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Distribution of sulfur dioxide over Indian subcontinent: Remote sensing observations and model reanalysis

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Abstract

Sulfur dioxide (SO_2) is a short-lived reactive trace gas, which plays vital roles in air quality, tropospheric chemistry and climate. Rapid changes in the technologies and expansion of energy sector is anticipated to have influenced the distribution of SO_2 across the Indian subcontinent, however continuous in situ observations are scarce. In this direction, we investigate the spatio-temporal distribution of SO_2 over the Indian region combining the remote sensing observations from space (OMI), in situ measurements (CPCB), and results from the global models (MERRA-2 and CAMS) for 2005–2015 period. The comparison of MERRA-2 results with surface SO_2 observations show model's ability to reproduce the general aspects of SO_2 distribution over this region. Analysis of the long-term MERRA-2 and CAMS simulations reveal an enhancement in the SO_2 levels, more pronounced over the industrial regions in the north, east and central India at rates in range of 0.1–0.3 ppbv yr^{-1} during 2005-2015 period. This concord well with the trends based on satellite-based observations and emission inventories over this region. These changes in SO_2 are further found to impact the regional distribution of the sulphate aerosols significantly.We estimated higher efficiency of sulphate formation during monsoon (0.30) than that during winter (0.15). This study highlights a need to conduct in situ observations of SO_2 over a network of stations especially in the identified regions of stronger SO_2 enhancements to understand the impacts on the regional air quality and climate.

Keywords: Sulfur, MERRA-2 reanalysis, CAMS, OMI, sulphate aerosols, modeling, South Asia, satellite-data

1. Introduction

SO_2 is a predominant sulfur containing air pollutant which also plays key roles in the tropospheric chemistry and aerosol formation. As a pollutant and precursor for sulphate aerosols, SO_2 affects the air quality, atmospheric visibility, and climate [1]. Sulphate aerosols induce a cooling on the atmosphere by backscattering solar radiation [2] and increasing the cloud albedo [3] and the lifetime of cloud [4]. It is produced mainly through the oxidation of SO_2 with OH radical in the gas-phase, and from conversion of SO_2 by H_2O_2 and O_3 in the aqueous phase [5]. Globally, anthropogenic sources are the major contributors (more than 70%) to the total SO_2 emissions such as the fossil-fuel combustion. Coal-fired power plants are suggested to be contributing ~ 46–69% of total SO_2 emissions in India [6, 7]. Additionally, strong contrasts in the variations of SO_2 were reported from different environments in Indian region [8, 9, 10]. However, these studies have been limited in space and time to derive the spatial distribution, evaluate the performance of atmospheric models, and to examine the trends covering the entire region.

In this direction, we investigated the distribution of SO_2 over the Indian region by analyses of the space-based (OMI) and ground-based measurements (CPCB) together with global model reanalysis (MERRA-2 and CAMS) during 2005-2015 period. Further production of sulphate aerosols over the Indian subcontinent is also studied.

2. Data and Methodology

2.1 Model datasets

SO_2 and sulphate fields based on the model reanalysis from the Modern-Era Retrospective analysis for Research and Applications, version 2 (MERRA-2) produced by NASA Global Modeling and Assimilation Office (GMAO), using Goddard Earth Observing System Model, version 5 (GEOS-5) [11] have been analyzed. MERRA-2 includes several improvements over its predecessor (MERRA-1) and provides a gridded, homogeneous record of the global atmosphere at a high spatial (½° latitude by ⅝° longitude by 72 model levels) and temporal resolution from 1980 to the present.

In addition, SO_2 and sulphate simulations from the Copernicus Atmosphere Monitoring Service (CAMS) model available at 80 km x 80 km resolution are used. CAMS reanalysis provides a consistent information on the atmospheric composition during 2003-2018 period based on an integrated atmospheric modelling and data assimilation system [12, 13]. CAMS model inputs included the anthropogenic emissions from MACCity, biomass burning emissions from the CAMS Global Fire Assimilation System, and biogenic emissions from the MEGAN model.

2.2 Satellite data: OMI

Remote-sensing based observations of planetary boundary layer (PBL) SO_2 column with grid size of $0.25^o \times 0.25^o$ obtained from the Ozone Monitoring Instrument (OMI) - Level-3 has been analyzed in this study. OMI instrument, onboard Aura satellite, is a nadir-viewing ultraviolet/visible spectrometer, and it provides global coverage everyday with spatial resolution of 13 km × 24 km at nadir with a local equator crossing time at 13:45 LT. Principal component analysis (PCA) based algorithm is utilized to compute SO_2 by minimizing the spectral interferences. More details on OMI based observations of SO_2 can be found elsewhere [14, 15].

2.3 Ground based measurements

Present study uses ground-based SO_2 data from seven different stations in the Indian region: Nainital [16]; Delhi [17]; Dibrugarh [18]; Kanpur [8]; Patna [9]; Kolkata [19]; Ahmedabad [20]. Moreover, annual SO_2 data for the year 2015 from the National Air Quality Monitoring Programme (NAMP) of CPCB (https://cpcb.nic.in/namp-data/) at 81 locations across the Indian region has also been utilized for model evaluation.

3. Results and Discussion

3.1 Model evaluation

Figure 1 shows the correlation between MERRA-2 model results and in situ measurements. MERRA-2 shows a good agreement with NAMP measurements over the Indian region with a correlation coefficient of 0.7. SO_2 abundances over most of the locations are also generally distributed between the y = 0.5x and y = 2x lines, showing an agreement between the model reanalysis and observations within a factor of 2. This evaluation result indicates the ability of the MERRA-2 in reproducing the observations of SO_2 over Indian region and that the reanalysis results can be used to investigate the spatio-temporal distribution of SO_2 and trends over this region.

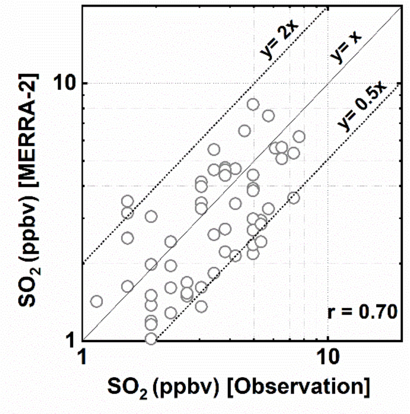

Figure 1: Correlation between model reanalysis (MERRA-2) and in situ observations of SO_2 over the Indian region during 2015.

3.2 Seasonal variation of SO_2

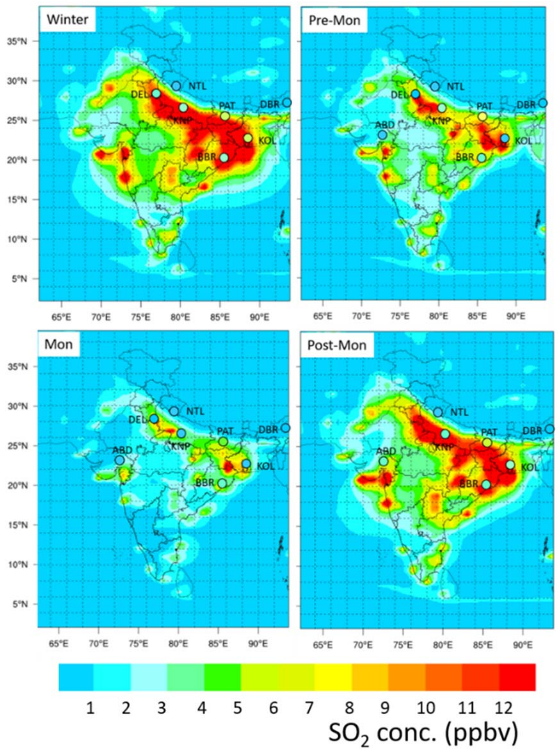

Figure 2: Mean distribution of SO_2 levels from model reanalysis (MERRA-2) during winter (DJF), pre-monsoon (MAM), monsoon (JJA) and post-monsoon (SON) during the period 2005–2015 over Indian region. Observations (circles) are shown on same color scale for a comparison.

The spatial distribution of MERRA-2 simulated surface SO_2 during four different seasons: winter (DJF), pre-monsoon (MAM), monsoon (JJA) and post-monsoon (SON) for the period 2005–2015 over Indian region is shown in the Figure 2. Model results clearly show peak SO_2 levels during winter, a reduction during pre-monsoon, lowest levels during monsoon and an increase again during post monsoon. These changes are mainly attributed to the variations in the boundary layer dynamics, transport, and emissions. Higher SO_2 levels are seen over the Indo-Gangetic Plain (IGP), east and central part of India whereas lower levels are seen over the southern region (latitude < ~16° N). High fuel usage and less efficient oxidation to sulphate can result in elevated SO_2 levels in the winter. Due to mixing with cleaner marine air masses and precipitation, SO_2 levels are lowest during summer-monsoon. Further, MERRA-2 SO_2 distributions show a general agreement with the previous studies reporting observations, except at a few stations (e.g. Delhi with MNB of 138%; Kanpur with MNB of 96% etc.). Overall, we find that MERRA-2 model shows an ability to simulate the SO_2 distribution over the Indian region. In following section, the MERRA-2 fields are analyzed to evaluate the long-term changes in SO_2 and the results are compared with CAMS reanalysis and satellite-based observations.

3.3 Long-term trend in SO_2 and sulphate

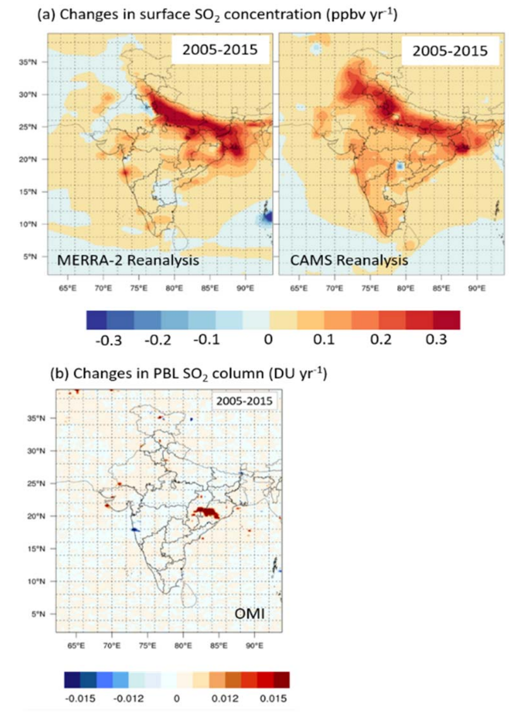

Figure 3: Long-term trends in **(a)** MERRA-2 and CAMS simulated surface SO_2 **(b)** OMI PBL-SO_2 column during 2005-2015 period over Indian region

The grid-by-grid trend analysis of surface SO_2 over Indian region during 2005-2015 using MERRA -2 and CAMS reanalysis is carried out. MERRA-2 shows a general enhancement in surface SO_2 levels over the industrial regions in north, east and central India at a rate of 0.1–0.3 ppbv yr^{-1} during 2005-2015 in agreement with results based on the CAMS reanalysis. Interestingly, OMI observations also reveal an increase in the PBL columnar SO_2 over east and central India especially over Chhattisgarh and Orissa (> 0.04 ppbv yr^{-1}, $P<0.05$) during 2005-2015 (Figure 3b), in agreement with the long term trends reported in other studies [15]. An enhancement in SO_2 emissions from energy sector (power generation) has been seen during 2005-2015 in the MACCity and CAMS inventories. A substantial increase in sulphate concentration is also observed over Indian subcontinent during 2005-2015 in line with the changes in the SO_2. Stronger increases are generally found over the industrialized areas in north, east and central India (0.3–0.5 μgm^{-3} yr^{-1}) (Figure 4).

We use the MERRA-2 results to compute the efficiency of sulphate formation from SO_2 during different seasons over Indian region (19-26 °N, 81-89 °E) using [sulphate/SO_x] ratio. Kaneyasu et al. (1995) [21] have considered ratio of sulphate aerosol to SOx > 0.1 as an indicator of the sulphate formation in the atmosphere which has been applied successfully in several studies [22]. Estimations using MERRA-2 shows a significant seasonal variation in sulphate formation with higher efficiency during monsoon (0.30) and lower during winter (0.15). Higher sulfate formation during monsoon is suggested to be due to stronger SO_2 oxidation with lager availability of atmospheric oxidants in higher temperature and humidity conditions. We recommend systematic observations of SO_2, sulphate, and meteorological parameters to evaluate these model-based estimations.

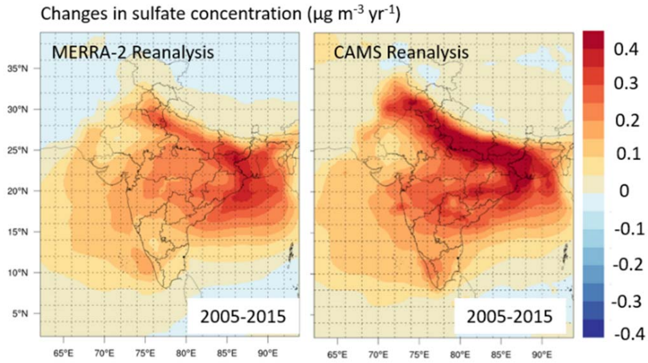

Figure 4: Trend in MERRA-2 and CAMS simulated sulphate concentrations during 2005-2015 period over Indian region

4. Summary and conclusions

In this study, the distribution and long-term (2005–2015) changes in SO_2 over Indian subcontinent has been investigated by combining space-based (OMI) and ground-based measurements (CPCB) with model reanalysis (MERRA-2 and CAMS). Model results are seen to capture the observed variations to an extent over this region. MERRA-2 reveals a significantly increasing trend in SO_2 during 2005-2015 with more pronounced enhancements over the industrial regions in India (0.1–0.3 ppbv yr^{-1}), in line with CAMS reanalysis and satellite-based observations. Further, a substantial increase (0.3–0.5 μgm^{-3} yr^{-1}) in sulphate concentrations are also simulated especially in the industrialized areas, in agreement with the SO_2 trend. Efficiency of sulphate formation is estimated to be 0.3 for monsoon and 0.15 for winter, implying that 30% total SO_2 getting converted to sulphate during monsoon. Long-term systematic observations together with development of time varying regional inventories are required for the assessment of the implications that these SO_2 changes will have over the Indian subcontinent.

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