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

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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$_2$SO$_4$ (Mauldin et al., 2012). The gas-phase sulfamic 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

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$_4$)$_2$SO$_4$ in PM$_{0.5-0.3}$ 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$_4$)$_2$SO$_4$ in PM$_{0.5-0.3}$ and PM$_{2.5}$ sulfate concentration indicates that the secondary formed (NH$_4$)$_2$SO$_4$ 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
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 & 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 ±2 μg m$^{-3}$ (one-hour average) and ±1 μg 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$_{0.3-0.5}$ ($R = 0.90$). This result indicates that the secondary formed PM$_{2.5}$ sulfate primary belonged to PM$_{0.5-0.3}$. 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$_{1.0-0.5}$ ($R = 0.91$) than that with the number concentration of PM$_{0.5-0.3}$ ($R = 0.88$). This means PM$_{2.5}$ was mainly dominated by not the number of PMs but PMs size. The number concentration of PM$_{5.2}$ 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 correlations among PM$_{2.5}$ sulfate, PM$_{2.5}$, PM$_{10}$, and number concentration of size-resolved PMs.

(a) Number conc. PM$_{5-2}$ (# L$^{-1}$)  
(b) Number conc. PM$_{1.0}$ (# L$^{-1}$)  
(c) Number conc. PM$_{0.5-0.3}$ (# L$^{-1}$)  
(d) PM$_{10}$ (μg m$^{-3}$)  
(e) PM$_{2.5}$ (μg m$^{-3}$)  
(f) PM$_{2.5}$ sulfate (μg m$^{-3}$)

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.
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$_{2.5}$ from March to June.
ern 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 μg m$^{-3}$ are reached. The cases exceed 35 μg m$^{-3}$ and 15 μg 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 μg 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$_{2.5}$ × 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 μg m$^{-3}$, 11.24 μg m$^{-3}$, and 15.53%, respectively. Therefore, it is fairly certain that the wash-out 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 μg m$^{-3}$ (yearly average), 129.0 μg m$^{-3}$ (yearly average), 67.6 μg m$^{-3}$ (yearly average), 81.7 μg m$^{-3}$ (wintertime average), respectively. PM$_{2.5}$ sulfate at same four megacities in China were 15.8 μg m$^{-3}$, 25.6 μg m$^{-3}$, 13.0 μg m$^{-3}$, 12.6 μg 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 μg 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$_2$SO$_4$ 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$_3$SCH$_3$) 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).](image-url)
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_2 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, 2008.

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

As discussed earlier, there was a strong correlation between PM_{2.5} sulfate and the number concentration of PM_{0.5-0.3}. 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_{0.5-0.3} and then that was plotted with the diurnal PM_{2.5} sulfate concentration.

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

\[
N.C.\text{,(NH}_4\text{)}_2\text{SO}_4 = N.C.\cdot\frac{C_{(\text{NH}_4\text{)}_2\text{SO}_4}}{\rho_{(\text{NH}_4\text{)}_2\text{SO}_4}} \cdot \left[ \sum \frac{C_{\text{spe.}}}{\rho_{\text{spe.}}} \right]^{-1}
\]

where \(N.C.\) is number concentration (\#/L), \(C\) is mass concentration (\(\mu g\) \(m^3\)), \(\rho\) is density (g \(cm^3\)), \(\text{spe.}\) 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 \(\text{spe.}\) 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:

\[
C_{(\text{NH}_4\text{)}_2\text{SO}_4} = C_{\text{SO}_4^{2-}} \cdot \frac{M.W.\text{,(NH}_4\text{)}_2\text{SO}_4}{M.W.\text{,SO}_4}
\]

\[
C_{\text{NH}_4\text{NO}_3} = C_{\text{NO}_3^{-}} \cdot \frac{M.W.\text{,NH}_4\text{NO}_3}{M.W.\text{,NO}_3}
\]

\[
C_{\text{NH}_4\text{Cl}} = C_{\text{Cl}^{-}} \cdot \frac{M.W.\text{,NH}_4\text{Cl}}{M.W.\text{,Cl}}
\]
\[ C_{\text{NH}_4\text{Cl}} = C_{\text{Cl}^-} \cdot \frac{M.W_{\text{NH}_4\text{Cl}}}{A.W_{\text{Cl}^-}} \]

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 \((\text{NH}_4)_2\text{SO}_4\) in PM\(_{0.5-0.3}\) (# L\(^{-1}\)) and PM\(_{2.5}\) sulfate concentration (\(\mu g\) m\(^{-3}\)).

Although their variations do not match perfectly, there is a close correspondence diurnal fluctuation. This result indicates that the secondary formed \((\text{NH}_4)_2\text{SO}_4\) 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 \((\text{NH}_4)_2\text{SO}_4\) 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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