Characterization of sulfate in the atmospheric particulate matter over Japan

Extended Abstract Number 236

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Introduction

• The sulfate is the major component of fine particulate matter, PM$_{2.5}$, and related to causing the human health adverse effect.

• In addition, it is also important on work as a cloud droplet and indirect effect to the global warming.

• In Japan, the environmental standard of PM$_{2.5}$ is promulgated in September 9, ‘09, and the reduction countermeasure will be required in future.

• To grasp the degree of transporting sulfate concentration is useful knowledge for predicting the future change.

• For understanding the concentration variation mechanism of the sulfate in PM$_{2.5}$, the long-term observation results of sulfate in Sakai, Osaka from 1986 to 2007 were discussed in detail.
The spatial distribution of sulfate at various sites in Japan

Figure  Mean concentrations of $(NH_4)_2SO_4$ at various sites in Japan.
(Observed by Japan ME)
Figure Comparison of sulfate and crustal aerosol concentrations in fine particulate matter at various sites in Japan. The numeral shows the percent of sulfate in the PM$_{2.5}$. 
Experimental Methods

Observation site:
Our Center at Sakai, Osaka, Japan

Filter sampling:
24-hour PM samples collected on a quartz fiber filter, 25 mm φ, at a suction rate of 20 l/min. since 1985

Chemical analysis:
OC and EC concentrations
by DRI model 2001 OC/EC carbon analyzer
Anion and cation concentrations
by Ion chromatography
Daily concentrations of SO$_4^{2-}$ in PM from January, 1986 to December, 2007 were obtained. 8000 days
Osaka Prefecture
Area: 1,894 km²
Population: 8.80 million

Fig. Map of sampling site.
Figure 2  Monthly mean concentration variations of nss- and ss-sulfate in PM observed at Sakai, Osaka, Japan in 1986 to 2007.
Table  The comparison of the mean concentration of nss- and ss-sulfate for 5 years in PM observed at Sakai, Osaka, Japan in 1986 to 2007 (µg/m³)

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<tbody>
<tr>
<td>nss-Sulfate</td>
<td>4.5 ± 1.3</td>
<td>5.2 ± 1.4</td>
<td>3.8 ± 1.1</td>
<td>4.4 ± 1.4</td>
<td>4.4 ± 1.4</td>
</tr>
<tr>
<td>ss-Sulfate</td>
<td>0.5 ± 0.2</td>
<td>0.5 ± 0.1</td>
<td>0.4 ± 0.1</td>
<td>0.6 ± 0.2</td>
<td>0.5 ± 0.2</td>
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± * : The standard deviation of monthly mean concentration during the period.

’86 – ’95 :  nss-SO₄²⁻ conc.  4 – 6 µ g/m³  ↑

’96 – ’00 :  nss-SO₄²⁻ conc.  6 – 3 µ g/m³  ↓

’02 – ’07 :  nss-SO₄²⁻ conc.  3 – 5 µ g/m³  ↑
Figure 1: Seasonal concentration variation of sulfate.
Figure 3  Seasonal concentration variations of carbon components and PM.

- OC: Organic Carbon
- EC: Elemental Carbon
- TC: Total Carbon
- PM/4
- OC/EC

Mean concentration (µg/m³)
Figure  
Comparison of carbon and sulfate ion concentration fluctuation by day of week.
SO$_2$ Emission

- The emission of SO$_2$ in China was 19.95 M-ton/y in 2000, and increased to 25.49 M-ton/y and 25.89 M-ton/y in 2005 and 2006, respectively*.
- Since the emission from the local enterprises was not included in emission statistics before 1995, it becomes the less estimation of about 40%**.
- In Japan it was 1 M-ton/y in 1980s and less after in 1990s.
- The emission from volcanoes around Japan islands is estimated with about 1 M-ton/y. Miyake-jima which erupted in July, 2000 reached the tens of thousands ton/day at the beginning, and it becomes 0.5 M-ton/y at the present.

* H.Koyanagi, Website (http://eco.nikkeibp.co.jp/article).
Figure 3 Variation of annual and seasonal mean concentrations of nss-sulfate observed at Sakai, Osaka. The sulfur dioxide emission (Mt-SO$_2$/year) in China also shown in this figure.
Figure 4  Daily concentration variation of nss- and ss-sulfate observed at Sakai in 2007.
NOAA HYSPLIT MODEL
Backward trajectories ending at 0000 UTC 13 Jul 07
GDAS Meteorological Data

Weather chart
NOAA HYSPLIT MODEL
Backward trajectories ending at 0000 UTC 14 Jul 07
GDAS Meteorological Data

Source: at 34.58 N 135.48 E

Meters AGL

1500 1500
1000 1000
500 500
100 100

18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00
07/13 07/12 07/11 07/10 07/09

Job ID: 319062  Job Start: Fri Dec 18 06:49:47 GMT 2009
Source 1 lat.: 34.58 lon.: 135.48 hgt.: 500, 100, 1500 m AGL
Trajectory Direction: Backward  Duration: 120 hrs  Meteo Data: GDAS1
Vertical Motion Calculation Method: Isobaric
Produced with HYSPLIT from the NOAA ARL Website (http://www.arl.noaa.gov/ready/)

Weather chart
Conclusion

- Generally, there is a tendency of the concentration reduction for the northeast from the southwest at site of Japanese islands, and it reduces almost by half.
- Annual mean varied from 3.3 to 6.0 µg/m³ for these twenty-two years. It has an increasing tendency in the period from '86 to '95, an decreasing tendency from '96 to 2000, and again an increasing tendency from '03 to '07.
- Nss-sulfate concentration is about 20% low in annual mean in September to January, and 20% high in April to July. The concentration fluctuation in the every year is big in June to August. In February, March and August, the concentration is almost the annual mean value. This variation pattern is quite different with those of carbon and PM.
- There is no the concentration difference by day of week. This result indicates that the human society activity in Japan does not influence the observed nss-sulfate concentration.
• Trend of variation between observed annual mean concentration of nss-sulfate and the emission of SO$_2$ in China is approximately alike.

• The back trajectories of high concentration days passed over the active volcanoes around Japan in a few days, and that the advection of air mass was mostly done from the China around Beijin were clarified.

• Especially, the high concentration is observed, when air mass from the continent is blocked by the seasonal rain front lingering near Japanese islands. On the contrary, the concentration remarkably decreases, when the air mass from the Pacific Ocean does the advection by low pressure and/or high pressure.

• Cooperation of the neighboring countries is necessary for the reduction countermeasure of the component like the sulfate of which the life time in the atmosphere is long.
Thank you