



Long-term variability in biogenic emission fluxes and ozone over South Asia: Integrating remote-sensing observations with modeling

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1. Abstract

The Indian subcontinent is a large geographical system which experiences diverse natural and anthropogenic processes influencing the regional air quality, atmospheric chemistry and climate. However, ground-based observations are often sparse and limited to a few chemical species and the models have larger uncertainties in simulating aforementioned processes for Indian conditions. In this regard, we combined the potentials of remote sensing data and modeling to study the long-term variations in natural emissions and surface ozone. While the mean ozone levels are higher over the northern India, the long-term trend is seen to be more pronounced (~ 0.4 ppbv y^{-1}) over the central-east regions in the vicinity of higher biogenic emissions of isoprene. In addition, tropospheric column ozone shows increasing trends over the northern Indian Ocean and surrounding larger regions including Southeast Asia. We find widespread enhancement in ozone across South Asia and stronger biogenic emissions over peninsular India during the warmest year (2016) as compared to the climatological mean distributions. Our study suggests the need to conduct comprehensive in-situ observations of biogenic volatile organic compounds for formulation of effective policies to mitigate the impacts of global warming on the air quality over South Asia.

2. Introduction

Anthropogenic emissions are the major driver of increasing levels of greenhouse gases in the Earth's atmosphere leading to the global warming. Besides anthropogenic activities, the biogenic emissions also substantially impact the atmospheric composition. Biogenic emissions, due to their strong dependence on air temperature, are anticipated to be higher in the future. The global surface temperature is reported to be higher by more than 1°C during 2011-2020, as compared to 1850 – 1900, making it the warmest decade to record [1], [2]. In fact 11 out of 15 warmest years of India are from the recent fifteen years (2007-2021) with 2016 being the warmest year since 1901 [2]. In lack of effective control measures, the global temperature may increase by more than 1.5°C by 2040 or before. Modeling studies have suggested that the air quality can worsen in a business as usual scenario over South Asia in the future [3], [4]. Nevertheless, studies focusing on biogenic emissions and their impacts on air quality, also considering potential role of aforementioned climate penalty, remain lacking over the South and Southeast Asian regions. Such studies are of paramount significance as these regions have been experiencing warming and more frequent heatwaves [5].

South and Southeast Asian regions have been undergoing rapid economic developments, substantial changes in the land use and land cover besides the regional climate warming [6]–[8]. Enhanced levels of ozone (O_3) as well as aerosol loadings have been observed over not only urban regions but also over rural areas and in the outflow towards the Indian Ocean [9], [10]. However, due to non-linear complex chemistry and diverse emissions it has been extremely challenging to quantify the drivers of air pollution and design most effective control measures. Recent field measurements in conjunction with photochemical modeling and case studies of COVID-19 lockdown highlighted the need to control volatile organic compounds (VOCs) to reduce ozone [11], [12]. VOC reductions are suggested to help additionally in coordinated mitigation of the fine particulate matter [13]. As discussed earlier, ongoing climate warming can favor high O_3 episodes particularly during heatwaves [1]. Global studies revealed that higher temperature increases the reaction rates and alters the biogenic emissions of isoprene, monoterpenes, etc. [14], [15]. Temperature increase from 20 to 40°C over Europe enhanced ozone by about 20 ppbv due to faster reaction rates directly, whereas isoprene emissions produced additional 11 ppbv of ozone [16]. The worsening of air quality due to climate change is considered as the climate penalty [15]. Therefore, the projected rise in temperature can offset the benefits of potential reductions in the anthropogenic emissions.

The studies evaluating the trends in biogenic emissions and ozone with climate warming however remain very limited over South and Southeast Asia, mainly due to lack of long-term systematic measurements across these regions. Considering the

discussed scenario, we have analyzed long-term variabilities in biogenic emissions, temperature, and ozone over recent two decades by exploiting the combined potentials of satellite data and modeling. In this study, we analyze the large-scale distribution over South and Southeast Asia and also the different sub-regions of the Indian subcontinent: 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) considering the diversity of emissions, topography, and meteorology as also applied in previous studies [17].

3. Data and Methodology

This study utilizes the monthly averaged biogenic isoprene emissions based on the CAMS-GLOB-BIOv3.1 inventory [18]. In this method, the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1) is used for calculating the biogenic emissions from vegetation and soils at regional and global scales (Fig 1a) as also applied in various chemistry-transport models [19]. The key inputs to the MEGAN model include meteorological conditions (e.g., solar radiation, air temperature, humidity), land use and land cover (Vegetation classification, leaf age and area, etc.), besides ambient carbon dioxide, and O₃ concentrations. The spatial resolution of emission inventory is 0.25° × 0.25° and the driving meteorology is based on recent reanalysis from the European Centre for Medium-Range Weather Forecast (ERA5). Further details about the biogenic emission calculation is described elsewhere[18].

The mean distribution of temperature, isoprene and ozone are obtained from the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis, available from 2003 - 2020. The CAMS model reanalysis (Fig 1b) is based on assimilation of a variety of remote-sensing measurements with integrated Forecasting System of the ECMWF [20].

The tropospheric column O₃ for 2005-2020 has been derived from Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS) instruments on board Aura satellite. Difference between total column O₃ measured by OMI, and stratospheric O₃ measured by MLS gives the tropospheric column O₃, after adjusting for the inter-calibration differences [21].

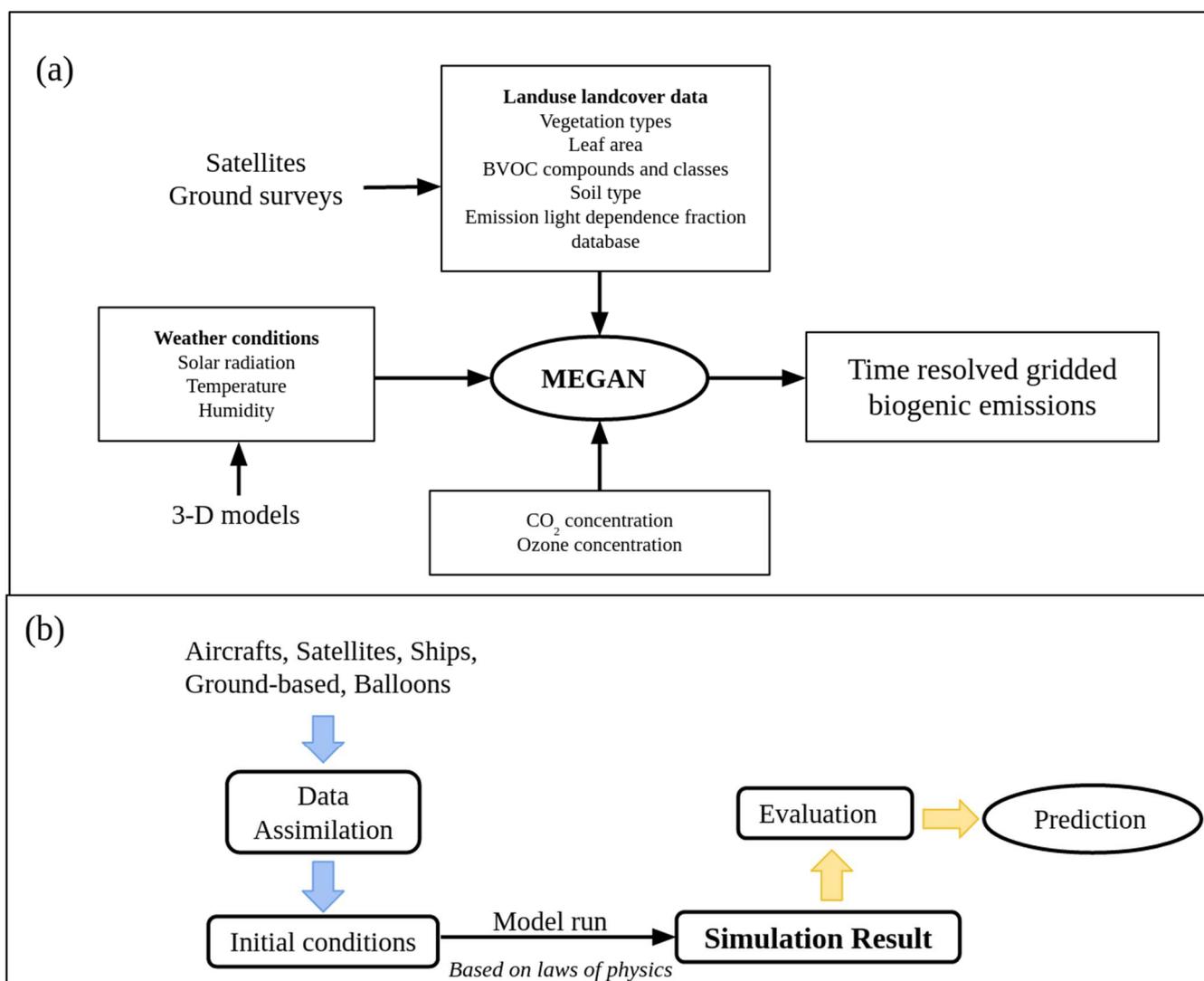


Figure 1: A simplified layout of the methodology for calculating (a) natural biogenic emissions and (b) meteorological and chemical fields in model reanalysis.

4. Results and discussions

Mean distributions of isoprene emissions during 2000-2020 over South Asia are shown for four different seasons: winters (DJF), pre-monsoon (MAM), monsoon (JJA) and post-monsoon (SON) in Figure 2. Isoprene hotspots are in general seen over forest and highly vegetated regions. Notably, the isoprene emissions are consistently higher (up to $2 \times 10^{-10} \text{ kg m}^{-2} \text{ s}^{-1}$) over the Southeast Asia during all seasons. The southern and central-east regions of India experience stronger isoprene emission (more than $1 \times 10^{-10} \text{ kg m}^{-2} \text{ s}^{-1}$) during the pre- and post-monsoon seasons. Enhanced isoprene emissions are also seen over northwest India during pre-monsoon.

The seasonal changes in O₃ are analyzed over five distinct sub-regions in India (Figure 3). The highest O₃ level (mean: ~ 48 ppbv) was simulated over the northern India during the pre-monsoon period. Afterwards, O₃ shows reduction during monsoon with lowest levels in August (mean: ~ 23 ppbv) over entire Indian region. In contrast, southern and western India experience higher levels of O₃ towards the end of post-monsoon with further increase until winter. These model results are in agreement with previous observational studies [10], [22], [23] suggesting transport of continental pollution emitted from vehicles, thermal power plants, industries, etc. which also enhance O₃ over the Arabian sea [8]. During the summer monsoon (JJA), clean marine air-masses from the Arabian sea and Indian ocean substantially dilute O₃ levels (lowest ~ 20 ppbv). Higher levels of NO₂ are seen near the emission sources and the enhancements are particularly strong during stagnant conditions in winters due to trapping of emissions in shallow boundary layer. Anthropogenic VOCs such as propane is also enhanced over the IGP and north-east regions in the South Asia [24].

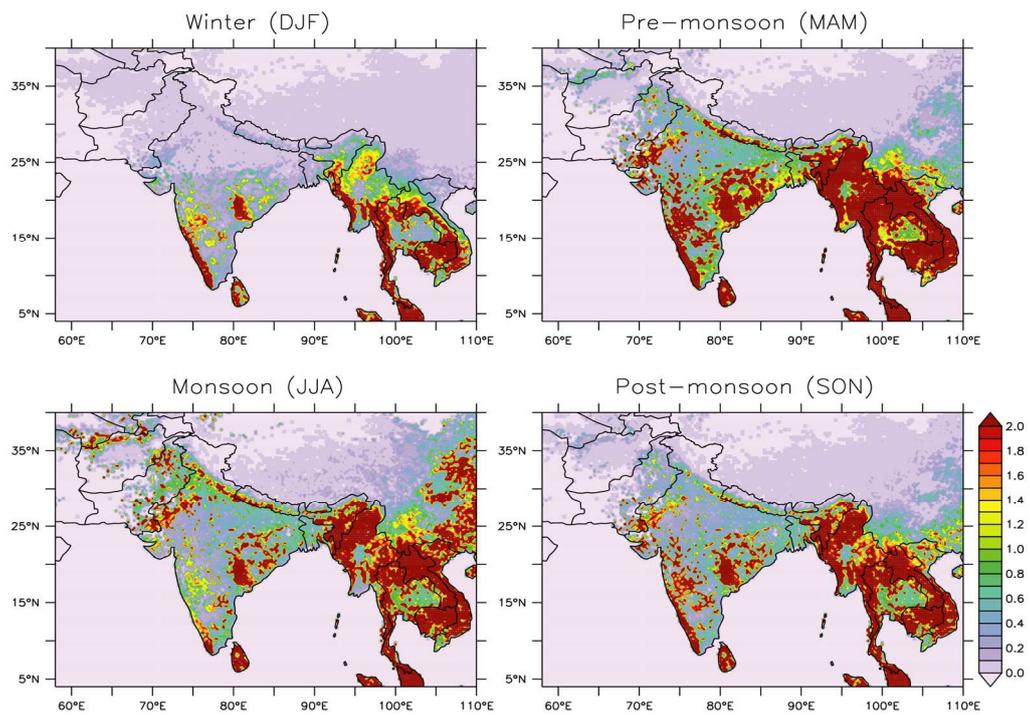


Figure 2: Isoprene emissions ($\times 10^{10} \text{ kg m}^{-2} \text{ s}^{-1}$) over South and Southeast Asia during different seasons based on CAMS-GLOB-BIOv3.1 inventory averaged over 2000–2020 period.

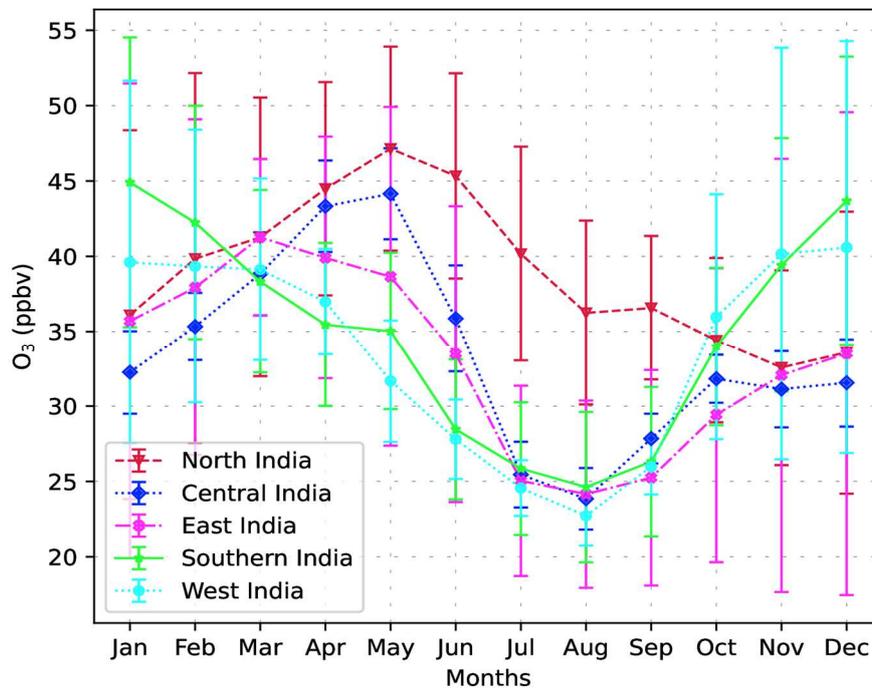


Figure 3: Seasonal cycle of surface O_3 over different sub-regions in India during 2003–2020 based on the CAMS reanalysis.

Long-term trends have been investigated over South Asia for surface O_3 (2003–2020) as well as tropospheric column O_3 (2005–2020), as shown in the figure 4. Model reanalysis shows a general positive trend in surface ozone over Indian region by 0–3

ppbv decade⁻¹. Within India, the annual trends were found to be higher (> 0.1 ppbv y^{-1}) over the central-east, north-west and eastern regions. Satellite-retrieved tropospheric column O_3 also shows statistically significant trends, mostly greater than 0.1 DU y^{-1} , over the Indian region. There is a consistency between increase at surface as well as in the tropospheric column, which suggests likely role of regional photochemistry in driving these trends. However, trends do differ spatially due to horizontal transport of ozone and its precursors and key role of stratospheric influences in the middle-upper troposphere. Generally, photochemistry involving regional pollution can have stronger roles near the surface whereas dynamics has greater roles in the free tropospheric altitudes. Further, satellites have relatively lesser sensitivity toward surface O_3 since the backscattered UV from surface O_3 does not reach satellite effectively. Our findings based on up-to-date satellite data and assimilated model results are seen to be in a good agreement with those estimated from in situ measurements over the different observational sites in India and surrounding oceanic regions [25], [26]. For example, O_3 levels were reported to be increasing by 0.4 ppbv y^{-1} over a tropical coastal station (Thumba, Thiruvananthapuram) in south India during 1997–2012 [25]. Increasing trend (1 – 2.2 ppbv decade⁻¹) was also seen over the Bay of Bengal during monsoon season [26]. Similarly, increasing O_3 trends were also reported in an urban location Delhi in the IGP (0.05 ppbv y^{-1}) and over Pune (0.04 ppbv y^{-1}) [27].

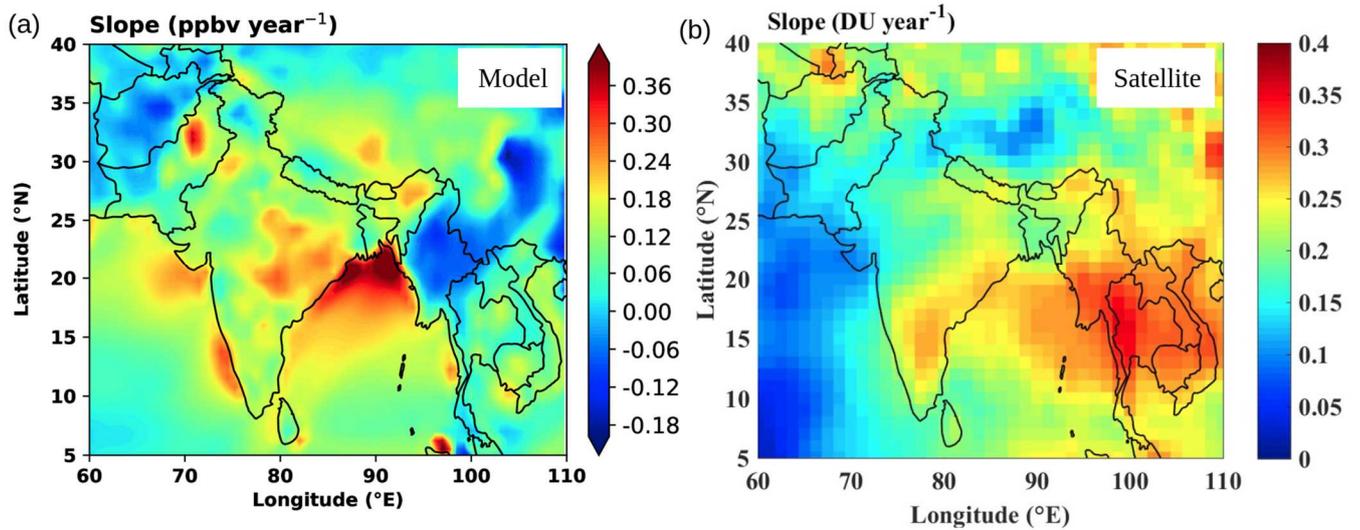


Figure 4: Long-term trends over South Asia in (a) surface O_3 during 2003–2020 derived from the CAMS reanalysis, and (b) tropospheric column O_3 during 2005–2020 derived from OMI/MLS satellite data.

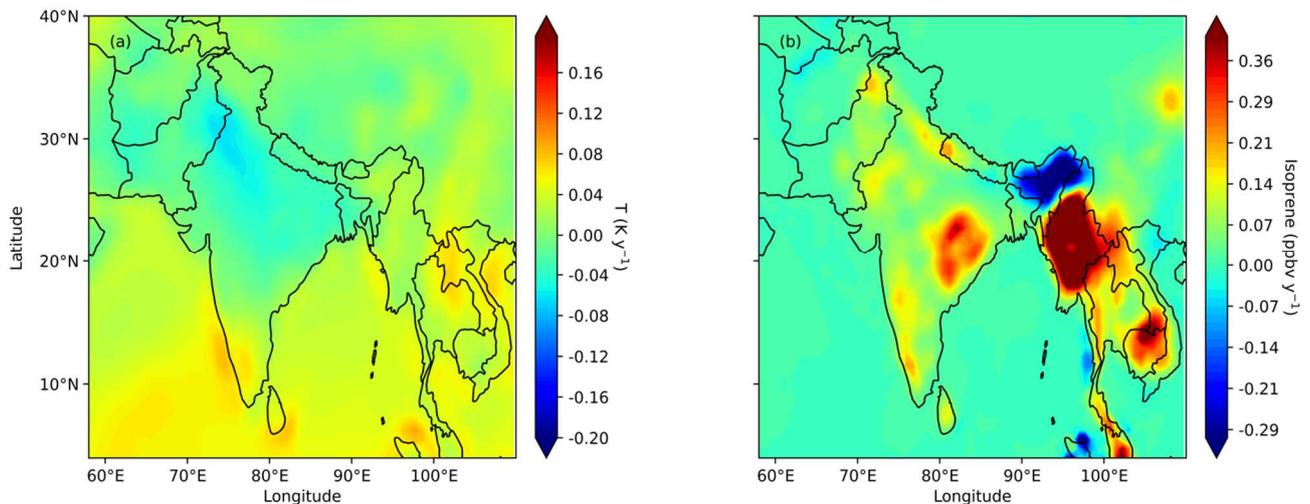


Figure 5: Long-term trends in (a) temperature and (b) isoprene during 2003–2020 over the South Asian region, derived from the CAMS reanalysis.

A gradual increasing trend in atmospheric temperature near surface is seen over Southern India, the northern Indian Ocean, Southeast Asia (Fig 5). Additionally, model results show increasing trends (up to 0.4 ppbv y^{-1}) in isoprene levels, most pronounced over central-east India. Strong enhancements in isoprene are also simulated over Southeast Asian region. Increasing

isoprene can be linked to effects of regional warming and changes in the land use land cover and requires further field and modeling studies in these regions. Further, it is interesting to note that both isoprene and O₃ have hotspots over central-east India suggesting a potential role of enhanced natural emissions in stronger O₃ build up. A recent study have reported that this part also experiences stronger SO₂ trends as seen in the satellite data ($\sim 0.2 \text{ DU y}^{-1}$) and CAMS reanalysis (up to 0.2 ppbv y^{-1}) [17].

A case study has been conducted to further examine the potential of climate warming in impacting atmospheric composition. Figure 6 shows the difference in temperature, isoprene and O₃ for the year 2016 (warmest year since 1901) with reference to their climatological mean values (2003–2020). The temperature was higher by up to about 1.3 K over the north-west and southern regions of India. Over northern-central and eastern regions also some general increase by 0.5–0.7 K were seen. Rise in temperature can enhance of the natural biogenic emissions of VOCs such as isoprene [28]. Interestingly, while there do not seem any warming over Southeast Asia, the isoprene levels do show large enhancements. Field observations would be required to evaluate these findings and to interpret possible role of changes in land use. Results also show a widespread enhancements in O₃ by about 3 ppbv levels are seen across India. The positive correlations are visible between temperature, isoprene and O₃ over the southern and central-east India. Recent studies have shown that strong enhancements in O₃ over IGP are correlated with the higher emissions of its precursors, including CO, NO₂ and anthropogenic VOCs [23], [24]. Regional climate warming can strengthen natural biogenic emissions besides directly intensifying the chemistry to yield more production of O₃ and secondary aerosols. Due to transport processes, the enhanced productions of O₃ over the hotspots have regional-scale impacts [8], [29].

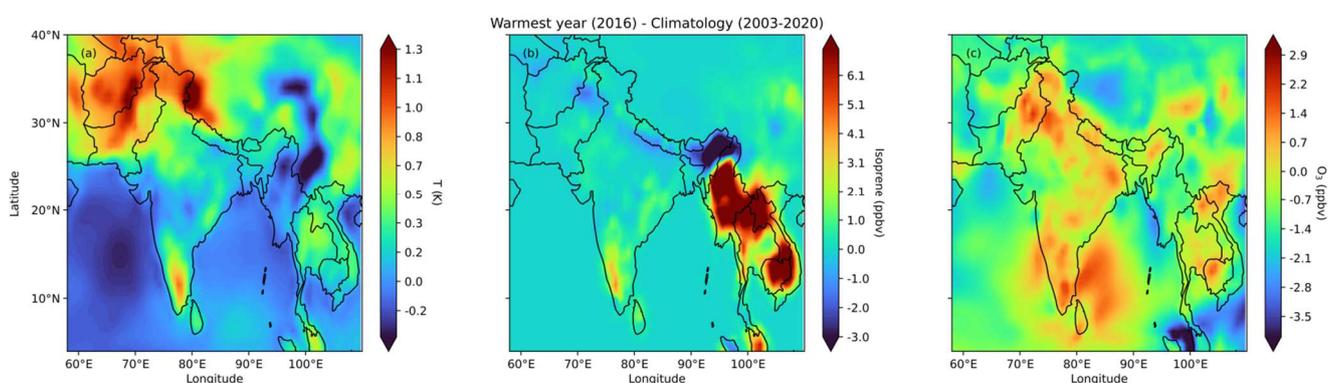


Figure 6: Spatial distribution of anomalies in (a) temperature, (b) isoprene, and (c) O₃ over South Asia for year 2016 as compared to the climatological mean derived from the CAMS reanalysis.

5. Summary and conclusions

In this study, the long-term spatio-temporal variability in isoprene and ozone over South and Southeast Asia has been analyzed by combining the remote-sensing datasets with modeling. The major findings of the study are summarized as follows:

- Distinct seasonal differences in isoprene are revealed, with substantially enhanced emissions ($> 1 \times 10^{-10} \text{ kg m}^{-2} \text{ s}^{-1}$) over southern and central-east India particularly during the pre- and post-monsoon seasons. In conjunction with emissions, isoprene trends ($> 0.14 \text{ ppbv y}^{-1}$) are more pronounced over central-east India and Southeast Asia.
- Satellite and model reanalysis show increasing trend in O₃ over South Asia with higher positive rates over Northern Indian Ocean and central-east Indian regions. Elevated levels of O₃ are associated with increasing temperature leading to more biogenic emissions plus intensification of photo-chemistry.
- A widespread enhancement in surface O₃ along with higher isoprene is seen over South Asia during the warmest year 2016 as compared with climatological mean highlighting importance of climate warming in influencing regional air composition.

Substantially enhanced natural emissions and O₃ are correlated with increasing regional warming over South Asia. There is a need to conduct systematic measurements of biogenic VOCs particularly over the identified hotspot regions to formulate policies to improve air quality and mitigate effects of regional climate change.

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