Spatiotemporal variations of tropospheric SO2 over China by SCIAMACHY observations during 2004–2009

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We validate SCIAMACHY SO2 data by MAXDOAS in China.
Different seasonal variation is found for different energy demand.
Tropospheric SO2 was partly under control from 2007 for the reduction policy.
SO2 decrease in several cities for pollution control for the 2008 Olympic Games.

1. Introduction

Changes in the abundance of sulphur dioxide have an impact on atmospheric chemistry and on the radiation field, and hence on climate. Consequently, global observations of sulphur dioxide are important for atmospheric and climate research. The lifetime of sulphur dioxide molecules in the troposphere is a few days (Eisinger and Burrows, 1998). The amount of SO2 is highly variable, above a low background concentration. Clean continental air contains less than 1 ppb of sulphur dioxide, which corresponds to a total column density <0.2 Dobson Units (DU) in a boundary layer of 2 km.

 Usually, atmospheric SO2 was monitored using the accurate but sparse surface SO2 measurements. But ground stations cannot be distributed equally over the globe or over a large area such as China. Recently satellite observations of the tropospheric SO2 have become available. The Global Ozone Monitoring Experiment (GOME) on ERS-2 (Eisinger and Burrows, 1998; Khokhar et al., 2005; Thomas et al., 2005), the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) on ENVISAT (Lee et al., 2008,
The GEOS–Chem model has been used with GEOS–Chem global 3-D model of tropospheric chemistry (Bey et al., 2001) to simulate the SO\textsubscript{2} distribution over China in different seasons. The model (version 4.23; see http://www-as.harvard.edu/chemistry/trop/geos/index.html) uses assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS) including winds, convective mass fluxes, mixed layer depths, temperature, precipitation, and surface properties. In this study, GEOS–Chem was used with GEOS–Chem data products at 4° × 5° global resolution. GEOS–Chem can also be run at 1° × 1° (and coming soon: 0.5° × 0.667°) resolution in nested grids for both China and North America.

3. Results and discussion

3.1. Validation of the satellite data products

The validation of the data products provided by a satellite instrument against ground-based data is among the most important tasks in any mission. Some validations of satellite SO\textsubscript{2} data from OMI have been done by aircraft data in Northeast of China with the long-term mean value is 0.65 DU with a standard deviation 1.1 DU (Krotkov et al., 2008), while the SCIAMACHY SO\textsubscript{2} data in China has not been validated yet. In this study, the ground-based measurements of SO\textsubscript{2} from the MAXDOAS instrument and GEOS–Chem model have been used for validation.

Fig. 1 presents a quantitative comparison between SCIAMACHY and MAXDOAS data during July to October 2008 in Beijing, which shows that SCIAMACHY data is consistent with the ground-based observations from the MAXDOAS instrument. The linearity regression coefficient is 0.92 and ratio 0.76 (P < 0.0001, N = 19).

Fig. 2 shows the results of a comparison of satellite data with predictions of the atmospheric model GEOS–Chem in two typical cases: January (high pollution) and July (low pollution), and these show that the distribution of tropospheric SO\textsubscript{2} in July monitored by satellite is very similar to model results. In January, the model result...
is some higher than satellite since the emission source used in the model overestimating SO2 concentration in East of China in winter season. Fig. 3 presents a quantitative comparison of the data for the east of China (110°–120°E, 30°–40°N) from satellite and model output, which also shows the model result consists with satellite in summer and a little higher than satellite result in winter. All the data show a good correspondence with correlation coefficient 0.88 and slope 1.4 (P < 0.0001, N = 12).

The consistency of SCIAMACHY data with the data from ground based remote sensing are available to give confidence in both the model and SCIAMACHY data over large areas not covered by surface observations. Based on the above primary validation results, the SCIAMACHY data can be used to study the tropospheric SO2 spatial and temporal distribution and trends over China.

3.2. Spatial and temporal distribution

Using the 2004–2009 SCIAMACHY data, the average distribution of SO2 over China is shown in the Fig. 4. The highest pollution levels occur in three regions: (1) east of China (110°–120°E, 30°–40°N); (2) Sichuan basin (104°–108°E, 27°–31°N); (3) Pearl River Delta region (113°–114°E, 22°–23°N). The clear region is the (4) west of China (80°–100°E, 30°–40°N). Fig. 5 exhibits the monthly average tropospheric SO2 vertical column density over the four areas. The seasonal variation can be seen clearly: that there is low value in summer and high in winter in most of China. But it show inverse seasonal variation in south of China (Pearl River Delta region) and the high pollution occur in summer or fall. Table 1 shows pollution levels averaged from 2004 to 2009, with: East ≈ Sichuan basin > Pearl River delta > West. And the heavy
pollution occurs in east of China (0.34 DU), which is about 4 times the value in west of China (0.09 DU). Fig. 5 also clearly shows that the SO2 load over East China is decreasing since the strong control for pollution emission in 2007 in preparation of 2008 Olympic Games in China, while the SO2 load in West China is increasing all the way during 2004–2009, which might reflect that anthropogenic activity increased due to promoting economic development in west of China. It also can be seen clearly that there is a distinct decrease over the three regions after 2007, except for west of China, that may be caused by the policy of the Chinese government to reduce SO2 emissions for the Olympic Games in 2008.

Fig. 3. Correlation of SCIAMACHY monthly average SO2 data with model results averaged in East of China over 2004–2009.

Fig. 4. Tropospheric SO2 vertical columns in DU averaged during 2004–2009 over China (1) east of China; (2) Sichuan basin; (3) Pearl River Delta region; (4) west of China.
3.3. Seasonal variation

Fig. 6 shows the distribution of tropospheric SO2 over China based on SCIAMACHY data averaged over the period 2004–2009 for the different seasons (spring: MAM, summer: JJA, autumn: SON, winter: DJF). It can be seen clearly that high pollution levels occur in spring and winter in North China. But in South China (Pearl River Delta region), high pollution levels occur during summer and autumn. Fig. 6 also illustrates that the SO2 pollution seems to move to the Yellow Sea and the Bohai Bay, and even further, to Japan and Korea, during the spring season because of the strong northwest winds and dust storms (Kim and Park, 2001; Ma et al., 2001; Iwasaka et al., 2003; Zhou et al., 1996; Nishikawa et al., 1991).

Fig. 7 shows the monthly variation of the four areas, with their characteristic seasonal variation. The very different seasonal variation for Pearl River delta, with high SO2 values occurring in summer and autumn, can be seen clearly.

Coal is the main energy source in China and its combustion is the main cause of the increase of atmospheric SO2 emissions in China. In wintertime, anthropogenic emissions are expected to be higher because of heating of buildings, as shown for China by Streets et al. (2003). This is in particular the case in North China, with its severe winters. In South China much less heating is needed in winter and the main coal consumption in that area comes from coal-fired power plants. During summer, the need for electricity is highest and so the SO2 pollution is higher in summer than in other seasons in South China (Wang, 2002).

3.4. Characteristics of tropospheric SO2 over megalocities in China

Table 2 lists the year average values of tropospheric SO2 concentrations for 14 typical major cities in China with a population of more than one million. It shows that the high SO2 pollution value averaged over 2004–2009 occur in the cities in North China and low value in the cities in west of China. Shijiazhuang is the heaviest pollution among all the 14 cities.

Fig. 8 exhibits the year average variation of tropospheric SO2 in those cities. All the cities increased before 2007 since total SO2 emission in China increased (Lu et al., 2010). Among 14 investigated cities, the SO2 observably decreased in the 11 cities after 2007. There are only three cities (Taiyuan, Yinchuan and Lanzhou) that show an increase in the SO2 levels after 2007, which can be found in Table 2 with RED colour.

The reasonable cause is that in 2006, the Chinese State Council issued a Decision on Implementing the Scientific Concept of Development and Stepping up Environmental Protection, which sets the goal for China’s environmental protection in the next five to fifteen years, and brings along desulphurization industry with new development opportunities. By the end of 2006, the operated flue gas desulphurization (FGD) unit capacity rose to 53 million kW, starting from 5 million kW at the end of 2000, accounting for some 14% of the thermal power installed capacity; of this 44 million kW was achieved by unit of 100,000 kW and above. Benefit from this policy carried out by Chinese government, is that the SO2 emissions in China were partly under control from 2006 (Qi et al., 2012). And in order to hold a nice Olympic Games in 2008 Chinese government further reduced the anthropogenic emissions from 2007 by shutting down many Polluting industries, especially in Olympic game cities (Beijing, Shanghai, Shenyang, Qingdao and Hongkong), such as high-emission vehicles were banned from the city's roads and the use of governmental and commercial vehicles were restricted and energy production in major coal-fired power plants was reduced, which cause the SO2 remarkably decrease after 2007. Especially in Beijing, that traffic within the ring roads was restricted.

### Table 1
Year average SO2 value for the four regions in China (2004–2009) (Unit: DU).

<table>
<thead>
<tr>
<th>Area</th>
<th>Lon/Lat</th>
<th>2004</th>
<th>2005</th>
<th>2006</th>
<th>2007</th>
<th>2008</th>
<th>2009</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>East</td>
<td>110–120 E, 30–40’ N</td>
<td>0.271 ± 0.045</td>
<td>0.326 ± 0.077</td>
<td>0.386 ± 0.075</td>
<td>0.433 ± 0.088</td>
<td>0.368 ± 0.153</td>
<td>0.275 ± 0.062</td>
<td>0.343 ± 0.083</td>
</tr>
<tr>
<td>Sichuan Basin</td>
<td>104–108 E, 27–31’ N</td>
<td>0.312 ± 0.131</td>
<td>0.335 ± 0.114</td>
<td>0.373 ± 0.134</td>
<td>0.406 ± 0.157</td>
<td>0.333 ± 0.158</td>
<td>0.332 ± 0.096</td>
<td>0.349 ± 0.132</td>
</tr>
<tr>
<td>Pearl River delta</td>
<td>113–114 E, 22–23’ N</td>
<td>0.206 ± 0.146</td>
<td>0.190 ± 0.094</td>
<td>0.223 ± 0.135</td>
<td>0.185 ± 0.073</td>
<td>0.121 ± 0.066</td>
<td>0.144 ± 0.064</td>
<td>0.178 ± 0.096</td>
</tr>
<tr>
<td>West</td>
<td>80–100 E, 30–40’ N</td>
<td>0.083 ± 0.013</td>
<td>0.091 ± 0.039</td>
<td>0.090 ± 0.025</td>
<td>0.101 ± 0.024</td>
<td>0.106 ± 0.038</td>
<td>0.108 ± 0.030</td>
<td>0.097 ± 0.028</td>
</tr>
</tbody>
</table>

Fig. 5. Monthly variations of tropospheric SO2 for different regions in China.
Fig. 6. Distribution of tropospheric SO$_2$ in DU over China based on SCIAMACHY data of 2004–2009 for the different season.

Fig. 7. Monthly variations of tropospheric SO$_2$ in different areas of China, averaged over 2004–2009.
to cars with even number plates on even days and with odd numbers on odd days (from 20 July). 300,000 high-emission vehicles were banned from the city's roads (1 July) and the use of governmental and commercial vehicles was restricted (by 50% from 23 June; by 70% after 1 July). Access to specific roads (the "Olympic Lanes") was prohibited for other than Olympic related traffic. Public transport capacity was increased with the introduction of new metro and bus lines. Polluting industry was shut down temporarily (20 July) or rebuilt outside Beijing. Energy production in major coal-fired power plants was reduced by 30% (20 July). All construction activities were put on hold (20 July). And now the car in Beijing is still restricted for one day each week. Fig. 9 clearly shows that the SO2 during Jul to Dec in 2008 is lower than the average from other years at the same period. In-situ measurement also shows a significant decrease from period before to after the control measures for the 2008 Olympic Games (Lin et al., 2012).

Since surrounding areas can contribute significantly to air pollution in Olympic game cities (Streets et al., 2007), similar measures have been taken in the surrounding areas, such as Heibei and Henan province, which are adjacent to Beijing and Qingdao respectively. So the SO2 value for Shijiazhuang city in Hebei province and Zhenzhou city in Henan province show distinctly decrease from 2007 (see Fig. 8).

Some cities, such as Taiyuan, Yinchuan and Lanzhou, showed an increase in tropospheric SO2 due to lack of industrial standards, laws & regulations, a lot of medium- and small-scale enterprises (the number of which increased from 2 in 2001 to 200 in 2006) compete maliciously in the market by price war, resulting in a steep decline of the gross margin of the desulphurization industry. Moreover, Taiyuan is one of the main coal mining areas in China. So those three cities still show an increase after 2007.

Fig. 10 shows the monthly variations of tropospheric SO2 in some typical megacities of China. It can be seen clearly that cities located in the north of China show lowest SO2 pollution levels in summer because of prominent anthropogenic activities and meteorological conditions (Chen et al., 2011). But the highest SO2 value doesn’t occur in December and January in Tianjin and Shanghai because of less effective data retrieved. In Guangzhou, the seasonal characteristic is reversed and high SO2 pollution occurs in summer. Because Guangzhou is located in the south of China, there is little or no heating needed in winter and the main coal consumers are coal power plants. During summer, the need for electricity is the highest of the whole year and so the SO2 pollution is higher in summer than in other seasons. Xie and Chen, 2003 showed that most of the coal power plants and thermal power industry are located to the south of Guangzhou city. The higher SO2

Table 2
Year average SO2 value for the major cities in China (2004–2009) (Unit: DU).

<table>
<thead>
<tr>
<th>City</th>
<th>Lon/Lat</th>
<th>2004</th>
<th>2005</th>
<th>2006</th>
<th>2007</th>
<th>2008</th>
<th>2009</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shijiazhuang</td>
<td>114.48E, 38.03N</td>
<td>0.493 ± 0.212</td>
<td>0.454 ± 0.182</td>
<td>0.669 ± 0.147</td>
<td>0.718 ± 0.279</td>
<td>0.521 ± 0.227</td>
<td>0.400 ± 0.211</td>
<td>0.543 ± 0.210</td>
</tr>
<tr>
<td>Zhenzhou</td>
<td>113.65E, 34.77N</td>
<td>0.346 ± 0.067</td>
<td>0.460 ± 0.108</td>
<td>0.467 ± 0.152</td>
<td>0.653 ± 0.189</td>
<td>0.481 ± 0.297</td>
<td>0.487 ± 0.253</td>
<td>0.482 ± 0.179</td>
</tr>
<tr>
<td>Tianjin</td>
<td>117.20E, 39.13N</td>
<td>0.366 ± 0.187</td>
<td>0.412 ± 0.142</td>
<td>0.559 ± 0.191</td>
<td>0.536 ± 0.153</td>
<td>0.424 ± 0.190</td>
<td>0.354 ± 0.150</td>
<td>0.442 ± 0.169</td>
</tr>
<tr>
<td>Taiyuan</td>
<td>112.57E, 37.87N</td>
<td>0.379 ± 0.224</td>
<td>0.415 ± 0.188</td>
<td>0.446 ± 0.136</td>
<td>0.453 ± 0.270</td>
<td>0.478 ± 0.151</td>
<td>0.366 ± 0.172</td>
<td>0.422 ± 0.190</td>
</tr>
<tr>
<td>Chongqing</td>
<td>106.54E, 29.59N</td>
<td>0.396 ± 0.278</td>
<td>0.331 ± 0.077</td>
<td>0.427 ± 0.152</td>
<td>0.512 ± 0.208</td>
<td>0.433 ± 0.189</td>
<td>0.378 ± 0.150</td>
<td>0.413 ± 0.176</td>
</tr>
<tr>
<td>Beijing</td>
<td>116.46E, 39.92N</td>
<td>0.381 ± 0.116</td>
<td>0.361 ± 0.168</td>
<td>0.406 ± 0.108</td>
<td>0.453 ± 0.132</td>
<td>0.330 ± 0.152</td>
<td>0.301 ± 0.128</td>
<td>0.372 ± 0.134</td>
</tr>
<tr>
<td>Shenyang</td>
<td>123.38E, 41.80N</td>
<td>0.276 ± 0.180</td>
<td>0.364 ± 0.154</td>
<td>0.278 ± 0.104</td>
<td>0.314 ± 0.192</td>
<td>0.305 ± 0.151</td>
<td>0.304 ± 0.099</td>
<td>0.307 ± 0.295</td>
</tr>
<tr>
<td>Wuhan</td>
<td>114.32E, 30.52N</td>
<td>0.288 ± 0.091</td>
<td>0.325 ± 0.113</td>
<td>0.376 ± 0.137</td>
<td>0.384 ± 0.131</td>
<td>0.223 ± 0.099</td>
<td>0.221 ± 0.106</td>
<td>0.303 ± 0.113</td>
</tr>
<tr>
<td>Chengdu</td>
<td>104.06E, 30.67N</td>
<td>0.419 ± 0.241</td>
<td>0.276 ± 0.118</td>
<td>0.319 ± 0.121</td>
<td>0.270 ± 0.121</td>
<td>0.238 ± 0.187</td>
<td>0.235 ± 0.132</td>
<td>0.293 ± 0.153</td>
</tr>
<tr>
<td>Shanghai</td>
<td>121.48E, 31.22N</td>
<td>0.305 ± 0.150</td>
<td>0.236 ± 0.097</td>
<td>0.277 ± 0.104</td>
<td>0.311 ± 0.158</td>
<td>0.279 ± 0.118</td>
<td>0.221 ± 0.077</td>
<td>0.272 ± 0.117</td>
</tr>
<tr>
<td>Yinchuan</td>
<td>106.27E, 38.47N</td>
<td>0.241 ± 0.181</td>
<td>0.175 ± 0.084</td>
<td>0.184 ± 0.137</td>
<td>0.235 ± 0.162</td>
<td>0.281 ± 0.225</td>
<td>0.177 ± 0.122</td>
<td>0.216 ± 0.152</td>
</tr>
<tr>
<td>Guangzhou</td>
<td>113.23E, 23.16N</td>
<td>0.258 ± 0.186</td>
<td>0.196 ± 0.100</td>
<td>0.243 ± 0.106</td>
<td>0.260 ± 0.097</td>
<td>0.143 ± 0.088</td>
<td>0.162 ± 0.093</td>
<td>0.219 ± 0.112</td>
</tr>
<tr>
<td>Lanzhou</td>
<td>103.73E, 36.03N</td>
<td>0.178 ± 0.119</td>
<td>0.155 ± 0.091</td>
<td>0.218 ± 0.147</td>
<td>0.194 ± 0.106</td>
<td>0.212 ± 0.147</td>
<td>0.163 ± 0.128</td>
<td>0.187 ± 0.123</td>
</tr>
<tr>
<td>Urumuqi</td>
<td>87.68E, 43.70N</td>
<td>0.197 ± 0.192</td>
<td>0.145 ± 0.109</td>
<td>0.158 ± 0.097</td>
<td>0.214 ± 0.140</td>
<td>0.195 ± 0.174</td>
<td>0.132 ± 0.108</td>
<td>0.174 ± 0.137</td>
</tr>
</tbody>
</table>

Fig. 8. Year variations of tropospheric SO2 in 14 major cities of China.
pollution in summer is caused by the southerly winds which then dominate the city, while in winter the main wind direction is from the north.

4. Conclusion

The tropospheric SO$_2$ columns measured by SCIAMACHY during 2004–2009 have been used to study the spatial and temporal distribution of tropospheric SO$_2$ over China. The main results of this study can be summarised as fellows.

(1) Validation of the SCIAMACHY SO$_2$ data shows that there is consistency between SCIAMACHY and MAXDOAS data, and results from GEOS–Chem. SCIAMACHY data can therefore be used to study the spatial and temporal distribution of tropospheric SO$_2$ spatial and trends therein for China.

(2) The geographic annual average distribution of tropospheric SO$_2$ over China was studied. Heavy SO$_2$ pollution occurs in the east of China and Sichuan basin because of prominent anthropogenic activities. A low tropospheric SO$_2$ column exit in western China because there is less anthropogenic activity.

(3) A typical seasonal variation with high pollution in winter and low in summer in the northwest of China has been found, while the inverse seasonal characteristic appear in the south of China for the energy demand and meteorological conditions.

(4) The characteristics of tropospheric SO$_2$ over the major cities in China were explored and the results show that tropospheric SO$_2$ was partly under control from 2007 because of the policy
from China government for reduction in SO2 emissions in 2006. And the SO2 value shows remarkably decrease in most of major cities after 2007 because strong control for the pollution emission for 2008 Olympic games in China.

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