Environmental Science & Technology

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Ozone Monitoring Instrument Observations of Interannual Increases in SO_2 Emissions from Indian Coal-Fired Power Plants during 2005–2012

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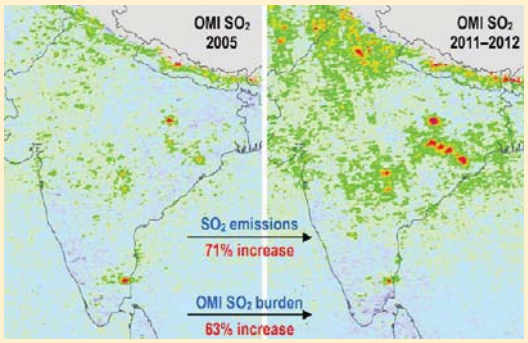
Supporting Information

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.[1−7] On the basis of the most recent emission estimates,[4,8,9] 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.[4] 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.[10,11] 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[1−3,5−7] or presented the year-by-year emission trend for the most recent years.[4,12−14] Following our previous work on NO_x emissions in India,[15] 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.[16,17] This is inconsistent with the fast increase in

Received: September 5, 2013
Revised: October 17, 2013
Accepted: November 25, 2013

   dx.doi.org/10.1021/es4039648 | Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Indian SO_2 emissions, and an independent data source is needed to verify the real SO_2 situation in India. Now, satellite remote sensing instruments such as the Ozone Monitoring Instrument (OMI) make routine SO_2 observations at high temporal and spatial resolution, providing valuable information about surface SO_2 sources.[18] Compared to nitrogen dioxide (NO_2) retrievals, satellite SO_2 signals are very noisy. In the past few years, satellite SO_2 instruments have been only used to detect the global distributions,[19,20] plumes,[21,22] and trends[23,24] of very large anthropogenic SO_2 sources. Oversampling smaller pixels from the center of the swath to fine grids (~3 km wide), de Foy et al.[22] and Russell et al.[25] 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 SO_2 plumes[26] and quantified the SO_2 emissions from individual U.S. power plants[27] that were typically undetectable using standard analysis techniques. Using the same method, SO_2 emissions from Canadian oil sands were also studied.[28,29] In this work, we apply a similar oversampling technique but with improved OMI retrievals to India and, for the first time, study the relationship between OMI SO_2 observations and SO_2 emissions over Indian coal-fired power plants during the OMI era of 2005−2012. We also show that OMI has the capability to monitor the interannual trend of SO_2 emissions from these point sources.

METHODS AND DATA SETS

Unit-Based SO_2 Emission Inventory. In our previous work, we developed a bottom-up, unit-based inventory for NO_x emissions from Indian public coal-fired power plants during 1996−2010.[15] In this study, we adapt the system to calculate annual SO_2 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 NO_x 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, SO_2 control technology, the exact time when the unit came into operation and/or retired, etc. Total SO_2 emission (E) from coal-fired power plants for year i is estimated by the following equation:

$$E_i = \sum_j \sum_k \sum_l 2 \cdot G_{i,j,k} \cdot \mathrm{SCC}_{i,j,k} \cdot S_{j,k} \cdot (1 - \mathrm{SR}_j) \cdot (1 - \eta_l) \quad (1)$$

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 SO_2 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[30] 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[30] and the International Energy Agency (IEA)[31] 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.,[13] GEO, IEA Coal Power Database,[32] and various power project reports. Where plant-specific S values were not available, the state-average S values reported by Reddy and Venkataraman[14] 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.[4,6] Using the default values recommended by the IPCC,[33] the SR ratios were assumed to be 5% for hard-coal-fired power units and 30% for lignite-fired ones. The effects of combustion technology and boiler age on SR ratios were not taken into account since relevant information is not available. Currently, SO_2 emissions are not regulated in India for coal-fired power plants. The Indian government only stipulates that space must be provided for installation of a flue-gas desulfurization (FGD) device for new generating units with capacity 500 MW and above, in case stringent regulations are implemented in the future.[34,35] 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 SO_2 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.[35] The capacity of these two plants accounts for ~2% of the total coal-based capacity in India during 2005−2012.

OMI SO_2 Retrievals and Processing. The OMI is a nadir-viewing imaging 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 SO_2, with daily global coverage since October 2004.[36] 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) SO_2 data in the OMSO2 Level-2 product, developed specifically for anthropogenic SO_2 pollution sources.[37,38] 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-

dx.doi.org/10.1021/es4039648 | Environ. Sci. Technol. XXXX, XXX, XXX−XXX

namically removed these affected pixels based on the RA flags provided in the OMSO2 Level-2 data. We also rejected pixels with SO_2 observations higher than 5 Dobson Unit (DU, 2.69 × 10^{16} molecules cm^{-2}) to exclude cases of transient volcanic SO_2.[27]

For valid pixels, a series of further corrections were applied to the originally retrieved PBL SO_2 values. First, OMI PBL SO_2 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").[20] The corrected slant columns were then further divided by the GEOS-Chem model precomputed clear-sky monthly local AMF to obtain the SO_2 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 SO_2 Level-3 product (i.e., OMSO2e at a resolution of 0.25 degree), and they significantly improve the quality of OMI SO_2 retrievals.[20,39]

The smallest size of the OMI pixel is 13 × 24 km^2, which is too large to observe the SO_2 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 SO_2 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 SO_2 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

SO_2 Emissions of Indian Coal-Fired Power Plants during 2005−2012. Figure 1 shows the annual trend of SO_2 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, SO_2 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 SO_2 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, SO_2 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 SO_2 products can only capture strong anthropogenic point or area plumes,[18] the fact that the majority of SO_2 emission increment in India is from newly installed large power units provides a favorable condition for observing the interannual increasing trend of Indian SO_2 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,

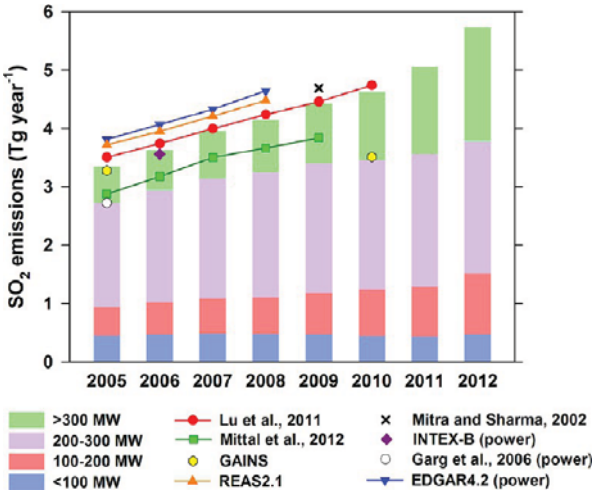


Figure 1. SO_2 emission estimates of Indian coal-fired power plants by unit size (bars) during 2005−2012 and their comparison with previous studies. Emissions of INTEX-B,[7] Garg et al.,[1] and EDGAR4.2[2] are estimates for the whole thermal power sector (i.e., including coal-, oil-, and gas-fired power plants).

Chhattisgarh, Maharashtra, and Tamil Nadu are the six states with power-plant emissions >500 Gg SO_2 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 SO_2 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 SO_2 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 SO_2 emissions are ~±13% around the mean. They are lower than in our previous work[4,40] 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 ones[4] calculated from the IEA national statistics. The discrepancies with other studies are caused by various factors, including the use of higher sulfur content[3] or lower coal consumption,[1,6] the exclusion of captive power plants,[13] and the inclusion of oil-fired and gas-fired plants[2] in their calculations.

SO_2 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 SO_2 maps, and it was reported that comparable SO_2 sources have similar mean OMI SO_2 values.[27] This implies that top-down OMI SO_2 observations can be used to estimate surface emissions and

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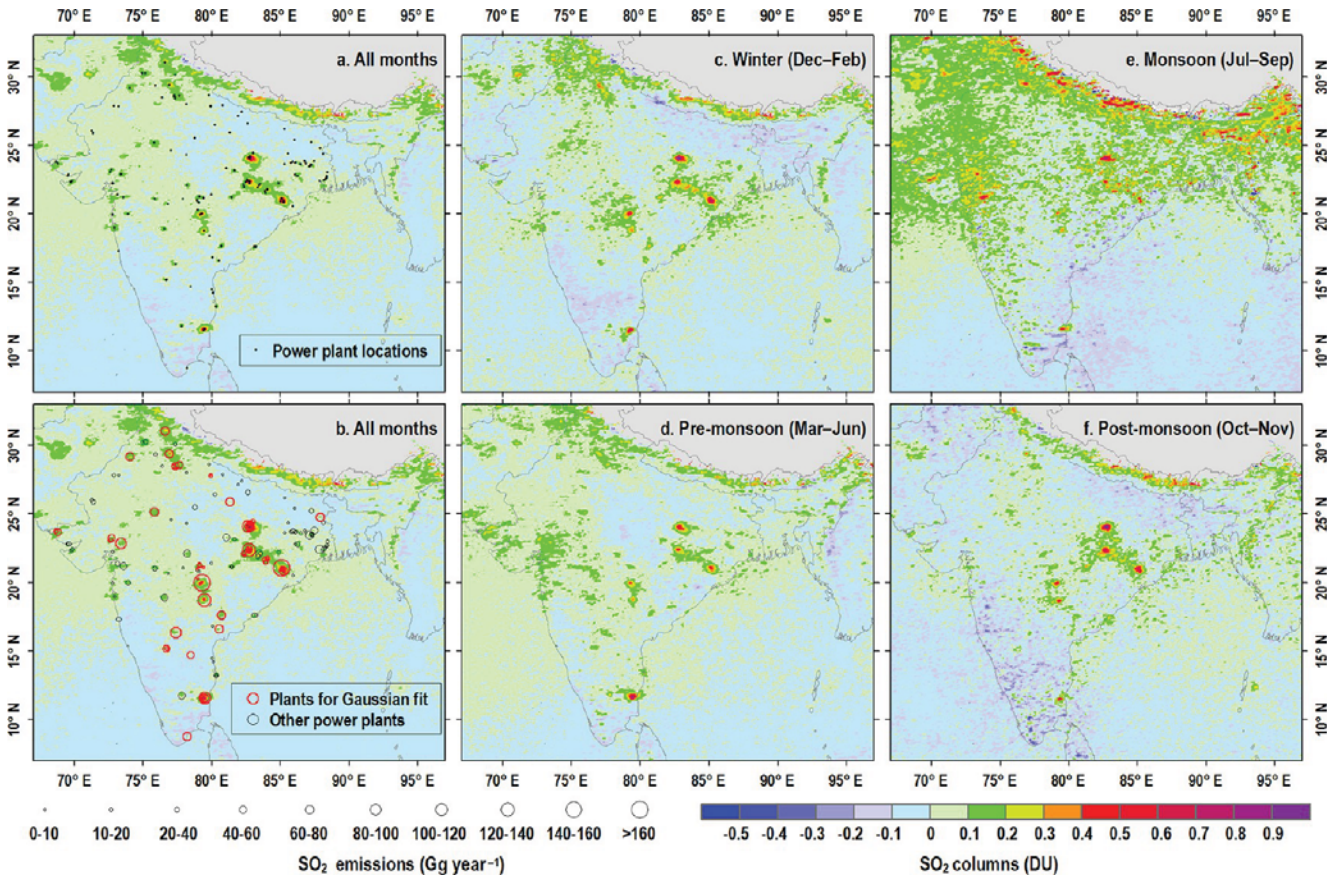

Figure 2. Locations of (a) Indian coal-fired power plants, (b) their average SO_2 emissions, and (a, b) annual and (c–f) seasonal averages of OMI SO_2 columns over India during 2005–2012. (b) Power plants in red are selected for the 2D Gaussian fit.

track their changes. Figure 2 displays the spatial distribution of mean OMI SO_2 over India from 2005 to 2012. A number of satellite SO_2 hot spots are observed, and they match the locations (Figure 2a) and the amounts of SO_2 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, SO_2 in the atmosphere has a chemical lifetime of around one day,[41] much longer than NO_2 (several hours). As shown in Figure 2, however, elevated SO_2 values are found only within ~50 km of the point sources, a distance which is smaller than for OMI NO_2 observations.[15] This further demonstrates that the current OMI SO_2 products are very noisy and only large point[27] or area[28] sources with intensive average emissions can be detected by OMI.

Figure 2c–f also shows the spatial distribution of average OMI SO_2 columns by season[42] over India during 2005–2012. Similar to our previous findings in OMI NO_2 seasonal maps,[15] July to September is the worst period to observe SO_2 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 NO_2, however, we do not observe significant seasonal variations in OMI SO_2 over India probably because the seasonality of the SO_2 lifetime is weaker. In previous studies to quantify the SO_2 emissions from U.S. power plants[27] and Canadian oil sands,[28,29] only May–August SO_2 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.[27] and Canada[28] that averaging over 3 years (4 summer months per year) of data typically produces a statistically significant mean OMI SO_2 map at the 95% confidence level to identify intensive SO_2 sources. Therefore, if all-year data can be used, we can generate reliable yearly OMI SO_2 maps (e.g., Figure 3) for India instead of requiring "3-year long-term" mean maps for the U.S.[27] and Canada.[28] Using such maps, we can also observe the interannual trend of Indian SO_2 emissions, and this will be discussed in detail in the next section.

To quantify the total SO_2 amount over a source, Fioletov et al.[27] used a two-dimensional (2D) Gaussian function to fit OMI SO_2 signals within a time window and radius:

$$\begin{aligned}\mathrm{OMI}_{SO_2} &= \alpha \cdot f(x, y) \\ &= \alpha \cdot \frac{1}{2\pi\sigma_x\sigma_y\sqrt{1-\rho^2}} \exp\left(-\frac{1}{2(1-\rho^2)}\left[\frac{(x-\mu_x)^2}{\sigma_x^2} + \frac{(y-\mu_y)^2}{\sigma_y^2} - \frac{2\rho(x-\mu_x)(y-\mu_y)}{\sigma_x\sigma_y}\right]\right)\end{aligned} \quad (2)$$

where x and y are the coordinates of the valid OMI pixel center; μ_x and μ_y are the coordinates of the center of the fitted 2D Gaussian function; and σ_x, σ_y, and ρ 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

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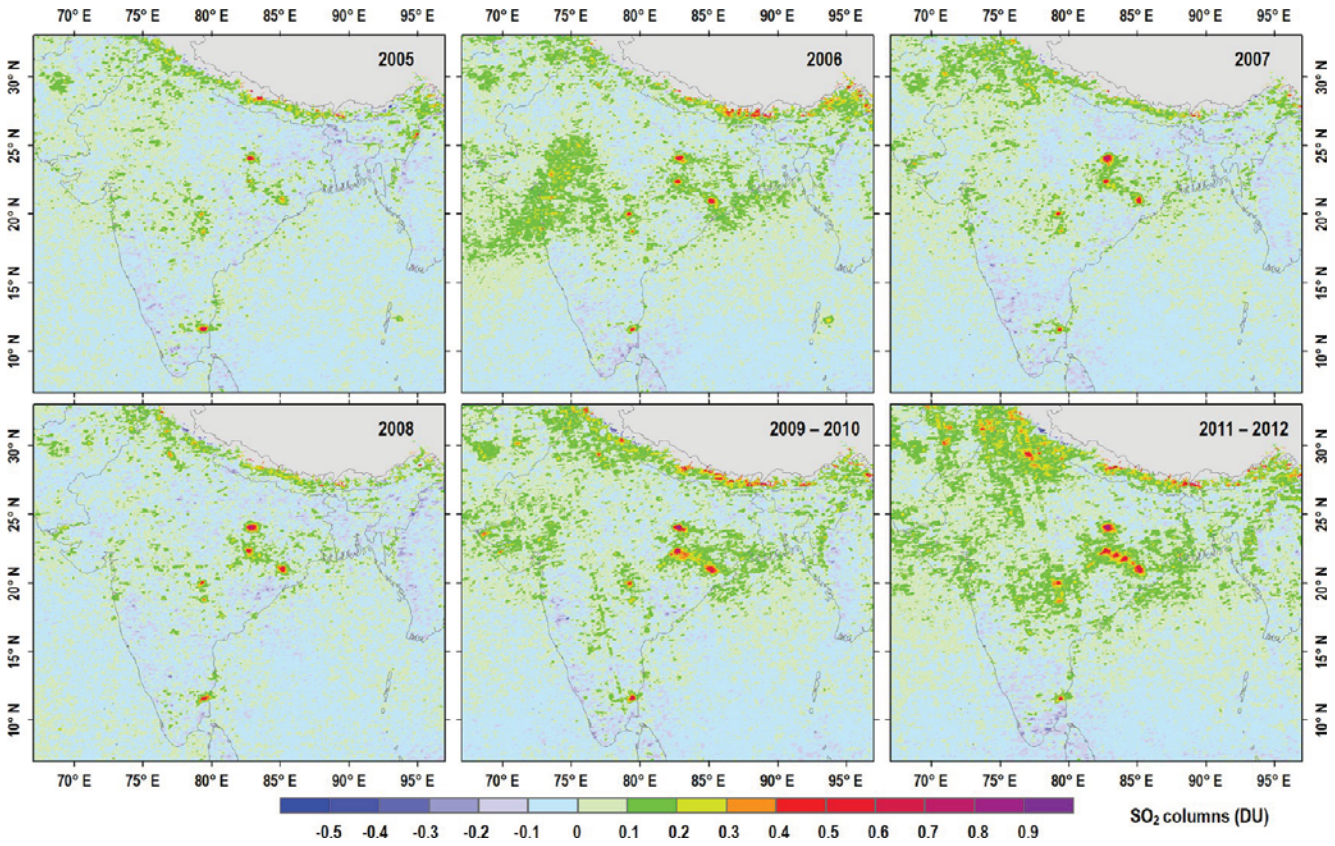

Figure 3. Spatial distribution of yearly OMI SO_2 columns over India during 2005−2012. OMI SO_2 maps for years 2009−2012 are combined biennially to make the satellite sample amounts similar to years 2005−2008 because, due to the row anomaly, about half of the track positions were operational for 2009−2012 compared to 2005−2008.

parameter α 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 σ_x and σ_y are in km, then α 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 ∼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 fit to obtain the amount of OMI-observed SO_2 (i.e., α) near the power plant area in that year. Figure 4 shows the scatter plot of annual SO_2 emissions against fitted α (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 α 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 (∼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.

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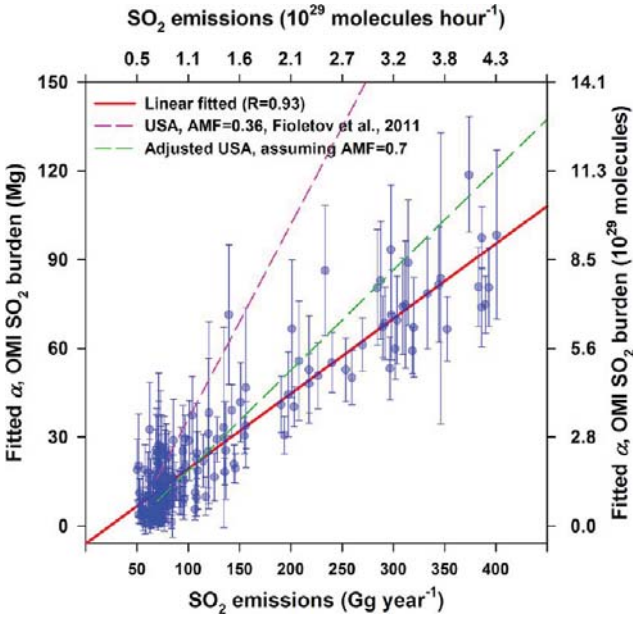

Figure 4. Scatter plot of annual SO_2 emissions from 23 power plant regions against fitted α (i.e., the OMI-observed SO_2 burden) for 2005−2012. Each point represents a yearly fitted α for a source. Actual OMI SO_2 measurements are used for the fitting. Error bars express the 95% confidence intervals.

Interannual Trends of SO_2 over Indian Coal-Fired Power Plants during 2005−2012. OMI SO_2 retrievals can be further used to study the interannual trend of SO_2 emissions from Indian coal-fired power plants, because one year of data is sufficient to generate a high-quality oversampled OMI SO_2 map for India. Figure 3 shows the yearly OMI SO_2 maps over India during 2005−2012. A clear increase in SO_2 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 SO_2 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 SO_2 products can only detect the elevated SO_2 values within a radius of ~60 km, the sum of OMI signals over the hotspot centers was continuously increasing during 2005−2012. The interannual variations of the OMI-observed SO_2 burden (i.e., the sum of fitted α) and SO_2 emissions of all selected power plant regions are shown in Figure 6. The total amount of SO_2 observed by the OMI increased by 63% from 2005 to 2012, consistent with the dramatic increase of SO_2 emissions in these regions (R = 0.93). The high correlation coefficient further demonstrates the close relationship between the OMI SO_2 observations and the bottom-up emissions over coal-fired power plant regions.

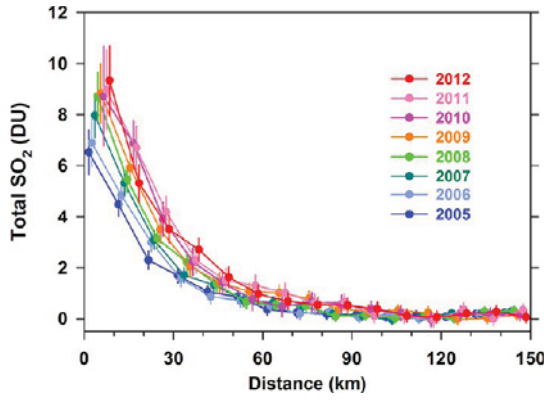

Figure 5. Yearly sum of OMI SO_2 for all power plant regions as a function of the distance between the source and the pixel center. Actual OMI SO_2 measurements are used for the calculation. Error bars are the 95% confidence intervals of the sum.

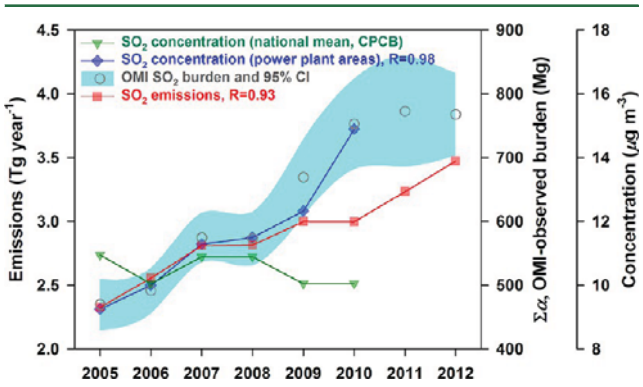

Figure 6. Interannual trend of SO_2 emissions from selected Indian coal-fired power plant regions, the OMI-observed SO_2 burden (the sum of fitted α and the corresponding 95% confidence intervals), national mean SO_2 concentrations reported by the CPCB of Government of India, and annual average SO_2 concentrations at selected coal-fired power plant regions. R values shown are the correlation coefficients with the OMI-observed SO_2 burden.

Figure 6 also shows the trend of national mean SO_2 concentrations reported by the Central Pollution Control Board (CPCB), Government of India.[17] These data were derived from the CPCB's National Air Quality Monitoring Programme (NAMP), a network of 320+ operating stations in 120+ cities/towns during 2005−2010 (http://cpcbedb.nic.in). The national mean of SO_2 levels reported by the Indian Government shows a decreasing trend, which is explained by recent policy measures such as a reduction of the sulfur content of diesel, the use of cleaner fuels in metropolitan areas, a change in domestic fuel from coal to liquefied petroleum gas (LPG), etc.[16,17] However, this declining national concentration trend reported officially differs markedly from the trends of both OMI SO_2 observations and national SO_2 emissions.[2−6,13] The reason for the difference is that most of the CPCB-NAMP stations are situated in large cities having few power plants. For further analysis, we selected those CPCB-NAMP stations that are located in our coal-fired power plant regions and have continuous SO_2 records between 2005 and 2010. In total, there are 70 stations in 27 cities that meet these two conditions (see Table S2 of the SI for the station list), and the annual average SO_2 concentrations for these 70 stations are shown in Figure 6. Clearly, the trend of surface SO_2 levels in the power plant regions is in excellent agreement with the OMI observations (R = 0.98) and both of them increased by ~60% during 2005−2010. The above analysis clearly indicates that the network of the CPCB-NAMP stations needs to be optimized to reflect the true SO_2 situation across the country. Current government reports and statistics may give a misleading impression that the SO_2 situation is improving. In general, there are too many monitors located in city centers where SO_2 pollution might be reduced due to some local control measures, whereas there are

dx.doi.org/10.1021/es4039648 | Environ. Sci. Technol. XXXX, XXX, XXX−XXX

too few monitors in emission-increasing areas that truly dominate national emissions.

We also noticed a difference between the changes in SO_2 emissions and the changes in OMI SO_2 observations (or surface SO_2 concentrations) between 2005 and 2012. As shown in Figure 6, the OMI SO_2 burden and the annual average SO_2 concentrations in coal-fired power plant regions increased by more than 60% during 2005−2012, while SO_2 emissions from these selected regions increased by only 49%. This is a similar situation to what we previously found for NO_2 over Indian power plant areas after 2005:[15] the changes in surface NO_2 are greater than the changes in NO_x emissions. For NO_2, we attributed this to possible changes in the overall NO_x chemistry over Indian power plant areas. We hypothesized that the continuous increase in NO_x emissions enhances OH radical production through chemical feedback and thus decreases the NO_x lifetime before 2005 but consumes OH radicals and thus increases the NO_x lifetime after 2005. For SO_2, it is possible that the NO_2-induced decreasing OH radical concentration after 2005 also reduced the oxidation of SO_2 and decreased the conversion efficiency of SO_2 to sulfate. As a result, the SO_2 concentrations increased at a greater rate than the emissions. Large industrial emitters near the power-plant regions, which are not taken into account in this study, may also contribute to the discrepancy. Further model simulations are needed to explore the synergistic effects of the high growth of both NO_x and SO_2 emissions on local atmospheric chemistry, as well as the consequent impacts on human health, air quality, and regional and global climate.

■ ASSOCIATED CONTENT

Supporting Information

(1) Electricity generation and coal consumption of Indian coal-fired power plants (Figure S1); (2) SO_2 emissions by state and by region (Table S1); (3) air quality monitoring stations selected to calculate the average SO_2 concentrations in coal-fired power plant regions (Table S2). This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was sponsored by the National Aeronautics and Space Administration (NASA) as part of the Air Quality Applied Sciences Team (AQAST) program. The India emission inventory was partially funded in support of the Ganges Valley Aerosol Experiment (GVAX) by the Office of Biological and Environmental Research in the U.S. Department of Energy, Office of Science. Argonne National Laboratory is operated by UChicago Argonne, LLC, under Contract No. DE-AC02-06CH11357 with the U.S. Department of Energy.

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