Ozone Monitoring Instrument Observations of Interannual Increases in SO$_2$ Emissions from Indian Coal-Fired Power Plants during 2005–2012

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Supporting Information

ABSTRACT: Due to the rapid growth of electricity demand and the absence of regulations, sulfur dioxide (SO$_2$) emissions from coal-fired power plants in India have increased notably in the past decade. In this study, we present the first interannual comparison of SO$_2$ emissions and the satellite SO$_2$ observations from the Ozone Monitoring Instrument (OMI) for Indian coal-fired power plants during the OMI era of 2005–2012. A detailed unit-based inventory is developed for the Indian coal-fired power sector, and results show that its SO$_2$ emissions increased dramatically by 71% during 2005–2012. Using the oversampling technique, yearly high-resolution OMI maps for the whole domain of India are created, and they reveal a continuous increase in SO$_2$ columns over India. Power plant regions with annual SO$_2$ emissions greater than 50 Gg year$^{-1}$ produce statistically significant OMI signals, and a high correlation ($R = 0.93$) is found between SO$_2$ emissions and OMI-observed SO$_2$ burdens. Contrary to the decreasing trend of national mean SO$_2$ concentrations reported by the Indian Government, both the total OMI-observed SO$_2$ and annual average SO$_2$ concentrations in coal-fired power plant regions increased by >60% during 2005–2012, implying the air quality monitoring network needs to be optimized to reflect the true SO$_2$ situation in India.

INTRODUCTION

Sulfur dioxide (SO$_2$) is a major air pollutant that enters the atmosphere through natural (e.g., volcanic eruptions, wildfires) and anthropogenic (e.g., combustion and release of sulfur-containing fuels and materials) processes. SO$_2$ and its atmospheric products (e.g., sulfate, sulfuric acid) are detrimental to human health, harmful to ecosystems, and responsible for several environmental problems such as acid deposition, smog formation, and visibility degradation. As fine particles, sulfate aerosols have major impacts on climate because they can reflect incoming solar radiation and act as cloud condensation nuclei (CCN) that modify the microphysical properties of clouds. SO$_2$ is also a regional and global issue because sulfate has a long atmospheric lifetime and can be transported long distances. Driven by the rapid economic development, SO$_2$ emissions from India have been continuously increasing over the past two decades. On the basis of the most recent emission estimates, India has surpassed the U.S. to be the world’s second largest SO$_2$ emitting country, after China, since 2010. The coal-fired power sector is the single largest contributor, accounting for ~50% of the national SO$_2$ emissions and ~70% of the emission increment during 1996–2010. For power plants, an accurate emission inventory at the plant/unit level is critical for atmospheric chemical transport models, as it can improve the model simulations significantly. Although there have been some published studies reporting SO$_2$ emissions from the Indian coal-fired power sector, few of them used actual activity rates and emission factors at the plant/unit level or presented the year-by-year emission trend for the most recent years. Following our previous work on NO$_x$ emissions in India, we use a unit-based methodology to develop the SO$_2$ inventory for the Indian coal-fired power sector in this study.

Compared to primary carbonaceous aerosols (i.e., black carbon and organic carbon), SO$_2$ problems are not given sufficient attention by the Indian Government. Currently, there is no SO$_2$ emission control regulation for coal-fired power plants because of the relatively low sulfur content of coals. On the basis of Indian government official reports, there was only one out of 361 monitoring stations recording an annual mean SO$_2$ concentration higher than the national air quality standard of 50 μg m$^{-3}$ in 2010, and there has been a decline in SO$_2$ levels since 2000. This is inconsistent with the fast increase in
Indian SO₂ emissions, and an independent data source is needed to verify the real SO₂ situation in India. Now, satellite remote sensing instruments such as the Ozone Monitoring Instrument (OMI) make routine SO₂ observations at high temporal and spatial resolution, providing valuable information about surface SO₂ sources. Compared to nitrogen dioxide (NO₂) retrievals, satellite SO₂ signals are very noisy. In the past few years, satellite SO₂ instruments have been only used to detect the global distributions, plumes, and trends of very large anthropogenic SO₂ sources. Oversampling smaller pixels from the center of the swath to fine grids (~3 km wide), de Foy et al. and Russell et al. identified plume features that were not visible on coarser grids. Averaging a large number of OMI pixels around the sources over a period of three years, Fioletov et al. recently identified the SO₂ plumes and quantified the SO₂ emissions from individual U.S. power plants that were typically undetectable using standard analysis techniques. Using the same method, SO₂ emissions from Canadian oil sands were also studied. In this work, we quantified plant-level activity rates derived from a series of annual reports of industrial companies through various annual reports of industrial companies and/or verification through direct field observations.

**METHODS AND DATA SETS**

**Unit-Based SO₂ Emission Inventory.** In our previous work, we developed a bottom-up, unit-based inventory for NOₓ emissions from Indian public coal-fired power plants during 1996–2010. In this study, we adapt the system to calculate annual SO₂ emissions and extend the inventory to the year 2012. In particular, we include a large number of captive (privately owned) coal-fired power plants that were not taken into account in the previous NOₓ study. In total, there are 165 coal-fired power plants containing more than 720 units in the updated database. On the basis of the data availability, unit-level or plant-level information was collected, including geographical location, boiler size (i.e., capacity) and type, coal type (i.e., hard coal or lignite) and sulfur content, electricity generation, specific coal consumption, SO₂ control technology, the exact time when the unit came into operation and/or retired, etc. Total SO₂ emission (E) from coal-fired power plants for year i is estimated by the following equation:

\[
E_i = \sum_j \sum_k \sum_l 2 \cdot G_{j,k} \cdot SCC_{j,k} \cdot S_{j,k}(1 - SR_i)(1 - \eta_i)
\]

where j, k, and l represent coal type, generation unit, and emission control technology, respectively; G is the electricity generation; SCC is the specific coal consumption per unit electricity generation; S is the sulfur content; SR is the sulfur retention in ash; and η is the removal efficiency of SO₂ control technology.

For public coal-fired power plants, most of the unit-level or plant-level activity rates were derived from a series of the Performance Review of Thermal Power Stations published by the Central Electricity Authority (CEA), Ministry of Power of India. For captive plants, such information was collected through various annual reports of industrial companies operating these plants. The exact locations of power plants were obtained from the Global Energy Observatory (GEO, http://globalenergyobservatory.org/index.php) and/or verified directly through Google Earth. As shown in Figure S1 of the Supporting Information (SI), the electricity generation and coal consumption in Indian coal-fired power plants increased dramatically by 61% and 64%, respectively, from 2005 to 2012. The predominant sizes of generating units are between 200 and 300 MW, and these units contribute about 50% of both the electricity generation and the coal consumption. In terms of the incremental growth, however, units with capacity >300 MW are the biggest contributors, accounting for 70% of the electricity generation growth and 63% of the coal consumption growth. The activity rates derived from this study match the official statistics from the Government of India and the International Energy Agency (IEA) very well (differences <5%), further indicating that the current database covers nearly all public and most captive coal-fired generating units in India.

Plant-specific coal sulfur contents (S) were compiled from a large number of reports, including Mittal et al., GEO, IEA Coal Power Database, and various power project reports. Where plant-specific S values were not available, the state-average S values reported by Reddy and Venkataraman were used. The national average S value of coals (including lignite) was calculated to be 0.55% in 2005, increasing to 0.58% in 2012, consistent with previous studies at the national level. Because the installation and operation of FGD is costly, it is understandable that the Indian coal-fired power plants would only install FGD devices if and when they are mandatory. In this work, we assume the SO₂ emissions are uncontrolled for all plants except the Trombay and Dahanu power plants, where seawater-based FGD devices (η = 90%) were reported to be operating. The capability of these two plants accounts for ~2% of the total coal-based capacity in India during 2005–2012.

**OMI SO₂ Retrievals and Processing.** The OMI is a nadir-viewing charge-coupled device spectrometer that has been measuring the earthshine radiance and the solar irradiance in the ultraviolet–visible range and providing aerosol and trace-gas observations, including SO₂, with daily global coverage since October 2004. It is aboard the National Aeronautics and Space Administration (NASA)’s EOS/Aura satellite, which is in a sun-synchronous ascending polar orbit at 705 km altitude with 13:45 local equator-crossing time. In this study, we use the planetary boundary layer (PBL) SO₂ data in the OMSO2 Level-2 product, developed specifically for anthropogenic SO₂ pollution sources. Daily pixel retrievals were filtered to remove data with large solar zenith angle (>70 degrees) or relatively high radiative cloud fraction (>0.2) and terrain height (>2 km). Pixels at swath edges (10 pixels on each side) were excluded to limit the across-track pixel width to ~40 km. Since June 2007, some cross-track positions were affected by row anomalies (RA) due to the partial external blockage of the radiance port on the OMI instrument (http://www.knmi.nl/omi/research/product/rowanomaly-background.php). We dy-
namically removed these affected pixels based on the RA flags provided in the OMSO2 Level-2 data. We also rejected pixels with SO2 observations higher than 5 Dobson Unit (DU, 2.69 × 10^{16} molecules cm^{-2}) to exclude cases of transient volcanic SO2.27

For valid pixels, a series of further corrections were applied to the originally retrieved PBL SO2 values. First, OMI PBL SO2 vertical columns were converted back to slant columns by multiplying by the predefined air mass factor (AMF) of 0.36. Then, the latitude-dependent Pacific average slant columns were subtracted from all valid pixels on a daily basis to remove the negative offset and the sharp gradient at 30°N that is caused by the switch of the a priori ozone profile at ±30° latitude during the retrieval process (the so-called “Pacific sector correction”).29 The corrected slant columns were then further divided by the GEOS-Chem model precomputed clear-sky monthly local AMF to obtain the SO2 vertical columns.20 Local AMF correction, Pacific sector correction, and dynamic correction for row anomaly are the three most critical steps in developing the gridded OMI SO2 Level-3 product (i.e., OMSO2e at a resolution of 0.25 degree), and they significantly improve the quality of OMI SO2 retrievals.20,39

The smallest size of the OMI pixel is 13 × 24 km^2, which is too large to observe the SO2 spatial distribution near a point source. Averaging a large number of individual pixels centered within a several km radius from the source, Fioletov et al.27 found that statistically significant signals can be obtained for sources with annual SO2 emissions greater than ~70 Gg year^{-1}. Using the same approach, we oversampled the valid pixels with corrected vertical columns at a 2 km × 2 km grid for the whole domain of India to analyze the interannual variations of the SO2 spatial distribution over coal-fired power plants. It is reported that large-scale biases still remain after the above corrections and processing, especially in the regions with broad pollution or persistent volcanic degassing.39 To remove them, local bias corrections were applied on a yearly basis, as recommended by Fioletov et al.27

■ RESULTS AND DISCUSSION

SO2 Emissions of Indian Coal-Fired Power Plants during 2005–2012. Figure 1 shows the annual trend of SO2 emissions from Indian coal-fired power plants by unit size from 2005 to 2012. A detailed emission summary by state and by region is provided in Table S1 of the SI. During 2005–2012, SO2 emissions increased rapidly by 71% from 3354 to 5738 Gg, with an annual average growth rate (AAGR) of 8.0%. This dramatic change reflects rapid economic and social development and no SO2 emission control in the Indian power sector.4,15 In terms of boiler size, coal-fired units with capacity >200 MW are the major contributors, accounting for ~73% of the total emissions. In particular, SO2 emissions from units with capacity larger than 300 MW increased by 1327 Gg from 2005 to 2012, contributing 56% to the emission increment. Since the current OMI SO2 products can only capture strong anthropogenic point or area plumes,18 the fact that the majority of SO2 emission increment in India is from newly installed large power units provides a favorable condition for observing the interannual increasing trend of Indian SO2 emissions with OMI retrievals, and this will be discussed in the next section.

Geographically, the distribution of coal-fired power plants is relatively uniform across the country except for the northeastern states (Figure 2a). Uttar Pradesh, Orissa, Gujarat, Chhattisgarh, Maharashtra, and Tamil Nadu are the six states with power-plant emissions >500 Gg SO2 in 2012, and together, they accounted for 60% of total emissions in that year. The emission increment is highest in Chhattisgarh (330 Gg in eight years), followed by Gujarat (318 Gg) and Orissa (300 Gg). Delhi is the only state where the SO2 emissions in the power sector have decreased in the past few years, mainly due to the conversion of coal-fired plants to gas-fired ones and the relocation of big power plants outside of the city of New Delhi.

The uncertainties of emission estimates were determined by a Monte Carlo approach with the Crystal Ball software, and 10 000 simulations were performed, as in previous work.4,15 We applied normal distributions with uncertainties (hereinafter, 95% confidence intervals) of 5% and 10% to coal consumption data and sulfur contents, respectively.4,15 For SR ratios and the FGD SO2 removal efficiency, uniform distributions were assumed in the range of minimum and maximum values reported in the literature.4,6,14,40 Results show that the uncertainties of estimated SO2 emissions are ~±13% around the mean. They are lower than in our previous work4,40 because of the detailed unit-based methodology and reliable unit/plant-level information used. Figure 1 also compares the estimated emissions to other inventories.1–7,13 Clearly, all of the estimates show increasing trends. The AAGR of our emissions during 2005–2010 is 6.7%, which is in line with values of 6.2%–7.5% in other inventories.2–4,13 The current estimates are in good agreement with our previous ones4 calculated from the IEA national statistics. The discrepancies with other studies are caused by various factors, including the use of higher sulfur content2 or lower coal consumption,16 the exclusion of captive power plants,13 and the inclusion of oil-fired and gas-fired plants9 in their calculations.

SO2 Emissions of Indian Coal-Fired Power Plants Observed by OMI. The spatial oversampling and local bias correction techniques described in the methodology section can produce high-resolution OMI SO2 maps, and it was reported that comparable SO2 sources have similar mean OMI SO2 values.27 This implies that top-down OMI SO2 observations can be used to estimate surface emissions and
track their changes. Figure 2 displays the spatial distribution of mean OMI SO2 over India from 2005 to 2012. A number of satellite SO2 hot spots are observed, and they match the locations (Figure 2a) and the amounts of SO2 emissions (Figure 2b) of large coal-fired power plants reasonably well. Note that there are some data artifacts at the southern foot of the Himalayas that may be caused by the unreliable retrievals over regions of extensive snow cover and high terrain height. Theoretically, SO2 in the atmosphere has a chemical lifetime of around one day, much longer than NO2 (several hours). As shown in Figure 2, however, elevated SO2 values are found only within ∼50 km of the point sources, a distance which is smaller than for OMI NO2 observations. This further demonstrates that the current OMI SO2 products are very noisy and only large point or area sources with intensive average emissions can be detected by OMI.

Figure 2c–f also shows the spatial distribution of average OMI SO2 columns by season over India during 2005–2012. Similar to our previous findings in OMI NO2 seasonal maps, July to September is the worst period to observe SO2 from OMI in India, because of the insufficient satellite observations caused by the frequent cloud coverage and heavy rainfall in the monsoon season. Different from OMI NO2, however, we do not observe significant seasonal variations in OMI SO2 over India probably because the seasonality of the SO2 lifetime is weaker. In previous studies to quantify the SO2 emissions from U.S. power plants and Canadian oil sands, only May–August SO2 data were used as they have higher signal-to-noise ratio than other months due to small solar zenith angle, low snow cover, strong actinic flux, etc. However, the fact that India is in low latitudes makes all satellite measurement conditions favorable all year round, meaning that the all-year satellite data are of high quality over India. It was found in both the U.S. and Canada that averaging over 3 years (4 summer months per year) of data typically produces a statistically significant mean OMI SO2 map at the 95% confidence level to identify intensive SO2 sources. Therefore, if all-year data can be used, we can generate reliable yearly OMI SO2 maps (e.g., Figure 3) for India instead of requiring “3-year long-term” mean maps for the U.S. and Canada. Using such maps, we can also observe the interannual trend of Indian SO2 emissions, and this will be discussed in detail in the next section.

To quantify the total SO2 amount over a source, Fioletov et al. used a two-dimensional (2D) Gaussian function to fit OMI SO2 signals within a time window and radius:

\[
\text{OMI}_{i,j} = \alpha f(x, y) = \frac{1}{2\pi\sigma_x\sigma_y(1-\rho^2)} \exp\left(-\frac{1}{2(1-\rho^2)} \left(\frac{(x-\mu_x)^2}{\sigma_x^2} + \frac{(y-\mu_y)^2}{\sigma_y^2} - \frac{2\rho(x-\mu_x)(y-\mu_y)}{\sigma_x\sigma_y}\right)\right)
\]

where \(x\) and \(y\) are the coordinates of the valid OMI pixel center; \(\mu_x\) and \(\mu_y\) are the coordinates of the center of the fitted 2D Gaussian function; and \(\sigma_x\) and \(\sigma_y\) are the parameters determining the shape of the fitted 2D Gaussian function. Since the integration of \(f(x, y)\) over the whole surface equals one, the
parameter $\alpha$ physically means the total number of SO$_2$ molecules observed (or the observed SO$_2$ burden) near the source. If OMI$_{SO_2}$ is in units of molecules km$^{-2}$ and $\sigma_x$ and $\sigma_y$ are in km, then $\alpha$ is in units of molecules and can be converted to mass units.

In this work, we follow the same methodology. To quantitatively compare the SO$_2$ emissions from coal-fired power plants with OMI SO$_2$ vertical columns, we first select 23 power plant regions based on the observed satellite signals. Any adjacent plants sharing the same SO$_2$ hotspot location are combined into a single source with total emissions set equal to the sum of their emissions and the location set at their emission-weighted center. Shown as red circles in Figure 2b, in total, 65 coal-fired power plants are included in this analysis, and they together accounted for $\sim$69% of the total SO$_2$ emissions during 2005–2012. We also calculated the proportion of their emissions to total SO$_2$ emissions within a radius of 50 km from weighted centers to ensure they are the dominant contributor to local emissions. Here, gridded emissions of other sources were taken from our previous work$^4$ for years 2005–2010 and scaled to 2012 based on the GAINS inventory.$^6$ Results show that coal-fired power plants contribute $>75\%$ to local SO$_2$ emissions for all 23 regions (and $>90\%$ for 16 regions), clearly indicating that the enhanced OMI SO$_2$ signals over these regions are primarily caused by emissions from coal-fired power plants.

For each power plant area, all the valid OMI pixels within a 60 km radius in a year were used for the 2D Gaussian fitted to obtain the amount of OMI-observed SO$_2$ (i.e., $\alpha$) near the power plant area in that year. Figure 4 shows the scatter plot of annual SO$_2$ emissions against fitted $\alpha$ (i.e., the annual averaged OMI SO$_2$ burden) over all selected power plant areas for the period 2005–2012. We found good agreement between SO$_2$ emissions and OMI-observed SO$_2$ burden over power plant areas in India ($R = 0.93$). Generally, power plant regions with annual SO$_2$ emissions $>50$ Gg year$^{-1}$ produce statistically significant $\alpha$ values. This value is somewhat lower than the threshold of 70 Gg year$^{-1}$ derived for the eastern U.S. by Fioletov et al.$^{27}$ probably because the additional treatments (including local AMF correction, Pacific correction, and dynamic removal of anomalous pixels) to the OMI level-2 product improved the quality of the SO$_2$ retrievals. It should be noted that the slope of the regression line ($\sim$2.2 h) does not represent the chemical lifetime (typically a few days) but represents the effective OMI-observed dispersion time due to advection, deposition, and chemical conversion of SO$_2$ in the atmosphere.$^{27}$ Figure 4 also shows the fitted results obtained by Fioletov et al.$^{27}$ in the U.S. Since they used OMI SO$_2$ retrievals with a constant AMF of 0.36, their results cannot be compared with ours directly; however, we adjusted Fioletov et al.’s fitted line with a local AMF of about 0.7, which is averaged from the GEOS-Chem precomputed local AMF over India.$^{20}$ After correction, the slope of the fitted line for India is still smaller than that for the U.S., implying that the average dispersion of SO$_2$ in India is faster than in the summertime in the U.S. Higher temperatures, more precipitation, a more oxidizing atmosphere, and different wind patterns in India may all contribute to the shorter dispersion time.
Interannual Trends of SO2 over Indian Coal-Fired Power Plants during 2005–2012. OMI SO2 retrievals can be further used to study the interannual trend of SO2 emissions from Indian coal-fired power plants, because one year of data is sufficient to generate a high-quality oversampled OMI SO2 map for India. Figure 3 shows the yearly OMI SO2 maps over India during 2005−2012. A clear increase in SO2 columns can be seen in nearly all hotspots, and new hotspots have come into existence in the past few years, reflecting the fast construction of large power plants/units in India. The yearly sum of OMI SO2 for all power plant regions as a function of the distance between the sources and the pixel centers is given in Figure 5. Although the current OMI SO2 products can only detect the signals over the hotspot centers, a fast increase of SO2 columns can be continuously observed during 2005−2012. The interannual variations of the OMI-observed SO2 burden (i.e., the sum of fitted $\alpha$ and the corresponding 95% confidence intervals), national mean SO2 concentrations reported by the CPCB of Government of India, and annual average SO2 concentrations at selected coal-fired power plant regions are shown in Figure 6. The total amount of SO2 observed by the OMI increased by 63% from 2005 to 2012, consistent with the dramatic increase of SO2 emissions in these regions ($R = 0.93$). The high correlation coefficient further demonstrates the close relationship between the OMI SO2 observations and the bottom-up emissions over coal-fired power plant regions.

Figure 4. Scatter plot of annual SO2 emissions from 23 power plant regions against fitted $\alpha$ (i.e., the OMI-observed SO2 burden) for 2005−2012. Each point represents a yearly fitted $\alpha$ for a source. Actual OMI SO2 measurements are used for the fitting. Error bars express the 95% confidence intervals.

Figure 5. Yearly sum of OMI SO2 for all power plant regions as a function of the distance between the source and the pixel center. Actual OMI SO2 measurements are used for the calculation. Error bars are the 95% confidence intervals of the sum.

Figure 6. Interannual trend of SO2 emissions from selected Indian coal-fired power plant regions, the OMI-observed SO2 burden (the sum of fitted $\alpha$ and the corresponding 95% confidence intervals), national mean SO2 concentrations reported by the CPCB of Government of India, and annual average SO2 concentrations at selected coal-fired power plant regions. $R$ values shown are the correlation coefficients with the OMI-observed SO2 burden.
too few monitors in emission-increasing areas that truly dominate national emissions.

We also noticed a difference between the changes in SO2 emissions and the changes in OMI SO2 observations (or surface SO2 concentrations) between 2005 and 2012. As shown in Figure 6, the OMI SO2 burden and the annual average SO2 concentrations in coal-fired power plant regions increased by more than 60% during 2005–2012, while SO2 emissions from these selected regions increased by only 49%. This is a similar situation to what we previously found for NO2 over Indian power plant areas. We hypothesized that the NO2-induced decreasing OH radical concentration situation to what we previously found for NO2 over Indian power plant areas increased by only 49%. This is a similar conversion efficiency of SO2 to sulfate. As a result, the SO2 emissions in China and sulfur trends in East Asia since 2000. Atmos. Chem. Phys. 2010, 10, 6311–6331.

The authors declare no competing financial interest.

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REFERENCES

(16) Air Quality Monitoring, Emission Inventory and Source Apportionment Study for Indian Cities - National Summary Report; Central Pollution Control Board: Delhi, India, February 1, 2011.
(17) National Ambient Air Quality Status & Trends in India - 2010; Central Pollution Control Board, Ministry of Environment & Forests: Delhi, India, January 1, 2012.


(39) Aura OMI Sulphur Dioxide Level 3 Best Pixel Global (0.25 deg Lat/Lon grids) Data Product-OMSO2e; National Aeronautics and Space Administration; http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omso2e_v003.shtml (June 1, 2013).

