OMI measured increasing SO$_2$ emissions due to energy industry expansion and relocation in Northwestern China

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Abstract

The rapid economy growth makes China the largest energy consumer and sulphur dioxide (SO$_2$) emitter in the world. In this study, we estimated the trends and step changes in the planetary boundary layer (PBL) vertical column density (VCD) of SO$_2$ from 2005 to 2015 over China measured by the Ozone Monitoring Instrument (OMI). We show that these trends and step change years coincide with the effective date and period of the national strategy for energy development and relocation in northwestern China and the regulations in the reduction of SO$_2$ emissions. Under the national regulations in the reduction SO$_2$ emissions in eastern and southern China, SO$_2$ VCD in the Pearl River Delta (PRD) of southern China exhibited the largest decline during 2005-2015 at a rate of -7% yr$^{-1}$, followed by the North China Plain (NCP) (-6.7% yr$^{-1}$), Sichuan Basin (-6.3% yr$^{-1}$), and Yangtze River Delta (YRD) (-6% yr$^{-1}$), respectively. The Mann–Kendall (MK) test reveals the step change points of declining SO$_2$ VCD in 2009 for the PRD and 2012-2013 for eastern China responding to the implementation of SO$_2$ control regulation in these regions. In contrast, the MK test and regression analysis also revealed increasing trends of SO$_2$ VCD in northwestern China, particularly for several "hot spots" featured by growing SO$_2$ VCD in those large-scale energy industry parks in northwestern China. The enhanced SO$_2$ VCD is potentially attributable to increasing SO$_2$ emissions due to the development of large-scale energy industry bases in energy-abundant northwestern China under the national strategy for the energy safety of China in the 21st century. We show that these large-scale energy industry bases could overwhelm the trends and changes in provincial total SO$_2$
emissions in northwestern China and contributed increasingly to the national total SO$_2$ emission in China. Given that northwestern China is more ecologically fragile and uniquely susceptible to atmospheric pollution as compared with the rest of China, increasing SO$_2$ emissions in this part of China should not be overlooked and merit scientific research.

1. Introduction

Sulfur dioxide (SO$_2$) is one of the criteria air pollutants emitted from both anthropogenic and natural sources. The combustions of sulfur-containing fuels, such as coal and oil, are the primary anthropogenic emitters, which contributed to the half of total SO$_2$ emissions (Smith et al., 2011; Lu et al., 2010; Stevenson et al., 2003; Whelpdale et al., 1996). With the rapid economic growth in the past decades, China has become the world’s largest energy consumer accounting for 23% of global energy consumption in 2015 (BIEE, 2016). Coal has been a dominating energy source in China and accounted for 70% of total energy consumption in 2010 (Kanada et al., 2013). The huge demand to coal and its high sulfur content make China the largest SO$_2$ emission source in the world (Krotkov et al., 2016; Su et al., 2011), which also accounted for two-third of Asia’s total SO$_2$ emission (Ohara et al., 2007). From 2000 to 2006, the total SO$_2$ emission in China increased by 53% at an annual growth rate of 7.3% (Lu et al., 2010). To reduce SO$_2$ emission, from 2005 onward the Chinese government has issued and implemented a series of regulations, strategies, and SO$_2$ control measures, leading to a drastic decrease of SO$_2$ emission, particularly in eastern
and southern China (Lu et al., 2011; Li et al., 2010).

Recently, two research groups led by NASA (National Aeronautics and Space Administration) and Lanzhou University of China published almost simultaneously the temporal and spatial trends of SO$_2$ in China from 2005 to 2015 using the OMI retrieved SO$_2$ PBL column density after the OMI is launched for 11 years (Krotkov et al., 2016; Shen et al., 2016). The results reported by the two groups revealed widespread decline of SO$_2$ in eastern China for the past decade. Shen et al noticed, however, that in contrast to dramatic decreasing SO$_2$ emissions in densely populated and industrialized eastern and southern China, the OMI measured SO$_2$ in northwestern China appeared not showing a decreasing trend. This is likely resulted from the energy industry relocation and development in energy-abundant northwestern China in the past decades under the national strategy for China's energy development and safety during the 21st century. Concern is raised for the potential impact of SO$_2$ emissions on the ecological environment and health risk in northwestern China because high SO$_2$ emissions could otherwise damage the rigorous ecological environment in this part of China, featured by very low precipitation and sparse vegetation coverage which reduce considerably the atmospheric removal of air pollutants (Ma and Xu, 2017).

To assess and evaluate the risks of the ecological environment and public to the growing SO$_2$ emissions in northwestern China, it is necessary to investigate the spatiotemporal distributions of SO$_2$ concentrations and emissions. However, the ground measurements of ambient SO$_2$ are scarce temporally and spatially in China,
and often subject to large errors and uncertainties. Owing to the rapid progresses in the remote sensing techniques, satellite retrieval of air pollutants has become a powerful tool in the assessment of emissions and spatiotemporal distributions of air pollutants. In recent several years, OMI (Dutch Space, Leiden, The Netherlands, embedded on Aura satellite) retrieved SO$_2$ column concentrations have been increasingly applied to elucidate the spatiotemporal variation of global and regional SO$_2$ levels and its emissions from large point sources, and evaluate the effectiveness of SO$_2$ control policies and measures (Krotkov et al., 2016; McLinden et al., 2015, 2016; Ialongo et al., 2015; Fioletov et al., 2015, 2016; Wang et al., 2015; Li et al., 2010). The decadal operation of the OMI provides the relatively long-term SO$_2$ time series data with high spatial resolution which are particular useful for assessing the changes and trends in SO$_2$ emissions induced by national regulations and strategies. The present study aims to (1) assess the spatiotemporal variations of SO$_2$ and its trend under the national strategy for energy industry development in northwestern China by making use of the OMI-measured SO$_2$ data during 2005-2015; (2) to further examine the usefulness of the satellite remote sensing of air quality.

2 Data and methods

2.1 Satellite data

We collected the level 3 OMI daily planetary boundary layer (PBL) SO$_2$ vertical column density (VCD) data in Dobson units (1 DU=2.69×10$^{16}$ molecules cm$^{-2}$) produced by the principal component analysis (PCA) algorithm (Li et al., 2013). The
spatial resolution is 0.25° × 0.25° latitude/longitude, available at Goddard Earth Sciences Data and Information Services Center (http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omso2_v003.shtml). This algorithm yields one-step SO₂ VCD. However, as Fioletov et al. (2016) noted, the PCA retrieved SO₂ VCD was virtually derived by adoption of an effective air mass factor (AMF) of 0.36 which is best applicable in the summertime in the eastern United States (US). The algorithm may cause systematic errors if anthropogenic emission sources are located in different latitudes and under complex topographic and underlying surface conditions. For instance, Wang (2014) has shown that AMF ≈ 0.57 in eastern China. In the present study, we have adopted the AMFs values in China provided by Fioletov et al. (2016) to adjust OMI measured VCD in the estimation of the SO₂ emission burden of major point sources in northwestern China.

2.2 SO₂ monitoring, emission, and socioeconomic data

Figure 1 is a China map which highlights 6 provinces in northwestern China, including Shaanxi, Gansu, Qinghai, Ningxia, Xinjiang, and Inner Mongolia. Traditionally, Inner Mongolia is not classified as a northwestern province in China. Given that the most energy resources in Inner Mongolia are located in its western part of this province (Fig. 1), here we include this province in northwestern China. North China Plain (NCP), Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin are also shown in the map. To evaluate and verify the spatial SO₂ VCD from OMI, we collected ground SO₂ monitoring data of 2014 through 2015 at 188 sampling sites (cities) across China (Figure 1), operated by the National Environmental Monitoring Center, available at
http://www.aqistudy.cn/historydata. The statistics between OMI retrieved SO\(_2\) VCD and monitored monthly and annually averaged SO\(_2\) air concentrations during 2014-2015 at 188 operational air quality monitoring stations across China are presented in Table S1 of Supplement. Figure S1 is the correlation diagram between SO\(_2\) VCD and sampled data. As shown in Table S1 and Fig. S1, the OMI measured SO\(_2\) VCDs agree well with the monitored ambient SO\(_2\) concentrations across China at the correlation coefficient of 0.85 (p<0.05) (Table S1). Figure 2 further compared annually averaged SO\(_2\) VCDs and SO\(_2\) air concentrations from 2005 to 2015 in 6 capital cities in Urumqi (Xinjiang), Yinchuan (Ningxia), Beijing (BTH and NCP), Shanghai (YRD), Guangzhou (PRD), and Chongqing (Sichuan Basin), respectively. The mean SO\(_2\) concentration data were collected from provincial environmental bulletin published by the Ministry of Environmental Protection of China (MEPC) (http://www.zhb.gov.cn/hjzl/zghjzkgb/gshjzkgb). Results show that the annual variation of mean SO\(_2\) VCDs match well with the monitored data except for Urumqi, the capital of Xinjiang Uygur Autonomous region. The OMI retrieved SO\(_2\) VCDs in Shanghai and Chongqing are higher than the measured SO\(_2\) concentrations from 2010 to 2015 but the both show consistent temporal fluctuation and trend. The measured SO\(_2\) concentrations peaked in 2013 in Yinchuan whereas the SO\(_2\) VCD reached the peak in 2012 and decreased thereafter. OMI measured SO\(_2\) VCDs in Urumqi show different yearly fluctuations compared with its annual concentrations. The measured SO\(_2\) concentrations in Urumqi decreased from 2011 to 2015 whereas the OMI measured SO\(_2\) VCDs did not illustrate obvious changes. It is not clear the
causes leading to such the inconsistence. Measured concentrations might be subject
to errors or not properly reported. Since the monitored SO₂ concentrations were
collected in the urban area spatially averaged over 8 monitoring sites across the city
whereas the OMI measured SO₂ VCD was averaged over all model grid points
(0.25×0.25 latitude/longitude resolution) in Urumqi city. This could also result in the
inconsistence between SO₂ VCD and measured data. However, such the error
appeared not occurring in other cities.
SO₂ anthropogenic emission inventory in China with a 0.25° longitude by 0.25°
latitude resolution for every two years from 2008 to 2012 was adopted from Multi
resolution Emission Inventory for China (MEIC) (Li et al., 2017, available at
http://www.meicmodel.org). The comparison between annual OMI SO₂ VCD and SO₂
emissions in China is presented in Fig. 3. As shown, the annual variation in SO₂
VCDs also agrees reasonably well with SO₂ emission data except for Midong. The
OMI measured SO₂ VCD in the PRD and Sichuan Basin decreased from 2008 to 2012
but SO₂ emission changed little. Compared with the other five marked regions, the
satellite measured SO₂ VCD in Midong declined in 2010 and inclined in 2012.
However, SO₂ emissions in Midong increased in 2012 at about factor of 11 and 8
higher than that in 2008 and 2010. It should be noted that the MEIC SO₂ emission
inventory from the bottom-up approach might be subject to large uncertainties due to
the lack of sufficient knowledge in human activities and emissions from different
sources (Li et al., 2017; Zhao et al., 2011; Kurokawa et al., 2013). From this
perspective, the satellite remote sensing provides a powerful tool in monitoring SO₂
emissions from large point sources and the verification of emission inventories

(Fioletov et al., 2016; Wang et al., 2015).


### 2.3 Trends and step change

The long-term trends of SO$_2$ VCD were estimated by linear regressions of the gridded annually SO$_2$ VCD against their time sequence of 2005 through 2015. The gridded slopes (trends) of the linear regressions denote the increasing (positive) or decreasing (negative) rates of SO$_2$ VCD (Wang et al., 2016; Huang et al., 2015; Zhang et al., 2015, 2016).

The Mann-Kendall (MK) test was also employed in the assessments of the temporal trend and step change point year of SO$_2$ VCD time series. The MK test is a nonparametric statistical test (Mann, 1945; Kendall, 1975), which is useful for assessing the significance of trends in time series data (Waked et al., 2016; Fathian et al., 2016). The MK test is often used to detect a step change point in the long term trend of a time series dataset (Moraes et al, 1998; Li et al., 2016; Zhao et al., 2016). It is suitable for non-normally distributed data and censored data which are not influenced by abnormal values (Yue and Pilon, 2004; Sharma et al 2016; Yue and...
Wang., 2004; Gao et al. 2016; Zhao et al., 2015). Recently, MK-test has also been used in trend analysis for the time series of atmospheric chemicals, such as persistent organic pollutants, surface ozone (O₃), and non-methane hydrocarbon (Zhao et al., 2015; Assareh et al., 2016; Waked et al., 2016; Sicard et al., 2016). Here the MK test was used to identify the temporal variability and step change point of SO₂ VCD for 2005-2015 which may be associated with the implementation of the national strategy and regulation in energy industry development and emission control during this period of time. Under the null hypothesis (no trend), the test statistic was determined using the following formula:

\[ S_k = \sum_{i=1}^{k} r_i \text{ } (k=2, 3, ..., n) \]  

(1)

where \( S_k \) is a statistic of the MK test, and

\[ r_j = \begin{cases} +1, & x_j > x_i \\ 0, & x_j \leq x_i \end{cases} \text{ } (j=1, 2, ..., i-1) \]  

(2)

where \( x_i \) is the variable in time series \( x_1, x_2, ..., x_n \), \( r_i \) is the cumulative number for \( x_i > x_j \). The test statistic is normally distributed with a mean and variance given by:

\[ E(S_k) = k(k-1)/4 \]  

(3)

\[ Var(S_k) = \frac{k(k-1)(2k+5)}{72} \]  

(4)

From these two equations one can derive a normalized \( S_k \), defined by

\[ UF_k = \frac{S_k - E(S_k)}{\sqrt{Var(S_k)}} \text{ } (k=1, 2, ..., n) \]  

(5)

where \( UF_k \) is the forward sequence, the backward sequence \( UB_k \) is calculated using the same function but with the reverse data series such that \( UB_k = -UF_k \).
In a two-sided trend test, a null hypothesis is accepted at the significance level if 

\[
\left| U_F_k \right| \leq (U_F)_{1-\alpha/2}, \quad \text{where} \quad (U_F)_{1-\alpha/2} \quad \text{is the critical value of the standard normal distribution, with a probability of} \ \alpha. \quad \text{When the null hypothesis is rejected (i.e., when any of the points in} \ U_F_k \ \text{exceeds the confidence interval} \ \pm 1.96; \ P=0.05), \ \text{an significant increasing or decreasing trend is determined.} \ U_F_k > 0 \ \text{often indicates an increasing trend, and vice versa.} \quad \text{The test statistic used in the present study enables us to discriminate the approximate time of trend and step change by locating the intersection of the} \ U_F_k \ \text{and} \ UB_k \ \text{curves.} \quad \text{The intersection occurring within the confidence interval} \ (-1.96, 1.96) \ \text{indicates the beginning of a step change point} \quad (\text{Moraes et al., 1998; Zhang et al., 2011; Zhao et al., 2015}).
\]

3 Results and discussion

3.1. Spatiotemporal variation in OMI measured SO$_2$

Given higher population density and stronger industrial activities, eastern and southern China are traditionally industrialized and heavily contaminated regions by air pollutions and acid rains caused by SO$_2$ emissions. **Figure 4a** shows annually averaged OMI SO$_2$ VCD over China on a 0.25° × 0.25° latitude/longitude resolution averaged from 2005 to 2015. SO$_2$ VCD was higher considerably in eastern and central China, and Sichuan Basin than that in northwestern China. The highest SO$_2$ VCD was found in the NCP, including Beijing-Tianjin-Hebei (BTH), Shandong, and Henan province. The annually averaged SO$_2$ VCD between 2005-2015 in this region reached 1.36 DU. This result is in line with previous satellite remote sensing
retrieved SO$_2$ emissions in eastern China (Krotkov et al 2016; Lu et al., 2010; Bauduin et al., 2016; Jiang et al 2012; Yan et al., 2014). However, in contrast to the spatial distribution of decadal mean SO$_2$ VCD (Fig. 4a), the slopes of the linear regression relationship between annual average OMI-retrieved SO$_2$ VCD and the time sequence from 2005 to 2015 over China show that the negative trends overwhelmed industrialized eastern and southern China, particularly in the NCP, Sichuan Basin, the YRD, and PRD, manifesting significant decline of SO$_2$ emissions in these regions. SO$_2$ VCD in the PRD exhibited the largest decline at a rate of 7% yr$^{-1}$, followed by the NCP (6.7% yr$^{-1}$), Sichuan Basin (6.3% yr$^{-1}$), and the YRD (6% yr$^{-1}$), respectively. Annual average SO$_2$ VCD in the PRD, NCP, Sichuan Basin, and YRD decreased by 52%, 50%, 48%, and 46% in 2015 compared to 2005 (Fig. 5), though the annual fluctuation of SO$_2$ VCD shows rebounds in 2007 and 2011 which are potentially associated with the economic resurgence stimulated by the central government of China (He et al., 2009; Diao et al., 2012). The reduction of SO$_2$ VCD after 2011 in these regions reflects virtually the response of SO$_2$ emissions to the regulations in the reduction of SO$_2$ release, the mandatory application of the flue-gas desulfurization (FGD) on coal-fired power plants and heavy industries, and the slowdown in the growth rate of the Chinese economy (CSC, 2011a; Wang et al., 2015, Chen et al., 2016).

As also shown in Fig. 4b, in contrast to widespread decline of SO$_2$ VCD, there are two "hot spots" featured by moderate increasing trends of SO$_2$ VCD, located in the China's Energy Golden Triangle (EGT, Shen et al., 2016, Ma and Xu, 2017) and
Urumqi-Midong regions in northwestern China. The annual growth rate of SO$_2$ VCD from 2005 to 2015 are 3.4% yr$^{-1}$ in the EGT and 1.8% yr$^{-1}$ in Urumqi-Midong, respectively (Fig. 4b). Further details are presented in Table 1. SO$_2$ VCDs in these two regions peaked in 2011 and 2013 which were 1.6 and 1.7 times of that in 2005 (Fig. 5). The raising SO$_2$ VCDs in the part of the EGT have been reported by Shen et al. (2016). The second hot spot is located in Midong industrial park, about 40 km away from Urumqi, the capital of the Xinjiang Uygur Autonomous Region. The both EGT and Midong industrial parks are featured by extensive coal mining, thermal power generation, coal chemical, and coal liquefaction industries. The reserve of coal, oil and natural gas in the EGT is approximately 1.05×10$^{12}$ ton of standard coal equivalent, accounting for 24% of the national total energy reserve in China (CRGECR, 2015). It has been estimated that there are deposits of 20.86 billion tons of oil, 1.03 billion cubic meters of natural gas, and 2.19 trillion tons of coal in Xinjiang, accounting for 30%, 34% and 40% of the national total (Dou, 2009). Over the past decades, a large number of energy-related industries have been constructed in northwestern China, such as the EGT and Midong chemical industrial parks in order to enhance China’s energy security in the 21st century and speed up local economy. Rapid development of energy and coal chemical industries in Ningxia Hui Autonomous region and Xinjiang of northwestern China alone resulted in the significant demands to coal mining and coal products. The coal consumption, thermal power generation, and the gross industrial output increased by 2.7, 3.5, and 6.6 times in Ningxia from 2005 to 2015, and by 2.7, 4.2 and 6.6 times in Xinjiang.
during the same period (NBSC, 2005, 2015). As a result, SO\textsubscript{2} emissions increased markedly in these regions, as shown by the increasing trends of SO\textsubscript{2} VCD in the EGT and Midong (Fig. 4b). Figure 6 illustrates the fractions of OMI measured annual SO\textsubscript{2} VCD and SO\textsubscript{2} emissions averaged over the 6 provinces of northwestern China in the annual national total VCD (Fig. 6a) and emissions (Fig. 6b) from 2005 to 2015. The both SO\textsubscript{2} VCD and emission fractions in northwestern China in the national total increased over the past decade. By 2015, the mean SO\textsubscript{2} VCD fraction in 6 northwestern provinces has reached 38% in the national total. The mean emission fraction was about 20% in the national total. It should be noted that there were large uncertainties in provincial SO\textsubscript{2} emission data which often underestimated SO\textsubscript{2} emissions from major point sources (Li et al., 2017; Han et al., 2007). In this sense, OMI retrieved SO\textsubscript{2} VCD fraction provides a more reliable estimate to the contribution of SO\textsubscript{2} emission in northwestern China to the national total.

The annual percentage changes in SO\textsubscript{2} VCD from 2005 onward are consistent well with per capita SO\textsubscript{2} emissions in China (Fig. 7). As aforementioned, while the annual total SO\textsubscript{2} emissions in the well developed BTH, YRD, and PRD were higher than that in northwestern provinces, the per capita emissions in all provinces of northwestern China, especially in Ningxia and Xinjiang, were about factors of 1 to 6 higher than that in the BTH, YRD, and PRD, as shown in Fig. 7. In contrast to declining annual emissions from the BTH, YRD, and PRD, the per capita SO\textsubscript{2} emissions in almost all western provinces have been growing from 2005 onward.

3.2 Trend and step changes in OMI measured SO\textsubscript{2} by MK test
Given that in the MK test the signs and fluctuations of $U_{F_k}$ are often used to predict the trend of a time series, this approach is further applied to quantify the trends and step changes in annually SO$_2$ VCD time series in those highlighted regions (a-f) in Fig. 4b from 2005 to 2015. Results are illustrated in Fig. 8. As shown, the forward and backward sequences $U_{F_k}$ and $U_{B_k}$ intersect at least once from 2005 to 2015. These intersections are all well within the confidence levels between -1.96 and 1.96 at the statistical significance $\alpha=0.01$. A common feature of the forward sequence $U_{F_k}$ in eastern and southern China provinces is that $U_{F_k}$ has been declining and become negative from 2007 to 2009 onward (Fig. 8a-d), confirming the downturn of SO$_2$ atmospheric emissions and levels in these industrialized and well developed regions in China. In the EGT and Midong areas of northwestern China (Fig. 4b), however, the $U_{F_k}$ values for SO$_2$ VCD are positive and growing, illustrating clear upward trends of SO$_2$ VCD over these two large-scale energy industry parks, revealing the response of SO$_2$ emissions to the energy industry relocation and development in northwestern China. To guarantee the national energy security and to promote the regional economy, the EGT energy program has been accelerating since 2003 under the national energy development and relocation plan (Zhu and Ruth, 2015; Chen et al., 2016), characterized by the rapid expansion of the Ningdong energy and chemical industrial base (NECIB) which is located about 40 km away from Yinchuan, the capital of Ningxia (Shen et al., 2016). By the end of 2010, a large number of coal chemical industries, including the world largest coal liquefaction and thermal power plants, have been built and operated, and the total installed capacity of thermal power
generating units has reached 1.47 million kilowatts (Zhao, 2016). Under the same national plan, the Midong industrial park in Xinjiang started to construction and operation from the early to mid-2000s which has almost the same industrial structures as those in the EGT, featured by coal-fired power generation, coal chemical industry, and coal liquefaction.

For those regions with declining trends of SO$_2$ VCD, their step change points in the NCP, YRD and Sichuan Basin occurred between 2012 and 2013. These step change points coincide with the implementation of the new Ambient Air Quality Standard in 2012, which set a lower ambient SO$_2$ concentration limit in the air (MEPC, 2012), and the Air Pollution Prevention and Control Action Plan in 2013 by the State Council of China (CSC, 2013a). This Action Plan requests to take immediate actions to control and reduce air pollution in China, including cutting down industrial and mobile emission sources, adjusting industrial and energy structures, and promoting the application of clean energy in the BTH, YRD, PRD and Sichuan Basin. The step change in SO$_2$ VCD over the PRD occurred in the earlier year of 2009-2010 and from this period onward the decline of SO$_2$ VCD speeded up, as shown by the forward sequence $U/k$ which became negative since 2007 and was below the confidence level of -1.96 after 2009, suggesting significant decreasing VCD from 2009 (Fig. 8c). In April 2002, the Hong Kong Special Administrative Region (HKSAR) Government and the Guangdong Provincial Government reached a consensus to reduce, on a best endeavor basis, the anthropogenic emissions of SO$_2$ by 40% in the PRD by 2010, using 1997 as the base year.
By the end of 2010, all thermal power units producing more than 0.125 million kilowatts electricity in the PRD were equipped with the FGD. During the 11th Five-Year Plan (2006-2010), the thermal power units with 1.2 million kilowatts capacity have been shut down. SO$_2$ emission was reduced by 18% in 2010 compared to that in 2005 (NBSC, 2006, 2011). This likely caused the occurrence of the step change in SO$_2$ VCD over 2009-2010.

The statistical significant step change points of SO$_2$ VCD in the EGT and Midong took place in 2006 and 2009, differing from those regions with decreasing trends of SO$_2$ VCD in eastern and southern China. The first step change point in 2006-2007 corresponds to the increasing SO$_2$ emissions in these two large-scale energy bases till their respective peak emissions in EGT (2007) and Midong (2008). The second step change point in 2009 coincides with the global financial crisis in 2008 which slowed down considerably the economic growth in 2009 in China, leading to raw material surplus and the remarkable reduction in the demand to coal products.

3.3 OMI SO$_2$ time series and step change point year in northwestern China

Since almost all large-scale coal chemical, thermal power generation, and coal liquefaction industries were built in energy-abundant and sparsely populated northwestern China over the past two decades, particularly since the early 2000s, those large-scale industrial parks and bases in this part of China likely play an important role in the growing SO$_2$ emissions in northwestern provinces. We further
examine the OMI retrieved SO$_2$ VCD to confirm and evaluate the changes in SO$_2$
emissions in northwestern China which should otherwise respond to these
large-scale energy programs under the national plan for energy relocation and
expansion. **Figure 9** displays the MK test statistics for SO$_2$ VCD in the 6 provinces
in northwestern China from 2005-2015. The forward sequence $UF_k$ suggests
decreasing trends in Shaanxi and Gansu provinces and a moderate increase in
Qinghai province. In Xinjiang and Ningxia where the most energy industries were
relocated and developed for the last decade (2005-2015), as aforementioned, $UF_k$
time series estimated using SO$_2$ VCD data illustrate clear upward trends. Compared
with those well developed regions in eastern and southern China, the $UF_k$ values of
SO$_2$ VCD in these northwestern provinces are almost all positive, except for Shaanxi
province where the $UF_k$ turned to negative from 2008, and Gansu province where
the $UF_k$ value become negative during 2012-2013.

The step change points identified by the MK test for SO$_2$ VCD in northwestern
China appear associated strongly with the development and use of coal energy. As
shown in **Fig. 9**, the intersection of the forward and backward sequences $UF_k$ and
$UB_k$ within the confidence levels of -1.96 (straight green line) to 1.96 (straight
purple line) can be identified in 2006 and 2007 in Ningxia and Xinjiang, respectively,
(corresponding well to the expansion of two largest energy industry bases from 2003
onward in Ningxia (NECIB) and Midong energy industry park in Xinjiang. The step
change point of SO$_2$ VCD in 2012 in Gansu province coincides with fuel-switching
from coal to gas in the capital city (Lanzhou) and many other places of the province
initiated from 2012 (CSC, 2013b). The MK derived step change point in Shaanxi province occurs in 2010 which is a clear signal of marked decline of fossil fuel products in northern Shaanxi where, as the part of the EGT (Ma and Xu, 2017) of China, the largest energy industry base in the province is located, right after the global financial crisis.

It is interesting to note that the forward sequences $U_{F_1}$ of SO$_2$ VCD (Fig. 9e and f) in Ningxia and Xinjiang exhibit the similar fluctuations as that in Ningdong (NECIB) and Midong energy industrial bases (Fig. 8e and f), manifesting the potential associations between the SO$_2$ emissions in these two large-scale energy industrial parks (major point sources) and provincial emissions in Ningxia and Xinjiang, respectively. This suggests that large-scale energy industrial parks and bases might likely overwhelm or play an important role in the SO$_2$ emissions in those energy-abundant provinces in northwestern China. To assess the connections between the major point sources in the two energy industrial parks and the provincial emissions, we made use of OMI measured SO$_2$ VCD to inversely simulate the SO$_2$ emission burdens in Xinjiang and Ningxia. We used the source detection algorithm (McLinden et al., 2016) and the approach, which fits OMI-measured SO$_2$ vertical column densities to a three-dimensional parameterization function of the horizontal coordinates and wind speed, proposed by Fioletov et al. (2015, 2016), to estimate the SO$_2$ source strength in the two industrial parks and its contribution to the provincial total SO$_2$ burdens. Figure 10 illustrates mean SO$_2$ burdens from 2005 to 2015 in northern Xinjiang (Fig. 10a) and Ningxia (Fig. 10b). The largest burdens can be seen
clearly in the Midong energy industrial base and the NECIB in these two minority autonomous regions of China. Lower SO$_2$ emission burdens are illustrated in mountainous areas of northern Xinjiang. Figure 11 illustrates the annual variations of estimated SO$_2$ emission burdens ($10^{26}$ molecules) in the NECIB and Midong energy industrial parks (scaled on the left Y axis) and their respective fractions (%), scaled on the right Y axis) in the total provincial SO$_2$ burdens in Ningxia and Xinjiang, respectively. The SO$_2$ burden increased from 2005 and reached the maximum in 2011 in the NECIB and declined thereafter, in line with the annual SO$_2$ VCD fluctuations (Fig. 5) in this industrial park which is, as aforementioned, attributable to the economic rebound in 2011 in China. Of particular interest is the large fraction of the estimated SO$_2$ emission burden in the NECIB in Ningxia (Fig. 11a), showing that this industrial park alone contributed to about 40-50% emission burdens to the provincial total SO$_2$ emission burden. Likewise, the SO$_2$ emission burden enhanced from 2005 and peaked in 2013 in Midong energy industrial park (Fig. 11b). The emission burden in this park contributed about 25-35% to the provincial total SO$_2$ emission burden.

Compared with the NECIB, the SO$_2$ emission burden is higher in the Midong industrial park but has the lower fraction in the provincial total emission burden. Covered by large area of desert and Gobi (Junngar Basin) underlying surfaces, there are only a few of SO$_2$ emission sources in vast northern Xinjiang region (total area of Xinjiang is $1.66 \times 10^6$ km$^2$), leading to the small ratio of the major point source (Midong) to total emission sources in Xinjiang. Nevertheless, overall our results manifest that, although there were only a small number of SO$_2$ point sources in these
two energy industrial parks, the SO$_2$ emissions from these parks made significant contributions to provincial total emissions. Given that the national strategy for China's energy expansion and safety during the 21st century is, to a large extent, to develop large scale energy industrial parks in northwestern China, particularly in Xinjiang and Ningxia (Zhu and Ruth, 2015; Chen et al., 2016) where the energy resources are most abundant in China, we would expect that the rising SO$_2$ emissions in northwestern China would increasingly be attributed to those large scale energy industrial parks and contributed increasingly to the national total SO$_2$ emission in China.

Table 1 presents the annual average growth rates of SO$_2$ VCD, industrial (second) Gross Domestic Product (GDP), and major coal-consuming industries in northwestern China and three developed areas (BTH, YRD, PRD) in eastern and southern China. The positive growth rates of SO$_2$ VCD can be observed in the three province and autonomous regions (Qinghai, Ningxia, and Xinjiang) of northwestern China. Although the growth rates of SO$_2$ VCD in other two provinces (Gansu and Shaanxi) are negative, the magnitudes of the negative growth rates are smaller than those in the BTH, YRD, and PRD, except for Zhejiang province in the YRD. This regional contrast reflects both their economic and energy development activities, and the SO$_2$ emission control measures implemented by the local and central governments of China. Although China has set a national target of 10% SO$_2$ emission reduction (relative to 2005) during 2006-2010 and 8% (relative to 2010) during 2011-2015 (CSC, 2007; CSC, 2011b), under the Grand Western Development Program of China, the regulation for SO$_2$ emission control was waived in those
energy-abundant provinces of northwestern China in order to speed up the large scale energy industrial bases and local economic development, and improve local personal income. In addition, although FGDs were widely installed in coal-fired power plants and other industrial sectors since the 1990s, by 2010 as much as 57% of these systems were installed in eastern and southern China (Zhao et al., 2013). The capacity of small power generators which were shut-down in western China was merely about 10808 MW, only accounting for about 19% of the capacity of total small power plants which were eliminated in China (55630 MW) during the 11th Five-Year Plan period (2006-2010) (Cui et al., 2016). As shown in Table 1, the SO₂ emission reduction plans virtually specified the zero percentage of SO₂ emission reductions in Qinghai, Gansu, and Xinjiang and lower reduction percentage in the emission reduction in Ningxia and Inner Mongolia as compared to eastern and southern China during the 11th (2006-2010) and 12th (2011-2015) Five-Year Plan. As a result, the average growth rate for thermal power generation, steel production, and coal consumption from 2005 to 2015 in northwestern China reached 14.1% yr⁻¹, 35.7% yr⁻¹, and 11.9% yr⁻¹, considerably higher than the averaged growth rates over eastern and southern China (5.9% yr⁻¹ in the BTH, 0.8% yr⁻¹ in the YRD, and 2.3% yr⁻¹ in the PRD).

4 Conclusions

The spatiotemporal variation in SO₂ concentration during 2005-2015 over China was investigated by making use of the PBL SO₂ column concentrations
measured by the Ozone Monitoring Instrument. The highest SO$_2$ VCD was found in the NCP, the most heavily polluted area by SO$_2$ and particulate matters (PM) in China, including Beijing-Tianjin-Hebei, Shandong, and Henan province. Under the national regulation for SO$_2$ control and emission reduction, the SO$_2$ VCD in eastern and southern China underwent widespread decline during this period. However, the OMI measured SO$_2$ VCD detected two "hot spots" in the EGT (Ningxia-Shaanxi-Inner Mongolia) and Midong (Xinjiang) energy industrial parks, in contrast to the declining SO$_2$ emissions in eastern and southern China, displaying an increasing trend with the annual growth rate of 3.4% yr$^{-1}$ in the EGT and 1.8% yr$^{-1}$ in Midong, respectively. The trend analysis further revealed enhanced SO$_2$ emissions in most provinces of northwestern China likely due to national strategy for energy industry expansion and relocation in energy-abundant northwestern China. As a result, per capita SO$_2$ emission in northwestern China has exceeded industrialized and populated eastern and southern China, making increasing contributions to the national total SO$_2$ emission. The estimated SO$_2$ emission burdens in the Ningdong (Ningxia) and Midong (Xinjiang) energy industrial parks from OMI measured SO$_2$ VCD showed that the SO$_2$ emissions in these two industrial parks made significant contributions to the provincial total emissions. This indicates, on one side, that the growing SO$_2$ emissions in northwestern China would increasingly come from those large scale energy industrial parks under the national energy development and relocation plan. On the other side, this fact also suggests that it is likely more straightforward to control and reduce SO$_2$ emissions in northwestern China because
the SO$_2$ control measures could be readily implemented and authorized in those state-owned large-scale energy industrial bases.

The Supplement related to this article is available online

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Table 1: Annual growth rate for OMI SO$_2$ VCD and economic activities for individual provinces and municipality during 2005-2014 (% yr$^{-1}$), and SO$_2$ emission reduction plan during the 11th and 12th Five-Year Plan period (%).

<table>
<thead>
<tr>
<th>Region</th>
<th>OMI SO$_2$ VCD</th>
<th>coal consumption</th>
<th>Industrial GDP</th>
<th>Thermal power generation</th>
<th>steel production</th>
<th>SO$_2$ emission reduction plan (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2006-2010$^a$</td>
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<tr>
<td>Inner Mongolia</td>
<td>0.94</td>
<td>11.29</td>
<td>20.48</td>
<td>14.07</td>
<td>8.38</td>
<td>-3.8</td>
</tr>
<tr>
<td>Shaanxi</td>
<td>-3.41</td>
<td>13.14</td>
<td>19.96</td>
<td>13.01</td>
<td>14.48</td>
<td>-12</td>
</tr>
<tr>
<td>Gansu</td>
<td>-0.09</td>
<td>6.69</td>
<td>14.19</td>
<td>8.89</td>
<td>9.92</td>
<td>0</td>
</tr>
<tr>
<td>Qinghai</td>
<td>0.69</td>
<td>11.20</td>
<td>18.70</td>
<td>9.88</td>
<td>12.37</td>
<td>0</td>
</tr>
<tr>
<td>Ningxia</td>
<td>0.95</td>
<td>11.79</td>
<td>17.44</td>
<td>15.04</td>
<td>152.71</td>
<td>-9.3</td>
</tr>
<tr>
<td>Xinjiang</td>
<td>1.57</td>
<td>17.21</td>
<td>14.21</td>
<td>23.39</td>
<td>16.27</td>
<td>0</td>
</tr>
<tr>
<td>Beijing</td>
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<td>-6.13</td>
<td>9.13</td>
<td>5.99</td>
<td>-48.52</td>
<td>-20.4</td>
</tr>
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<td>15.84</td>
<td>6.01</td>
<td>10.19</td>
<td>-9.4</td>
</tr>
<tr>
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<td>4.16</td>
<td>12.37</td>
<td>6.22</td>
<td>10.70</td>
<td>-15</td>
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<tr>
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<td>0.86</td>
<td>-0.92</td>
<td>-26.9</td>
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<tr>
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<td>12.51</td>
<td>7.49</td>
<td>13.35</td>
<td>-18.0</td>
</tr>
<tr>
<td>Guangdong</td>
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<td>6.15</td>
<td>12.03</td>
<td>5.92</td>
<td>6.87</td>
<td>-15.0</td>
</tr>
</tbody>
</table>

$^a$ and $^b$ represents proposed reduction in SO$_2$ emission in 2010 relative to 2005, and 2015 relative to 2010, respectively. The value for PRD refers to the proposed target for Guangdong Province.
Figure Captions

Figure 1 Provinces, autonomous regions, and selected regions in China in this investigation. Northwestern China, defined by pink slash, includes Inner Mongolia, Shaanxi, Gansu, Qinghai, Ningxia, and Xinjiang province. Light green shadings with cross highlight Beijing-Tianjin-Hebei (BTH) and the light green color stands for the North China Plain (NCP, including BTH), defined by light green color, including BTH, Shandong, and Henan province. The Sichuan Basin, Yangtze River Delta (YRD), and Pearl River Delta (PRD) is defined by yellow, pink, and blue color. Red triangle indicate 188 monitoring sites across China.

Figure 2 Annually averaged SO\(_2\) VCD (DU), scaled on the right-hand-side Y-axis and measured annual SO\(_2\) air concentration (µg/m\(^3\)), scaled on the left-hand-side Y-axis, in Beijing, Shanghai, Chongqing, Guangzhou, Yinchuan, and Urumqi.

Figure 3 Annually averaged SO\(_2\) VCD (DU), scaled on the right-hand-side Y-axis and annual emissions (thousand ton/yr) of SO\(_2\) on the left-hand-side Y-axis in the NCP, YRD, PRD, Sichuan Basin, EGT, and Midong.

Figure 4 Annual averaging OMI-retrieved vertical column densities of SO\(_2\) (DU) and their trends from 2005 to 2015 on 0.25° × 0.25° latitude/longitude resolution in China. (a) Annual mean SO\(_2\) vertical column densities; (b) slope (trend) of linear regression relationship between annual average OMI-retrieved SO\(_2\) VCD and the time sequence from 2005 to 2015 over China. The positive values indicate an increasing trend of SO\(_2\) VCD from 2005 to 2015, and vice versa. The blue circle highlights the six selected regions where SO\(_2\) VCD displayed dramatic change for further assessment of the long term trends and step change points in SO\(_2\) VCD. These six regions are NCP (a), YRD (b), PRD (c), Sichuan Basin (d), Energy Golden Triangle (EGT, e), and Midong (f).

Figure 5 Percentage changes in annual mean OMI SO\(_2\) VCD in the four highlighted regions in eastern and southern China and two large-scale energy industry parks in the EGT and Midong region in Figure 4b (relative to 2005).

Figure 6 Annual fractions of OMI retrieved SO\(_2\) VCD and emissions averaged over 6 northwestern provinces in the national total SO\(_2\) VCD from 2005 to 2015 and emission from 2005 to 2014. (a) fraction of annual mean SO\(_2\) VCD; (b) fraction of annual mean emission. Fractions of SO\(_2\) VCD are calculated as the ratio of the sum of annually averaged SO\(_2\) VCD in northwestern China to the sum of annually averaged SO\(_2\) VCD in the national total from 2005 to 2015 (%).

Figure 7 Per capita SO\(_2\) emission in six provinces of northwestern China and three key eastern regions (tons/person). The value for PRD refers to the per capita SO\(_2\) emission for Guangdong province.

Figure 8 Mann-Kendall (MK) test statistics for annually SO\(_2\) VCD in those highlighted regions (Figs. 1 and 4b) from 2005-2015. The blue solid line is the forward sequence \(UF_k\) and the red solid line is the backward sequence \(UB_k\) defined by Eq (5). The positive values for \(UF_k\) indicate an increasing trend of SO\(_2\) VCD, and vice versa. Two straight solid lines stand for confidence interval between -1.96 (straight green line) and 1.96 (straight purple line) in the MK test. The bold black line in the middle highlights zero value of \(UF_k\) and \(UB_k\). The bold black line in the
middle highlights zero value of $U_{F_k}$ and $U_{B_k}$. The intersection of $U_{F_k}$ and $U_{B_k}$ sequences within the intervals between two confidence levels indicates a step change point.

Figure 9 Mann-Kendall (MK) test statistics for annually SO$_2$ VCD in six provinces in northwestern China from 2005-2015. The blue solid line is the forward sequence $U_{F_k}$ and the red solid line is the backward sequence $U_{B_k}$ defined by Eq (5). The positive values for $U_{F_k}$ indicate an increasing trend of SO$_2$ VCD, and vice versa. Two straight solid lines stand for confidence interval between -1.96 (straight green line) and 1.96 (straight purple line) in the MK test. The intersection of $U_{F_k}$ and $U_{B_k}$ sequences within intervals between two confidence levels indicates a step change point.

Figure 10 Mean SO$_2$ burden estimated by the OMI measured SO$_2$ VCD (DU) using a new emission detection algorithm (Fioletov et al., 2016). (a) SO$_2$ burden in northern Xinjiang; (b) SO$_2$ burden in Ningxia.

Figure 11 Annual SO$_2$ burdens (10$^{26}$ molecule) in the Ningdong and Midong energy industrial parks and their fractions in provincial total SO$_2$ burden. (a). SO$_2$ burden (blue bar) in Ningdong and its fraction (red solid line) in the total provincial SO$_2$ burden in Ningxia; (b). SO$_2$ burden (blue bar) in Midong and its fraction (red solid line) in the total provincial SO$_2$ burden in Xinjiang. The left Y-axis stands for SO$_2$ emission burden and the right Y-axis denotes the fraction (%).
Figure 1
Figure 2

Chongqing

Measured SO2
SO2 VCD

Measured SO2 (μg m⁻³)
SO2 VCD (DU)

Year

Beijing

Shanghai

Guangzhou

Urumqi

Yinchuan

Shanghai

Beijing

Urumqi

Yinchuan

Chongqing

Guangzhou

Measured SO2
SO2 VCD

Measured SO2 (μg m⁻³)
SO2 VCD (DU)

Year

Year

Year

Year

Year

Year

Year

Year

Year

Year

Year

Year
Figure 3

(a) North China Plain

(b) Yangtze River Delta

(c) Pearl River Delta

(d) Sichuan Basin

(e) Energy Golden Triangle

(f) Midong

SO$_2$ emission (thousand ton yr$^{-1}$)
Figure 4

(a) 2005-2015


Change from 2005 SO2 VCD (%)

Year

North China Plain
Energy Golden Triangle
Pearl River Delta
Sichuan Basin
Midong
Yangtze River Delta

Figure 5

Change from 2005 SO2 VCD (%)

Year

North China Plain
Energy Golden Triangle
Pearl River Delta
Sichuan Basin
Midong
Yangtze River Delta
Figure 6

(a) Fraction of SO$_2$ VCD of northwestern China (%)

(b) Fraction of SO$_2$ emission from northwestern China (%)

Figure 7

Per capita SO$_2$ emissions (tons/person)

- Ningxia
- Xinjiang
- Qinghai
- Inner Mongolia
- Gansu
- Shaanxi
- BTH
- YRD
- PRD

Year
Figure 8

(a) North China Plain
(b) Yangtze River Delta
(c) Pearl River Delta
(d) Sichuan Basin
(e) Energy Golden Triangle
(f) Midong

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Discussion started: 29 March 2017
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Figure 9

(a) Inner Mongolia  
(b) Shaanxi

(c) Gansu  
(d) Qinghai

(e) Ningxia  
(f) Xinjiang

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Figure 10

Figure 11