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High Time-resolution Characterization of $PM_{2.5}$ Sulfate Measured in a Japanese Urban Site

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ABSTRACT

The high time-resolution monitoring data are essential to estimate rapid changes in chemical compositions, concentrations, formation mechanisms, and likely sources of atmospheric particulate matter (PM). In this study, $PM_{2.5}$ sulfate, $PM_{2.5}$, PM_{10}, and the number concentration of size-resolved PMs were monitored in Fukuoka, Japan by good time-resolved methods during the springtime. The highest monthly average $PM_{2.5}$ sulfate was found in May (8.85 μg m^{-3}), followed by April (8.36 μg m^{-3}), March (8.13 μg m^{-3}), and June (7.22 μg m^{-3}). The cases exceed the Japanese central government's safety standard for $PM_{2.5}$ (35 μg m^{-3}) reached 10.11% during four months campaign. The fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ varied from 12.05% to 68.11% with average value of 35.49% throughout the entire period of monitoring. This high proportion of sulfate in $PM_{2.5}$ is an obvious characteristic of the ambient $PM_{2.5}$ in Fukuoka during the springtime. However, the average fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ in three rain events occurred during our intensive campaign fell right down to 15.53%. Unusually high $PM_{2.5}$ sulfate ($>$30 μg m^{-3}) marked on three days were probably affected by the air parcels coming from the Chinese continent, the natural sulfur in the remote marine atmosphere, and a large number of ships sailing on the nearby sea. The theoretical number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ was originally calculated and then compared to $PM_{2.5}$ sulfate. A close resemblance between the diurnal variations of the theoretically calculated number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ and $PM_{2.5}$ sulfate concentration indicates that the secondary formed $(NH_4)_2SO_4$ was the primary form of sulfate in $PM_{2.5}$ during our monitoring period.

Key words: Sulfate, Particulate matter, $PM_{2.5}$, Particle number concentration, Fukuoka

1. INTRODUCTION

As the primary gaseous pollutant, sulfur dioxide (SO_2) with nitrogen oxides (NO_x) is emitted from the burning processes of fossil fuels and can be oxidized in the atmosphere to form gas-phase sulfate via reaction with OH radical to yield H_2SO_4 (Mauldin *et al*., 2012). The gas-phase sulfuric acid, furthermore, preferentially form ammonium sulfate under most conditions, though ammonium nitrate is favored by low temperature and high humidity.

Ammonium sulfate is a major component of $PM_{2.5}$ with ammonium nitrate, carbonaceous mass, and crustal material. Through the one year monitoring study at Hong Kong, Huang *et al*. (2014) reported that sulfate was the most abundant component in the $PM_{2.5}$ mass (32.0% in average, ranging from 24.3-41.1%).

It is well known that $PM_{2.5}$ containing secondary water soluble particles including ammonium sulfate can affect the eyes and nasal mucosa and causes respiratory diseases (Seaton *et al*., 2005; Lebowitz, 1996). Dockery *et al*. (1993) investigated the health risk of fine PM including sulfate and they reported that higher ambient levels of fine PM and sulfate were associated with increased mortality from not only respiratory but cardiac diseases. Especially, since ammonium sulfate is water-soluble, it can be dissolved in sweat and pass through the skin into the bloodstream, and then it can lead to more harmful health risk (Garrod *et al*., 1998).

Sulfate aerosols have also potential to mitigate the effect of rising surface temperatures by scattering the solar radiation (Keith, 2000). As one of the global scale issues associated with sulfate, acid rain can be also considered. The sulphate ion joins with hydrogen atoms in the air and becomes sulphuric acid. This then falls back down to earth as acid rain.

In order to fully understand the health risk and other local/global effects of sulfate particle as well as to control the sources of direct $PM_{2.5}$, its spatio-temporal distributions and transport with the patterns of source emissions must be comprehensively investigated. Especially, the availability of high time-resolved $PM_{2.5}$ sulfate measurement provides the opportunity to better characterize and improve its exposure estimate. Moreover, a good time-resolved monitoring for $PM_{2.5}$ sulfate will provide us with valuable knowledge regarding its health effect as well as formation process, and transport.

In general, the mass concentration of ambient particulate sulfate has been routinely measured by collecting filter samples (usually over 24 hr) typically followed by semi or bulk analyses using an ion chromatographic technique. In recent, several semicontinuous instrumentation methods, capable of providing real-time sulfate measurements, have been developed (Drewnick *et al*., 2003). However, there are a limited number of such reasonable real-time monitoring studies.

In this study, we investigated the characteristics of the highly time-resolved $PM_{2.5}$ sulfate measured simultaneously with $PM_{2.5}$, PM_{10}, and number concentration of size-resolved PMs in a Japanese urban site.

2. MATERIALS AND METHODS

2. 1 Description of the Monitoring Site

An intensive monitoring of $PM_{2.5}$ sulfate, $PM_{2.5}$, PM_{10}, and number concentration of size-resolved PMs were performed simultaneously at a ground-based site in Fukuoka, Japan from Mar. 19 to Jun. 16, 2008. The city of Fukuoka located in the northwest part of Fukuoka Prefecture is the Kyushu's largest and one of the Japan's ten most populated cities. Because of its closeness to the Asian mainland, added to various local sources (see Fig. 4), this area can be directly exposed to the outflow of air masses from the Asian continent especially during springtime. A ground-based sampling station (with a height of 15 m above ground) is located at the Fukuoka Women's University building (33.40°N; 130.26°E) and is marked as a filled circle in Fig. 5.

2. 2 Monitoring of Sulfate and PMs

Highly time-resolved $PM_{2.5}$ sulfate was measured continuously with the ambient particulate sulfate monitor (8400S) manufactured by Rupprecht and Patashnick Co. $PM_{2.5}$ was collected on a platinum strip and then a higher flash temperature was employed for 10-14 msec to reduce sulfate SO_2. Finally, the evolved SO_2 (i.e., product of sulfate reduction) was quantified by pulsed fluorescence. The sampling cycle length was slightly shorter (485 sec). A detailed description of design, operation, and data reduction and processing can be found elsewhere (Drewnick *et al*., 2003). The quality assurance of this $PM_{2.5}$ sulfate monitor (8400S) was reasonably proven through the intercomparision and evaluation of four semi-continuous $PM_{2.5}$ sulfate instruments by Drewnick *et al*. (2003). In their field study, an excellent agreement among four $PM_{2.5}$ sulfate instruments (R^2 and the recoveries were 0.87-0.94 and 0.98-1.06, respectively) was created.

In order to measure $PM_{2.5}$ mass concentration, a light scattering $PM_{2.5}$ monitors (Dust scan Scouts, Rupprecht & Patashnick Co. Model 3020) was simultaneously operated. Details on this $PM_{2.5}$ monitoring system was previously described (Ma and Kim, 2013).

PM_{10} mass concentration was measured by the tapered element oscillating microbalance (TEOM®, Series 1405) monitor. PM_{10} mass concentration data were reported in micrograms per cubic meter at standard averaging time of 10 min. This monitor has excellent short-term precision with $\pm 2\ \mu g\ m^{-3}$ (one-hour average) and $\pm 1\ \mu g\ m^{-3}$ (24-hour average).

The number concentrations of size-selective PMs (i.e., 0.3-0.5 μm, 0.5-1.0 μm, and 2.0-5.0 μm) were also monitored by optical particle counters (OPC) (RION, KC-01D).

3. RESULTS AND DISCUSSION

3. 1 Inter-correlation among $PM_{2.5}$ Sulfate, $PM_{2.5}$, PM_{10}, and Number Concentration of Size-resolved PMs

The relationships among $PM_{2.5}$ sulfate, $PM_{2.5}$, PM_{10}, and number concentration of size-resolved PMs were examined using the correlation coefficient (Fig. 1). $PM_{2.5}$ sulfate has a strong relationship with the number concentration of $PM_{0.5\text{-}0.3}$ ($R=0.90$). This result indicates that the secondary formed $PM_{2.5}$ sulfate primary belonged to $PM_{0.5\text{-}0.3}$. Highly linear correlation ($R=0.82$) was also found between $PM_{2.5}$ sulfate and $PM_{2.5}$. It is therefore suggested that $PM_{2.5}$ was strongly influenced by $PM_{2.5}$ sulfate, namely, sulfate was one of primary components in $PM_{2.5}$. Meanwhile, $PM_{2.5}$ has a higher correlation with the number concentration of $PM_{1.0\text{-}0.5}$ ($R=0.91$) than that with the number concentration of $PM_{0.5\text{-}0.3}$ ($R=0.88$). This means $PM_{2.5}$ was mainly dominated by not the number of PMs but PMs size. The number concentration of $PM_{5\text{-}2}$ has no meaningful relationship with PM_{10}, which is interesting since a more direct relationship might be expected.

3. 2 Time-series Variation of $PM_{2.5}$ Sulfate and $PM_{2.5}$

Fig. 2 shows the timely variation of hourly average $PM_{2.5}$ sulfate and $PM_{2.5}$ during four serial months from March to June. The hourly variations of $PM_{2.5}$ sulfate

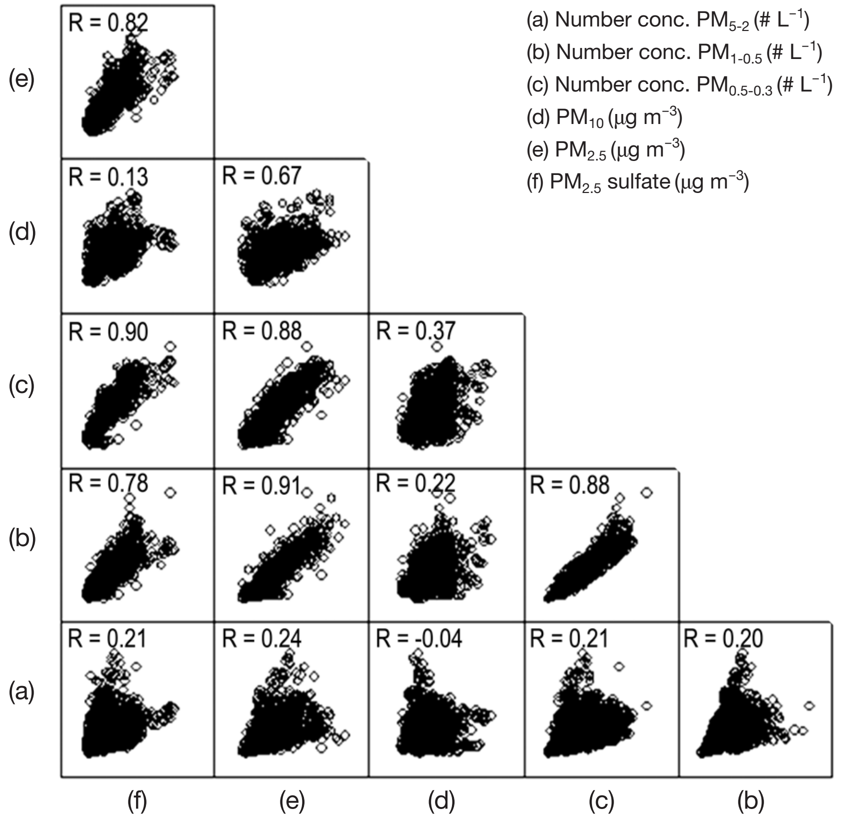


Fig. 1. Inter correlationships among $PM_{2.5}$ sulfate, $PM_{2.5}$, PM_{10}, and numberconcentration of size-resolved PMs.

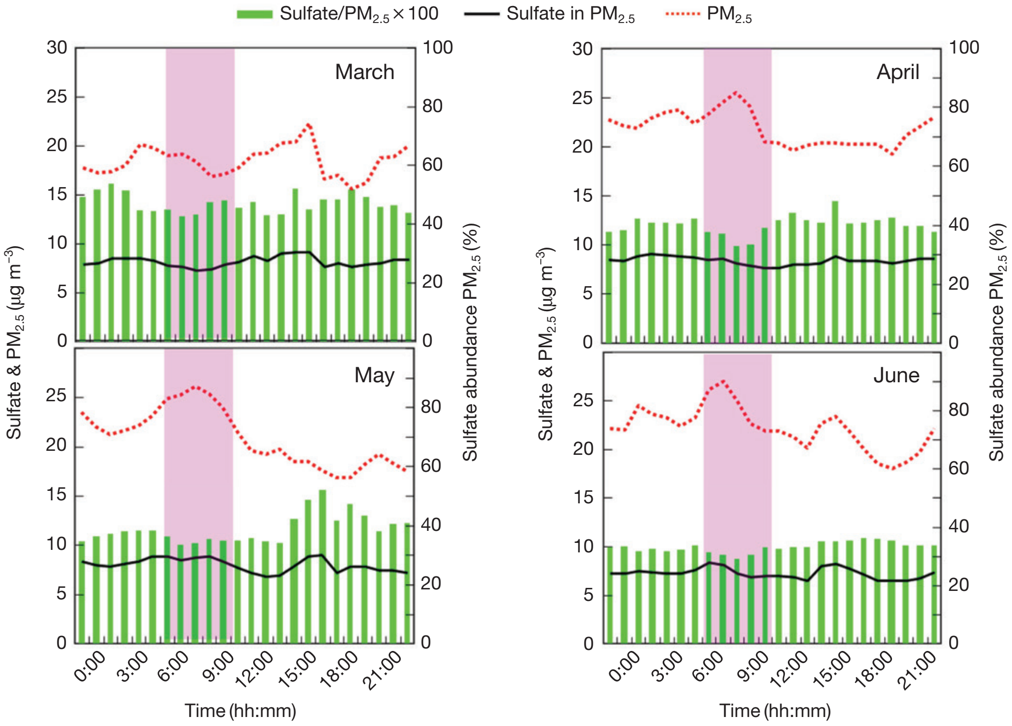
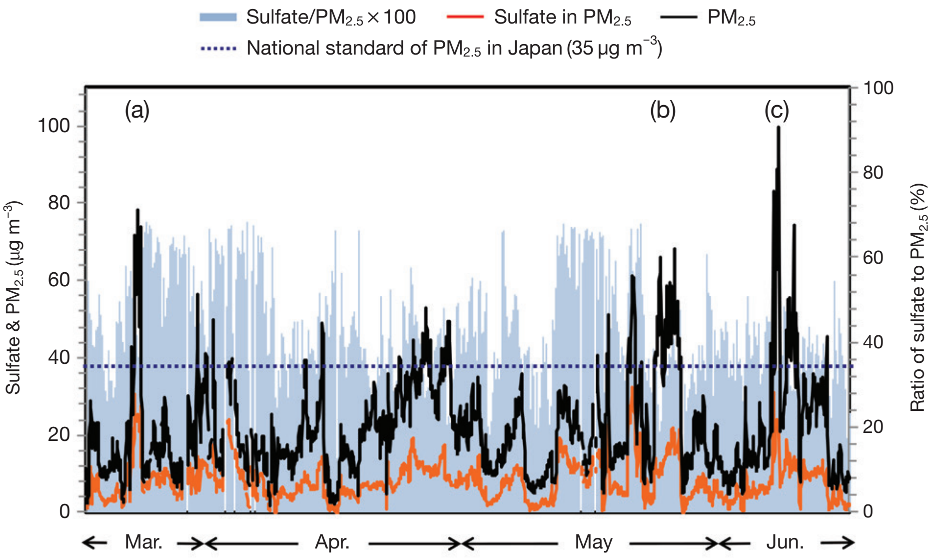


Fig. 2. Timely fluctuation of hourly-averaged $PM_{2.5}$ sulfate in $PM_{2.5}$ and $PM_{2.5}$ in four serial months from March to June.

Table 1. Statistical summary of $PM_{2.5}$ sulfate, $PM_{2.5}$, and the fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ monitored at 10-min intervals.

		Average	Min.	Max.	S.D.[a]
March (n=1,584)[b]	$PM_{2.5}$ sulfate ($\mu g\ m^{-3}$)	8.13	7.29	9.17	0.51
	$PM_{2.5}$ ($\mu g\ m^{-3}$)	18.34	15.62	22.22	1.57
	Sulfate fraction (%)[c]	47.09	42.54	53.91	3.27
April (n=4,176)	$PM_{2.5}$ sulfate ($\mu g\ m^{-3}$)	8.36	7.56	9.12	0.41
	$PM_{2.5}$ ($\mu g\ m^{-3}$)	21.84	19.28	25.51	1.73
	Sulfate fraction (%)	39.99	32.77	47.86	3.18
May (n=4,320)	$PM_{2.5}$ sulfate ($\mu g\ m^{-3}$)	8.85	6.85	9.01	0.66
	$PM_{2.5}$ ($\mu g\ m^{-3}$)	20.88	16.91	26.13	2.91
	Sulfate fraction (%)	39.07	33.52	52.04	4.96
June (n=2,160)	$PM_{2.5}$ sulfate ($\mu g\ m^{-3}$)	7.22	6.44	8.34	0.05
	$PM_{2.5}$ ($\mu g\ m^{-3}$)	22.17	18.05	27.04	2.27
	Sulfate fraction (%)	33.14	29.02	36.35	1.85

[a]: standard deviation
[b]: data number taken at 10-min intervals
[c]: $PM_{2.5}$ sulfate/$PM_{2.5}$ × 100



(a) Mar. 25, (b) May 23, (c) Jun. 9

Fig. 3. Trends in time-series variation of $PM_{2.5}$ sulfate and $PM_{2.5}$ from March to June.

and $PM_{2.5}$ show a severe temporal fluctuation. As might be expected, the maximum level of $PM_{2.5}$ was marked during the morning rush-hour and beginning of work from 06:00AM to 09:00AM during the whole period with the exception of March. Meanwhile, in the case of $PM_{2.5}$ sulfate, there was no meaningful diurnal fluctuation through the whole period. The highest monthly average $PM_{2.5}$ sulfate was found in May (8.85 $\mu g\ m^{-3}$), followed by April (8.36 $\mu g\ m^{-3}$), March (8.13 $\mu g\ m^{-3}$), and June (7.22 $\mu g\ m^{-3}$). Meanwhile, the fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ was marked the highest level in March (47.09%), followed by April (39.99%), May (39.07%), and June (33.14%). Details on monthly $PM_{2.5}$ sulfate, $PM_{2.5}$, and the fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ monitored at 10-min intervals during spring season are statistically summarized in Table 1.

Fig. 3 shows the trends in time-series variation of $PM_{2.5}$ sulfate and $PM_{2.5}$ measured at an interval of 10 minutes from March to June. The drastic variations of high-time resolved $PM_{2.5}$ sulfate and $PM_{2.5}$ were still found in the whole monitoring period. The Japanese central government's safety standard for $PM_{2.5}$ is a 35 $\mu g\ m^{-3}$ hourly-average of over a 24-hour period, and a 15 $\mu g\ m^{-3}$ annually. Many local governments in western Japan, especially Fukuoka, are providing detailed hourly $PM_{2.5}$ data on their websites and have adopted stricter policies. They also issue a $PM_{2.5}$ warning to their residents when $PM_{2.5}$ levels of 35 $\mu g\ m^{-3}$ are reached. The cases exceed 35 $\mu g\ m^{-3}$ and 15 $\mu g\ m^{-3}$ reached 10.11% and 51.21%, respectively during our four months campaign. The temporal variation of $PM_{2.5}$ sulfate (average 7.02 $\mu g\ m^{-3}$) looks very similar to that of $PM_{2.5}$. The fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ varied from 12.05% to 68.11% with the average value of 35.49%. The values of [$PM_{2.5}$ sulfate/$PM_{2.5} \times 100$] over 50% accounted for 14.78% of full data throughout the entire period of monitoring.

Meanwhile, average $PM_{2.5}$ sulfate, $PM_{2.5}$, and the fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ during three rainy periods were 2.17 $\mu g\ m^{-3}$, 11.24 $\mu g\ m^{-3}$, and 15.53%, respectively. Therefore, it is fairly certain that the washout of pollutants, especially water-soluble sulfate, by precipitation relieves temporarily the air pollution in urban area.

Yang *et al.* (2011) characterized the $PM_{2.5}$ speciation in representative megacities across China. According to their study, $PM_{2.5}$ in Beijing, Chongqing, Shanghai, Guangzhou were 118.5 $\mu g\ m^{-3}$ (yearly average), 129.0 $\mu g\ m^{-3}$ (yearly average), 67.6 $\mu g\ m^{-3}$ (yearly average), 81.7 $\mu g\ m^{-3}$ (wintertime average), respectively. $PM_{2.5}$ sulfate at same four megacities in China were 15.8 $\mu g\ m^{-3}$, 25.6 $\mu g\ m^{-3}$, 13.0 $\mu g\ m^{-3}$, 12.6 $\mu g\ m^{-3}$, respectively. The $PM_{2.5}$ of Fukuoka in this study is fairly lower in comparison with the lowest $PM_{2.5}$ level among the cities in China. However, the fraction of $PM_{2.5}$ sulfate to $PM_{2.5}$ with average value of 35.49% is overwhelmingly higher than those of Chinese cities ranged from 6.9% to 19.8%. This result indicates that the high proportion of sulfate in $PM_{2.5}$ is an obvious characteristic of the ambient $PM_{2.5}$ in Fukuoka during the springtime. Therefore, it can be said that reductions in sulfate, fundamentally SO_2, lead to net reductions in $PM_{2.5}$ mass concentration in Fukuoka during the spring season.

3. 3 Discussions on Unusually High $PM_{2.5}$ Sulfate

In Fig. 3, there were three event days unusually high $PM_{2.5}$ sulfate ((a) Mar. 25, (b) May 23, and (c) Jun. 9). The $PM_{2.5}$ sulfates in these three episodical days were over 30 $\mu g\ m^{-3}$. In order to discuss on this unusually high $PM_{2.5}$ sulfate, we drawn the back trajectories at 500 and 1000 m altitudes on three event days of highly recorded $PM_{2.5}$ sulfate using the National Oceanic Atmospheric Administration (NOAA) HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) dispersion-trajectory model "backwards" (http://www.arl.noaa.gov). The result of back trajectories (the left of Fig. 4) is displayed in Fig. 4 with the source distribution in the highlighted region of Fukuoka (the right of Fig. 4).

In the cases of Mar. 25 (i.e., a dust storm occurred), the air parcels were coming from the Chinese continent. Dupont *et al.* (2015) reported that the metal oxides present in mineral dust can act as atmospheric photo catalysts promoting the formation of fine H_2SO_4 and sulfate particles. Therefore, the episodically high $PM_{2.5}$ sulfate on Mar. 25 was probably promoted by mineral dust and/or the local sources distributed at upwind region.

Meanwhile, the prevailing wind on May 23 was originated from the south-western part of Kyusyu Island and the Pacific Ocean. The oceanic emission of dimethylsulfide (DMS: CH_3SCH_3) is known to be a major pri-

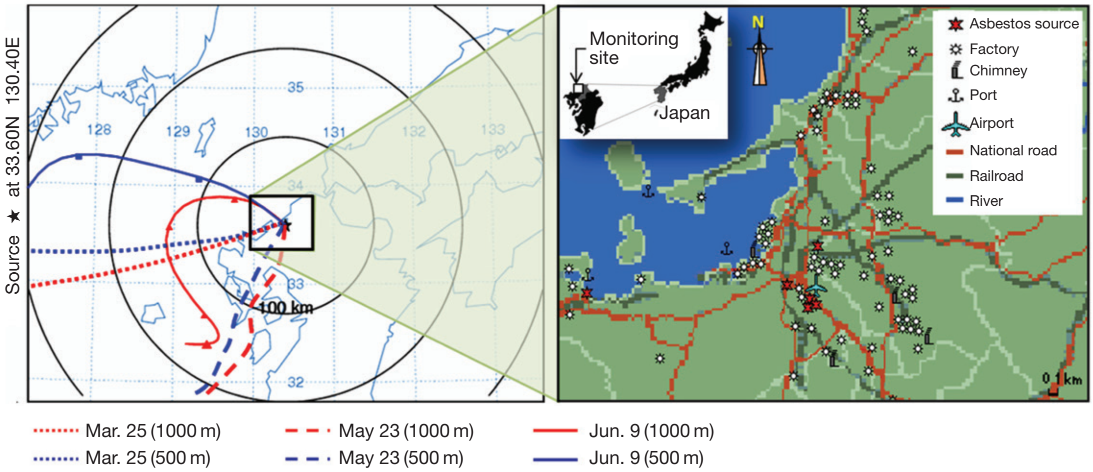


Fig. 4. Back trajectories at 500 and 1000 m altitudes on three event days of highly recorded $PM_{2.5}$ sulfate (left) and the source distribution in the highlighted region of Fukuoka (right).

mary source of natural sulfur in the remote marine atmosphere (Luria and Sievering, 1991; Bates *et al*., 1987). Therefore the highest mass concentration of $PM_{2.5}$ sulfate observed on May 23 was probably influenced by the sulfate aerosol formed by the heterogeneous and homogeneous oxidations of SO_2 in the remote marine atmosphere.

The air parcel of Jun. 9 arrived at our sampling site after passing through the Korean Strait, where a large number of ships including oil tanker and large transport ships are sailing. Ships are major sources of atmospheric pollutants. In Europe, the emissions of SO_2 and NO_x from shipping are projected to exceed all the land-based emissions by 2020 (European Commission, 2005). In the Baltic Sea and the North Sea, the average SO_2 emissions out of 22 ships were 54 ± 13 kg h^{-1} and 42 ± 11 kg h^{-1}, respectively (Berg *et al*., 2012). Consequently, the heightened $PM_{2.5}$ sulfate level on Jun. 9 was possibly due to the liner shipping, coastal liner, and fishery on the Korean Strait and Japanese coast.

Aso volcano (32.88°N, 131.11°E) and Sakurajima volcano (31.35°N, 130.39°E) located in the central Kyushu Island and the bosom of Kagosima bay of Kyusyu, respectively are the world's most active volcanoes. In recent years, they have been the sites of frequent ash eruption. Volcanic eruption also typically releases SO_2, sulfate, and other sulfur-containing species (Iino and Terada, 2003). It is therefore very meaningful to examine the transport and dispersion of the volcanic ashes on May 22 because the prevailing wind on May 23 was originated from the south-western part of Kyusyu Island. We ran the time dependent 3-dimensional Volcanic Ash Forecast Transport And Dispersion (VAFTAD) model developed by the National Oceanic Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) (Heffter and Stunder, 1993). The VAFTAD calculates transport and dispersion of volcanic aerosol particles from an ash column extending from the volcano summit to the column top. The input data for model calculation are eruption time, coordinates (start point), and height of source. The forecast meteorological fields for VAFTAL model are routinely generated from National Centers for Environmental Prediction (NCEP) daily runs of the Global Forecast System (GFS) model and the North American Mesoscale (NAM) model. More detailed model description of the VAFTAD was given by Heffter and Stunder (1993).

Fig. 5 shows the visual ash clouds forecasted by the NOAA's volcanic ash model at surface-FL550 during 6 h from eruption at two volcanoes on May 22. As displayed in Fig. 5, the VAFTAD simulation result indicates that both volcanic ash plumes bounded for the east part of each volcano for a while after eruption and then it moved toward to the southeast. Thus, the present monitoring site appears not to be affected by the volcanic ash plumes.

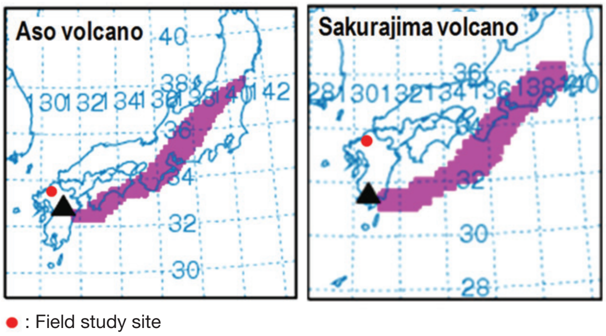


Fig. 5. Visual ash clouds forecasted by the NOAA's volcanic ash model at surface-FL550 during 6 h from eruption at two volcanoes on May 22, 2008.

3.4 Theoretically Calculated Number Concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$

As discussed earlier, there was a strong correlation between $PM_{2.5}$ sulfate and the number concentration of $PM_{0.5\text{-}0.3}$. It is very meaningful to examine the chemical forms of $PM_{2.5}$ sulfate probably classified as $(NH_4)_2SO_4$, NH_4HSO_4, and H_2SO_4. For this reason, we tried to calculate the theoretical number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ and then that was plotted with the diurnal $PM_{2.5}$ sulfate concentration.

The theoretical number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ was calculated by the equation below.

$$N.C._{(NH_4)_2SO_4} = N.C._{PM_{0.5\text{-}0.3}} \cdot \frac{C_{(NH_4)_2SO_4}}{\rho_{(NH_4)_2SO_4}} \cdot \left[\sum_{i}^{n} \frac{C_{spe.}}{\rho_{spe.}}\right]^{-1}$$

where *N.C.* is number concentration (# L^{-1}), *C* is mass concentration ($\mu g\ m^{-3}$), ρ is density ($g\ cm^{-3}$), *spe.* is each composition of five-major species ($n = 5$) of $PM_{2.5}$.

Higo *et al*. (2013) reported that the major components of $PM_{2.5}$ collected at 14-outdoor sites in Fukuoka were sulfate, nitrate, elemental carbon (EC), and organic carbon (OC) and their sum accounted for 82% of $PM_{2.5}$. Therefore, in this study, EC, OC, $(NH_4)_2SO_4$, NH_4NO_3, and NH_4Cl were selected as the *spe.* in above equation.

The concentration of ammonium salts i.e., $(NH_4)_2SO_4$, NH_4NO_3, and NH_4Cl were theoretically calculated by follows:

$$C_{(NH_4)_2SO_4} = C_{SO_4^{2-}} \cdot \frac{M.W._{(NH_4)_2SO_4}}{M.W._{SO_4}}$$

$$C_{NH_4NO_3} = C_{NO_3^-} \cdot \frac{M.W._{NH_4NO_3}}{M.W._{NO_3}}$$

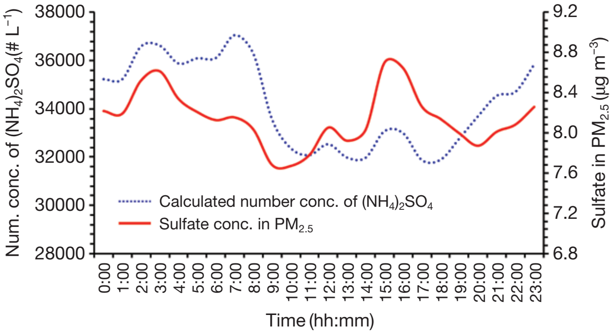


Fig. 6. Diurnal variation of the theoretically calculated number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ (# L^{-1}) and $PM_{2.5}$ sulfate concentration ($\mu g\ m^{-3}$).

$$C_{NH_4Cl} = C_{Cl^-} \cdot \frac{M.W._{NH_4Cl}}{A.W._{Cl}}$$

where *M.W.* and *A.W.* are molecular weight and atomic weight, respectively.

Fig. 6 shows the diurnal variations of the theoretically calculated number concentration of $(NH_4)_2SO_4$ in $PM_{0.5\text{-}0.3}$ (# L^{-1}) and $PM_{2.5}$ sulfate concentration ($\mu g\ m^{-3}$).

Although their variations do not match perfectly, there is a close correspondent diurnal fluctuation. This result indicates that the secondary formed $(NH_4)_2SO_4$ was the primary form of sulfate in $PM_{2.5}$ during our monitoring period. Moreover, $PM_{2.5}$ sulfate ion was usually fully neutralized to $(NH_4)_2SO_4$ during our monitoring campaign.

4. CONCLUSIONS

A clear understanding of the uncertainty related to $PM_{2.5}$ sulfate, which plays an important role in the ambient atmosphere at both emission source and receptor sites, is essential to urban air quality improvement. In addition, a good time-resolved monitoring for $PM_{2.5}$ sulfate and relevant PMs will provide us with valuable knowledge regarding their health effect as well as formation process, and transport. In the present study, several comprehensive evaluations of the $PM_{2.5}$ sulfate together with relevant species were performed. As discussed earlier, air pollution in the countries of East Asia is not only a domestic issue anymore. Accordingly, in order to find the cross-border solution to the long-range transboundary air pollution, the relevant nations (i.e., China, Japan, and South Korea) have to agree more actively to share data on pollutants. Consequently, the results obtained in this study would help policy makers to develop effective regional emission control strategies toward improving the air quality of urban areas in East Asia.

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