Center (SAPRC)-99 scheme (Carter, 2000) for gas-phase chemistry; this scheme uses 72 chemical species and 214 chemical reactions, including 30 photochemical reactions. For aerosol calculations, we applied the third-generation CMAQ aerosol module (AERO3), which includes the Secondary Organic Aerosols Model (SORGAM) (Schell et al., 2001) as a secondary organic aerosol model, ISORROPIA (Nenes et al., 1998) as an inorganic aerosol model, and the piecewise parabolic method (PPM) (Binkowski and Shankar, 1995) as the regional particulate model.

We conducted two sets of numerical experiments. First, we performed simulations for 1 January 1999–31 December 2008 (control run). Second, we conducted a perturbation run with emissions from China set to zero to estimate the contribution from Chinese anthropogenic emissions. We defined the Chinese contribution as the difference between the control run and the perturbation run. Both runs used the same meteorological field and initial and boundary conditions for chemical tracers. Meteorological fields for each year were generated by RAMS with initial and boundary conditions defined by the National Centers for Environmental Prediction – National Center for Atmospheric Research (NCEP–NCAR) Reanalysis 1 datasets (http://www.cdc.noaa.gov/cdc/data.ncep.reanalysis.html) (Kalnay et al., 1996; Kistler et al., 2001). The reanalysis datasets have spatial resolution of 2.5°×2.5° and temporal resolution of 6 h. The initial fields of chemical compounds were prepared by the initial conditions processor (ICON) of the CMAQ modeling system (Byun and Schere, 2006). The influence of the initial conditions was eliminated during the 3-month spin-up period. The monthly averaged lateral boundary conditions for most chemical tracers were obtained from a global chemical transport model: Chemical AGCM for Study of Atmospheric Environment and Radiative Forcing (CHASER; Sudo et al., 2002).

For these simulations, we prepared datasets for anthropogenic emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOC), black carbon, organic carbon, and ammonia (NH_3) using the Regional Emission Inventory in Asia (REAS, ver. 1.1) (Ohara et al., 2007). The

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REAS datasets include most anthropogenic sources such as fuel combustion and industrial processes for 1981–2003. We extended the datasets until 2005 using the same methodology as that used by Ohara et al. (2007). We included new data related to energy consumption and industrial activities (e.g., International Energy Agency, 2007; United Nations, 2005, 2006). For parameters such as emission factors and removal efficiencies, we adopted those from 2003. The emissions were fixed at 2005 level for the remainder of the period. Seasonal variation is not considered in the REAS database. For volcanic SO_2 emissions, excluding the Miyakejima Volcano, and emissions from biomass burning, we used climatological inventories from Streets et al. (2003). For the Miyakejima Volcano, which erupted in the summer of 2000, we used the annual mean SO_2 emissions for 2002 from Kazahaya et al. (2003).

This modeling system has been used for analyzing particulate sulfate and sulfur depositions (Katayama et al., 2008), particulate nitrogen and nitrogen depositions (Uno et al., 2007b), and tropospheric ozone (Yamaji et al., 2006, 2008) over eastern Asia, including Japan. In these studies, the simulated results show good agreement with observations.

3 Results and discussion

3.1 Temporal variations of submicron aerosols

Figure 2 shows interannual variation of monthly averages on submicron aerosols at Mt. Tateyama. Horizontal bars in the panel show the month of the insufficient data number. The largest spikes in May–June 2003 might have resulted from the Siberian boreal forest fires as detected at Mt. Fuji (3776 m), ca. 160 km southeast of Mt. Tateyama (Kaneyasu et al., 2007). Although data gaps and such spikes hamper seasonal and interannual variations, winter minima show a slight increasing tendency. Seasonal variations with higher volume concentrations in spring to early summer were also noticed.

Figure 3 presents average monthly seasonal variation of submicron aerosol volume.

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The volume concentration of submicron aerosols was high, with large variation in late spring to early summer (April–July) and low in winter (November–February) as reported previously (Osada et al., 2003). In the analysis of seasonal variation of submicron aerosols in that study, the variation was attributed to the change in the dominant air mass system around Japan based on backward air trajectories from Mt. Tateyama. In winter (November and February), the dominant air flow was derived from the far west of Japan with faster movements. In early summer (June), stagnant slow air flow was detected around the coastal area of the Yellow Sea and around the Japanese islands. The Yellow Sea coastal area is a vast source region of anthropogenic SO_2 (Streets et al., 2003; Ohara et al., 2007). Consequently, meteorological conditions in early summer are suitable to form submicron SO_4^{2-} particles through conversion of anthropogenic SO_2 during slow transport from the coastal area of the Yellow Sea and around Japan (Uno et al., 1998; Osada et al., 2003).

Figure 4 portrays semi-seasonal (combination of 1–3 months) interannual variations of submicron aerosols. Increasing trends of submicron aerosols at a 5% significance level were found for monthly combinations on December–January and March–April. No clear trend was apparent in other combinations of months. As Fig. 2 shows, a slight increasing trend in winter minimum corresponds to an increasing trend in the combination of December–January. Although it is difficult to notice in Fig. 2 because of missing data and spikes in early summer such as in 2003, an increasing trend in spring (March–April) is also discernible in Fig. 4.

Factors relating to the increasing trend in winter to spring observed at Mt. Tateyama might be classified into two points: the cause of the increase and the reason for seasonality. Both are examined in the next section.

3.2 Factors relating to increasing trend and seasonal preference in winter–spring

Aerosol concentrations might be decreased by precipitation scavenging near the site. Our observation was made at a high-elevation site: in-cloud nucleation scavenging

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might reduce submicron aerosol concentration near the site. Although no year-round record of precipitation at the site exists, data at a meteorological station (Kamiichi, about 25 km northwest of Murododaira) near the site were used to elucidate the inter-annual trend of precipitation amount. Figure 5 shows monthly precipitation at Kamiichi for December-January and March-April. The data show no significant trend, suggesting that local precipitation scavenging is not a major factor of the increasing trend of submicron aerosols at Mt. Tateyama.

Increasing SO_2 emissions in eastern Asia, especially in China, might engender the increase of SO_4^{2-} aerosols over Japan, which is located leeward of China. Figure 6 portrays yearly SO_2 emissions in China (red vertical bars in the upper panel; Ohara et al., 2007 with update to 2005), simulated nssSO_4^{2-} aerosol concentrations with all sources (blue line), the Chinese contribution (red line in the middle panel), and the fraction (%) of nssSO_4^{2-} aerosols derived from China (green line in the lower panel) at Mt. Tateyama based on numerical experiments by the chemical transport model. An increasing trend is evident in yearly SO_2 emissions in China, at least before 2005. The yearly rate of increasing SO_2 emission was slow in the early period (–2002) but accelerated after 2002. Rapid industrial development in China has necessitated consumption of fossil fuels including sulfur-containing materials on an enormous scale. However, increasing rates of SO_2 emission in China will be reduced through application of recent pollution control technology and phasing out of small thermal power plants. Consequently, SO_2 emissions in China are difficult to estimate for recent years. However, at least it can be said that the yearly rate of increase in SO_2 emissions has remained positive during 1999–2005.

Estimated SO_4^{2-} aerosol concentrations of the total and Chinese components showed clear seasonal variations: high in summer and low in winter. The simulated seasonal variation agrees well with that of submicron aerosols at Mt. Tateyama. A slightly increasing trend in winter minimum was also well simulated in the model results. The contribution of Chinese SO_2 emissions to SO_4^{2-} aerosol concentrations at Mt. Tateyama was high (60–80%) in winter–spring and low (20–80%) in other seasons.

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The higher Chinese contribution engenders a more direct link of SO_2 emission trends in China to the production of SO_4^{2-} aerosols, implying that SO_4^{2-} aerosol concentrations might be enhanced discernibly during winter–spring. This agrees well with observed seasonality of the increasing trend of submicron aerosols during winter–spring.

Observed increases in submicron volume concentrations were compared directly with simulated SO_4^{2-} at Mt. Tateyama, as portrayed in Fig. 7. Assuming that all aerosols consist only of ammonium sulfate (density=1.8 g cm^{-3}), the corresponding ranges of vertical axes are portrayed in Fig. 7 for aerosol volume (red, right axis) and nssSO_4^{2-} concentrations (blue, left axis). Although chemical components (nitrate and organic, etc.) other than $(NH_4)_2SO_4$ must exist in submicron aerosols (e.g. Krivacsy et al., 2001 at a high elevation site), increasing trends of simulated SO_4^{2-} concentrations agree well with that for submicron volume concentrations, especially for the earlier period (before 2005) of simulation. According to chemical analyses of ionic constituents of free tropospheric aerosols at Mts. Tateyama and Norikura (Osada et al., 2007), the sum of SO_4^{2-} and NH_4^+ comprised about 80% of the total ionic weight. Therefore, the enhanced concentration of aerosol SO_4^{2-} neutralized by NH_4^+ at the site will increase the volume of aerosols. The OPC data used for this study cover the limited size range of 0.3–1.0 μm. Therefore, comparison of absolute values between observed and simulated data are not so meaningful, but agreement of the increasing tendency is important.

On the other hand, interannual variation of the aerosol volume presented in Fig. 7 provides an insight into SO_2 emission trends in China. As described earlier, submicron volume concentrations showed an increasing trend until 2007 or 2008. However, the volume concentrations were almost constant during 2007–2008, and might have started to decrease in 2009. According to official news releases from the Ministry of Environmental Protection in China (MEP, 2009), SO_2 emissions in China seemed to decrease slightly for 2008 because of installation of desulfurization facilities and the progressive shutting down of small power plants. That recent SO_2 reduction in China accords with our observation, but a more extensive record is needed for future studies to confirm this trend.

4 Summary and conclusions

Submicron (0.3–1.0 μm) aerosol data at Mt. Tateyama for the recent decade showed clear seasonal and interannual variations. Monthly average volume concentrations of submicron aerosols were high in spring to early summer and low in winter. Significant increasing trends at the 5% level were found for volume concentrations during December–January and March–April seasons. No trend was found in local precipitation. Simulated SO_4^{2-} concentration at Mt. Tateyama from the results of regional aerosol modeling with SO_2 emission inventory up to 2005 showed similar seasonal variation and winter–spring increasing trends to those of observed aerosol concentration. A higher contribution of Chinese derived SO_4^{2-} concentration was estimated for winter–spring by the model analysis. In fact, SO_2 emissions in China have been increasing rapidly. Therefore, transport of anthropogenic SO_4^{2-} from China engenders an increasing trend of submicron aerosols, especially in winter to spring in this area.

Acknowledgements. We are indebted to the staff of Tateyama Kurobe Kanko (TKK) and Tateyama Caldera SABO Museum for assisting our work at Mt. Tateyama. This work was performed with the support of a Grant-in-Aid for Scientific Research in Priority Areas, Grant No. 18067005 (W-PASS), provided by the Ministry of Education, Culture, Sports, Science and Technology, Japan, and by Grants-in-Aid for Scientific Research (C) 1368061, (B) 20310009, and (A) 20244078 from the Ministry of Education, Culture, Sports, Science and Technology. This research is a contribution of IGBP/SOLAS activity. This work was supported by the Global Environment Research Fund of the Ministry of the Environment, Japan (S-7).

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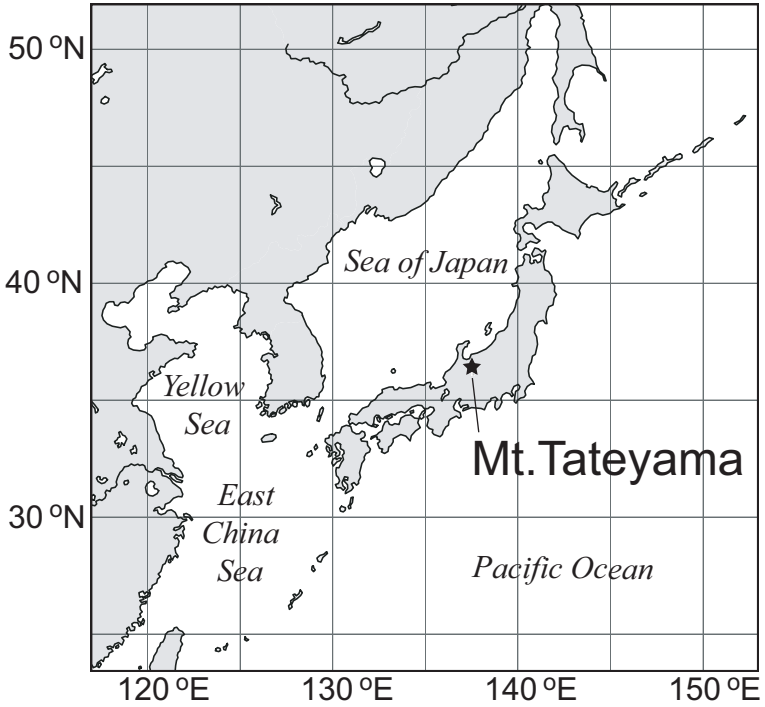

Fig. 1. Map of Mount Tateyama, Japan.

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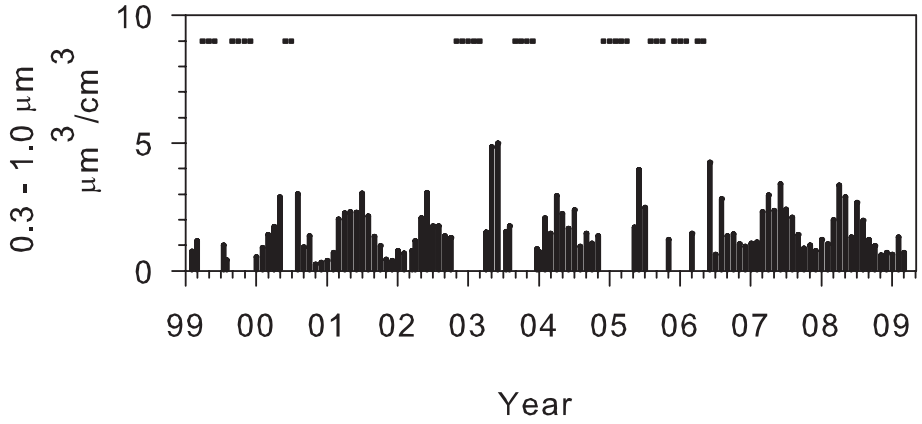

Fig. 2. Monthly volume concentration of submicron aerosols at Mt. Tateyama. Horizontal bars represent periods of missing data.

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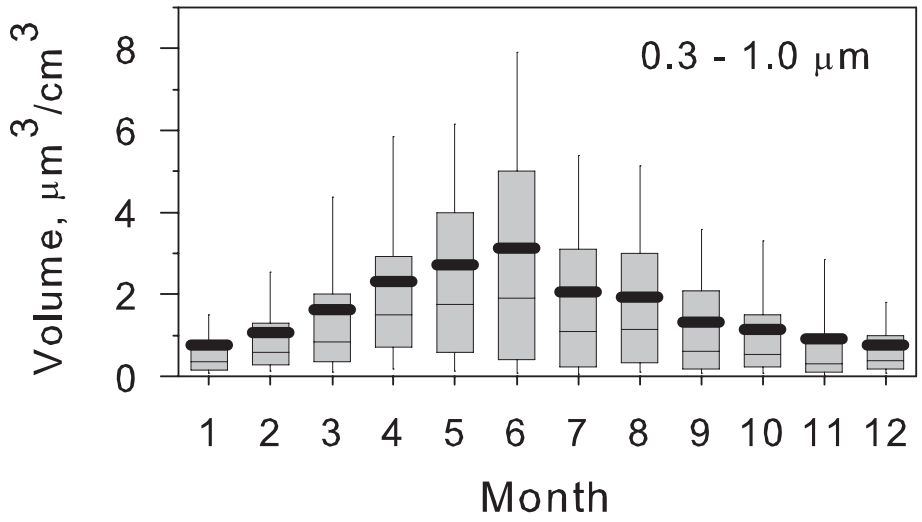

Fig. 3. Monthly box plot of submicron aerosol volume concentration. The lower boundary of the box shows the 25th percentile, the line within the box marks the median, and the upper boundary of the box shows the 75th percentile. Whiskers above and below the box represent the 90th and 10th percentiles. The mean is also portrayed as a thick line.

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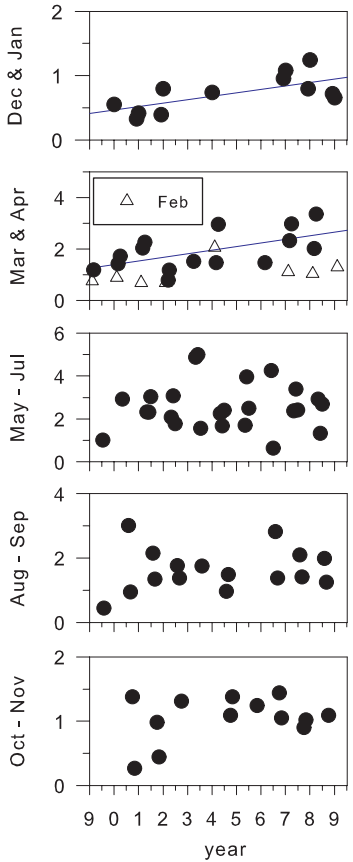

Fig. 4. Interannual variations of seasonally combined monthly submicron aerosol volume data at Mt. Tateyama.

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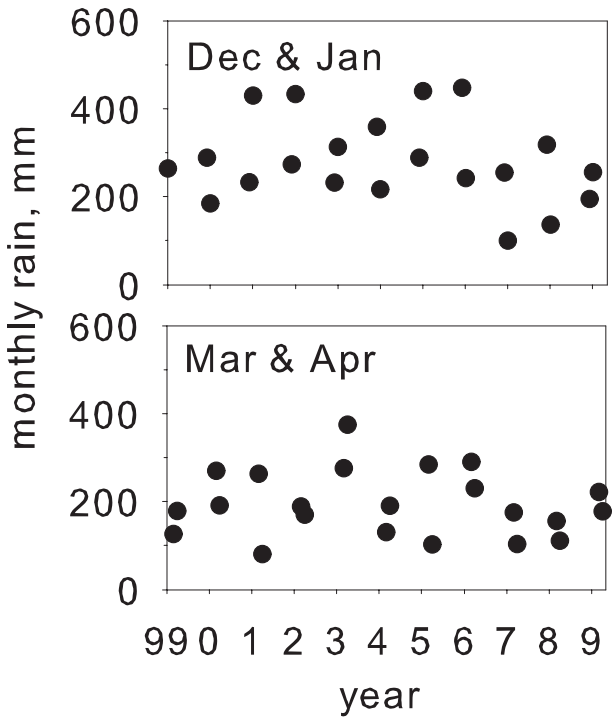

Fig. 5. Interannual variations of monthly precipitation amount at Kamiichi, near Mt. Tateyama.

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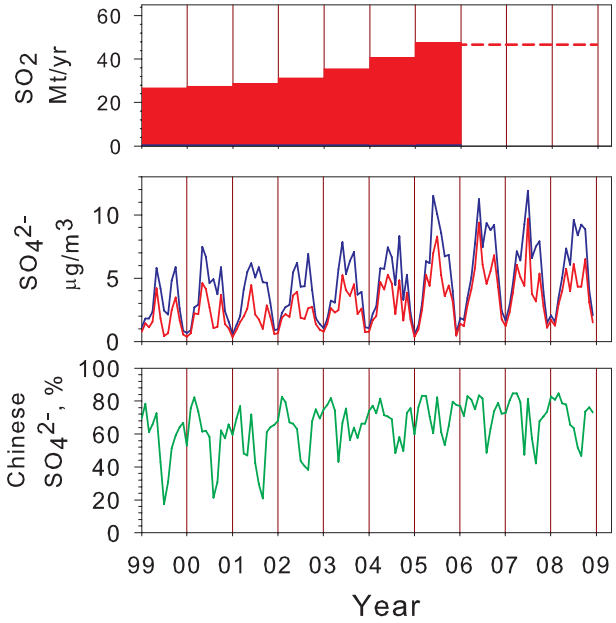

Fig. 6. Yearly SO_2 emission in China (red vertical bars in the upper panel; Ohara et al., 2007 with update to 2005), simulated SO_4^{2-} aerosol concentrations with all sources (blue line) and Chinese contribution (red line in the middle panel), and the fraction (%) of SO_4^{2-} aerosols derived from China (green line in the lower panel) at Mt. Tateyama based on numerical experiments using the chemical transport model.

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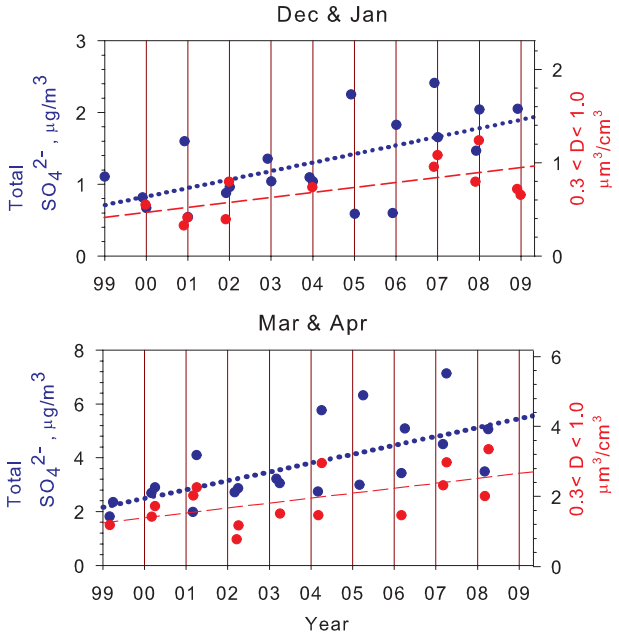

Fig. 7. Simulated monthly SO_4^{2-} concentration and observed submicron volume concentrations at Mt. Tateyama for December–January and March–April. Dotted and dashed lines represent linear fittings for simulated SO_4^{2-} and measured volume concentrations.

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