A New and Detailed Assessment of the Spatiotemporal Characteristics of the SO₂ Distribution in the Pearl River Delta Region of China and the Effect of SO₂ Emission Reduction

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ABSTRACT

Signal enhancement technology (sub-pixel interpolation) is used to obtain SO₂ column concentrations for Guangdong Province in China from 2005 to 2016. The high resolution (2 km × 2 km) data used was obtained via a remote sensing satellite (Ozone Monitoring Instrument) and verified by comparing it with average annual SO₂ data recorded in ground monitoring stations. The correlation was found to be up to 0.95. Moreover, the data was cross-correlated with national and regional inventories of pollution sources. The results show that the regional characteristics of the spatial distribution obtained are consistent and the detailed characteristics are highly coincidental. Based on this, the new and detailed spatiotemporal variation was analyzed and the effect of emission reduction in urban agglomerations on the SO₂ concentration in the Pearl River Delta (PRD) region of China investigated. The results demonstrate that the distribution of SO₂ pollution in the PRD has been transformed over the period studied. In the early stages, it had a traditional high-concentration type of distribution (with agglomeration areas like Guangzhou and Foshan as high-concentration pollution centers) and this has changed to the currently-observed low-concentration decentralized type of distribution (mainly distributed along administrative boundaries). In the last 10 years, significant SO₂ emission reduction has occurred in prefecture-level cities, e.g., Foshan, Zhongshan, and Guangzhou (with emission-reduction amplitudes of 71%, 65%, and 57%, respectively). Foshan and Zhongshan are the top two prefecture-level cities in the PRD region in terms of significant reduction in rate of SO₂ contribution. The SO₂ contribution rate fell from 17% to 13% in Foshan and from 16% to 10% in Zhongshan. However, the relative contribution rates in Zhaoqing and Huizhou increased from 7% to 11% and from 6% to 10%, respectively. The size of the emission reduction and changes in SO₂ contribution rates in the prefecture-level cities in the PRD region show that the government's efforts to improve air quality have had a significant effect.

Keywords: Pearl River Delta; Satellite remote sensing; Sulfur dioxide; Assessment of emission reduction.

INTRODUCTION

Sulfur dioxide (SO₂) is a gas found in trace amounts in the atmospheric boundary layer, mainly as a result of anthropogenic pollution and natural emission, e.g., volcanic eruption. However, in China, it mainly comes from anthropogenic sources (Cheng *et al.*, 2017; Fang *et al.*, 2017; Wang *et al.*, 2018). SO₂ has a life cycle of 1–3 days

in the atmosphere and is ultimately oxidized into sulfate aerosols. These form haze and rain that is acidic which harms our health, changes the atmospheric environment, and damages ecological systems (Hadei *et al.*, 2017; Li *et al.*, 2018).

Since the beginning of the 21st century, China has seen a rapid increase in industrialization and urbanization. Thus, a huge amount of energy is being consumed each year. Subsequently, about 1×10^9 tons of fossil fuel are being used in industrial production so that large amounts of SO₂ are being emitted due to the burning of coal and metal smelting (Song and Yang, 2014). Emission levels obtained using a 'bottom-up' (a method to measure emission levels of

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different regions in the increasing order of their administrative levels) approach demonstrate that SO₂ emission in China has increased from 2.17×10^7 tons in 2000 to 3.32×10^7 tons in 2006 (an increase of 53%). In the same period, the amount of SO₂ emitted generating thermal power rose from 1.06×10^7 to 1.86×10^7 tons. Emission rose by 85% in north China, but only 28% in the south (Lu *et al.*, 2010).

Guangdong (in South China) is a frontier province with a large economy brought about by the reform and opening up of China. It must be noted that Guangdong province includes 21 prefecture-level cities. Nine of these (Guangzhou, Foshan, Shenzhen, Dongguan, Zhongshan, Jiangmen, Huizhou, Zhaoqing, and Zhuhai) are located in the Pearl River Delta (PRD) which is a highly urbanized region. The amount of SO₂ emitted in Guangdong province rose from 9.05×10^5 tons in 2000 to 1.29×10^6 tons in 2005. The corresponding increase in average annual SO₂ concentration, as monitored by ground monitoring stations, was from 20 to $27 \mu\text{g m}^{-3}$ (Wang *et al.*, 2012). These significant quantities of SO₂ led to the regional atmosphere becoming seriously polluted (Wang *et al.*, 2017; Ni *et al.*, 2018) and deterioration of the air quality in urban areas (Lu *et al.*, 2016; Liu and Wang, 2017; Yang *et al.*, 2017; Itahashi *et al.*, 2019).

In order to control air pollution and reduce emission of SO₂, the Chinese Government has implemented various emission reduction policies since 2005, e.g., introducing flue gas desulfurization (FGD) and shutting down small generator units. As a result, the proportion of thermal power plants capable of FGD increased from 12% in 2005 to 82.6% in 2010 (Zhao *et al.*, 2013; Liu *et al.*, 2019). Because of the government's continued implementation of emission reduction policies, SO₂ emission has been much more effectively controlled in China.

In particular, significant emission reduction has been achieved in the PRD region. In 2005, the rate of growth of SO₂ emission slowed down to an obvious extent and emission in 2006 began to decrease after reaching a peak (Lu *et al.*, 2010). Furthermore, SO₂ emission from thermal power generation fell by 14.3% from 2005 to 2010 (Zhao *et al.*, 2013) and the average annual SO₂ concentration, as monitored on the ground, decreased from $83 \mu\text{g m}^{-3}$ in 1996 to $21 \mu\text{g m}^{-3}$ in 2015 (Zhao *et al.*, 2018). An inventory of atmospheric pollution emission in the PRD region demonstrates that SO₂ emission has fallen by 51% from 2006 (1.12×10^6 tons) to 2012 (5.52×10^5 tons) (Yang *et al.*, 2015; Zheng *et al.*, 2009). At the same time, air quality monitoring in the PRD region, Hong Kong, and Macau suggests that the average annual SO₂ concentration declined by 61.7% (from about 47 to $18 \mu\text{g m}^{-3}$).

The above mentioned research on the variation and assessment of SO₂ emission reduction in the last 20 years has been mainly based on 'bottom-up' inventories, that is, the emissions are calculated according to weighting factors related to spatial distributions and monitoring data from traditional (discrete) ground stations. Using such emission inventories, one can only acquire data for discrete grids which leads to a large degree of uncertainty. Because of the rapid rate of industrialization and urbanization, and the introduction of new emission reduction policies, emission

inventories will soon be inconsistent with actual emissions and will show obvious time lags (Zheng *et al.*, 2009; Wang *et al.*, 2016; Xu *et al.*, 2017). In addition, as these discrete ground monitoring stations are rather sparse and cover a limited amount of space, they are not able to characterize spatial distributions very well (Lee *et al.*, 2011; Sindhwani *et al.*, 2015; Cheng *et al.*, 2018).

Remote sensing satellites are an emerging means of detecting air quality and can be used to complement conventional techniques such as ground monitoring, airborne observation, and bottom-up collection of the emission inventories. Moreover, they have some unique advantages, such as wide covering range, long term sequencing, and up-bottom monitoring. Over the past 20 years, a new generation of spaceborne hyperspectral remote sensors have been developed. Indeed, numerous spaceborne sensors, including the Global Ozone Monitoring Experiment (GOME) and GOME-2, Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) (Khattak *et al.*, 2014), Ozone Monitoring Instrument (OMI) (Khokhar *et al.*, 2016), and Ozone Mapping and Profiler Suite (OMPS) have been launched. Of particular interest among these is the OMI carried by the Aura satellite launched by the National Aeronautics and Space Administration (NASA) of the United States.

OMI has a high spatial and spectral resolution and can be used to remotely monitor various pollutants, including SO₂. Its early applications included monitoring volcanic eruptions around the world (Krueger *et al.*, 2009; Vicente *et al.*, 2009), gas emission from large-scale thermal power plants (Zhang *et al.*, 2009; Li *et al.*, 2010), and analyzing the temporal and spatial variation of high-concentration pollutants over large regions (Krotkov *et al.*, 2008; Zhang *et al.*, 2010). The success of these applications proves that OMI, with its high detection sensitivity and accuracy, is not only capable of detecting volcanic eruptions, but also detecting anthropogenic SO₂ emission on a large scale. Furthermore, as the transit time of OMI has lengthened, its function has been extended to long-term monitoring and evaluation of SO₂ emission from large smelting plants and coal-fired power plants (Carn *et al.*, 2007). Thus, it has been able to verify that the deployment of flue gas desulfurization (FGD) devices has had a significant effect on the control and reduction of SO₂ emission (Li *et al.*, 2010). In more recent years, scholars around the world have used low-resolution SO₂ data acquired using OMI to study the spatiotemporal distribution characteristics of SO₂ in provincial and municipal regions (Zhao *et al.*, 2011; Li *et al.*, 2016), estimate dry deposition (Du *et al.*, 2015), derive emission inventories (Kourtidis *et al.*, 2018), and evaluate the effect of emission reduction (Song and Yang, 2015).

In this study, we increase the spatial resolution of the OMI data grids from $0.25^\circ \times 0.25^\circ$ ($28 \text{ km} \times 28 \text{ km}$) to $2 \text{ km} \times 2 \text{ km}$ by employing the latest sub-pixel interpolation method. After validating the results in comparison to ground and emission inventory data, we first systematically study the characteristics of the new and more detailed spatiotemporal distribution of SO₂ after it reached its peak

surface concentration in 2006. We then evaluate and compare the contributions made by prefecture-level cities to emission reduction in the PRD region. The research aims to provide rigorous scientific support for the emission reduction policies implemented as part of the national ‘13th Five-Year Plan’ to prevent and control atmospheric pollution and thus contribute to China’s ‘Blue Sky Protection Campaign’.

MATERIALS AND METHODS

Satellite Data and Quality Control

OMI was jointly built by the Netherlands and Finland and is one of four sensors carried by the Aura earth observation satellite system launched by NASA on July 15, 2004. It can be used to monitor O₃, aerosols, cloud coverage, and levels of trace gases, like SO₂ and NO₂, in the atmosphere (Levelt *et al.*, 2006). As it is a hyperspectral sensor, it can scan 2,600 km of its orbit at ultraviolet, visible, and near-infrared wavebands, and it can make observations at spatial resolutions from 13×24 to 28×150 km². The data it produces is widely employed in research aimed at monitoring and warning of impending volcanic eruption, and for monitoring, evaluating, and forecasting levels of urban pollution.

Principal component analysis (PCA) is the latest business-standard algorithm applied to determining SO₂ levels in OMI’s boundary layer data (Li *et al.*, 2013). The inversion algorithm selects hyperspectral data from 310.5 to 340 nm and extracts the principal components relating to O₃ absorption, surface reflectivity, instrument noise, and ring effects. These are used to establish a relational expression for Jacobi matrix inversion and thus directly acquire the total vertical column amount of atmospheric SO₂. The PCA algorithm reduces the OMI SO₂ inversion error and improves the product quality and application range of the data. Thus, inversion products are more suitable for monitoring anthropogenic SO₂ pollution. Therefore, in this study, we adopt the SO₂_PBL (planetary boundary layer) data of V003 OMI L2 orbit products covering 2005 to 2016 (<https://daac.gsfc.nasa.gov/>).

In order to obtain the SO₂ distribution close to the ground and at high resolution, it is essential to improve the accuracy of the SO₂ data. Thus, the pixel data was strictly screened according to the following quality requirements: (1) Since 2007, 60 of OMI’s CCD sensors have been successively influenced by scattered light and blocking of their visual fields, thus producing row anomalies (RA). Therefore, the XTrackQualityFlags field is used to eliminate abnormal values. (2) In order to eliminate large errors caused by large observation angles of the pixels on the edge of the scanning orbit, the data corresponding to the first and last groups of ten pixels are eliminated. (3) The solar zenith angle is controlled to be in the range of 0–70°C. To eliminate the effect of cloud, the equivalent radiation cloud cover needs to be smaller than 0.2. To eliminate the effect of volcanic eruption, the total amount of SO₂ needs to be less than 10 DU. (4) The standard deviation of the noise in the total PBL SO₂ column data from the OMI is 1.5 DU. However,

long-term data sequences (e.g., three years) can be treated using statistical methods to reduce the standard deviation to below 0.3 DU (Krotkov *et al.*, 2008).

Ground Observation Data

The SO₂ concentrations recorded at 88 ground monitoring stations in the PRD were obtained from the Environmental Monitoring Center in Guangdong province, China. The observation and quality control of the data was performed according to the relevant national observation standards for atmospheric pollutants.

The inventory of SO₂ emission sources in the PRD region in 2008 with the resolution of $0.25^\circ \times 0.25^\circ$ was obtained from the Multi-resolution Emission Inventory (MEIC) website (<http://www.meicmodel.org>) established by Tsinghua University in China. The inventory of emission sources in the PRD in 2012 with the resolution of $3 \text{ km} \times 3 \text{ km}$ refers to information presented in the literature (Yang *et al.*, 2015).

Signal Enhancement Technology

One pixel of the OMI ranges from 13×24 to 28×150 km² from nadir to edge. Thus, a great deal of SO₂ emission information from ground pollution point sources is generally lost when OMI is directly used for ground monitoring.

To overcome this, Fioletov *et al.* (2011) utilized a statistical calculation method (sub-pixel interpolation) to enhance signals. The spatial resolution thus presented corresponded to a fine grid of points with each grid unit measuring $2 \text{ km} \times 2 \text{ km}$. High- and low-spatial frequency filtering was also applied to the OMI SO₂ L2 orbital data in long-term sequences, thereby reducing the noise in the signals and standard deviation of the data. Thus, weak SO₂ signals could be enhanced and displayed, something that could not be achieved using ‘ordinary’ processing methods. Overall, they were able to obtain the spatial distribution of SO₂ in eastern parts of the USA at sub-pixel levels ($2 \text{ km} \times 2 \text{ km}$). Based on a statistical analysis of the data of Fioletov *et al.* (2011) the total amount of SO₂ in sub-pixel columns was shown to be significantly statistically correlated with independent point sources with annual SO₂ emission levels of 60 kt or more (or multiple point sources within a range of 50 km). Correlation coefficients of up to 0.93 were obtained. The sub-pixel OMI SO₂ data further revealed that the SO₂ emission from the largest coal-fired power plants in the USA had fallen by 40% from 2005–2007 to 2008–2010. This is highly consistent with the fact that SO₂ levels decreased by 46% after implementing monitoring measures for pollution according to reports (<http://camddata.andmaps.epa.gov/gdm/index.cfm?fuseaction=emissions.wizard>).

Fioletov *et al.* (2011) actually selected two coal-fired power plants as their research objects. One, a thermal power plant in Georgia, corresponded to the largest source of SO₂ emission (170 kt annually). The other, in Belews Creek, North Carolina, corresponded to the 20th largest emission source (88 kt annually). Based on the statistics of the OMI SO₂ data from 2005 to 2007, the distance from the center of the OMI detection pixel to the position of the thermal power plants was used as a distance function. The

results showed that the total column amount of SO₂ followed Gaussian (normal) distributions with respect to distance from the thermal power plants. Thus, high SO₂ concentrations were observed adjacent to the thermal power plants and the further the distance from the plant, the smaller the total column amount of SO₂. At distances over 50 km, the total column amount of SO₂ tended to zero.

According to the abovementioned conclusions, it can be seen that if one attempts to detect point sources of SO₂ emissions using OMI then only 1–2 pixels will produce signals (pixel ranges from 13×24 to 28×150 km²). This is bound to result in many point sources of emission not being detected by the satellite. At the same time, the conclusions also indicate that statistically significant averages can be obtained if a large amount of independent pixel data is used that cover a range of a few kilometers adjacent to the point source. According to these statistics, using average values of OMI data taken over three years near point sources with annual emissions of 70 kt allows statistically significant results to be obtained at the 95% confidence level.

Guangdong province (20°E–26°N, 109°E–118°E) was selected as research region and, 333×500 grid points were calculated using the MATLAB software package (which has a function that yields the spherical distances between given points of latitude/longitude). As shown in Fig. 1, the dimension of each grid unit is 2 km $\times$ 2 km and the black grid points are the target objects for value assignment. Taking each of these target points as center, a circle of radius 12 km is also drawn in this diagram within which the OMI SO₂ orbital pixel data was obtained corresponding to a continuous period of three years. All data in the 12 km range from the center of the pixel to the center of the target grid point (after undergoing quality control) were selected and then mean values were obtained and assigned to the target grid points. According to the observational statistics that the signal to point-source distance obeys a Gaussian (normal) distribution, the data for the 2 km $\times$ 2 km grid points display the detailed characteristics of the sub-pixel spatial distribution. In the figure, the dimension of minimum pixel corresponding to OMI's nadir is used as a reference.

Fig. 1. Schematic diagram illustrating the application of the sub-pixel algorithm.

The range of the statistical radius determines the degree of smoothness of the data: the larger the radius, the lower the noise, i.e., the smoother the data (however, the spatial resolution is also correspondingly lower). After many calculation experiments, the radius of 12km is chosen as the best choice.

RESULTS AND DISCUSSION

Cross-validation with Ground Monitoring Stations in the PRD Region

The average annual SO₂ concentrations, as recorded by the ground monitoring stations in the PRD region, were compared to the corresponding average values of the total column amounts of SO₂ obtained using the sub-pixel method applied to the remote sensing data. As the principles and measurement units of the remote sensors are different from those used in the ground observations, the two sets of data were first normalized before making the comparison (i.e., both sets of data were assigned the reference value of 1.00 in the year 2006).

Fig. 2 shows the yearly change in the normalized average amounts of SO₂ determined according to the two methods. The two sets of SO₂ data are clearly highly correlated during the period 2006–2015. The two sets of data give a good fit to a linear function giving a slope of 0.97 and a correlation coefficient of 0.95. Both sets of data consistently show the same decreasing trend in SO₂ concentration over the years shown. The data points show approximately linear reductions in the average annual amounts of SO₂ corresponding to -0.063 (sub-pixel method) and -0.067 (ground monitoring method). That is, the SO₂ levels in the PRD region have decreased at an annual rate of 6–7% per year. By comparing the average annual SO₂ emissions in the PRD in 2015 and 2006, the sub-pixel method suggests the average decrease in SO₂ corresponded to 57% over this period, while that obtained using the ground monitoring data corresponds to 66% (a difference of 9%).

Cross-correlation of Regional Spatial Distributions and Detailed Characteristics

The high resolution (2 km $\times$ 2 km) SO₂ data obtained using the sub-pixel method can be used to derive spatial and regional SO₂ distributions for Guangdong province in a clearer and more detailed manner. In order to objectively cross-validate the spatial distributions and detailed characteristics of the SO₂ levels obtained via the sub-pixel method, a comparison was made of distributions generated using ground monitoring data, OMI SO₂ L2 data, and national and regional inventory data. Fig. 3 shows spatial SO₂ distributions for the whole of Guangdong province and PRD region according to the data from the different sources.

Figs. 3(a), 3(b), and 3(d) present the spatial SO₂ distributions for Guangdong province in 2008 generated using data from ground monitoring stations via nearest-neighbor interpolation, the official $0.25^\circ \times 0.25^\circ$ grid data from the OMI, and the sub-pixel method, respectively. Fig. 3(c) displays the results obtained using the national

Fig. 2. Normalized plots of the average annual SO₂ emissions in the PRD region obtained using the sub-pixel method and data from the ground monitoring stations from 2006 to 2015. The good correlation between the two sets of data is further illustrated via a linear regression plot.

Fig. 3. Spatial distribution of SO₂ pollution derived using data from different sources: ground monitoring, emission inventories, and remote sensing satellite. (a)–(d) present results for the whole of Guangdong province (2008 data), and (e) and (f) just those for the PRD region (2012 data).

inventory of SO₂ emission sources (2008) as estimated by Tsinghua University based on emission factors using different technologies, see <http://www.meicmodel.org>). Figs. 3(e) and 3(f) presents results for the PRD region only. The former was obtained by Yang *et al.* (2015) who used basic activity data on anthropogenic and natural sources in the atmosphere to establish a spatial allocation scheme for local pollutants and a time-slot allocation scheme to characterize industrial pollution discharge and thus generate a map of the regional SO₂ emissions in the PRD in 2012. In contrast, Fig. 3(f) presents the SO₂ distribution in 2012 as obtained using the sub-pixel technology.

Fig. 3(a) demonstrates that the uneven distribution of state-controlled ground-based monitoring stations means that the spatial distribution generated via interpolation is not particularly objective and does not give a good representation of the spatial distribution. For example, compared to the situation in Guangzhou and Foshan, there are five state-controlled monitoring stations in Shaoguan city that are too highly concentrated (red circle in Fig. 3(a)). The observed SO₂ values here are large (0.047–0.068 mg m⁻³), so that the overall regional SO₂ concentration in Shaoguan is depicted as being higher than that in Guangzhou and Foshan.

Due to the coarseness of the spatial grid in the official OMI product, Fig. 3(b) only displays the highs or lows of the regional pollution distributions (The largest value, 1.7 DU). The results obtained via sub-pixel interpolation in Fig. 3(d), however, have much higher spatial resolution and yield a larger extreme value (2.6 DU). Thus, they give a clearer and more detailed representation of the distribution characteristics and cover a larger range of extreme values. The emission inventory of atmospheric pollution distribution (Fig. 3(c)) reflects the structure of the atmospheric pollution emission over a range of various geographic regions and administrative boundaries (e.g., regions, cities, districts, and counties) and has spatiotemporal properties. The results obtained via sub-pixel interpolation were compared with the national and regional emission source inventories which revealed the similarities and differences in their spatial distributions and detailed characteristics. The comparison shows, for example, that the regional and spatial distribution of SO₂ in 2008 in Guangdong province are highly similar and in good agreement (the white circles in Figs. 3(c) and 3(d) highlight the areas in which the general degree of coincidence is particularly high). However, as far as detailed characteristics are concerned, the sub-pixel method is clearly superior to the national inventory method. These two methods both show that the spatial and regional distribution of SO₂ is dominated by the presence of one major center and three smaller regions in Guangdong province. But the national inventory data can only reveal the spatial and regional distributions – it cannot display the detailed characteristics of the distribution. Furthermore, from the perspective of time, the national inventory data for Guangdong province suggests the emission rates in 2012 and 2008 were 2.96×10^7 and 3.13×10^7 ton y⁻¹, respectively, corresponding to a reduction of 5.4%. This is obviously smaller than the 43% reduction suggested by the

sub-pixel method and 38% reduction suggested by the ground monitoring data. Thus, our preliminary analysis demonstrates that the national inventory does not properly update the SO₂ data in Guangdong province and so it does not objectively represent the true picture in the province.

Figs. 3(e) and 3(f) show a comparison of the results obtained for the PRD region using the sub-pixel interpolation and regional inventory methods. The two distributions match spatially to a high degree, both essentially highlighting various point sources, e.g., boilers, process sources, power plants, non-road sources. Moreover, the detailed characteristics of the distributions are obviously similar while they also show small differences. Both of them imply that the SO₂ distribution in the PRD region had transformed by 2012 into a more decentralized affair with several small pollution regions (compare this with the earlier 2008 distribution wherein the pollution was concentrated into one big region).

The detailed distribution characteristics shown Figs. 3(e) and 3(f) indicate that the polluted regions fall into two general classes: agglomeration areas consisting of urban residential areas and factories, and areas associated with regional administrative boundaries. Comparing the two spatial distributions, the polluted regions in the regional inventory (Fig. 3(e)) in the center and west of Guangzhou and east of Foshan correspond to those in the center and west of Foshan and north of Zhaoqing in the figure generated using the sub-pixel method (Fig. 3(f)), although their positions are slightly shifted to the east or west. In addition, the pollution sources in Shenzhen in Fig. 3(e) appear to have no corresponding sources in Fig. 3(f). The reasons for these differences in correspondence need to be further investigated, but this is not attempted here. However, it is clear that, on the whole, the SO₂ information obtained using the sub-pixel method gives a good representation of the characteristics of the regional and detailed pollution distributions.

Spatiotemporal Variation of SO₂ Concentration in the PRD Region

The sub-pixel method was used to generate spatial distributions for the SO₂ concentration in Guangdong province for four years in the period 2006–2016 (Fig. 4). In these diagrams, the same scale is used to specify the amount of SO₂ (0–3 DU) so that the diagrams are directly comparable and highlight the temporal and spatial changes in SO₂ occurring in the province during this period.

The figures clearly show that the PRD region was a high-concentration pollution zone in 2006 (the largest value, 3.1 DU). The three other regions (western, northern, and eastern Guangdong) corresponded to decentralized pollution regions at this time wherein the SO₂ concentrations were low. As Guangzhou was chosen to host the Asian Games in 2010, the PRD region began to trial a combination of pollution-controlling measures (relocation, desulfurization, and rectification of polluting enterprises) in 2009.

The subsequent reduction in emissions gradually began to change the characteristics of the SO₂ distribution in the PRD region. As demonstrated in Fig. 4, the pollution intensity in

Fig. 4. The spatial distributions of SO_2 in Guangdong province in 2006, 2009, 2013, and 2016, as obtained using the sub-pixel method.

the PRD region in 2009 had decreased significantly (the largest value, 2.2 DU) and the polluted region had become more dispersed. By 2016, the highly-polluted region of the PRD had disappeared from the Guangdong province altogether (the largest value, 1.1 DU). As a result, all four regions of Guangdong province (PRD region together with the northern, eastern, and western regions) could finally be classified as small decentralized pollution regions with low pollution concentrations.

As the most significant changes in Guangdong occur in the PRD region, it is worth examining this region in more detail. Fig. 5 shows the results obtained using the sub-pixel method for the PRD region for the same years shown in Fig. 4. However, this time the color scale used to represent SO_2 concentration is not fixed. Instead, the scale maximum employed is reduced in later years to highlight the areas of pollution that remain.

As found before, a large area of the PRD region can be seen to suffer from a high concentration of SO_2 pollution in 2006. Two circles of different color appear in the Fig. 5: one corresponds to the high-concentration of SO_2 accumulated from Guangzhou and Foshan (primary region with red circle); the other to the high-pollution zone to the west of Dongguan, Zhongshan, north of Jiangmen, and south of Zhaoqing (secondary region with white circle). By 2009, the adjustments made to the industrial structure and implementation of other pollution control and prevention measures led to a significant decrease in the intensity of pollution in the primary and secondary regions. By 2013, the regional distribution characteristics had essentially changed from its original type (concentrated in one mass) into one that was much more decentralized. The pollution distribution characterized by the primary and secondary regions had transited into a set of regional pollution

distributions along the administrative boundaries between Guangzhou and Dongguan, Foshan and Zhaoqing, as well as Foshan and Zhaoqing, together with decentralized ‘dots’ and zonal distributions along the other administrative boundaries. Moving next to 2016, it is apparent that the intensity of the pollution in the PRD region had changed slowly.

Extent of Emission Reduction in Nine Prefecture-level Cities in the PRD Region

In this section, we examine the long-term change in SO_2 pollution levels in nine prefecture-level cities in the PRD region. High-resolution SO_2 data was obtained for these cities using the sub-pixel method and three-year moving averages calculated. The resulting SO_2 levels are presented in Fig. 6 for the nine prefecture-level cities chosen.

Fig. 6(a) shows that the SO_2 pollution levels in each of these cities fell from 2006 to 2016. Moreover, the basic trends in each of the curves shown are the same. That is, the extent of emission reduction was generally consistent in these cities and the SO_2 levels in these cities essentially decreased year-on-year at the same rate. However, based on the curves shown in Fig. 6(a), 2011 appears to have been a transition year: from 2006 to 2011, the SO_2 level decreased significantly; thereafter, it dropped much more gently.

The differences between these prefecture-level cities, in terms of SO_2 , are clearly significant in 2006. The highest ranked city at this time was Foshan (with a total SO_2 concentration of 2.2 DU), followed by Zhongshan (2.1 DU), and Guangzhou (1.5 DU). At the other end of the scale, Huizhou (0.8 DU) was the least polluted city, followed by Zhaoqing (1.0 DU). The total SO_2 concentration in Foshan was therefore almost three times that found in Huizhou. In

Fig. 5. The spatial distributions of SO₂ in the PRD region in 2006, 2009, 2013, and 2016, as obtained using the sub-pixel method.

Fig. 6. (a) The changes in SO₂ concentration in nine prefecture-level cities in the PRD from 2006 to 2016, (b) Pie chart showing the relative contributions made by individual cities to SO₂ emissions in 2006, and (c) Pie chart showing the corresponding contributions in 2016.

contrast, in 2016, after emission reduction measures had been in place for 11 years, the differences between the SO₂ densities in the prefecture-level cities had become much smaller (to the extent that they all appear to be very similar).

The data can also be used to quantify the extent to which the emission levels have been reduced in particular cities. Thus, in Zhongshan, Foshan, and Guangzhou the SO₂ levels fell significantly in the period 2006–2016: by 71%, 65%, and 57%, respectively. The corresponding figures for Zhaoqing and Huizhou are less impressive: 37% and 25%, respectively. Therefore, the emission reduction measures implemented have had the largest effect on Guangzhou, Foshan, and Zhongshan. Zhaoqing and Huizhou, however, have seen much more modest improvements.

Fig. 6(b) shows the relative contributions made to the SO₂ emissions by the nine prefecture-level cities in the PRD region in 2006. (Here, ‘relative contribution’ means the contribution made by the city compared to the total contribution made by all nine cities – hence the contributions sum to 100%). The figure shows that Foshan (17%), Zhongshan (16%), and Guangzhou (12%) made the largest contributions in 2006, while Huizhou (6%), Zhaoqing (7%), and Shenzhen (9%) made the smallest contributions. Fig. 6(c) shows the corresponding situation 11 years later (2016). This time the largest contributions are made by Foshan (13%), and Guangzhou and Dongguan (each 12%) – the smallest contributions are made by Zhongshan, Huizhou, and Shenzhen (each 10%).

Figs. 6(b) and 6(c) show that the contributions made by two cities decreased significantly in response to 11 years of emission-reduction measures. Thus, Zhongshan saw its contribution fall from 16% to 10%, while that in Foshan dropped from 17% to 13%. (However, this means that other contributions increased. Thus, the contribution of Zhaoqing rose from 7% to 11% and that of Huizhou increased from 6% to 10%. This is because the emission reduction amplitudes in these two cities were relatively small, as discussed earlier.)

CONCLUSIONS

Using sub-pixel interpolation to enhance the signals obtained from a remote sensing satellite (OMI), the spatial resolutions of the grids used to specify the distribution of SO₂ were increased from $0.25^\circ \times 0.25^\circ$ to $2 \text{ km} \times 2 \text{ km}$. The main conclusions are as follows:

- (1) A linear fit between data from the sub-pixel method and ground monitoring stations in the PRD region yielded a correlation coefficient of 0.95 and a slope of 0.97. The two sets of data only has a difference of 9% from 2015 and 2006.
- (2) Cross-correlation with data from ground monitoring stations, emission inventories and OMI SO₂ L2 products shows that the values obtained using the sub-pixel method gives a clearly superior representation of the characteristics of the regional and detailed pollution distributions.
- (3) The temporal and spatial variation of the SO₂ distribution in Guangdong province was successfully

obtained. In 2006, there was clearly a highly-concentrated region of pollution centered on the PRD region and there were other small decentralized pollution regions of low concentration elsewhere (western, eastern, and northern Guangdong province). By 2016, the whole of Guangdong, consisted of four decentralized pollution regions of low concentration at this stage.

- (4) A more detailed examination of the SO₂ distribution in the PRD region demonstrated that there were a primary circle of high-concentration and a zonal secondary circle of pollution in 2006. By 2013, the original pattern had transformed into dots and zonal distributions characterized by low pollution levels located near the administrative boundaries.
- (5) After 11 years of subsequent emission reduction, the SO₂ levels in Zhongshan, Foshan, and Guangzhou had been significantly reduced (by 71%, 65%, and 57%, respectively). Meanwhile, the reduction in Zhaoqing and Huizhou was much less (by 37% and 25%, respectively). In addition, SO₂ two of the prefecture-level cities saw their contributions decrease significantly: Foshan saw its contribution drop from 17% to 13% and Zhongshan’s dropped from 16% to 10%. However, two other prefecture-level cities saw their contributions increase: Zhaoqing from 7% to 11%, and Huizhou from 6% to 10%.

The reduction in SO₂ emission in the prefecture-level cities, and the contributions they made to pollution in the PRD region, indicate that the government’s efforts to improve the air quality in the region have been very effective and highly significant changes have been accomplished (The State Council, 2013).

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