Assessment of Spatiotemporal Variability and Rising Trends of Surface Ozone Over India

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Abstract

With rising anthropogenic activities, surface ozone levels have increased across different parts of the world including India. Studies have shown that surface ozone shows distinct characteristics across India, however these studies are mostly based on isolated locations. Any comprehensive and spatiotemporally consistent study on surface ozone is lacking thus far. Keeping these facts in mind, we utilize ground-based observations and reanalysis datasets to investigate the surface ozone variability, seasonality, and linkages with meteorology over India. A validation exercise shows that the Copernicus Atmosphere Monitoring Service Reanalysis (CAMSRA) reasonably compares against the ground-based observations. Results show that the CAMSRA ozone is in good agreement with the observations across India, where it shows better correlations ($r > 0.7$) over southern regions and relatively lesser (> 0.5) over northern and eastern regions indicating larger variability and spread over these regions. We further quantify this agreement in terms of range, mean absolute error (MAE), and root mean square error (RMSE). Time series analysis shows that the CAMSRA captures seasonal variations irrespective of location. Spatial distribution of surface ozone shows higher (lower) concentrations of about 40-60 ppb (15-20 ppb) during pre-monsoon (monsoon) months over a broad region covering northern and western parts, and peninsular India. A prominent increase during May is noted over the northern region especially over Indo-Gangetic Plains (IGP). These seasonal variations are linked to solar radiation (SR), temperature, low-level circulation, and boundary layer height (BLH). Furthermore, Principal Component Analysis (PCA) is performed to understand the dominant patterns of spatiotemporal variability for different seasons. It is seen that the first (second) mode shows a high percentage variance explained ranging between 30-50% (10-20%). The time series of PCA-1 mode indicates an overall increasing trend across India with a notable increase over south and central India. The second mode indicates prominent variability over the IGP (southern India) in the pre-monsoon (post-monsoon) season, which shows significant interannual variability. During the monsoon season, an interesting dipole pattern is seen which closely resembles the active and break spell patterns of the Indian summer monsoon. Overall, the spatiotemporal variations in surface ozone are closely tied to meteorology while the rising trends indicate the potential role of increasing precursors across India.

1. Introduction

Surface ozone has become a widely studied constituent in recent times due to its emergence as a short-lived ($\sim$ few days) secondary pollutant and its damaging impacts on human health and crop production (e.g. Mills et al. 2007; Avnery et al. 2011; Ghude et al. 2014; Lelieveld et al. 2015). It plays a pivotal role in tropospheric chemistry through controlling the oxidation processes as the source of hydroxyl radicals (OH) (Levy, 1971; Logan, 1985) and through its reaction with many organic compounds (Atkinson, 1990). Ozone in the troposphere is a secondary air pollutant product of photochemical reactions involving carbon monoxide (CO) and volatile organic compounds (VOCs) in the presence of nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) termed as ozone precursors (Chameides and Walker, 1973; Fishman et al. 1979; Crutzen, 1995). These photochemical reactions are dependent on solar radiation and usually, the rate of reaction is highest during the summer months. As compared to surface ozone, the ozone concentrations are higher in the upper tropospheric levels where it is affected more by the large-scale circulations and exchanges between stratosphere-troposphere (Brunamonti et al. 2018; Kumar et al. 2021). The surface ozone has a relatively shorter life span and exhibits

significant spatiotemporal heterogeneity (Ojha et al. 2012; Nair et al. 2018). Recent studies have reported an increasing surface ozone trend over different parts of the world including Asia, where a rise in concentrations poses a stiff challenge to more than 5 billion people (Monks et al. 2015; Singh et al. 2021b). The fast-paced developing economies such as China and India have experienced enhanced anthropogenic emissions which have led to increased ozone concentrations in recent decades (Cooper et al. 2014; Sun et al. 2016; Wang et al. 2017). This rise has been noted through various precursors e.g., biomass burning, fossil fuel combustion, and lightning, etc.

Understanding the spatiotemporal variability of surface ozone over India is important as it ranks amongst the toxic air pollutants and any scientific literature discussing these aspects remains elusive. A few studies have investigated the surface ozone variability over different parts of India and the surrounding Indian Ocean region (e.g., Nair et al. 2011; Peshin et al. 2017; Tyagi et al. 2016; Tiwari et al. 2015; Girach et al. 2017; Anshika et al. 2021). Most of these studies have been focused on particular observational locations and therefore a comprehensive understanding of spatial distribution and variability is yet to be explored. The long-term analysis of surface ozone over India has been limited by the availability of reliable and consistent records. Although satellite-based measurements provide tropospheric surface ozone concentrations, they are often of coarse temporal and spatial resolution (Ojha et al. 2012). Model-based analysis has reported significant biases along with limitations in capturing small and regional scale variability (see Sharma et al. 2017 and references therein). A recent study by Hakim et al. (2019) analyzed the observations and outputs from model simulations to understand the annual cycle of surface ozone over India. However, the analysis was constrained by the limited number of observational locations and time. Therefore, it requires spatially well sampled and reliable estimates to correctly examine surface ozone variability and impacts. Some studies have thus highlighted the need for an increased network of ground-based stations (Schultz et al. 2017).

In this regard, ozone records based on station observations coordinated by the Central Pollution Control Board (CPCB) India, provide a good source for validating remotely sensed and reanalysis datasets and study the subregional scale variability over different parts of India. Another important aspect is that India has complex geography/topography and is subjected to strong seasonal variations in circulation patterns. Significant seasonal changes in the circulation patterns are seen during different seasons e.g., from pre-monsoon to monsoon, monsoon to post-monsoon followed by post-monsoon to winter months annually. Driven primarily by the incoming solar radiation, strong changes in circulation and convection leads to the annual cycle dominated by the monsoon type of climate (Singh et al. 2021a). Therefore, the region is subjected to complex interactions between the regional meteorology and anthropogenic emissions which can combinedly contribute to the spatial and temporal variability of surface ozone.

With a rise in population and anthropogenic activities, it is important to understand the surface ozone variability over the region. Though studies have documented its variability over isolated locations, any comprehensive and consistent study is lacking thus far. It is imperative to analyze the distribution and seasonal behavior of ozone over India and understand its variability at longer time scales. Keeping these facts in mind, the present study primarily examines surface ozone variability across India based on the observed and reanalyzed datasets. We first compare the reanalysis data with in situ high resolution station

records over different geographical subregions (north, east, west and south). A comprehensive validation exercise is carried to infer the reliability of Copernicus Atmosphere Monitoring Service (CAMS) data and quantifications are presented and discussed. Through the time series analysis, we study the seasonality of surface ozone over different locations across India. Further, we use the CAMS data to assess the spatiotemporal variability of surface ozone across India. Additionally, we analyze the associated meteorology and briefly discuss the linkages of meteorological variables with seasonal variations in surface ozone. Basically, the present study aims towards bringing out the statistics of surface ozone over the Indian region as seen through the lens of seasonal variability, its relationship with meteorological variables of interest, and identifying the dominant modes of variability. The organization of the paper is as follows: Sect. 2 discusses the data and methodology; Sect. 3 discusses the results of analyses followed by the summary and conclusions in Sect. 4.

2. Data And Methodology

In the present study, we have utilized ground-based observations and reanalysis datasets over India to address the surface ozone variability. The details of ground-based observations and the reanalysis data products is described in the following subsections.

2.1 Central Pollution Control Board (CPCB) station observations

CPCB measures ambient air quality through a network of stations across India under the National Air Quality Monitoring Program. Under this program, the surface ozone is regularly monitored through automatic monitoring stations out of which we have considered about 20 ground-based stations over different parts of India for the period 2018–2019. The CPCB data is available in public domain (<https://app.cpcbcr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/caaqm-comparison-data>). We choose the stations objectively so that the data is well distributed in space across India and also there is continuity in time. This is done to understand the seasonal cycle and validate the reanalysis-based dataset. It also helps in analyzing small-scale regional influences. Therefore, we use data only from those stations with at least > 80% records available during the analysis period. The site specifics including latitude, longitude, and altitude (height above the sea level) of stations used in the analysis are provided in Table 1.

Table 1

List of ground-based stations, their location (latitude, longitude and height above the mean sea level), and the metrics of comparison used for the validation of CAMSRA data.

Station	Latitude	Longitude	Height AMSL (m)	Ozone range Obs (ppb)	Ozone range CAMSRA (ppb)	CC	RMSE	MAE
Ambala	30.38	76.78	275	3.1- 68.39	6.7- 75.77	0.48	14.47	12.86
Lodhi Road Delhi	28.59	77.23	213	2.75- 69.87	10.41- 80.3	0.53	13.54	12.35
Faridabad	28.04	77.31	203	3.13- 68.24	9.1-72.6	0.48	13.04	11.34
Gurugram	28.46	77.01	229	1.93- 68.83	10.4- 80.2	0.44	14.21	12.05
Ayanagar	28.45	77.14	261	5.1- 70.3	6.8- 70.77	0.62	22.31	20.82
Ajmer	26.46	74.64	479	3.6- 63.15	17.8- 68.16	0.69	16.75	15.46
Kanpur	26.45	80.32	131	2.2- 55.96	9.97- 81.38	0.42	21.94	20.66
Varanasi	25.34	83	79	2.45- 75.24	7.35- 81.9	0.39	17.22	14.37
Gaya	24.79	85	120	2.45- 71.69	9.29- 75.31	0.37	19.42	16.83
Udaipur	24.58	73.69	597	3.4- 43.98	10.26- 49.79	0.57	13.94	12.55
Ahmedabad	23.02	72.58	52	5.22- 71.95	12.93- 81.84	0.46	13.86	15.13
Aurangabad	19.88	75.34	582	2.48- 67.36	15.89- 61.1	0.76	13.21	11.71
Visakhapatnam	17.72	83.29	11	1.74- 83.77	14.65- 71.16	0.70	22.11	20.65
Solapur	17.67	75.9	467	7.11- 65.68	16.11- 65.16	0.76	10.58	8.28
Hyderabad	17.36	78.47	508	2.1- 46.96	8.56- 60.35	0.86	10.12	9.72
Rajamundry	17	81.8	41	8.1- 59.89	9.66- 62.78	0.78	16.17	13.37
Amaravati	16.51	80.52	24	3.86- 69.59	13.72- 72.22	0.36	22.22	19.61

Station	Latitude	Longitude	Height AMSL (m)	Ozone range Obs (ppb)	Ozone range CAMSRA (ppb)	CC	RMSE	MAE
Tirupathi	13.63	79.42	153	1.4– 51.56	11.55– 57.09	0.72	13.96	12.81
Bengaluru	12.98	77.6	924	1.9– 53.03	12.03– 51.83	0.72	13.37	12.31
Thiruvananthapuram	8.48	76.95	12	2.83– 51.16	11.35– 60.26	0.68	15.26	14.28

2.2 Reanalysis data

The long-term surface ozone variability is assessed through the CAMS data. The CAMS reanalysis (referred hereafter as CAMSRA), is produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) and consists of three-dimensional time-consistent atmospheric composition fields, including both aerosols and chemical species (Inness et al. 2019). Consistent ozone fields are produced by utilizing a range of instruments e.g., ozone retrievals are obtained from the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY instrument, the Ozone Monitoring Instrument, and the Global Ozone Monitoring Experiment-2, ozone profile data from the Michelson Interferometer for Passive Atmospheric Sounding and Microwave Limb Sounder, and ozone partial columns from the Solar Backscatter Ultraviolet radiometer. The CAMSRA dataset starts from the year 2003 intending to add more data in time. CAMSRA data is shown to be useful in capturing the climatology, trends, and also in evaluating chemistry-based models (Inness et al. 2019). For the present study, we utilize the CAMSRA derived surface data of ozone for the period 2003–2019, archived at 0.75° x 0.75° horizontal resolution. Before using CAMSRA data for the analysis, it is validated against the CPCB ground station-based observations. Additionally, we use the reanalyzed meteorological fields from the fifth generation European Centre for Medium-Range Weather Forecasts (ERA5) (Hersbach et al. 2020) to understand the association of surface ozone with meteorology.

3. Results

3.1 Validation of CAMSRA

Analysis of globally averaged CAMSRA surface ozone shows that it compares well against the ground-based surface observations available from the World Meteorological Organization (WMO) coordinated Global Atmosphere Watch (GAW) programme (see Inness et al. 2019 and references therein). The study also reports a good agreement across the European region. In general, the surface ozone fields from CAMSRA and observations match well within 10% for most years with biases being negative during the first half of the year and positive during the second half. The study robustly concluded that CAMSRA is more consistent than the previous reanalysis and has smaller biases compared with the independent ozone observations. However, we notice that there are only a few stations available over India under the WMO GAW program with differing periods of data availability. Therefore, we first validate the daily averaged surface ozone values for

the period of the recent two years (2018 and 2019) from the latest generation CAMSRA against CPCB ground-based measurements and quantify the robustness of agreement. The stations used for this purpose are shown in Fig. 1 and also listed in Table 1. These ground-based (20) stations are located across India and are useful in understanding the surface ozone variability from the subregional scale perspective also (which would be explored in the later sections). The results of the correlation analysis are shown in Fig. 2. The stations in Fig. 2 (and Table 1) are arranged in such a way that the latitude decreases as we move downwards. We notice that the surface ozone concentrations vary in the range $\sim 2\text{--}80$ ppb for both CAMSRA and CPCB observations for most of the stations. However, a relatively larger spread in these values over the northern and eastern parts of India e.g., over Ambala, Delhi, Faridabad, Gurugram, Varanasi, and Gaya. We compute the correlation coefficient (CC) and note that CAMSRA based surface ozone estimates are well correlated with the stations located in the southern and the western parts (correlation coefficient > 0.6) e.g. over Ayanagar, Ajmer, Aurangabad, Visakhapatnam, Solapur, Hyderabad, Rajamundry, Tirupati, Bengaluru, and Thiruvananthapuram. Similar correlations have been documented by the recent studies over different locations across India (see Ojha et al. 2019; Anshika et al. 2021 and references therein). The scatter analysis performed here indicates a reasonable agreement between CAMSRA and ground-based observations over India as a whole. Apart from CC, we have also computed other metrics of comparison in terms of range, Mean Absolute Error (MAE), and Root Mean Square Error (RMSE). Table 1 also lists these values obtained for the 2018–2019 period. It is seen that CAMSRA shows good agreement with the observations in terms of correlation and range. However, when we compare day-to-day variations quantitatively, we note that CAMSRA overestimates absolute surface ozone concentrations across all the stations. In particular, the CAMSRA produces an elevated minimum in surface ozone concentrations. The northern stations show observations varying in the range $\sim 2\text{--}70$ ppb while the CAMSRA ranges about $\sim 6\text{--}80$ ppb. Over this region, the MAE (RMSE) varies between 11 ppb (13 ppb) for Faridabad and 20.8 ppb (22 ppb) for Ayanagar. Over the eastern region, we note smaller CC, and high MAE and RMSE. Here the surface ozone ranges between 2.5–75 ppb (7–82 ppb) for observations (CAMSRA). The MAE and RMSE vary between 14–17 ppb and 17–20 ppb respectively. These large variations over the northern and eastern regions are indicative of significant day-to-day variability over the region. The stations located in the western region show better agreement as compared to those in eastern and northern regions. The range of surface ozone variability over the western region is noted to be 3–72 ppb and 10–81 ppb from CPCB and CAMSRA respectively. Among the western stations, maximum agreement is noted over Udaipur, with the least MAE (~ 12.5 ppb) and RMSE (~ 13.9 ppb). Although the correlation is highest over Ajmer, we also note that the MAE and RMSE are also on the higher side. As we go further south, we note higher CC and a decrease in MAE and RMSE. The CC for almost all the southern station locations are higher than those of other regions (except Amaravati) with the highest CC value of about 0.86 over Hyderabad. The range of surface ozone variability over the region is found to be $\sim 2\text{--}83$ ppb and $\sim 8.5\text{--}72$ ppb for CPCB ground-based observations and CAMSRA respectively. Though some of these stations show higher ozone maximum e.g., Visakhapatnam shows the maximum surface ozone concentrations of about 83.7 ppb in the observations, the MAE and RMSE are still lower as compared to the stations located in other regions. The lowest (highest) MAE of about 9.7 ppb (19.6 ppb) is noted over Hyderabad (Amaravati). Similar RMSE values are also noted for the two stations i.e., about 10.1 ppb and 22 ppb for Hyderabad and Amaravati respectively. It is interesting to note that the range of surface ozone over the coastal station Thiruvananthapuram is slightly lesser ($\sim 3\text{--}51$ ppb for observations and $\sim 11\text{--}60$ ppb for

CAMSRA) as compared to those in the higher latitudes, but the MAE (15.26 ppb) and RMSE (14.28 ppb) are still higher. This is indicative of significant differences between the day-to-day variations between the two datasets.

Next, we analyze the time series of surface ozone obtained from CAMSRA and compare it with ground-based observations as shown in Fig. 3. For this, we have considered four representative ground-based stations taken from each of the four boxes shown in Fig. 1. These stations are located over northern, eastern, western, and southern parts of India and help represent distinct variability over polluted and coastal locations. From the time series, one can clearly notice that CAMSRA captures the seasonal cycle of surface ozone. Despite overestimations as seen in the validation analysis, the seasonal cycle is well captured at each station irrespective of the location. Over the polluted northern station Delhi, we find that the surface ozone usually peaks in May with max concentrations crossing 60 ppb while the minimum is seen during August when the concentrations drop below 20 ppb every year. The elevated levels of surface ozone during May are due to strong insolation which supports the photochemical formation of ozone with the aid of its precursors (e.g. Anshika et al. 2021). The observed decrease during the monsoon and winter seasons might be linked to the washing out of precursors due to monsoon rains and western disturbances respectively (e.g. Attada et al. 2020). The surface ozone variability over another polluted eastern station Varanasi is similar to that of Delhi (except the winter minimum) where it peaks in May (~ 60 ppb) and drops to a minimum during August (~ 20 ppb). The western station at Udaipur shows the maximum (minimum) surface ozone during April (September) with values ranging between 35 ppb to 40 ppb (15 ppb to 20 ppb). Similar maxima and minima in surface ozone over Udaipur were also reported by Yadav et al. (2016) wherein they found that the highest values in the pre-monsoon and winter seasons were associated with the elevated levels of both carbon monoxide (CO) and NO_x (NO + NO₂). Daily concentrations of CAMSRA ozone at Thiruvananthapuram also show a clear seasonality with the values varying between a maximum of about 50 ppb and a minimum of 20 ppb. The station-based records show similar seasonality, but with lesser amplitudes compared to the CAMSRA. As compared to the other three stations, the Thiruvananthapuram station shows a shifted peak in time which is observed during the late winter and early summer months (~ 50 ppb) while the minimum is seen during the later part of the monsoon season (~ 20 ppb). A similar surface ozone cycle was also reported by Nair et al. (2018) wherein the authors attribute the minimum to moisture-laden strong westerly/south-westerly winds which bring cleaner air mass, and a decrease in available solar radiation under cloudy monsoon conditions. The study ascribes an increase in surface ozone to the transport of continental pollutants (mostly precursors of ozone) by easterly/north-easterly winds during the winter to early summer months. Analysis of surface ozone time series indicates that the CAMSRA has the capability for reproducing the seasonality irrespective of the location. It reasonably captures the temporal evolution of the highest and lowest concentrations of surface ozone when compared with the ground-based observations. Also, we notice frequent occurrences of higher concentrations as we move ahead in time, which probably indicates an increasing trend in the surface ozone.

Though we have discussed these metrics based upon stations located across India, it is also important to note that surface ozone is significantly affected by local factors. Therefore, even though two locations may be close in space, the variability may differ significantly. This analysis quantifies that the southern stations are in better agreement with the CAMSRA while the stations over northern and eastern subregions show

relatively poorer agreement. Such differences may be caused by increased emissions of precursors over the region (Verma et al. 2017). It is to be kept in mind that these values might fall closer on the seasonal and annual time scales if we use long-term observations. Nevertheless, CAMSRA reasonably captures the minima, maxima, and seasonality of surface ozone over most of the locations. Ojha et al. (2019) carried out a similar analysis for an observatory located in Dehradun and showed the comparison between ground-based observations and CAMSRA. While comparing the noontime surface ozone they found that it varies between 11.6–83.7 ppb and 46.4–111.6 ppb for observations and CAMSRA respectively. They obtained a CC of 0.86 and a mean bias (RMSE) of about 29.5 ppb (31 ppb) and concluded that CAMSRA was able to capture the day-to-day variations in noontime ozone. We however realize that such results may differ regionally which would be explored in the later sections. Nevertheless, our comprehensive analysis brings out the extent of coherency between CAMSRA and CPCB station observations across India and proves that CAMSRA can be reasonably used to examine the daily, monthly and seasonal variability in the absence of ground-based observations. The validation and time series analysis performed here equips us to utilize long-term (2003–2019) CAMSRA data to understand the spatiotemporal variability of surface ozone and its linkages with the meteorology over the region. Hereafter the analysis of surface ozone would be based on the CAMSRA while that of meteorological parameters would be based on ERA5 reanalysis.

3.2 Climatological spatial distribution of surface ozone

We use the long-term CAMSRA dataset and obtain the monthly climatology which is shown in Fig. 4. The maxima in surface ozone are usually seen over a broad region covering northern and western parts, and peninsular India during the pre-monsoon months (March-May) while minimum over these regions is observed during monsoon months (June-August) after which it starts increasing again. Increase during the May month is more prominent over the Northern Indian region especially over Indo-Gangetic Plains (IGP). The highest surface ozone concentrations vary mostly in the range between 40–60 ppb while the lowest values range between 15–25 ppb. During the pre-monsoon season, it occasionally crosses 70 ppb over the northern stations. However, there are regions e.g., the western and southern tips where the maximum and minimum do not co-occur in time with the larger domain. Over the eastern and western arcs, we observe pronounced surface ozone concentrations during the winter months. The reason behind higher concentrations during the pre-monsoon months is linked to the availability of solar radiation and precursors which supports photochemical ozone production. This aspect would be discussed in subsequent sections (see Fig. 6). The observed increase during the winter months might be due to the stagnant atmospheric conditions (see Verma et al. 2017). We note a marked decrease in surface ozone during the monsoon months over southern and south-western parts with concentrations dropping close to 15 ppb. It is seen that with the progression of the monsoon, low values spread over most parts of India. Studies have reported that the onset of the South Asian summer monsoon in late May and early June brings westerly and south-westerly winds from the Arabian Sea to the Bay of Bengal resulting in rainy weather conditions over the Indian region (Gadgil, 2003; Yadav and Singh 2017). The resulting monsoon precipitation efficiently removes ozone precursors (see Kumar et al. 2012), wherein cloudy conditions and low temperatures hamper ozone production through photochemical reactions. We note that the surface ozone minimum over the north-eastern region persists until the post-monsoon months as the region continuously experiences rains during this time. These characteristics are also noted when we analyze the seasonal mean behavior of surface

ozone across India during different seasons (see Fig. S1) viz. winter (December-January-February), pre-monsoon (March-April-May), monsoon (June-July-August), and post-monsoon (September-October-November). To understand the spatial variability of surface ozone we also analyze the standard deviation (SD) climatology (Fig. 5). This helps in identifying the regions which experience more variability in surface ozone concentrations. High variability is seen over northern (and eastern coastal regions) parts of India especially over IGP regions during May and June with values above 5 ppb. During May, higher variability is also seen over the southern peninsula. The lowest variability with values of about 2–3 ppb across India is seen during the core monsoon months (July-August). The variability gradually increases during the post-monsoon months and continues for the winter months. Clearly, the ozone variability is influenced by the monsoon with a minimum during the rainy months and higher values at other two ends viz. the pre- and post-monsoon months.

We further analyzed the area-averaged climatological concentrations over four geographical regions namely Northern India (NI), Eastern India (EI), Western India (WI), and Southern India (SI) shown as boxes in Fig. 1 (see Fig. S2). Climatologically, we note that the highest concentrations of ozone over NI (EI) are observed in May with values around ~ 46 ppb (~ 49 ppb). Interestingly we note that the minimum is observed during January (August) for NI (EI), which shows a distinct ozone seasonality over these regions. The climatological mean is smallest over WI where the highest (lowest) concentrations occur during April (August), with a value close to ~ 36 ppb (~ 24 ppb). Among the four regions, the southern region stands out in terms of seasonality. Here the maximum (~ 48 ppb) surface ozone is observed during the winter month, however, the minimum (~ 26 ppb) is noted during the monsoon season (August). Overall, the analysis indicates the distinct seasonality over different subregions of India during the study period.

3.3 Linkages with regional meteorology

In this subsection, we discuss the meteorological parameters of interest to surface ozone production and variability. We have evaluated the climatology of surface temperature and solar radiation (SR) and the results are shown in Fig. 6. An obvious one-to-one correspondence is seen between solar radiation and temperature. In both parameters we note a clear seasonality with maximum values during pre-monsoon months where temperature (SR) values vary between 34–38°C (800–1000 W/m²) across India. These pre-monsoon conditions are conducive for ozone formation which can be seen in the spatial distribution of surface ozone (Fig. 3) wherein higher solar radiation (and temperature) helps in the chemical production of ozone through the photochemical reactions. The photochemical production is further enhanced by an increase in ozone precursors (e.g. NO_x) (Jacob and Winner, 2009; Doherty et al. 2013; Pusede et al. 2015), and natural emissions such as biogenic NMVOCs and soil NO_x emissions (Lu et al. 2018) during the pre-monsoon season.

With the arrival of monsoon, a gradual decrease in temperature (SR) from June is seen and the lowest values of less than 20°C (~ 550 W/m²) start appearing during the winter months (from November) over northern parts. The temperature (SR) further decreases over the northern regions of India with values ranging between 8–18°C (300–500 W/m²) during the peak winter months. The north-south gradients in temperature and SR become more prominent as we move from post-monsoon to winter months. During the monsoon months, cloudy conditions and associated precipitation lead to a reduction in solar radiation and surface

temperature, thereby reducing photochemical ozone production. The winter-time high concentration of surface ozone over the northern, eastern, and IGP regions is linked primarily to an increase in CO levels. The CO increase during this season is caused by a combination of factors which include direct emissions (Hakim et al. 2019) and contributions from secondary sources such as the oxidation of Volatile Organic Compounds (Grant et al. 2010).

Next, we analyze the seasonal cycle of surface wind circulation (shown by streamflow) and boundary layer height (BLH) evolution across India. These factors are important as they give an idea about the potential transport and mixing of ozone in the troposphere. It can be seen that pre-monsoon months are associated with increased BLH and continental north-westerly (NW) winds (as seen in the streamflow patterns) over the western and northern parts of India (Fig. 7). Higher BLH co-occurs with higher surface ozone concentrations while NW stream flow brings dry air which further raises the temperature over the region making the conditions conducive for surface ozone production (Yadav et al. 2016). The south-westerly (SW) flow during the monsoon season brings moist oceanic air from the Arabian sea and cloudy conditions over the region which limits the photochemical production and reduces the BLH. The appearance of surface ozone minimum over the southern tip and west coast is consistent with the flow patterns in the early phase of the monsoon. A further drop in surface ozone over the western parts and peninsular India is seen during the core monsoon months of July and August. During this phase, the Bay of Bengal branch of monsoon circulation brings moist air over the eastern and north-eastern parts which result in cloudy conditions and reduces the surface ozone production. The post-monsoon to winter increase in surface ozone seems to co-occur with low BLH and the advent of north-easterly (NE) flow. It potentially results in the transport of precursors over the region and helps in ozone transport and formation. In general, low BLH favors the accumulation of pollutants while high BLH supports the vertical mixing of the precursors which can help in the production assisted by strong insolation and surface temperatures (Yadav et al. 2016). Low BLH in winter months may result in accumulation and stagnation of pollutants near the surface, which also prevents the mixing of ozone poor surface air with the ozone rich air present at higher altitudes. Our analysis indicates that monthly (or seasonal) evolution of surface ozone is influenced by the availability of SR and higher surface temperature which aids in photochemical production, along with enhanced or suppressed convective mixing and transport by winds.

3.4 Dominant modes of surface ozone variability over India

In order to further examine the spatial and temporal variability of surface ozone, we have performed the Principal Component Analysis (PCA) and the results have been shown in Fig. 8–11. The usage of PCA is advantageous as it reduces dimensionality without losing important information in large datasets. This will be used to explain the relationship between dominant modes of spatial variability and their temporal evolution. Figure 8 shows the variance explained by different independent modes obtained from the PCA over the period 2003–2019. For the four different seasons considered, the first mode shows a high percentage variance explained ranging between 30–50% while the variance explained by the second mode ranges between 10–20%. Hence the first two modes together explain large variance (40–70%) within the data. Therefore, we primarily discuss the spatial and temporal patterns of the first two modes only. For instance, Fig. 9 shows the spatial patterns of the first mode of PCA (PCA-1). It indicates that during the pre-

monsoon (MAM) conditions, much of the variance is explained from the south and central Indian region while in other seasons much of the variance can be seen from the central and eastern Indian region. Specifically, during the monsoon, the core monsoon region shows a high variance of surface ozone, which might be linked to cloudy and rainy conditions. Further, the spatial distribution of PCA-1 is also closely related to the climatological distribution of surface ozone shown in Fig. 4. The spatial distribution corresponding to the second mode of PCA (PCA-2) is shown in Fig. 10. It is interesting to note the pre-monsoon spatial distribution of PCA-2, where the IGP region shows high variance. During the monsoon period (JJA) and post-monsoon season (SON), we see a dipole type of pattern. For the monsoon season, this pattern is seen over the west coast and the central Indian region, while for the post-monsoon season it is seen over south India and northwest India respectively. These spatial patterns of the first two modes explain much of the spatial variability seen in the climatological surface ozone distribution shown in Fig. 4. It is also interesting to understand the temporal evolution of these two modes which is shown in Fig. 11. In Fig. 11(a), one can see the time series of PCA-1 mode which indicates an increasing trend irrespective of the season. This implies that over much of the south and central Indian region the surface ozone has increased during 2003–2019. The time evolution of the second mode indicates some contrasting trends in different seasons. For instance, during the pre-monsoon (post-monsoon) season where much of the ozone variability is associated with the IGP (southern India), we do not see any significant trend instead we observe a significant interannual variability. While it is interesting to note the dipole spatial pattern of PCA-2 associated with the monsoon season (Fig. 10), it shows a significant decreasing trend (Fig. 11b). Interestingly the second mode during JJA which shows a decreasing trend and is associated with a dipole spatial pattern, resembles the regions of active (organized) and break (weak) monsoon spells during the season (Rajeevan et al. 2010; Singh et al. 2021a). Overall, much of the variance is primarily explained by the PCA-1, which indicates an increasing trend of surface ozone over the Indian region.

4. Summary And Conclusion

Surface ozone is one of the major toxic secondary pollutants and is important to understand its distribution and variability. In the present study, we have examined these aspects of surface ozone over India using CPCB station observations and CAMSRA datasets during 2003–2019. Before using CAMSRA, we have validated CAMSRA against 20 ground observations across different subregions of India. We have performed the PCA and investigated the spatiotemporal variability of surface ozone and linkages with the meteorological parameters. Following are the main highlights and conclusions of this study:

1. The validation analysis indicates that the CAMSRA surface ozone is in good agreement with the station-based observations. It is noted that the correlation coefficients for different subregions at 95% level of significance are: ~ 0.6 for southern and the western parts, and ~ 0.4 for northern and eastern parts of India. The range, MAE, and RMSE have also been computed for all the stations used in the analysis. We find that the correlation is significant, and there is a fair agreement in the range. However, we note that CAMSRA overestimates the surface ozone particularly the lower part of the spectrum. Overall, the validation analysis through CC, MAE, and RMSE brings out the extent of coherency between CAMSRA and CPCB data, and results indicate that it can be reasonably used for the analysis of long-term surface ozone variability and trends over India.

2. Time series analysis of daily averaged surface ozone from CAMSRA and observations shows strong seasonality in surface ozone concentrations. Despite overestimations, we note that CAMSRA compares well against observations at each station irrespective of the location. Over the polluted northern station e.g., Delhi, surface ozone usually peaks in May with maximum concentrations frequently reaching beyond 60 ppb while the minimum is seen during August when the concentrations drop below 20 ppb every year. Surface ozone variability over another polluted eastern station e.g., Varanasi is similar to that of Delhi (except the winter minimum), where it peaks in May (~ 60 ppb) and drops to a minimum during August (~ 20 ppb). The time series over the western region e.g., Udaipur shows the maximum (minimum) surface ozone during April (September) with values ranging between 35 ppb to 40 ppb (15 ppb to 20 ppb). The southern subregion e.g., Thiruvananthapuram shows a clear seasonality with values varying between (maximum of) 50 ppb and (minimum of) 20 ppb.
3. Spatiotemporal distribution of surface ozone shows maximum over a broad region covering northern and western parts, and peninsular India during pre-monsoon months (March-May), while minimum concentrations are observed during the monsoon months (June-August). An increase during the pre-monsoon month of May is more prominent over the Northern Indian region, especially over the IGP. The highest surface ozone concentrations vary mostly in the range between 40–60 ppb while the lowest values range between 15–25 ppb. During the pre-monsoon season, it crosses 70 ppb occasionally over the northern parts. The observed increase in surface ozone concentrations during the pre-monsoon months is due to increased SR and temperature which favors the photochemical ozone formation in the presence of precursors.
4. Analysis of meteorological parameters e.g., temperature, SR, winds (streamflow), and BLH show coherent seasonality where enhanced levels of surface ozone during the pre-monsoon months are linked with high surface temperature (SR) varying between 34–38°C (800–1000 W/m²). These conditions favor ozone formation through photochemical reactions (which is confirmed from the ozone distribution maps). During the monsoon months, cloudy conditions and precipitation lead to a reduction in solar radiation and surface temperature thereby reducing photochemical ozone production.
5. Analysis of PCA shows that the first two modes together explain large variance in the surface ozone with variance explained ranging between 30–50% and 10–20% respectively. The first mode indicates that during the pre-monsoon (MAM) conditions, much of the variance is explained by the south and central Indian region while in other seasons it is mostly contributed by the central and eastern Indian region. The time series of PCA-1 mode indicates an increasing trend irrespective of the seasons. The second mode during JJA shows a decreasing trend which is associated with a dipole spatial pattern resembling the regions of active and break monsoon spells during the season. The time evolution of the first and second modes shows a contrasting trend with significant interannual variability.
6. The second mode during the pre-monsoon (post-monsoon) is majorly associated with the IGP (southern) region. The temporal evolution of the second mode indicates large interannual variations indicative of strong meteorological controls on the surface ozone variations over these regions.

Declarations

Ethical approval and consent to participate: Authors consciously assure that the manuscript is an original work and follows the ethical standards of research.

Consent for publication: Not applicable.

Availability of data and materials: All the data sets used in this present study are available publicly and the same has been provided in the manuscript as well as in the acknowledgement section.

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Authors' contributions: Kumar and Singh conceptualized the problem and wrote the manuscript. RKK, BBS, KRK, KNK and VS performed the analysis. AR provided the scientific inputs on the discussions and edited the manuscript. All authors contributed towards the discussions and interpretation of the results.

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Figures

Figure 1

Location of ground-based observational stations. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 2

Scatter analysis highlighting the correlation (value mentioned in the brackets) between ground-based observations and CAMSRA surface ozone over different locations (as shown in Fig. 1) across India.

Figure 3

Surface ozone time series obtained from the daily averaged data over four different representative stations spread across India chosen each from the Northern, Eastern, Western and Southern box as shown in Figure 1.

Figure 4

Climatological spatial distribution of monthly surface ozone based on CAMSRA during the period 2003-2019. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 5

Same as Figure 4, but for standard deviation. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 6

Same as Figure 4 but for solar radiation (shaded) and temperature (contour). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 7

Same as Figure 4 but for Planetary Boundary Layer Height (shaded) and stream flow (contour). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 8

The variance explained by different independent modes obtained from the Principal Component Analysis (PCA) analyzing the seasonal surface ozone over the period 2003-2019.

Figure 9

Spatial pattern of PCA mode 1 (PCA-1) for different seasons: (a) Pre-monsoon, (b) Monsoon, (c) Post-monsoon, and (d) Winter. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 10

Similar to Figure 9 but for PCA mode 2 (PCA-2). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 11

Temporal evolution of the first two modes during different seasons corresponding to: (a) PCA-1, and (b) PCA-2

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