

# TREND SPATIAL & TEMPORAL DISTRIBUTION, AND SOURCES OF THE TROPOSPHERIC SO<sub>2</sub> OVER CHINA BASED ON SATELLITE MEASUREMENT DURING 2004~2009

Xingying Zhang<sup>(1) (2)\*</sup>, Jos van Geffen<sup>(3)</sup>, Peng Zhang<sup>(1)</sup>, Jing Wang<sup>(1)</sup>

(1) Key Laboratory of Radiometric Calibration and Validation for Environmental Satellites, China Meteorological Administration (LRCVES/CMA), National Satellite Meteorological Center, China Meteorological Administration, Beijing 100081, China

(2) State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), The Institute of Atmospheric Physics, CAS, Beijing 100029, China

(3) Belgian Institute for Space Aeronomy (BIRA-IASB), Brussels, Belgium

## ABSTRACT

In this study, SCIAMACHY/ENVISAT SO<sub>2</sub> data products have been validated by ground based remote sensing instrument MAXDOAS in China. The spatial and temporal distribution of tropospheric SO<sub>2</sub> over China is discussed in this study. The result shows that the SO<sub>2</sub> load over East China is decreasing since strong control for pollution emission in 2007 for preparation of 2008 Olympic Games in China, while the SO<sub>2</sub> load in West China is increasing all the way during 2004-2009, which might reflect that the anthropogenic activity was added to promote the economy development in west of China.

Typical seasonal variation with high pollution levels in winter and low in summer is found in the northwest of China, while the inverse seasonal variation is found for the south of China. The characteristics of tropospheric SO<sub>2</sub> over megacities in China were explored and the results show that tropospheric SO<sub>2</sub> was partly under control from 2006 because of the policy from China government for reduction in SO<sub>2</sub> emissions in 2005.

**Keywords:** tropospheric SO<sub>2</sub>, satellite measurement

## INTRODUCTION

Sulphur dioxide (SO<sub>2</sub>) enters the atmosphere as a result of both natural phenomena and anthropogenic activities. Emission sources are combustion of fossil fuels, oxidation of organic material in soils, volcanic eruptions, and biomass burning. The lifetime of sulphur dioxide molecules in the troposphere is a few days<sup>[1]</sup>.

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. In addition, SO<sub>2</sub> at high concentrations has negative effects on human health, in particular in combination with fog (smog).

Usually, atmospheric SO<sub>2</sub> was monitored using the accurate but sparse surface SO<sub>2</sub> 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 SO<sub>2</sub> have become available. The Global Ozone Monitoring Experiment (GOME) on ERS-2<sup>[1]</sup>, the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) on ENVISAT<sup>[2,3]</sup>, the Ozone

Monitoring Instrument (OMI) on EOS/Aura<sup>[4,5]</sup>, and the GOME-2 instrument on MetOp-A<sup>[6]</sup> have demonstrated their ability to observe columns of SO<sub>2</sub> in the troposphere using measurements in the ultraviolet.

Acid rain and sulphur dioxide (SO<sub>2</sub>) pollution in China are very severe problems – ambient concentrations in some regions are several times higher than air quality standards allow – and have a significant impact on human health, ecosystems, and cultural resources. Since 1995 the Chinese government has placed great emphasis on controlling acid rain and SO<sub>2</sub> pollution. In this study, we report on tropospheric SO<sub>2</sub> data from SCIAMACHY for 2004-2009 over China for the first time.

## 1 Methodology and data sources

### 1.1 SCIAMACHY instrument and SO<sub>2</sub> data retrieval

The SCIAMACHY instrument<sup>[7]</sup> is an 8 channel grating spectrometer measuring in nadir, limb, and occultation (both solar and lunar) geometries. SCIAMACHY covers the spectral region from 220 to 2400 nm with a spectral resolution of 0.25 nm in the UV, 0.4 nm in the visible and less in the NIR. The instrument was launched on ENVISAT in a sun-synchronous orbit on March 1st, 2002 and is in nominal operation since August 2002. With a resolution of 60 x 30 km<sup>2</sup>, SCIAMACHY reaches the scale of SO<sub>2</sub> urban plumes for major cities. Such satellite observations, together with infrared satellite

observations of tropospheric carbon monoxide<sup>[8]</sup>, nitrogen dioxide<sup>[9]</sup>, ozone, other gaseous species, and measurements of aerosols by dedicated instruments<sup>[10,11]</sup>, offer new perspectives to study air pollution.

Using the Differential Optical Absorption Spectroscopy (DOAS) technique<sup>[12]</sup>, a number of atmospheric trace gases can be retrieved from the spectra, including O<sub>3</sub>, NO<sub>2</sub>, SO<sub>2</sub>, BrO, OCIO, HCHO, and H<sub>2</sub>O. In the absence of clouds, a large part of the photons observed by SCIAMACHY in nadir have penetrated down to the troposphere, and global maps of tropospheric concentration fields can therefore be derived from these measurements.

### 1.2 Ground-based remote sensing data

In order to get a quantitative comparison with the satellite data, the ground-based remote sensing data from a MAXDOAS instrument located on the roof of a building in Beijing, above 40m, is used. The MAXDOAS instrument is produced by Key Laboratory of Environmental Optical and Technology, Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences. MAXDOAS instruments are highly sensitive to absorbers in the lowest few kilometers of the atmosphere and vertical profile information can be retrieved by combining the measurements with Radiative Transfer Model (RTM) calculations. The potential of the technique for a wide variety of studies of tropospheric trace species and its (few) limitations are discussed elsewhere<sup>[13-15]</sup>.

### 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<sub>2</sub> data have been done in some areas outside China [16], while the SO<sub>2</sub> data in China has not been validated yet. In order to validate the SCIAMACHY SO<sub>2</sub> data in China, the ground-based measurements of SO<sub>2</sub> from the MAXDOAS instrument have been used for this work.

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.

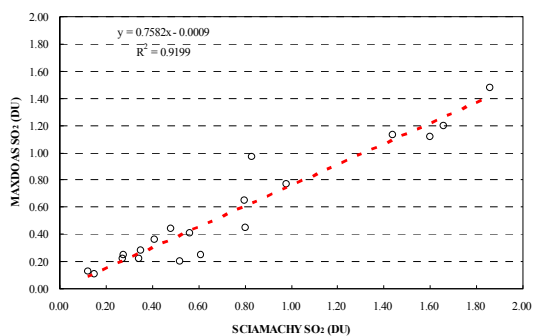


Figure 1. Correlation of SCIAMACHY and MAXDOAS measurements in Beijing

The consistency of SCIAMACHY data with the data from ground based remote sensing are available to give confidence in 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 SO<sub>2</sub> spatial and temporal distribution and trends over China.

#### 3.2 Spatial and temporal distribution

Using the 2004-2008 SCIAMACHY data, the average distribution of SO<sub>2</sub> over China is shown in the Fig. 2. The highest pollution levels occur in the three regions: (1) east of China; (2) Sichuan basin; (3) Pearl River Delta region. The clear region exists in the (4) west of China.

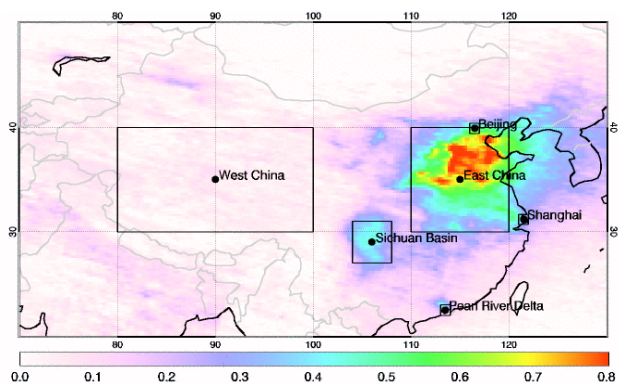


Figure 2. Tropospheric SO<sub>2</sub> vertical columns in DU averaged during 2004 - 2008 over China

Fig. 3 exhibits that the monthly average tropospheric SO<sub>2</sub> vertical column density over the four areas. The seasonal variation could 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 exists in summer.

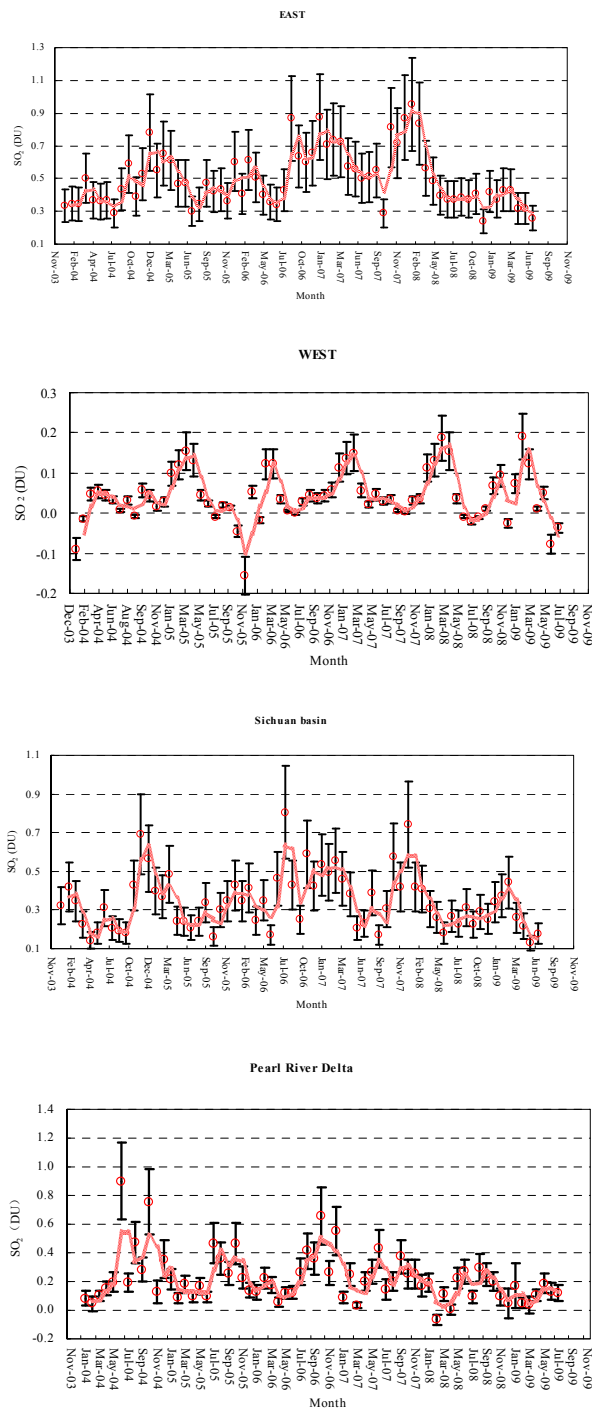


Figure 3. Monthly variations of tropospheric  $SO_2$  for different regions in China

Fig. 3 also clearly shows that the  $SO_2$  load over East China is decreasing since strong control for pollution emission in 2007 for preparation of 2008 Olympic Games in China, while the  $SO_2$  load in West China is increasing all the way

during 2004-2008, which might reflect that the anthropogenic activity was added to promote the economy development in west of China.

### 3.3 Seasonal variation

Fig. 4 shows the distribution of tropospheric  $SO_2$  over China based on SCIAMACHY data averaged over the period 2004-2008 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. 4 also illustrates that the  $SO_2$  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<sup>[17-20]</sup>.

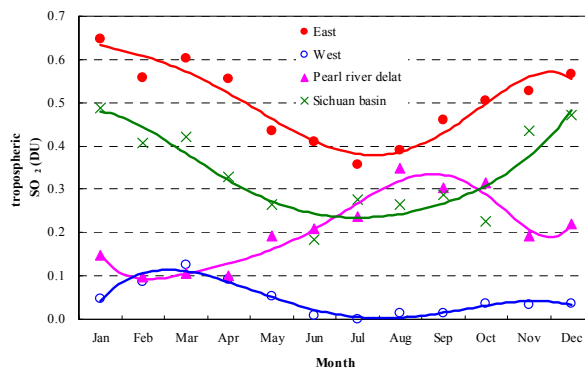


Figure 5. Monthly variations of tropospheric  $SO_2$  in different areas of China, averaged over 2004-2009

Fig.5 shows the monthly variation of the four areas, with their characteristic seasonal variation. It could be seen more detailed that it's very different seasonal variation for Pearl delta, that the high  $SO_2$  value present in summer and autumn.

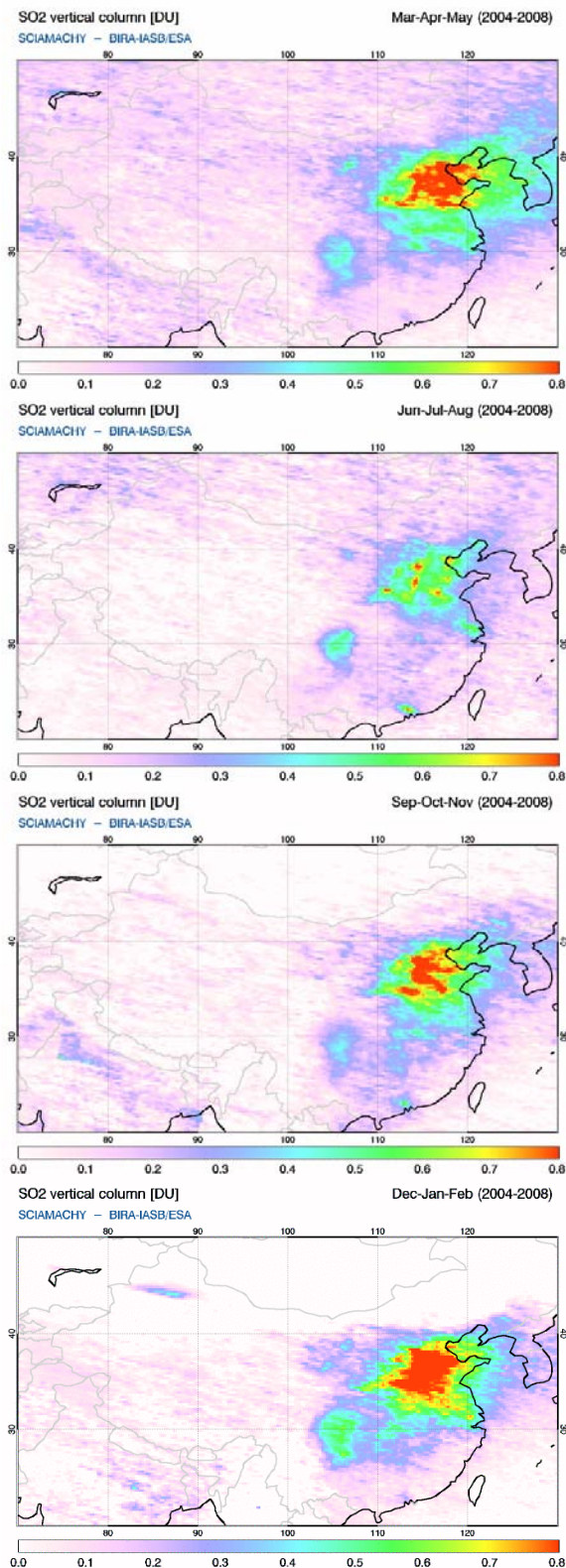


Figure 4. Distribution of tropospheric SO<sub>2</sub> in DU over China in 2004-2008 for the different season

Coal is the main energy source in China and its combustion is the main cause of the increase of atmospheric SO<sub>2</sub> 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)<sup>[21]</sup>. 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-fire power plants. During summer, the need for electricity is highest and so the SO<sub>2</sub> pollution is higher in summer than in other seasons in South China<sup>[22]</sup>.

### 3.4 Characteristics of tropospheric SO<sub>2</sub> over megacities in China

Fig. 6 exhibits the year average variation of tropospheric SO<sub>2</sub> in those cities. Among 14 investigated cities, the SO<sub>2</sub> observably decreased in the 11 cities after 2007. And among these 11 cities, there are 6 cities decreased from 2006. There are only three cities (Taiyuan, Yinchuan and Lanzhou) that show an increase in the SO<sub>2</sub> levels after 2007, which can be found in table 2 with RED colour.

The reasonable cause is that in 2005, 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

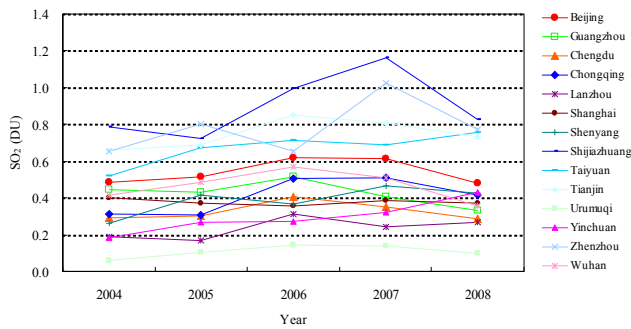


Figure 6. Year variations of tropospheric  $SO_2$  in 14 typical megacities of China

protection in the next five to fifteen years, and brings along desulphurization industry with new development opportunities. By the end of 2005, 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, the  $SO_2$  emissions in China were partly under control from 2006. And in order to hold a nice Olympic Games in 2008 Chinese government further step down 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  $SO_2$  remarkably decrease after 2007. Especially in Beijing, that traffic within the ring roads was restricted 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. 7 clearly shows that the  $SO_2$  during Jul to Dec in 2008 is lower than the average from 2004 to 2007 at the same period.

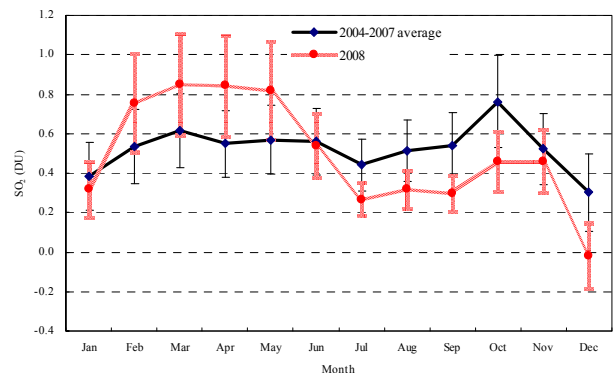


Figure 7. Monthly variations of tropospheric  $SO_2$  in Beijing

Since surrounding areas can contribute significantly to air pollution in Olympic game cities [23], hence similar measures have been taken in the surrounding areas, such as Hebei and Henan province, which are adjacent to Beijing and Qingdao respectively. So the  $SO_2$  value for Shijiazhuang city in Hebei

province and Zhenzhou city in Henan province show distinctly decrease from 2007 (see Fig. 4).

Some cities, such as Taiyuan, Yinchuan and Lanzhou, showed an increase in tropospheric SO<sub>2</sub> for being 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 the main coal producing area in China. So those three cities still show an increase after 2007.

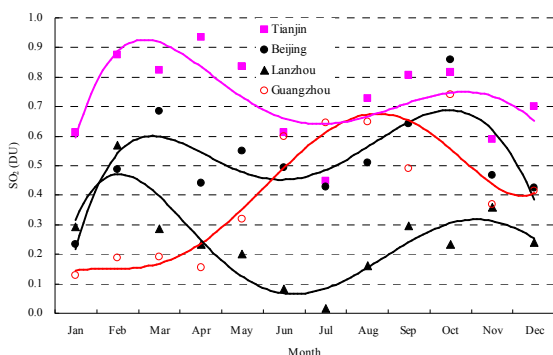


Figure 8. Monthly variations of tropospheric SO<sub>2</sub> in typical megacities of China, averaged over 2004-2009

Fig. 8 shows the monthly variations of tropospheric SO<sub>2</sub> in some typical megacities of China. It can be seen clearly that cities located in the north of China show lowest SO<sub>2</sub> pollution levels in summer because of prominent anthropogenic activities and meteorological conditions. The highest SO<sub>2</sub> value doesn't presents in December and January because the industry activity remarkably decreases during the Spring Festival. But in Guangzhou, the seasonal characteristic is reversed and high SO<sub>2</sub> 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 SO<sub>2</sub> pollution is higher in summer than in other seasons. Xie et al. (2003)<sup>[24]</sup> showed that most of the coal power plants and thermal power industry are located to the south of Guangzhou city. The higher SO<sub>2</sub> 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<sub>2</sub> columns measured by SCIAMACHY during 2004 to 2009 have been used to study the spatial and temporal distribution of tropospheric SO<sub>2</sub> over China for the first time. The main results of this study can be summarised as follows.

- (1) Validation of the SCIAMACHY SO<sub>2</sub> data shows that there is consistency between SCIAMACHY and MAXDOAS data. SCIAMACHY data can therefore be used to study the spatial and temporal distribution of tropospheric SO<sub>2</sub> spatial and trends therein for China.
- (2) The geographic annual average distribution of tropospheric SO<sub>2</sub> over China was studied. Heavy SO<sub>2</sub> pollution occurs in the east of China and in the Sichuan basin because of prominent anthropogenic activities. In western China, a low tropospheric SO<sub>2</sub> column has been found, because there is nearly no anthropogenic activity in that region.
- (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 given locations and meteorological conditions. Guangzhou suffers high SO<sub>2</sub> pollution in summer, since most of the coal power plants and the thermal power industry are located to the south of Guangzhou and southerly winds dominate during the summer time.

(4) The characteristics of tropospheric SO<sub>2</sub> over megacities in China were explored and the results show that tropospheric SO<sub>2</sub> was partly under control from 2006 because of the policy from China government for reduction in SO<sub>2</sub> emissions in 2005. And the SO<sub>2</sub> value shows remarkably decrease in most of the megacity after 2007 because strong control for the pollution emission for 2008 Olympic games in China.

#### **Acknowledgment**

This work was supported by National Natural Science Funds of China (Grant No. 40905056), the fund from State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (Grant No. LAPC-KF-2008-11), the key projects from the 11th five-year plan of national scientific and technological (Grant No.2008BAC34B04-2), the European Union project “AMFIC”(Grant No.FP6-2005-Space-1), and the National Basic Research Program of China “973” project (Grant Nos. 2005CB422200x and 2006CB403702).

#### **Reference**

1. Eisinger, M., and J. P. Burrows, 1998, Tropospheric sulfur dioxide observed by the ERS-2 GOME instrument, *Geophys. Res. Lett.*, 25, 4177–4180.
2. Lee, C., Richter, A., Weber, M., and Burrows, J. P., 2008, SO<sub>2</sub> Retrieval from SCIAMACHY using the Weighting Function DOAS (WFDOAS) technique: comparison with Standard DOAS retrieval, *Atmos. Chem. Phys.*, 8, 6137–6145.
3. Loyola, D., Van Geffen, J., Valks, P., Erbertseder, T., Van Roozendael, M., Thomas, W., Zimmer, W. and Wißkirchen, K., 2007, Satellite-based detection of volcanic sulphur dioxide from recent eruptions in Central and South America, *Advances in Geosciences* 14, 35-40.
4. Krotkov, N.A., Carn, S.A., Krueger, A.J., Bhartia, P.K. and Yang, K., 2006, “Band Residual Difference Algorithm for Retrieval of SO<sub>2</sub> From the Aura Ozone Monitoring Instrument (OMI),” *IEEE Trans. Geosc. Remote Sens.*, 44 (no. 5), May 2006, 1259–1266.
5. Carn, S.A., N.A. Krotkov, A.J. Krueger, K. Yang, and P.F. Levelt, 2007, Sulfur dioxide emissions from Peruvian copper smelters detected by the Ozone Monitoring Instrument, *Geophys. Res. Lett.* 34, L09801, doi: 10.1029/2006GL029020.
7. Bovensmann, H., J. P. Burrows, M. Buchwitz, J. Frerick, S. Noel, V. V. Rozanov, K. V. Chance, and A. P. H. Goede, 1999, SCIAMACHY: Mission objectives and measurement modes, *J. Atmos. Sci.*, 56, 127–150.



8. Emmons, L. K., et al. (2004), Validation of measurements of pollution in the troposphere (MOPITT) CO retrievals with aircraft in situ profiles, *J. Geophys. Res.*, 109(D3), D03309, doi:10.1029/2003JD004101.
9. Van der A, R. J., D. H. M. U. Peters, H. Eskes, K. F. Boersma, M. Van Roozendaal, I. De Smedt, and H. M. Kelder, 2006, Detection of the trend and seasonal variation in tropospheric NO<sub>2</sub> over China, *J. Geophys. Res.*, 111, D12317, doi:10.1029/2005JD006594.
10. Chu, D. A., Y. J. Kaufman, G. Zibordi, J. D. Chern, J. Mao, C. Li, and B. N. Holben, 2003, Global Monitoring of Air Pollution over land from EOS-Terra MODIS, *J. Geophys. Res.*, 108(D21), 4661, doi: 10.1029/2002JD003179.
11. Wang, J., and S. A. Christopher, 2003, Intercomparison between satellites derived aerosol optical thickness and PM<sub>2.5</sub> mass: Implications for air quality studies, *Geophys. Res. Lett.*, 30(21), 2095, doi: 10.1029/2003GL018174.
12. Platt, U., 1994, Differential Optical Absorption Spectroscopy (DOAS), in *Air Monitoring by Spectroscopic Techniques*, M.W. Sigrist (ed.), Chemical Analysis Series 127, John Wiley & Sons, New York.
13. Honninger G., von Friedeburg C., and U. Platt, 2004, Multi axis differential optical absorption spectroscopy (MAX-DOAS), *Atmos. Chem. Phys.*, 4, 231–254,
14. Hanlim Lee, Young J. Kim, Chulkyu Lee, and Yeo Sook Kim, 2007, Remote Measurement of Atmospheric Trace Gases and Aerosol Using Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) and Imaging DOAS (I-DOAS) Techniques, The 10th International Conference on Atmospheric Sciences and Applications to Air Quality, May 14 – 16, 2007, Hong Kong, China.
15. Wang, P., Richter, A., Bruns, M., Burrows, J. P., Scheele, R., Junkermann, W., Heue, K.-P., Wagner, T., Platt, U., and Pundt, I., 2006, Airborne multi-axis DOAS measurements of tropospheric SO<sub>2</sub> plumes in the Po-valley, Italy, *Atmos. Chem. Phys.*, 6, 329–338
16. Krotkov, N. A., B. McClure, R. R. Dickerson, S. A. Carn, C. Li, P. K. Bhartia, K. Yang, A. J. Krueger, Z. Li, P. F. Levelt, H. Chen, P. Wang, and D. Lu, 2008, Validation of SO<sub>2</sub> retrievals from the Ozone Monitoring Instrument over NE China, *J. Geophys. Res.*, 113, D16S40, doi: 10.1029/2007JD008818.
17. Byung-Gon Kim, Soon-Ung Park, 2001, Transport and evolution of a winter-time Yellow sand observed in Korea, *Atmospheric Environment*, 35, 3191-3201
18. Iwasaka, Y.; Shi, G.-Y.; Shen, Z.; Kim, Y. S.; Trochkin, D.; Matsuki, A.; Zhang, D.; Shibata, T.; Nagatani, M.; Nakata, H., 2003, Nature of atmospheric aerosols over the desert areas in the Asian continent: chemical state and number concentration of particles measured at Dunhuang, China. *Water, Air, & Soil Pollution: Focus*, 3(2), 129-145.
19. Zhou M. K. Okada, F. Qian, P.M. Wu, L. Su, B.E. Casareto, T. Shimohara, 1996, Characteristics of dust-storm particles and their long-range transport from China to Japan---case studies in April 1993. *Atmospheric research*, 40, 19-31

20. Nishikawa, Masataka; Kanamori, Satoru; Kanamori, Nobuko; Mizoguchi, Tsuguo, 1991, Kosa aerosol as eolian carrier of anthropogenic material. *Science of the Total Environment*, 1991, 107, 13-27
21. Streets D G, Bond T C, Carmichael G R, et al., 2003, An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res.*, 108 (D21), 8809, doi: 10.1029/2002JD003093
22. Wang Zhi xuan(2002), Integrated Countermeasures and Suggestions to SO<sub>2</sub> Emission Control of Thermal Power Plants in China, *ELECTRICITY*, 13(1), 23-26
23. Streets, D.G., J. S. Fu, C. J. Jang, J. Hao, K. He, X. Tang, Y. Zhang, Z. Wang, Z. Li, Q. Zhang, L. Wang, B. Wang, and C. Yu (2007), Air quality during the 2008 Beijing Olympic Games, *Atmos. Environ.*, 41, Issue 3, January 2007, Pages 480-492, ISSN 1352-2310, DOI: 10.1016/j.atmosenv.2006.08.046
24. Xie M., and Chen Y.S., 2003, SO<sub>2</sub> Pollution of Guangzhou in Recent Years: Its Characteristics and Sources, *Environmental Science and Technology*, Vol. 26, No. 2, 18-21. (In Chinese)