The Trend of Sulfur Dioxide Emissions in China after 2000

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ABSTRACT

With the rapid development of the economy, the sulfur dioxide (SO_2) emission from China since 2000 is of increasing concern. In this work, we first presented the change of SO_2 emission in China since 2000 using a technology-based methodology. Then, we compared the SO_2 emission data with a variety of official statistics, ground-based measurements and satellite observations of sulfur related quantities over East Asia. The results indicate that from 2000 to 2006, total SO_2 emission in China increased by 53%, from 21.7 Tg to 33.2 Tg. The emission growth rate slowed around 2005, and emissions began to decrease after 2006 mainly due to the wide application of flue-gas desulfurization (FGD) devices in power plants. The trend of estimated SO_2 emission in China is consistent with the trends of SO_2 concentration and acid rain pH and frequency in China, as well as with the increasing trends of background SO_2 and sulfate concentration in East Asia. A longitudinal gradient in the percentage change of urban SO_2 concentration in Japan is found during 2000-2007, indicating that the transport of increasing SO_2 from the Asian continent partially counteracts the local reduction of SO_2 emission downwind. Both surface solar radiation (SSR) measurements and satellite aerosol optical depth (AOD) products show that China and East Asia excluding Japan underwent a continuous dimming after 2000. The arrested growth in SO_2 emissions since 2006 is also reflected in the decreasing trends of SO_2 and SO_4^{2-} concentrations, acid rain pH values and frequencies, and AOD over East Asia.

INTRODUCTION

Sulfur dioxide (SO_2) and its atmospheric products (e.g., sulfate, sulfuric acid) have adverse effects on human health, and can affect the atmospheric environment from local to regional and global scales (e.g., acid deposition, direct and indirect radiative forcing). China is one of the largest SO_2-generating countries in the world. It is reported that the anthropogenic SO_2 emission in China contributed to about one-fourth of the global emission and more than 90% of East Asia emission since the 1990s [1-3]. In the past few years, the effect of SO_2 emission in the Asian continent on the regional and global environment has been studied by using both chemical transport models and intensive field experiments (e.g., ACE-Asia, TRACE-P, and INTEX-B), and most recently by using satellite observations directly. The results of these studies strongly suggested that the generation of sulfur species was increasing over time in this region, and sulfur-containing air pollutants are transported long distances from the Asian continent to the Northwestern Pacific, North America, and the rest of the northern hemisphere.

The SO_2 emission in China has changed dramatically since 2000. During the Chinese 10th Five-Year Plan period (2001-2005), the State Environmental Protection Administration (SEPA)—changed to

the Ministry of Environmental Protection (MEP) in March 2008—set a goal to reduce the national SO_2 emission level in 2000 (20 Tg/year) by 10% by the year 2005. However, this goal was not achieved. Due to the massive increase in fossil-fuel consumption resulting from rapid economic growth, the lag of the introduction of desulfurization equipment, and the low efficiency of the installed desulfurization devices, the national SO_2 emission in 2005 actually rose to 25.5 Tg/year, 42% higher than the goal. Furthermore, these SO_2 emission values reported annually by the China MEP are believed to be underestimated due to the neglect of emission from rural industries and biofuels [4, 5]. Although there have been previous studies that reported estimates of SO_2 emission from China [1, 4-6], few of them focused on trends during the period after 2000. The Regional Emission Inventory in Asia (REAS) [1] only presented historical SO_2 emission in China up to 2003, and it was reported to overestimate the SO_2 emission due to not reflecting the recent spread of desulfurization devices in China [7]. Other studies such as Zhang et al. [5], Klimont et al. [8], and Zhao et al. [9] estimated SO_2 emission either for a single year or without annual time series. In a rapidly developing country like China, new technologies are continuously coming into the market, causing dramatic changes in the technology distribution as well as the emission factor distribution in relevant sectors. Therefore, it is necessary to present the historical SO_2 emission based on the dynamic technology penetration and the dramatic growth in activity rates after 2000.

In this paper, we first present the change of SO_2 emission in China since 2000 by using a technology-based methodology specifically for China. The SO_2 emission data are then compared with a variety of official environmental statistics, ground-based measurements, and satellite observations of sulfur related quantities over East Asia, such as SO_2 and SO_4^{2-} concentrations, surface solar radiation (SSR), aerosol optical depth (AOD), etc. The purpose of this work is to:

- quantify releases of SO_2 from China since 2000;
- provide a better understanding of the impact of SO_2 emission in China across the East Asian region; and
- support the latest NASA field experiment on Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS).

METHOD

Estimation of Anthropogenic SO_2 Emission in China

Basic methodology

In this study, a detailed technology-based methodology specifically for China is used to estimate the SO_2 emission. Emissions are estimated based on activity data for 31 provinces in mainland China. Hong Kong, Macao, and Taiwan are not included. The emission sources are classified into four major sectors: power generation, industry, domestic, and transportation, including both fuel combustion and non-combustion sources. The provincial emission of SO_2 (E_i) is estimated from the following equation:

Equation (1) $$E_i = \sum_j \sum_k A_{i,j,k} \left[\sum_m X_{i,j,k,m} EF_{i,j,k,m} \right]$$

where E = SO_2 emission
A = activity rate (e.g. fuel consumption, material production)
X = fraction of fuel in a sector that is consumed by a specific technology
EF = net emission factor

i, j, k, m represent the province (municipality, autonomous region), the economic sector, the fuel or product type, and the technology type for combustion and industrial processes, respectively. For the fuel combustion sources, EF can be calculated by:

Equation (2) $EF_n = 2 \times S \times (1 - SR) \times (1 - \eta_n)$

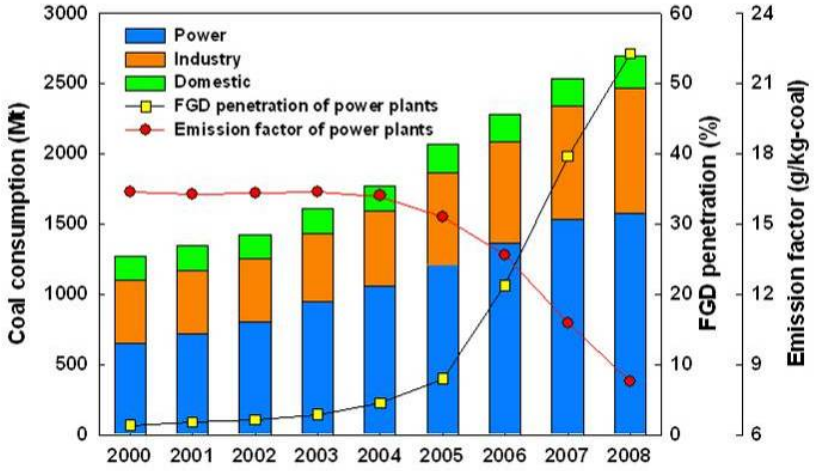
where η_n = removal efficiency of control technology n
S = sulfur content of fuel
SR = sulfur retention in ash

Based on this framework, we updated China's SO_2 emission to the year 2008 with the most recent statistics available, including fuel consumption, flue-gas desulfurization (FGD) penetration, technology renewal, human activity, and so on.

Activity rates and emission factors

Activity data for China during the period 2000-2008 were obtained from statistics published by a variety of government agencies. The fossil-fuel and biofuel consumption of stationary combustion sources by sector and by province were derived from the provincial energy balance tables of the China Energy Statistical Yearbook (CESY). As shown in Figure 1, the coal consumption in China increased from 1271 to 2689 Mt during 2000-2008, with an annual average increase rate of 9.8%; power plants contributed about 65% of this increase. For non-combustion sources, industrial production levels by province were obtained from the China Industry Economy Statistical Yearbook. For on-road vehicle sources, we categorized vehicles into seven types, corresponding to the classification scheme in the USEPA's MOBILE emission factor model [10]. The fuel consumption of each type was calculated as a product of the vehicle population, the annual average vehicle mileage traveled, and the fuel economy for each vehicle type. The vehicle populations by type were derived from the China Automotive Industry Yearbook. Details of the methodology have been described elsewhere [11].

Figure 1. Coal consumption, FGD penetration and net EF of power plants in China during 2000-2008.



Dynamic emission factors were used to reflect the dramatic changes in fuel property and technology penetration during 2000-2008. The sulfur contents (S) of coal consumed by combustion sources in each province were derived from Streets et al. [4], Ohara et al. [1], and Zhao et al. [9, 12]. The national average S of coal in China was 1.08% and 1.02% in 2000 and 2005, respectively. We used interpolation values to represent S in each year during 2000-2005, and assumed the sulfur contents did not vary after 2005 since no reliable data are available. The sulfur retention ratios (SR) were assumed to be 0.15 for power plants and 0.05-0.45 for other sectors, depending on the process type, combustion technology, and coal type. The sulfur contents of oil products were determined from the national standards, which are 0.22% and 0.05% for diesel and gasoline, respectively. Province-by-province FGD application rates of power plants were estimated by the ratio of average FGD installed capacity to average capacity of power plants in each year. Relevant data were obtained from the China MEP and the

China Electric Power Yearbook. FGD application rates can impact emission factors significantly. As shown in Figure 1, the FGD penetration of power plants increased from 4% in 2004 to 54% in 2008, and net emission factors decreased by 49%, correspondingly.

Data Sets

Table 1 lists the data sets used in this work. SO_2, SO_4^{2-} and AOD (or SSR) data from ground-based monitoring sites and satellites are compared to the SO_2 emission change in China after 2000. For ease of comparison between the trends of multifarious data sets, all the data sets are normalized based on the following equation [3]:

Equation (3) $$x'_{i,j} = (x_{i,j} - \overline{x_j}) / \sigma_j$$

where $\overline{x_j}$ = average value of data set j

σ_j = standard deviation of data set j

$x'_{i,j}$ = ith normalized value of data set j (i.e. $x_{i,j}$)

Table 1. Summary of data sets used in this study.

	Monitoring data	Satellite data
SO_2	A variety of statistical yearbooks (China) Environmental Bulletins of a variety of provinces and cities (China) EANET (Japan and South Korea) NIES (Japan)	SCIAMACHY OMI
SO_4^{2-}	EANET (Japan and South Korea)	
Acid rain	A variety of statistical yearbooks (China)	
AOD		MODIS, MISR
SSR	Subset of SSR observations reported by Wild et al. (2009) (East Asia)	

Monitoring data

The annual SO_2 concentrations, pH values, and frequencies of acid rain in Chinese cities are collected from a wide range of sources, including Environmental Bulletins of various cities and provinces, China Environment Yearbook, China Statistical Yearbook on Environment, China Statistical Yearbook for Regional Economy, etc. The SO_2 concentration data of Japan are from the air pollution database system operated by the Environmental Information Center of the National Institute for Environmental Studies (NIES, http://www.nies.go.jp/igreen/index. html). We also use the SO_2 and SO_4^{2-} monitoring data reported annually by the Acid Deposition Monitoring Network in East Asia (EANET). Here, data from 13 sites (1 urban, 3 rural, and 9 remote) in Japan and South Korea are used.

SSR changes over East Asia after the 1980s were reported to be in line with the changes in anthropogenic emission and consequent changes in AOD [2, 3]. Hence, SSR observations are used as a surrogate of AOD monitoring data. To simplify the comparison, SSR change due to aerosols at a given wavelength is calculated by the classical equation [3]:

Equation (4) $$I / I_0 = \exp(-\mathrm{AOD})$$

where I_0 = extraterrestrial (top-of-the-atmosphere) irradiance of the sun

I = surface solar irradiance

The SSR observation data used in this study are a subset of the data reported by Wild et al. [13]. For East Asia, all-sky values of SSR measurements are taken from the Global Energy Balance Archive (GEBA) at the Swiss Federal Institute of Technology (ETH) Zurich, and the World Radiation Data Center (WRDC) of the Main Geophysical Observatory in St. Petersburg. Since the time period studied in the present work is from 2000 to 2008, we only include stations with continuous reliable records up to at least 2005. In all, data from 28 stations in four countries are used in this work: Japan (14 sites), China (7 sites), Mongolia (3 sites), and South Korea (4 sites).

Satellite Data

The SO_2 satellite data are from both the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) aboard the European Space Agency's (ESA) Environmental Satellite (ENVISAT) and the Ozone Monitoring Instrument (OMI) aboard the NASA's Earth Observing System (EOS)/Aura satellite. For SCIAMACHY, we use the monthly level-3 product with grid cells of $0.25^o \times 0.25^o$ from the Support to Aviation Control Service (SACS, http://sacs.aeronomie.be/index.php). For OMI, the planetary boundary layer (PBL) SO_2 data in the OMSO2 Level-2G products were used, and they were acquired from NASA's Goddard Earth Sciences Data and Information Services Center (GES-DISC) at http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omso2g_v003.shtml. Daily retrievals were first filtered to remove data with large solar zenith angle ($>70^o$), or relatively high radiative cloud fraction (>0.3) and terrain height (>1.5 km), or anomalous scenes, and then averaged at $0.5^o \times 0.5^o$ resolution to reduce the noise. Annual mean SO_2 column amounts were then calculated from the daily data for the years 2005-2008.

AOD satellite retrievals are from the Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging Spectroradiometer (MISR). The AOD data have discontinuities in some mesh grid points, mainly in middle and high latitudes (i.e., bright land surfaces such as the desert and snow-covered surfaces), which were excluded in the analysis. In this study, the monthly level-3 products of MODIS (version 5) and MISR (version 31) are used, and they were acquired at http://disc.sci.gsfc.nasa.gov/giovanni. Global coverage in the absence of clouds is obtained in one to two days for MODIS and in six to nine days for MISR. Horizontal resolutions are $1^o \times 1^o$ and $0.5^o \times 0.5^o$ for MODIS and MISR, respectively.

RESULTS AND DISCUSSION

SO_2 Emission in China after 2000

Emission trend

Figure 2a shows the annual trend of SO_2 emission in China after 2000. Compared to the relatively stable or decreasing trend during 1995-1999 [1-3, 6], we estimate that the SO_2 emission in China increased by 53%, from 21.7 Tg in 2000 to 33.2 Tg in 2006, with an annual growth rate of 7.3%. This growth rate is in good agreement with annual growth rates of 6.3%-9.9% estimated by other researchers [1, 5, 8], and of 6.2%-9.6% derived from satellite constraints [14]. This dramatic change was driven by the rapid increase of fossil-fuel consumption (78% growth in total energy consumption) due to the economic boom (99% growth in GDP) during this period. Although the GDP and the total energy consumption in China were still increasing after 2006, the national SO_2 emissions began to decrease, due to the application of FGD technology and the phase-out of small, high-emitting power generation units. In 2006 China's MEP reaffirmed its commitment to reducing SO_2 emissions. It resolved in its 11th Five-Year Plan (2006-2010) to cut the national SO_2 emissions by 10% (i.e., to 22.9 Tg in 2010), relative to the 2005 level. To achieve this goal, emission reduction requirements were to be strictly enforced. As a result, FGD devices began to be widely installed in coal-fired power plants in China. By the end of 2008, the FGD penetration had risen to 60%, which is responsible for an estimated reduction of 13.3 Tg SO_2 in that year. However, it should be noted that the *actual* operation of FGD equipment is unknown and

certainly increases the level of uncertainty in emission after 2005. Recent developments in SO_2 satellite retrievals may be able to constrain the SO_2 emission values or even the FGD operation of large point sources (LPS) to verify SO_2 emission in the future.

Figure 2. Trends of SO_2 emission in China, 2000-2008. (a) SO_2 emission; (b) Normalized trends.

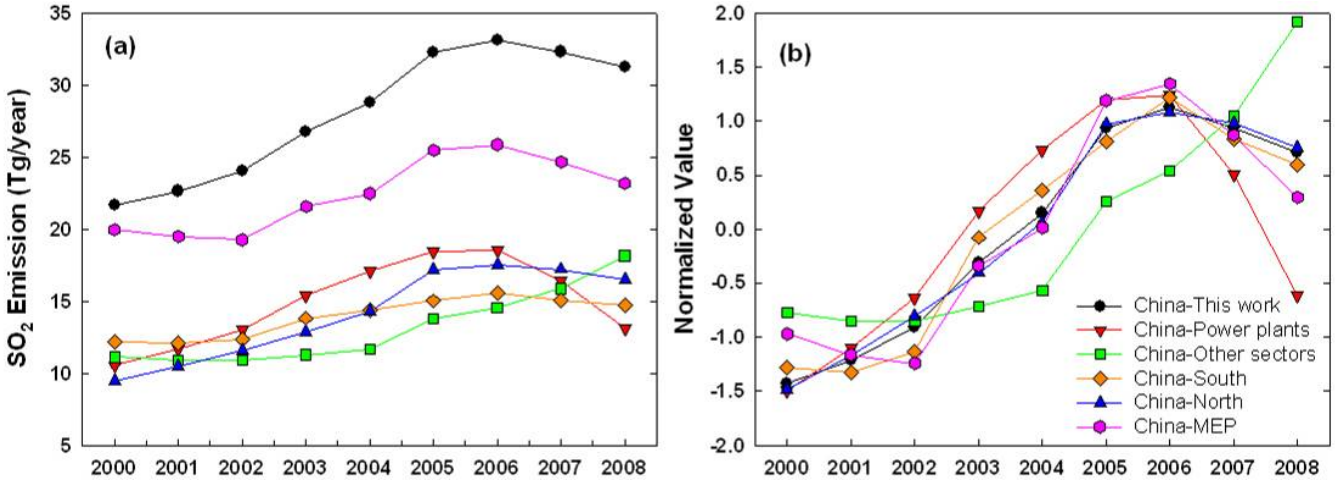


Figure 2a also shows the SO_2 emission of China by sector (power and other) and by geographical region, and the normalized values of all these data sets are displayed in Figure 2b. Power plants are the main sources of SO_2, contributing more than 50% to the total emission. Compared to a 31% increase in emissions in other sectors, SO_2 emission from power plants increased by 76%, from 10.6 Tg in 2000 to 18.6 Tg in 2006, which can be viewed in the context of a 116% increase of total thermal-based electricity generation during the same period. The trend of SO_2 emission from the power sector is in good agreement with the national emission estimate (Figure 2b, R=0.95), confirming that SO_2 emission from power plants dominates the national SO_2 emission. In particular, because of the decrease of power sector emission resulting from increasing FGD penetration, national SO_2 emission declined after 2006, despite the continuously increasing emission from other sectors.

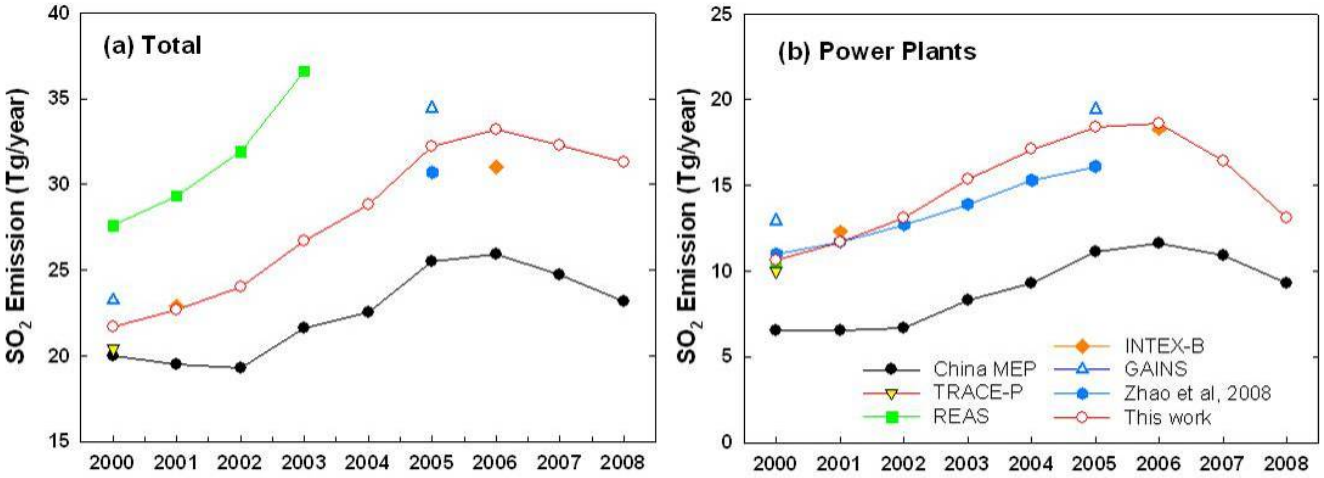
Geographically, the emissions from both north and south China are highly correlated with the total emission (R>0.99), implying that the change of SO_2 emission is nationwide. However, the extent of the increment is quite different geographically. From 2000 to 2006, SO_2 emission from north China increased by 85%, whereas that from south China increased by only 28%. This result mirrors the fact that more power plants were constructed in north China (e.g., Inner Mongolia, Shandong, Henan, and Hebei provinces), and some of those in Inner Mongolia have been identified by NO_x and SO_2 observations of OMI aboard NASA's Aura satellite[15, 16].

Comparison with previous studies

Figure 3 compares the SO_2 emission estimated in this study to other works. Our estimates agree well with those reported by the China MEP (R=0.96), but are about 10-30% higher, which may be caused by the neglect of SO_2 emission from rural industries and biofuels in the China MEP inventory [4, 5]. The trends of the two inventories are also similar at sector levels: a 76% increase of SO_2 emission was estimated in the power sector in this study during 2000-2006, while MEP presented a 78% increase. For TRACE-P [4] in 2000, the differences are mainly attributed to the improvements of methodology; and for INTEX-B [5] in 2006, differences are attributed to the extrapolated activity rates from 2004 data in the INTEX-B data, which were lower than the actual data reported in CESY. Our estimates are slightly lower than GAINS [8], and significantly lower than REAS [1]. This might be caused by a combined effect of the different amounts and distribution of fuel consumption between sectors and the implied emission factor assumptions. For emissions from power plants, estimates from different studies are similar, as the

uncertainties of activity data and emission factors are relatively small. Recently, Aikawa et al. [7] compared the measured sulfate (SO_4^{2-}) concentration with simulation results from a regional chemical transport model (CMAQ) at multiple sites over the East Asia Pacific Rim region. The SO_2 emissions used in the model were prepared based on both the REAS and the China MEP inventories. Their comparisons suggest that the REAS inventory overestimates, whereas the China MEP inventory underestimates the SO_2 emission from China. Our results fall midway between these two estimates.

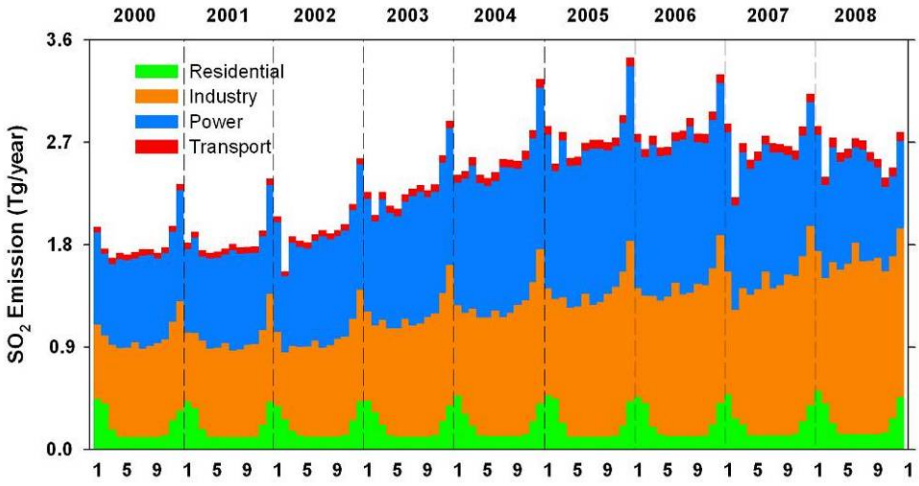
Figure 3. Comparison of SO_2 emission estimates for China, 2000-2008.



Seasonality of emissions

Year-specific monthly fractions for SO_2 emissions from each major sector during 2000-2008 are developed. For the residential sector, we follow the same methodology used in the TRACE-P inventory [4], assuming a dependence of stove operation on regional monthly mean temperatures, to generate monthly emissions. For the other sectors, monthly fractions are determined on monthly activity data of power generation, industrial GDP, sulfuric acid and coke production, volume of passenger and freight transported by ship, railway and aviation, and etc. Figure 4 presents monthly emissions in China during 2000-2008. Comparing with CO, BC and OC emissions, SO_2 emissions have weaker seasonality variations, because they are mainly from power and industrial sectors that have less of a season cycle. Generally, emissions are higher in November, December and January due to the increasing fuel consumption used in residential sector for heating, and are lower in February since the reduced industrial activity during the Chinese Spring Festival holiday.

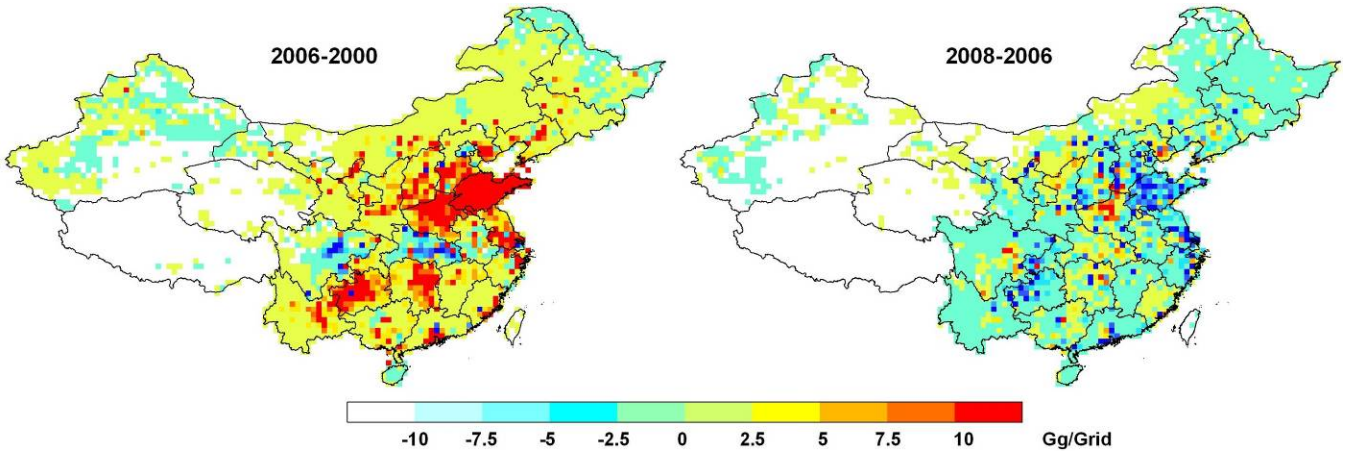
Figure 4. Monthly anthropogenic SO_2 emissions in China during 2000-2008.



Gridded emissions

Figure 5 shows the spatial distribution change of SO_2 emissions in China at a resolution of 30 min × 30 min during 2000-2006 and 2006-2008. Emissions are distributed using various year-by-year spatial proxies, including total, urban and rural population, Industrial GDP, crop land cover, and etc. All power generation units with capacity larger than 300 MW in China are identified as large point sources, while other plants are treated as area sources. Obviously, increments are highest in the eastern central China from 2000 to 2006 (e.g. Shandong, Hebei, and Henan provinces). Since this area is directly to the windward of Japan and South Korea, the dramatic change of SO_2 emission after 2000 may influence SO_2, SO_4^{2-} and AOD not only in China but also in other parts of East Asia, and this will be discussed further in the following sections.

Figure 5. SO_2 emission distributions change at 30 min × 30 min resolution.



Comparison of SO_2 Emission and Sulfur Related Quantities

SO_2 emission vs. SO_2 concentration in China

Figure 6 compares the change of SO_2 emission (by province) and SO_2 concentration (by major cities) in China from 2000 to 2006 (Figure 6a) and 2006 to 2008 (Figure 6b). The chromatic difference represents the percentage SO_2 emission or SO_2 concentration change. Due to the implementation of the revised Air Pollution Prevention and Control Law in 2000 and the relatively high SO_2 ambient level before 2000, the SO_2 concentrations in some big cities decreased dramatically during 2000-2006. However, the majority of cities still show a trend of increasing annual SO_2 concentration, which is generally consistent with the SO_2 emission increase during the same period. This result confirms the fact that SO_2 is a primary air pollutant with a relatively short lifetime, and its concentration broadly reflects the emission level of its surrounding area. For the period between 2006 and 2008, a nationwide decrease in SO_2 concentration also agrees well with the national emission decrease. The SO_2 change in China after 2000 can be seen even more clearly by using the satellite observations. As shown in Figure 7a and Figure 7b, a substantial increase (decrease) in the yearly average of boundary layer SO_2 is observed by both SCIAMACHY and OMI retrievals before (after) 2007. This is also consistent with the SO_2 emission change before and after 2006 as shown in Figure 5.

SO_2 emission vs. acid rain in China

Acid rain is a serious and growing problem in China [9]. Although NO_x, PM, NH_3, and other species also contribute to the acidification, SO_2 emission is still believed to be the major cause of the acid rain in China [17]. The increase of SO_2 emission between 2000 and 2006 ought to have worsened the acid rain situation. Figure 8 shows the acid rain frequencies and precipitation pH in cities under the National Acid Rain Monitoring Program of China during 2000-2008. The proportion of cities

experiencing acid rain with a frequency of >50% and >75% increased between 2000 and 2005; meanwhile the proportion of cities experiencing moderate and heavy acid rain (average pH of precipitation less than 5.0 and 4.5, respectively) increased too, indicating the acid rain problem was getting more severe year by year. After 2005, the acid rain problem in Chinese cities was alleviated in both frequency and average precipitation pH value. All these findings are consistent with the SO_2 emission trend and SO_2 concentration trend in China after 2000.

Figure 6. Comparison of the percentage change of SO_2 emission and SO_2 concentration in China. Squares represent provincial capitals, and circles represent other cities.

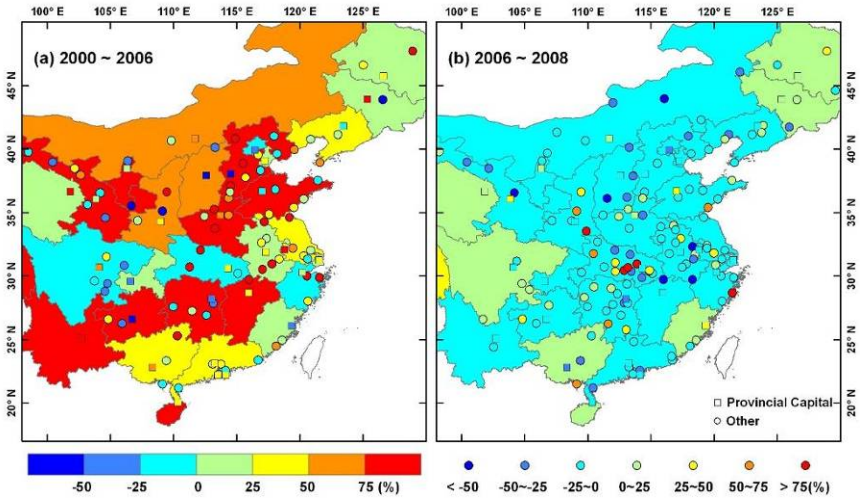


Figure 7. Differences in annual SO_2 vertical columns of boundary layer from satellite retrievals.

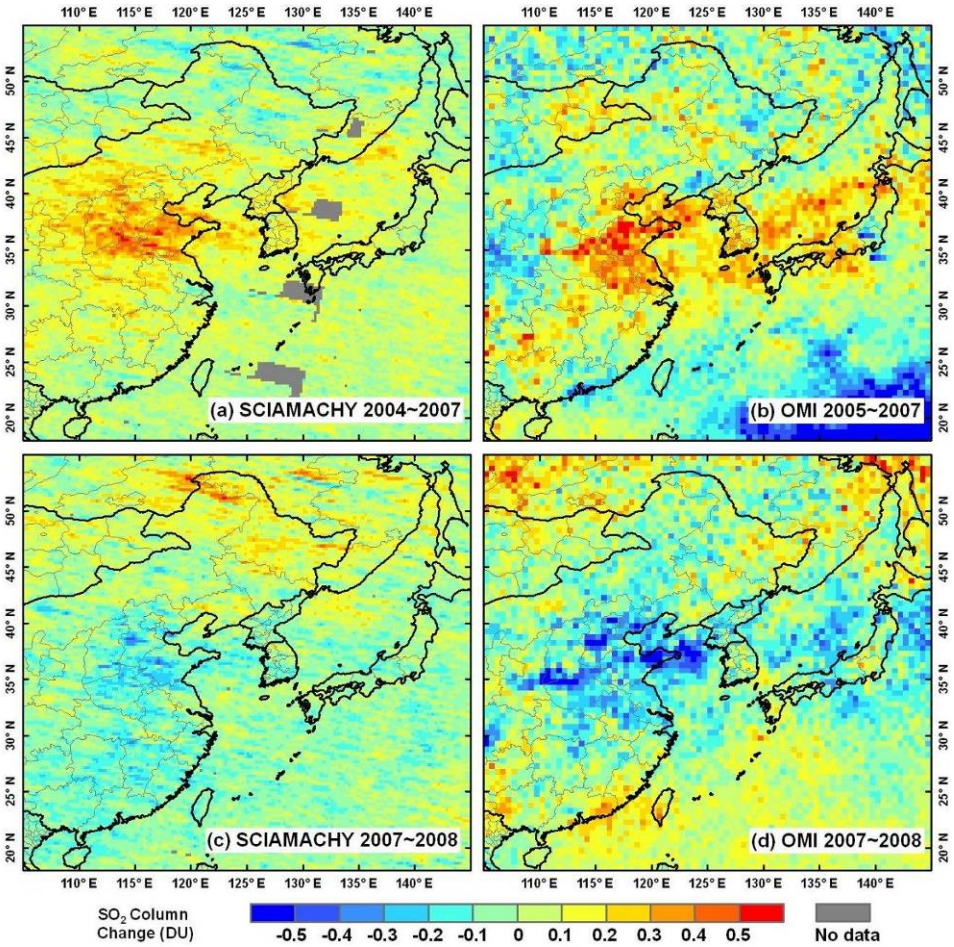
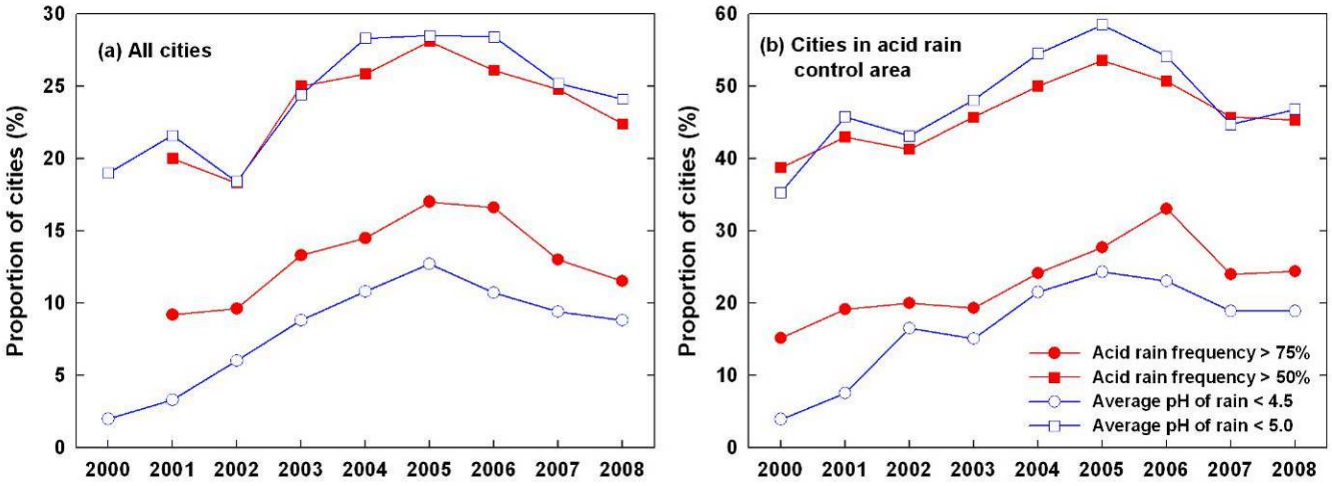


Figure 8. Acid rain situation in China.



SO_2 emission vs. SO_2 and SO_4^{2-} concentration in Japan and South Korea

Figure 9 shows the percentage change of annual SO_2 concentration at about 1000 sites in the Japan National Air Monitoring Network (from the database of Japan NIES) between 2000 and 2007. Unlike China, SO_2 concentrations in most cities of Japan tend to continuously decrease after 2000. This is in line with the decrease of both volcanic and local anthropogenic SO_2 emission in Japan during the period. However, some sites close to the Asian continent (e.g., sites in the southwest of Japan) showed an opposite increasing trend. The inset of Figure 9 plots the percentage change of SO_2 in each site versus each site's longitude. Although the data are scattered, the SO_2 percentage change is generally decreasing with longitude, indicating that the further the site is from the Asian continent, the greater the SO_2 concentration decrease. Recently, Aikawa et al. [7] found a significant longitudinal/latitudinal gradient in the measured and modeled SO_4^{2-} concentrations over the East Asian Pacific Rim region, and the SO_4^{2-} concentrations were higher at the sites closer to the Asian continent. On the other hand, they found no such distinct gradient in SO_2 concentrations. However, in this study, we found a longitudinal gradient in the percentage change of urban SO_2 concentration in Japan during 2000-2007. It is reasonable to believe that the longer lifetime of SO_4^{2-} enables it to be transported over longer distances, whereas the shorter lifetime of SO_2 makes it more correlated with local emissions. However, our result clearly demonstrates that, in spite of the relatively short tropospheric lifetime of SO_2, the transport of increasing SO_2 from the Asian continent partially counteracts the local reduction of SO_2 emission downwind, and even overrides it in some southwest areas of Japan.

Figure 10a displays the inter-annual variation of SO_2 average concentration over remote and other (urban and rural) EANET sites separately. Apparently, SO_2 concentrations at urban and rural EANET sites decreased after 2000, and this is in line with the trend in Japan NIES sites, which are mostly located in urban areas as well. The substantial increase in SO_2 concentration at remote sites during 2002-2007 is probably attributed to the SO_2 emission increase on the Asian continent. The background SO_2 increase can also be observed in the SO_2 retrievals of SCIAMACHY and OMI in Figure 7. As distinct from primary emitted gaseous SO_2, SO_4^{2-} in aerosols are mostly secondary air pollutants, and may be transported for longer distances than SO_2. Figure 10b shows that the sulfate concentrations in PM of the EANET sites have increasing tendencies after 2003, which is consistent with the SO_2 emission increase in China during this period. In addition, the increasing trend for most sites levels off and changes to a decreasing trend around 2005 to 2006, which is also in agreement with the SO_2 emission change in China after 2005.

Figure 9. Percentage change of annual SO_2 concentration in sites of Japan National Air Monitoring Network between 2000-2007. The inset plots the percentage change of SO_2 versus site longitude.

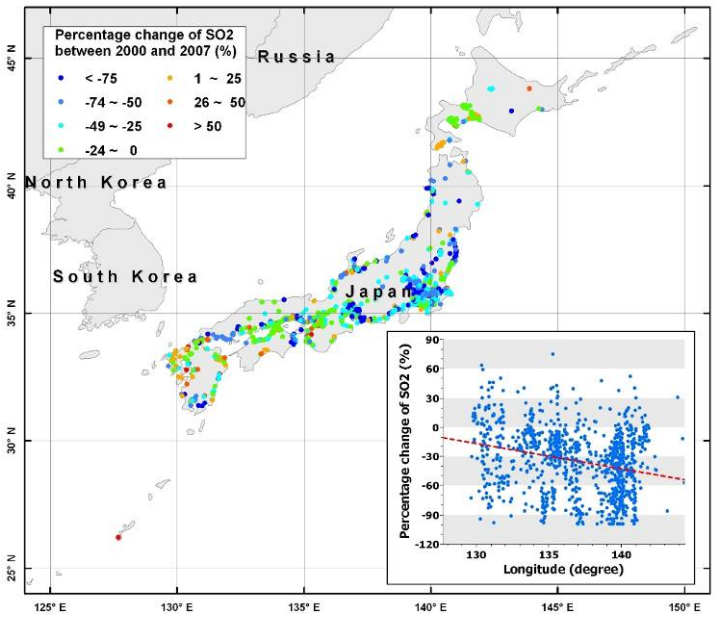
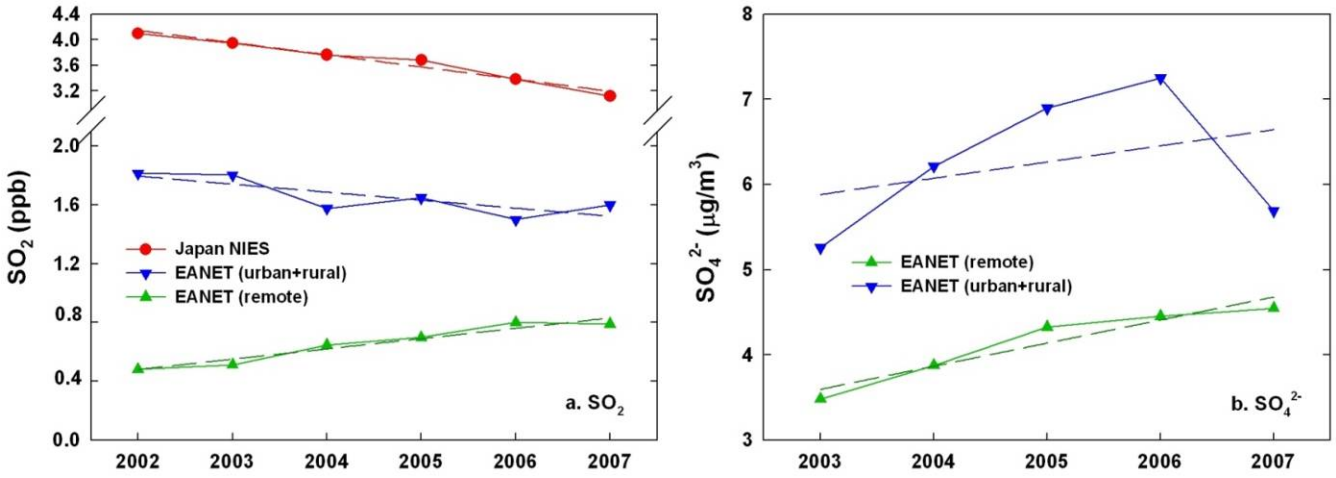


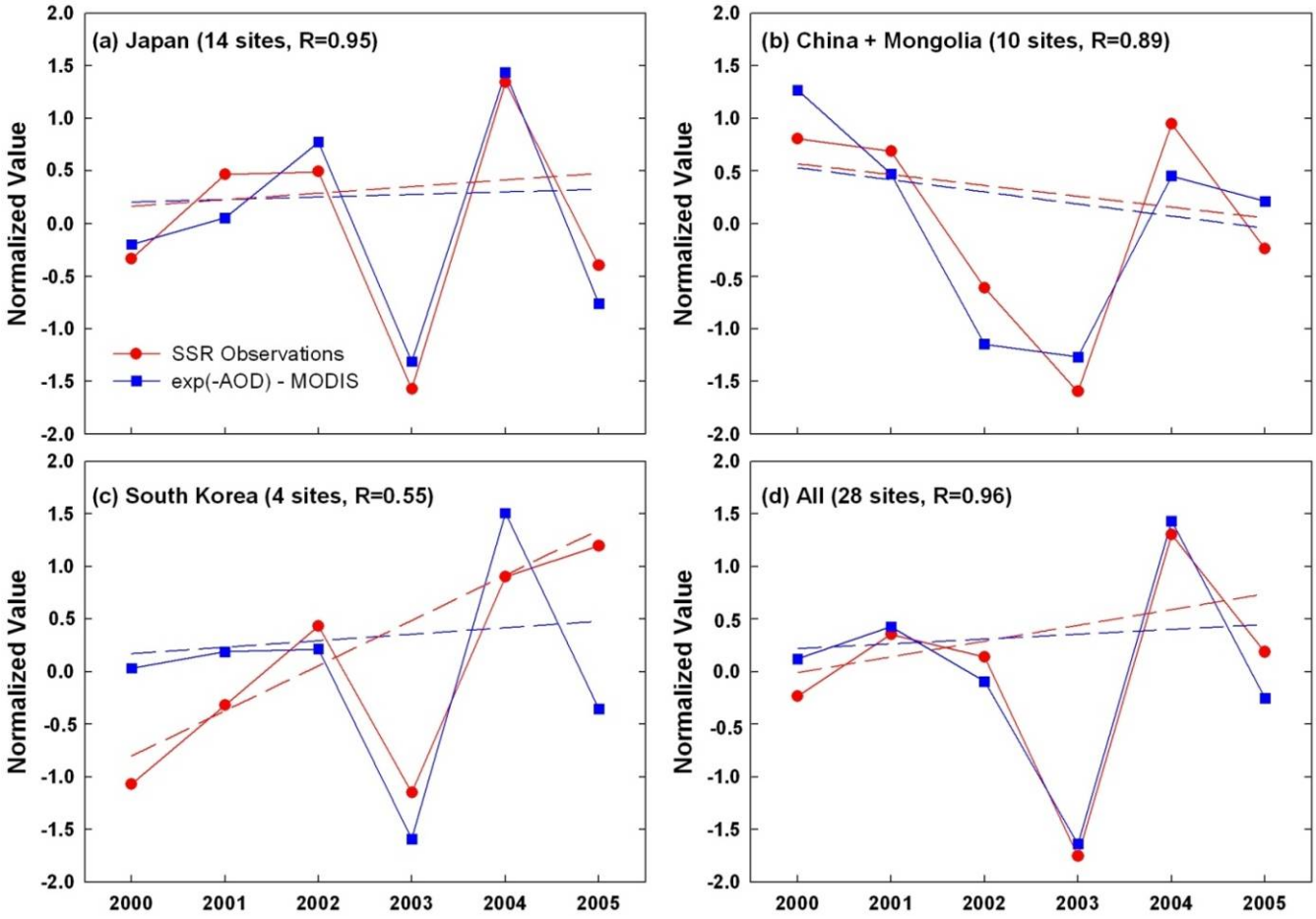
Figure 10. Inter-annual variation of SO_4^{2-} and SO_2 average concentrations at Japan NIES sites and EANET sites. Dashed lines represent the linear tendencies.



<u>SO_2 emission vs. SSR in East Asia</u>

Figure 11 compares the annual time series of normalized mean SSR observations and MODIS exp(-AOD) mean values for the 28 SSR sites, by region, in which the AOD data are extracted from the grid boxes of the MODIS monthly level-3 dataset where SSR sites are located. We divide the 28 SSR sites into three regions (Japan, China/Mongolia, and South Korea), and the annual time series for each region is determined by averaging over the complete records of the individual sites. As shown in Figure 11, the SSR observations and the MODIS exp(-AOD) at the 28 sites have nearly the same inter-annual variations, suggesting that the MODIS AOD data are consistent with solar radiation measurements in East Asia. As the linear tendencies of each dataset show in Figure 11, Japan and South Korea showed a brightening between 2000 and 2005, and China and Mongolia underwent a continuous dimming after 2000. The continuous dimming in China is consistent with the dramatic increase in SO_2 emission in East Asia after 2000 and provides further evidence to confirm the hypothesis that changes in AOD over time, particularly the regional transitions from dimming to brightening, are caused by the changing patterns of anthropogenic emissions of aerosols and aerosol precursors [2, 3].

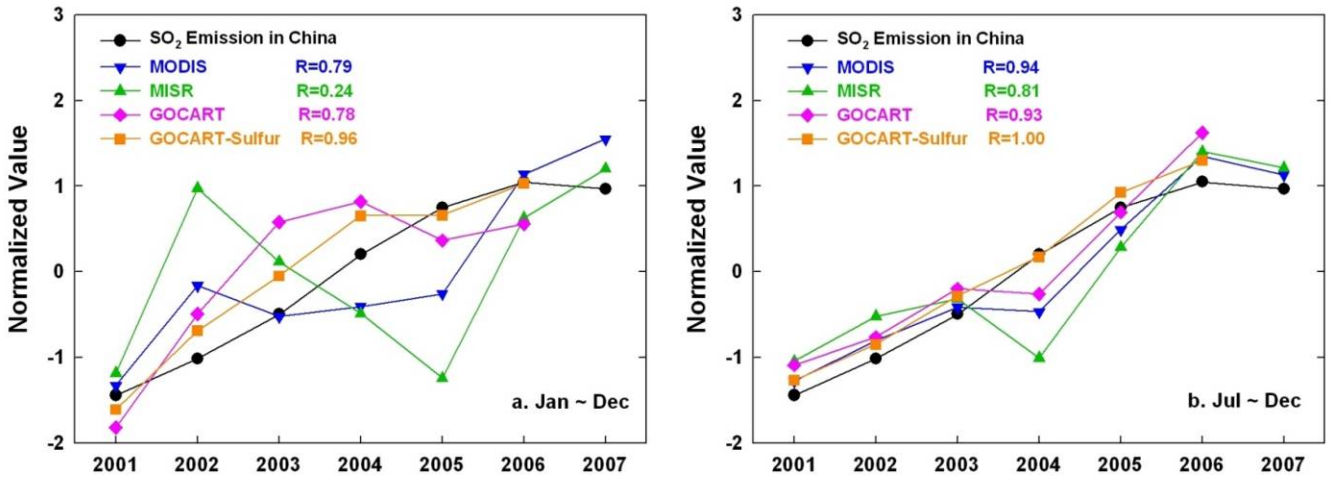
Figure 11. Evolutions of normalized annual mean SSR observations and exp(-AOD) from MODIS at 28 SSR observation sites. Dashed lines represent the linear tendencies excluding the year 2003 data.



SO_2 emission vs. AOD in East Asia

Figure 12 shows comparisons of three-year central moving average values of normalized SO_2 emission in China with AOD values averaged over East Asia from MODIS and MISR satellite retrievals, as well as with AOD simulated by the Goddard Chemistry Aerosol Radiation and Transport model (GOCART, obtained from Giovanni). As shown in Figure 12a, although the datasets have obviously different inter-annual variations, the tendencies are the same, showing an increase in AOD over East Asia after 2000. This is consistent with the work of van Donkelaar et al. [14]. They analyzed the MODIS and MISR AOD for 2000-2006 over eastern China, and found increases of 4.1%/year and 3.4%/year in MODIS and MISR AOD, respectively. It should be noted that this conclusion was drawn when they focused their analysis between July and December of each year. During this period, their GEOS-Chem model results showed sulfur contributed to 56% of total AOD in China. Therefore, we further compare the AOD trend of MODIS, MISR, GOCART, and SO_2 emission semiannually (from July to December, Figure 12b). Clearly, AOD trends agree much better with SO_2 emission in China during the period of July to December. *R* values between SO_2 emission and each AOD dataset increase significantly from 0.24-0.96 to 0.81-1.00, indicating that the effect of emission in China on AOD is more pronounced during the second half of the year. Additionally, the decline of both MODIS and MISR AOD in East Asia after 2006 corresponds well with the decrease of China SO_2 emission, further indicating the close relationship between SO_2 emission and AOD.

Figure 12. Three-year central moving average values of normalized China SO_2 emission, MODIS AOD, MISR AOD, GOCART AOD, and the sulfur portion of GOCART AOD over East Asia. Values are averaged (a) from January to December and (b) from July to December of each year. R values shown are the correlation coefficients of each dataset with SO_2 emission in China.



CONCLUSIONS

Using a detailed technology-based methodology specifically for China, we estimate that SO_2 emission in China increased by 53%, from 21.7 Tg in 2000 to 33.2 Tg in 2006, at an annual growth rate of 7.3%. Power plants are the main source of SO_2 in China, contributing more than 50% to the total emission. The growth of China's SO_2 emission began to slow down around 2005, and emission then began to decrease after 2006, mainly due to the application of FGD devices in power plants. The changes in emission since 2000 are not uniform geographically. From 2000 to 2006, SO_2 emission from north China increased by 85%, whereas that from south China increased by only 28%. We compared the SO_2 emission with a variety of official environmental statistics, ground-based measurements, satellite observations, and model results, and found that the changes in SO_2 emissions are reflected in trends in other sulfur-related quantities in China and throughout the East Asia region.

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KEY WORDS

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