Seasonal Evolution of Sulfur Dioxide Over the Indian Subcontinent

Lakhima Chutia, Narendra Ojha, Imran A. Girach, Binita Pathak, Lokesh K. Sahu, and Pradip K. Bhuyan

Abstract – Reanalysis, which combines the chemical-transport model with remote-sensing measurements, has shown potential to fill data gaps over observationally sparse regions of the globe. Here, the seasonal distribution of sulfur dioxide (SO_2) over the Indian region was analyzed for 2005–2015 period using Copernicus Atmosphere Monitoring Service (CAMS) reanalysis. CAMS reproduced general features and seasonality observed in surface SO_2 over this region. Elevated levels were revealed across the Indo-Gangetic Plain (IGP) and over eastern and central India. SO_2 shows a prominent seasonality over India, with a maximum typically during winter and a minimum during the summer monsoon. The winter maximum is attributed to weaker chemical sink, stagnant meteorological conditions, and elevated emissions, whereas wet scavenging, inflow of marine air, and stronger sulfate formation efficiency cause lower SO_2 during the monsoon. SO_2 levels exhibited an enhancement over IGP and central and eastern India, with rates in the range of 0.5 ppbv to 4 ppbv per decade. Like the distribution, trends also showed seasonal dependence, with weaker trends during the summer monsoon and stronger trends during the winter and postmonsoon. In situ observations of SO_2 and related species especially over the identified hot spots are recommended to validate satellite and model data sets and evaluate air-quality and climate implications.

1. Introduction

Sulfur dioxide (SO_2) is an air pollutant with adverse health effects, mostly produced by the combustion of fossil fuels [1]. As a key air pollutant and aerosol precursor, SO_2 affects air quality, tropospheric chemistry, and the climate [2]. The chemistry of SO_2 in the troposphere is linked with gas-to-particle conversion processes—e.g., SO_2 oxidation into sulfate aerosols—as well as formation from reduced sulfur compounds [3]. Sulfate aerosols substantially influence the earth's radiation budget by scattering solar radiation and acting as cloud condensation nuclei [4, 5]. The dominant pathway of sulfate formation is through aqueous-phase oxidation of SO_2 by hydrogen peroxide and ozone [3]. In the gas phase, SO_2 is oxidized by hydroxyl radicals (OH) to sulfur trioxide, which further leads to the formation of sulfuric acid [3].

The power-generation sector is the largest contributor (∼46%) to SO_2 emissions in India, followed by the industrial and residential sectors [6]. Due to its relatively shorter lifetime and diverse emission sources, SO_2 distribution shows strong spatial heterogeneity as reported from different environments in the Indian region [7–9]. The distribution is also affected by chemistry, emissions, and seasonal changes in meteorological conditions. Due to sparse in situ measurements, an accurate understanding of seasonal changes in SO_2 distribution and its chemical transformations is still an intriguing challenge over the Indian region. Limited studies based on satellite observations revealed enhancements in SO_2 loading over the Indian region, particularly over the states of Chhattisgarh and Odisha, during the 2005–2015 period [10]. Model reanalysis, combining the chemical-transport model and remote-sensing measurements, has shown the potential to fill data gaps over observationally sparse regions of the globe [11, 12]. However, studies evaluating model reanalysis data sets against observations, seasonality in SO_2 levels, and its conversion into sulfate using both observations as well as model reanalysis have been still lacking over the Indian region.

Here we consider a study period of 10 years (2005–2015) to investigate seasonal variability as well as seasonal trends in SO_2 over the Indian region by analyzing Copernicus Atmosphere Monitoring Service (CAMS; https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4-monthly?tab=form) model reanalysis and Ozone Monitoring Instrument (OMI, https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura_OMI_Level3/OMSO2e.003/) satellite observations. We have estimated the efficiency of sulfate formation for different seasons.

2. Data and Methodology

2.1 CAMS Model

CAMS reanalysis provides consistent gridded fields of atmospheric composition based on an integrat-

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ed atmospheric-modeling and data-assimilation system. CAMS employs the Carbon Bond 5 mechanism (CB05) chemistry module, comprising 54 chemical species and 126 chemical reactions [11, 13]. The aerosol and chemistry schemes in CB05 are independent in that both carry their own SO_2 variable [14]. Anthropogenic emissions of SO_2 are based on the Monitoring Atmospheric Composition and Climate/City ZEN EU projects (MACCity; https://eccad3.sedoo.fr) inventory available globally on a $0.5° \times 0.5°$ grid. The chemical fields simulated by the CAMS model available at 80 km $\times$ 80 km resolution have been analyzed in this study.

2.2 Observational Data Sets

The present study used ground-based SO_2 data reported from eight stations across the Indian subcontinent for evaluation of the model reanalysis: Nainital [15], Delhi [16], Dibrugarh [17], Kanpur [7], Patna [8], Kolkata [18], Ahmedabad [19], and Bhubaneswar [9]. Ground-based SO_2 measurements have generally been performed using online analyzers based on a UV fluorescence technique.

In addition, remote-sensing based observations of the planetary boundary layer (PBL) SO_2 column from the Ozone Monitoring Instrument (OMI) are analyzed here. OMI's onboard Aura is a nadir-viewing ultraviolet/visible spectrometer, which provides global coverage every day at a horizontal resolution of 13 km $\times$ 24 km, with a local equator crossing time at 13:45. The OMI PBL SO_2 product employs an algorithm based on principal-component analysis that reduces retrieval noise by a factor of 2 [20].

2.3 Study Region

In this study, we divided the Indian subcontinent into five different subregions—North India (NI: 26°–37.5° N, 70°–82° E), East India (EI: 18°–30° N, 82°–98° E), Central India (CI: 18°–26° N, 75°–82° E), West India (WI: 18°–26° N, 68°–75° E), and Southern India (SI: 8°–18° N, 73°–83° E)—based on the diversity of emissions, topography, and meteorology. Such classifications of different regions in the Indian subcontinent were also used in earlier studies, for example to study variations in ozone and volatile organic compounds [21, 22].

3. Results and Discussion

3.1 Model Evaluation

The performance of CAMS reanalysis was evaluated in various studies globally [11, 12]. Here we briefly describe the comparison of CAMS data with ground-based studies over the Indian region. CAMS SO_2 captures observed variations to an extent, with a correlation coefficient > 0.55. Seasonal changes in SO_2 have been reproduced by CAMS over the stations, including Ahmedabad, Patna, and Bhubaneswar. CAMS, in agreement with measurements, showed higher levels during winter/postmonsoon and lower levels during the summer monsoon. Observed SO_2 values were higher in the monsoon season as compared to the winter season at complex topographical stations like Dibrugarh and Nainital, which is not reflected in the model simulation. The differences between model and observational data are suggested to be associated with uncertainties in input emissions and lack of SO_2 assimilation in the model. Emission inventory data are not always consistent, particularly over rapidly developing countries such as India [23]. The lack of spatially resolved core activity data, emission factors, and removal efficiencies for different technologies create major uncertainties in emissions.

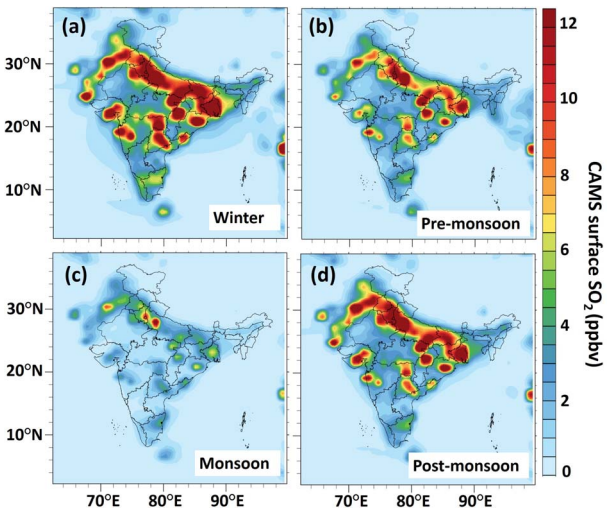

Figure 1. Mean distributions of surface SO_2 from CAMS reanalysis over the Indian region during (a) winter (DJF), (b) premonsoon (MAM), (c) monsoon (JJA) and (d) postmonsoon (SON) for 2005–2015 period.

Different CAMS simulations were evaluated and discussed elsewhere [11, 13, 24, 25]. In the following sections, we analyze CAMS data to investigate the seasonal distribution and trends in SO_2 over the Indian region.

3.2 Seasonal Distribution

Spatial distributions of surface SO_2 from CAMS reanalysis during winter (December-January-February; DJF), premonsoon (March-April-May; MAM), monsoon (June-July-August; JJA), and postmonsoon (September-October-November; SON) are shown in Figure 1. SO_2 levels are particularly higher over the Indo-Gangetic Plain (IGP) and the east and central parts of India compared to the southern region. Seasonally, SO_2 levels are high during winter, decline during the premonsoon, and attain their lowest values during the monsoon. A recovery of higher values can be seen again during the postmonsoon. Further, these regional and seasonal differences over different subregions of the Indian subcontinent are analyzed quantitatively in section 2.3 and [21]. The month-to-month changes

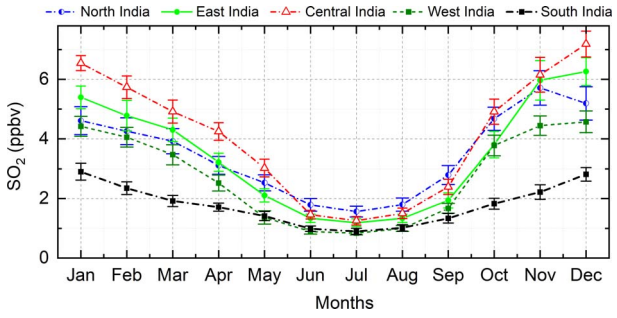

Figure 2. Monthly variation of SO_2 over different subregions of the Indian subcontinent during 2005–2015 based on CAMS reanalysis.

analyzed over five different subregions are shown in Figure 2.

The highest mean SO_2 mixing ratios (3 ppbv to 7.2 ppbv) can be noticed over most of the subregions during December, whereas the lowest levels are seen during July (0.8 ppbv to 1.5 ppbv). Monthly mean SO_2 levels (1.2 ppbv to 7.2 ppbv) are found to be higher over Central India, owing to the enhanced energy generation and industrialization, and lower (<3 ppbv) over Southern India.

The seasonal distributions of CAMS surface SO_2 and OMI PBL SO_2 (Figure 3) are more or less the same, but elevated loading particularly over Chhattisgarh and Odisha, is evident in the satellite observations. Differences between reanalysis and satellite are suggested to be due to differences in the vertical layers (surface versus boundary layer) and uncertainties in retrievals and model inputs. Nevertheless, stronger SO_2 levels during winter are seen in both reanalysis and satellite data. Large seasonal variability is due to the combined effect of multiple processes, such as chemistry, emissions, meteorological conditions, and boundary-layer dynamics [26]. Higher fuel usage and shallower boundary layer/stagnant condition contribute to higher levels of SO_2 during winter. Another important factor that causes higher levels of SO_2 during winter is the weak chemical sink, which could contribute to less-efficient SO_2 oxidation. During winter, OH radicals are expected to be low due to the lower photochemical activity resulting from low incoming solar flux. SO_2 levels are lowest during the summer monsoon due to efficient wet scavenging by precipitation and transport of cleaner air diluting the pollutants. Additionally, higher abundances of atmospheric oxidant (OH) can enhance the conversion of SO_2 to sulfate.

To further corroborate this result, CAMS data were analyzed to estimate the sulfate formation from SO_2 using the sulfate/SO_x ratio as an indicator of formation efficiency. The sulfate formation efficiency can be represented by $\in_{av} = RSO_x \times [\text{sulfate}/SO_x]$, where RSO_x is the remaining fraction of SO_x [27]. RSO_x is computed using the linear regression slope of the correlation between SO_x and CO from CAMS reanalysis and the emission ratio of SO_2 to CO from the MACCity inventory. Sulfate formation shows significant seasonal

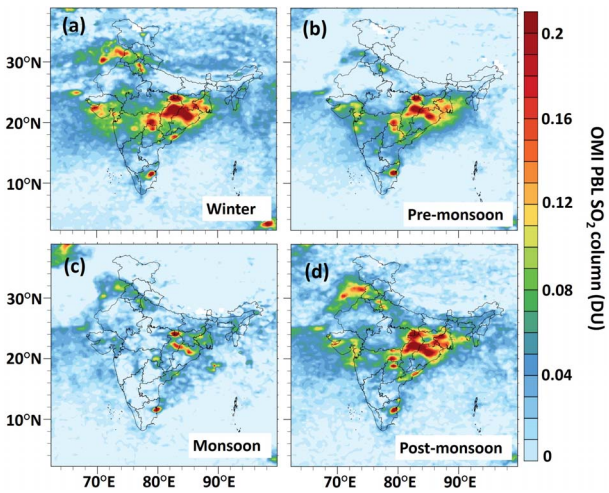

Figure 3. Mean distributions of PBL SO_2 over the Indian region derived from OMI during (a) winter (DJF), (b) premonsoon (MAM), (c) monsoon (JJA) and (d) postmonsoon (SON) for 2005–2015 period.

variation, with higher efficiency during monsoon season (0.30) and lower during the winter (0.15). This implies a 30% conversion of total SO_2 to sulfate during monsoon season and 15% conversion during winter. This would contribute to lower and higher values of SO_2 during monsoon season and winter, respectively. Higher sulfate formation during monsoon season is attributed to stronger SO_2 oxidation in the ample availability of atmospheric oxidants plus warm humid atmospheric conditions. Contrarily, during winter, limited atmospheric oxidants could reduce the formation efficiency.

3.3 Long-Term Trends

CAMS SO_2 trends over the Indian region were examined during four different seasons (Figure 4). SO_2 shows statistically significant increasing trends in the range of 0.5 ppbv to 4 ppbv per decade for all seasons over the Indian region. Increasing trends are found to be stronger over the IGP, including the industrial regions. Trends are particularly larger (1.5 ppbv to 4 ppbv per decade) during the winter and postmonsoon seasons, whereas the summer monsoon season exhibits a weaker trend (0.5 ppbv to 2 ppbv per decade) during 2005–2015, corroborating with its seasonal variation. There are some spots of decline during the winter season over some parts of West and Central India. CAMS seasonal trends are corroborated with the satellite-based trend in the PBL SO_2 column (figure not shown). OMI observations show a significant increase in SO_2 over East and Central India for all seasons, especially over Chhattisgarh and Odisha (> 0.2 decade/year, $p < 0.05$) during 2005–2015, consistent with the model reanalysis. However, a decline is seen in the PBL SO_2 column over some regions in North and West India during the premonsoon and postmonsoon seasons, which is in contrast with the model results.

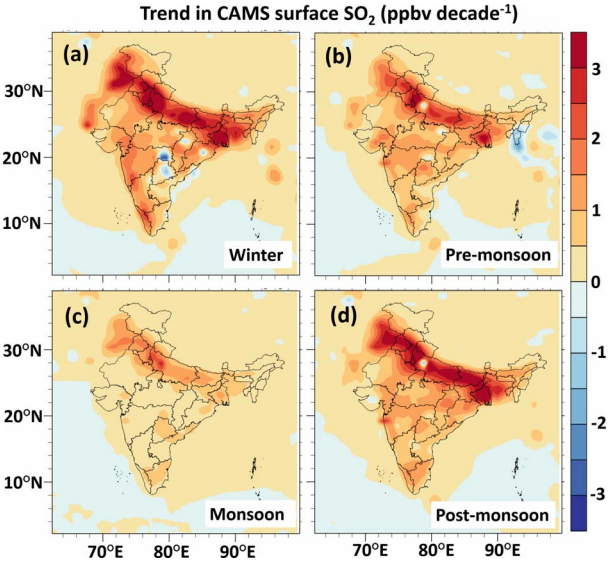

Figure 4. SO_2 trends over the Indian region during (a) winter (DJF), (b) premonsoon (MAM), (c) monsoon (JJA) and (d) postmonsoon (SON) for 2005–2015 period.

Overall, SO_2 enhancements are found to be in agreement with the changes in anthropogenic emissions over the study region, such as in the MACCity and CAMS inventories. This is attributed to an increase in the total installed capacity and electricity generation by coal-fired power plants during 2005–2015 over the Indian region. We additionally analyzed sulfate variation (not shown) from the CAMS reanalysis. In line with the SO_2 changes, sulfate concentrations show a substantial increase with particularly strong trends of 1 $\mu g/m^3$ to 5 $\mu g/m^3$ per decade over the IGP and East and Central India. Similar to the SO_2 trend, sulfate shows a stronger trend during winter and a weaker trend during monsoon season.

4. Summary and Conclusions

In this study, seasonality, long-term trends, and sulfate formation efficiency of SO_2 over the Indian region were investigated using model reanalysis (CAMS). The major findings of this study are the following:

- SO_2 showed distinct seasonal differences, with higher levels (∼3 ppbv to 8 ppbv) during winter and lower levels (∼1 ppbv to 2 ppbv) during monsoon season. Lower OH, stronger emissions, and stagnant meteorological conditions cause higher SO_2 during winter, whereas higher sulfate formation efficiency, wet scavenging, and inflow of marine air result in lower SO_2 during monsoon season.
- Significant SO_2 enhancements were seen particularly over industrial and energy-generation areas during 2005–2015, with weaker trends (0.5 ppbv to 2 ppbv per decade) during the summer/monsoon season and stronger trends (1.5 ppbv to 4 ppbv per decade) during the winter/postmonsoon season.
- Systematic observations together with development of up-to-date regional emission inventories are needed for assessments of air quality and climatic implications of SO_2 changes over South Asia and downwind regions.

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6. References

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