In-situ Measurement and CMAQ Simulation of SO$_2$ over central China

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**Background**

As a booming economy relying on coal, China has emitted ~22 Tg SO$_2$ in 2009 compared to ~5.7 Tg in the U.S. 2009. With SO$_2$ emission reduction measures in the 11th Five-Year Plans of China (2006-2010) and the clean air action for Beijing Olympics, the total SO$_2$ emission has decreased from ~26 Tg (2006) to ~22 Tg (2009).

In the atmosphere, SO$_2$ is oxidized to sulfuric acid ($H_2SO_4$) and sulfate aerosols ($SO_4^{2-}$) mainly through the following reactions:

- Gas phase: $OH + SO_2 \rightarrow HOSO_2 + M$
- Aqueous: $SO_2 + H_2O \rightarrow H_2SO_3 + M$
- $SO_2 + NO_3 \rightarrow SO_4^{2-} + NO + H_2O$

SO$_2$ and its descendant sulfate aerosols not only have profound impacts on the local environment and human health, but also cause regional problems due to long range transports. It is essential to study the SO$_2$ distribution and evolution in China to improve our understanding of air quality issues in the East Asia.

**SO$_2$ Measurement**

**In-situ measurement**

UV fluorescence Technology: SO$_2$ molecules are first excited to a high energy level by photon ($hv$) and decay to a lower energy state with releasing the fluorescence ($h\nu$). $SO_2 + hv \rightarrow SO_2 + h\nu$.

The intensity of fluorescence is proportional to the SO$_2$ concentration.

**Remote sensing**

- Aura OMI products: The Band Residual Difference (BRD) algorithm is developed at NASA-GESF/CUMBC SO$_2$ group. Daily SO$_2$ products are released and publicly available (http://rtiles.gsfc.nasa.gov). These products have 4 estimates of SO$_2$ column density: Planetary Boundary Layer (PBL), Lower Tropospheric (TRL), Middle Tropospheric (TRM), and Upper Tropospheric and Stratospheric (UTL) SO$_2$ column. In this study, OMIO PBL products are used.

Spring 2008 Campaign in Central China

A month-long aircraft campaign was conducted in spring 2008 (March 25 to April 22, 2008), Henan province of central China (Figure 1 right). Henan is the most populous region in China with more than 100 million residents, and has heavy industries including coal mines and power plants. Coal is also widely used for domestic cooking and heating.

A Y-7 turboprop transport aircraft (Figure 1 left) was used as the airborne measurement platform, and located at Zhengzhou Xincheng International Airport (ATA code: CGO 34.51N, 113.54E). The ambient air inlet was installed on the rear of the left wing. The cruise speed was 440 km/h and spirals from 900 to the 4000 m were conducted during research flights. SO$_2$ information within the PBL (lower than 1000 m) was retrieved from the measurements during the gradual descending to land at CGO.

**Summary of Research Flights**

- Figure 2 shows an example of SO$_2$ measurements during the flight in 04/05/2008. One spiral over Changyuan (114.68E, 35.20N) is conducted from 1000 to 4000 m showing up to 5 ppbv SO$_2$ and one descending over Zhongzhou (2000 m) observes up to 1.8 ppbv SO$_2$ within the PBL (lower than 1000 m).

During the campaign, total 8 flights have been carried out over different locations such as Changyuan (strong OMIO SO$_2$ signal region) and Yexian (weak OMIO SO$_2$ signal region). Here is the summary of major features of each flight:

<table>
<thead>
<tr>
<th>Date</th>
<th>Location</th>
<th>Elevation</th>
<th>SO$_2$ (lower than 1000 m)</th>
<th>SO$_2$ (1000~2000m)</th>
<th>SO$_2$ (2000~4000m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>04/05</td>
<td>Changyuan</td>
<td>3 ppbv</td>
<td>2 ppbv</td>
<td>1 ppbv</td>
<td>0.2 ppbv</td>
</tr>
<tr>
<td>04/09</td>
<td>Huojia</td>
<td>1 ppbv</td>
<td>0.5 ppbv</td>
<td>0.5 ppbv</td>
<td>0.05 ppbv</td>
</tr>
<tr>
<td>04/15</td>
<td>Shangcai</td>
<td>1 ppbv</td>
<td>0.5 ppbv</td>
<td>0.5 ppbv</td>
<td>0.05 ppbv</td>
</tr>
<tr>
<td>04/09</td>
<td>Weishi</td>
<td>1 ppbv</td>
<td>0.5 ppbv</td>
<td>0.5 ppbv</td>
<td>0.05 ppbv</td>
</tr>
</tbody>
</table>

**CMAQ Results**

The SO$_2$ distribution has highlty temporal and spatial variability. Figure 3 demonstrates the average SO$_2$ profile with high error bar near the surface. The average SO$_2$ column content is 0.78 Dobson Unit (DU).

The SO$_2$ burden over central China using CMAQ v4.6 with improvements of the SO$_2$ dry deposition rate and advection scheme. The comparison of in-situ and remotely-sensed measurements with CMAQ simulation shows a reasonably consistent picture of the tropospheric SO$_2$. CMAQ-MR results of sulfate aerosols and CO are in good agreement with the in-situ observations.

**Conclusion**

We simulated SO$_2$ over central China using CMAQ v4.6 with improvements of the SO$_2$ dry deposition rate and advection scheme. The comparison of in-situ and remotely-sensed measurements with CMAQ simulation shows a reasonably consistent picture of the tropospheric SO$_2$. CMAQ-MR results of sulfate aerosols and CO are in good agreement with the in-situ observations. The monthly average CMAQ SO$_2$ profile is calculated based on 3 sensitivity runs (Figure 7). The runs underestimate the SO$_2$ concentration in the PBL and overestimate the SO$_2$ in the free troposphere (FT). The run with MR correction has better evaulation of surface SO$_2$ concentration.

The budget of sulfur compounds from CMAQ simulation is calculated (Figure 8). The comparison of Mr and CMAQ SO$_2$ burden (9.4 km) and CMAQ (8.9 km) column maps demonstrates CMAQ captures the location of SO$_2$ plumes well but understimates the burden by ~20%, ~40% (50% for ashber) km.

The lifetime of SO$_2$ is computed as 36.5 hours.

The SO$_2$ burden in China is high over central China, probably due to emission export. The SO$_2$ column over central China is 0.60 g/km$^2$ (within the PBL) and 3.9 g/km$^2$ (up to 4000 m) with error less than 3%.

**Figure 1** Y-7 aircraft at CGO and the airborne measurement platform.