



Spatial variability of trace gases (NO₂, O₃ and CO) over Indian region during 2020 and 2021 COVID-19 lockdowns

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Abstract COVID-19 lockdown has given us an opportunity to investigate the pollutant concentrations in response to the restricted anthropogenic activities. The atmospheric concentration levels of nitrogen dioxide (NO₂), carbon monoxide (CO) and ozone (O₃) have been analysed for the periods during the first wave of COVID-19 lockdown in 2020 (25th March–31st May 2020) and during the partial lockdowns due to second wave in 2021 (25th March–15th June 2021) across India. The trace gas measurements from Ozone Monitoring Instrument (OMI) and Atmosphere InfraRed Sounder (AIRS) satellites have been used. An overall decrease in the concentration of O₃ (5–10%) and NO₂ (20–40%) have been observed during the 2020 lockdown when compared with business as usual (BAU) period in 2019, 2018 and 2017. However, the CO concentration increased up to 10–25% especially in the central-west region. O₃ and NO₂ slightly increased or had no change in 2021 lockdown when compared with the BAU period, but CO

showed a mixed variation prominently influenced by the biomass burning/forest fire activities. The changes in trace gas levels during 2020 lockdown have been predominantly due to the reduction in the anthropogenic activities, whereas in 2021, the changes have been mostly due to natural factors like meteorology and long-range transport, as the emission levels have been similar to that of BAU. Later phases of 2021 lockdown saw the dominant effect of rainfall events resulting in washout of pollutants. This study reveals that partial or local lockdowns have very less impact on reducing pollution levels on a regional scale as natural factors like atmospheric long-range transport and meteorology play deciding roles on their concentration levels.

Keywords COVID-19 lockdown · O₃ · NO₂ · CO · Satellite measurements · OMI · AIRS

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Introduction

Air pollution due to the conventional and unsustainable use of fossil fuels, poor urban planning and insufficient implementation of policies and measures have led to a disquieted societal situation in recent years. The accumulation of toxic air pollutants due to pollution can cause human health issues, deteriorate the ecosystem and even degenerate the economic development of the country by affecting the agricultural sector (Lal et al., 2020). According to the World

Health Organization (WHO), 91% of the world's population lives in an area where pollutant levels exceed WHO limits and resulting in an estimated 4.2 million deaths per year (WHO, 2022). However, it has been noticed that global pollution levels reduced drastically during the lockdowns implemented to combat the COVID-19 pandemic, which was first detected in the Wuhan province of China in December 2019 (Wu et al., 2020). The infectious viral outbreak has been so severe that strict restrictions have been imposed all over the world.

The government of India imposed a nationwide lockdown from 25th March 2020 to 31st May 2020 in a phase-wise manner to counter the viral spread (MHA, 2020). Phase 1 (25th March 2020–14th April 2020) has been the strictest among all. Phase 2 (15th April–3rd May), phase 3 (4th May–17th May) and phase 4 (18th May–31st May) lockdowns have been continued with few exceptions and relaxed public restrictions. This led to a sudden standstill in major economic activities such as transport, construction and industries, which eventually reduced pollutant emissions and improved the overall air quality. However, the post-lockdown period again witnessed degrading air quality as the pollutant levels started rising back to the pre-lockdown levels (Gopikrishnan et al., 2022) due to the restart of economic activities. The lockdown has provided us with a natural laboratory condition to study baseline pollutant levels in the presence of declined anthropogenic emissions. However, like other countries in the world, India too faced a second wave of mutant COVID-19 virus spread by late February 2021. This time, the government gave the liberty to the respective states to plan their lockdown strategies (depending on the severity of the spread) and a nationwide complete lockdown has not been imposed. Many states announced lockdowns from the first week of April 2021 to the second week of June 2021 (Indian Industry, 2022) (Table S1), where the restrictions have not been as strict as the lockdowns in 2020. Respective districts announced containment measures based on their COVID Test Positivity Rate (TPR) and the capacity of the healthcare system. Industries and offices have been allowed to function with 50% staff capacity in most of the states. Few states implemented only night curfews and weekend lockdowns to control the viral spread. Festival celebrations have been monitored by the authorities to keep a check on

COVID-appropriate behaviour by the public. Public transport has also been restricted to 50% occupancy (MoHFW, 2021). As a result, there have been dynamic changes in pollutant levels as the scenarios changed over time from 2020 to 2021.

Several studies have reported improvement in the air quality during the 2020 lockdown period in different countries using surface and satellite datasets. For example, Wuhan in China has the highest monthly NO₂ concentration reduction of 63%, while CO concentration reduction has been largest in Lima, Peru, in April 2020 (–61%), while all the other species decreased in concentration, O₃ on the other hand, increased during the lockdown in Wuhan, China (31–50%); Seoul, South Korea (2–33%); and Milan, Italy (12–86%) (Habibi et al., 2020). The studies, based in Almaty, Kazakhstan, show a reduction in NO₂ (–35%), CO (–49%) and an increase in O₃ (15%) (Kerimray et al., 2020). In the USA, reductions in NO₂ (–49%) and CO (–37%) have been observed. The PM reductions have been observed only where NO₂ declined the most and the impact on O₃ has been mixed and comparatively less (Chen et al., 2020). The study of NO₂ concentration data from the Sentinel-5P satellite in Iraq showed a 35–40% reduction and a 13% increase in ozone during the first lockdown (Hashim et al., 2021). While in Spain, a reduction in concentration has been observed for NO₂ (–51%), and BC (–41%), but O₃ levels increased (33 to 57%) (Tobías et al., 2020). Studies in Brazil observed a reduction of NO₂ (–54.3%), CO (–64.8%) and an increase in O₃ (30%) (Nakada & Urban, 2020). Karumuri et al. (2022) also found similar reduction (50–60%) in NO₂ and SO₂ over Arabian Peninsular using WRF-Chem model simulations.

The studies done at different parts of the Indian subcontinent also yielded a similar result. Based on the studies on the rural area of Gadanki in Andhra Pradesh (Jain et al., 2021), trace gas reduction is observed for NO₂ (–58.8%), CO (–10.1%) and O₃ (–7.1%). Here, O₃ concentration has decreased, unlike in urban areas, the NO_x is limited in the rural area and rich in VOC, any further reduction in NO_x will only decrease the concentration of O₃. Studies on CO levels at 925 hPa pressure level show a 5–10% increase over the Central India region (Alladi et al., 2021). While in the industrialized area of Gujarat, the surface data from Central Pollution Control Board (CPCB) indicates an improved Air Quality Index,

the concentration of NO_2 and CO have reduced by 30–84%, 3–55% and the O_3 concentration increased by 16–48% (Selvam et al., 2020). A 50–60% reduction in mean tropospheric columnar NO_2 level has been observed over India (Ratnam et al., 2020). Studies on Central, hilly, Indo-Gangetic Plain, North-East, Peninsular, North-West and major cities of India concluded a reduction of NO_2 and O_3 during lockdown by 5–35% and 4–18% when compared with the 2019, post-lockdown period from June to September 2020 witnessed a sudden increase in NO_2 up to 57% and O_3 had a slight decrease of 2% in levels across different part of the country. Compared to 2020 levels, an increase in NO_2 levels during the same lockdown period in 2021 has been observed, even though the lockdown has been prevalent because of the second wave of virus spread (Gopikrishnan et al., 2022).

NO_2 and CO are the primary pollutants emitted from vehicular, biomass burning and industrial sources. O_3 is a secondary pollutant whose levels depend on the concentration of NO_2 , CO and other VOC in the atmosphere (Zhang et al., 2019). Tropospheric O_3 is a strong greenhouse gas and a strong oxidant. It is responsible for the formation of photochemical smog and causes serious health issues in humans. The levels of these compounds are pivotal in determining the overall air quality of a particular region. This study will help in understanding the overall variability of O_3 and its dependency on NO_2 and CO during the reduced anthropogenic emission scenario.

This work has been unique in a way that it focuses on the inter-comparison of CO, NO_2 and O_3 level changes observed during lockdowns in 2020 (25th March to 31st May) and 2021 (partial lockdowns in different regions depending on the spread severity) using satellite datasets, with proper validation of satellite data and assessment of natural factors influence. To the best of our knowledge, no precedent studies directing the spatial variation of CO, NO_2 and O_3 over different regions of India during both lockdown periods have been conducted. This study provides an important impetus for an emission reduction strategy to address the pollution problem. It compares how the changes in anthropogenic emission during the two lockdowns (one being very strict across India and the other being partial depending on the spread severity) lead to the variability in criteria pollutants. Another key aspect that has been covered in this study is

checking the confidence levels of satellite observations in picking the sudden changes in pollutant variations during the episodic events, for which the satellite measurements have been verified by carrying out extensive statistical validation using surface measurements. The extent of meteorological influence has also been considered in addition to the anthropogenic inputs to draw conclusions on the variability of pollutants. Further, this study may help to incorporate combined dynamical changes observed in pollutant levels due to anthropogenic reduction and natural factors in the atmospheric models.

Datasets and site description

The time period of the data analysis spans over 5 years, starting from 2017 to 2021. The periods from 2017 to 2019 years have been referred to as BAU (business as usual), 2020 lockdown from 25th March to 31st May 2020 has been cited as LDN-1 and for 2021 lockdown or LDN-2, and data has been taken from 25th March 2021 to 15th June 2021. LDN-2 effects have been analysed state-wise according to the period in which lockdowns have been announced (supplementary Table S1). Indian subcontinent regions within a bounding box from 67 to 97° E and 8 to 36° N have been considered for the study. As the intensity of anthropogenic activities varies over each topographical region, spatial analysis has been conducted for the maximum possible resolution.

Atmospheric InfraRed Sounder (AIRS)

CO and O_3 have been obtained from the AIRS satellite, which is a hyperspectral infrared instrument of NASA's AQUA satellite. Level 3 data from 925 hPa pressure level with a spatial resolution of $1^\circ \times 1^\circ$ and a temporal resolution of 1 day have been used for the study. AQUA passes over the equator at 13:35 h ($\pm \sim 30$ min), and the ascending node over the Indian region is during the 13:00–14:00 h window. AIRS have a nadir footprint of 13.5 km and 2378 infrared channels ranging from 3.7 to 15.4 μm . The standard stage 2 validations, where accuracy has been determined for widely distributed locations and time using ground measurements, claim 15% and 20% accuracy for level 2 data of CO and O_3 , respectively (Pagano

et al., 2010). The data has been accessed from the NASA Goddard Earth Sciences Data and Information Services Centre (GES DISC) repository (<https://disc.gsfc.nasa.gov/>).

Ozone Monitoring Instrument (OMI)

NO₂ has been obtained from the OMI (Ozone Monitoring Instrument) of the AURA satellite which considers the Level-3 vertical column density (VCD_{trop}) with a spatial resolution of 0.25°×0.25° and a temporal resolution of 1 day. AURA passes over the equator at 13:45 h (±~30 min) and ascends over the Indian region during the 13:00 to 14:00 h window. OMI has a nadir footprint of 13 km×24 km for the optimal spectral range of 300–500 nm for ultraviolet and visible radiations. Differential optical absorption spectroscopy (DOAS) has been used to determine the slant column density, which is then converted to vertical column density (VCD_{trop}) using the air mass factor computed from radiative transfer models (Lamsal et al., 2022). The data has been taken from a public repository (<https://disc.gsfc.nasa.gov/>) managed by NASA GES DISC.

Surface measurements

The surface pollutants concentration from a few representative stations of each zone has been used to validate the satellite observations to furnish the results obtained with sufficient confidence. The surface data have been retrieved from the CPCB CAAQMS (Central Pollution Control Board—Continuous Ambient Air Quality Monitoring Stations) portal, which is a common data repository for all the state and organization-run surface stations across India (<https://app.cpcbcr.com/ccr/#/caaqm-dashboard-all/caaqm-landi ng/data>) and have been managed by CPCB Delhi. Only those stations have been selected that have at least 60% valid data during the lockdown period from 25th March to 31st May every year from 2017 to 2021, which includes a few missing records, erroneous constant values and outliers. The validation has been done for satellite data, which have been collected by the AIRS (NASA Aqua satellite) and OMI (NASA Aura Satellite) when it passes over India from 13:00 to 14:00 h time frame daily. Hence, the daily surface data have been taken for a time period of 13:00 to 14:00 h with the maximum possible temporal

resolution of 15 min. One-hour values are then averaged to remove any possible outliers. The hourly surface data for Gadanki in Andhra Pradesh have been sourced from National Atmospheric Research Laboratory (NARL), a centre which mainly specialises in atmospheric research. Gadanki is a rural site which is situated away from urban influences (Jain et al., 2021). The trace gas data from NARL have been available only from 2019 to the present.

Meteorological data

Planetary boundary layer (PBL) height, temperature and relative humidity (RH) have been taken from ERA5 Global reanalysis data sets. Hourly data have been taken with a spatial resolution of 0.25°×0.25° (~31 km) from the 925 hPa pressure level. ERA-5 is the fifth generation ECMWF (European Centre for Medium-Range Weather Forecasts) atmospheric reanalysis of global climate variables covering the period from 1979 to the present with a temporal resolution of 1 h and resolve the atmosphere using 137 pressure levels from surface to a height of 80 km where it is 0.01 hPa. (Hersbach et al., 2020) (<https://cds.climate.copernicus.eu/cdsapp#!/search?type=dataset>).

The daily rainfall data over the Indian region have been taken from the India Meteorological Department (IMD) website, where the data have been managed by the Climate Monitoring and Prediction group (https://www.imdpune.gov.in/Clim_Pred_LRF_New/Gridded_Data_Download.html). Daily gridded data with 0.25°×0.25° spatial resolution has been used for the analysis (Pai et al., 2014).

Fire counts

Fire occurrences that have been identified within 1 km×1 km spatial resolution tiles by the Moderate Resolution Imaging Spectro-radiometer (MODIS) from the TERRA and AQUA satellites have been used for this study (Giglio et al., 2016). The fire counts data for the 2020 and 2021 lockdown period have been accessed from FIRMS (Fire Information for Resource Management System) portal (<https://firms.modaps.eosdis.nasa.gov>). The fire occurrences detected in each 0.5°×0.5° grid over India have been summed to calculate the fire count for that area.

Methodology and validation of satellite observations

Methodology

The percentage change in pollutant levels has been estimated relative to the BAU years, 2017, 2018 and 2019, when there have been no lockdowns prevalent and anthropogenic activities contributed to the normal emission levels. The validation has been carried out by calculating Pearson’s correlation value while comparing daily satellite and surface measurements over the relevant period in BAU and lockdown years. The conversion of satellite data into a similar unit of surface data has been done to maintain uniformity while comparing. The daily satellite measurements of CO, NO₂ and O₃ have been averaged in a phase-wise manner for the lockdown period in 2020. For 2021 lockdown phase 1, periods have been chosen according to the initiation of restrictions in each state (supplementary Table S1). The percentage change in concentration during the lockdown period with the corresponding BAU period has been calculated using Eq. (1) and plotted spatially.

$$\text{Percentage change} = \frac{(\text{CLDN} - \text{CBAU}) \times 100}{\text{CBAU}} \quad (1)$$

Meteorological data have been analysed by comparing the average phase-wise variation of temperature, PBL height, rainfall and relative humidity during the lockdown period, a net quantitative study on the changes observed during lockdown with BAU has been carried out to conclude the overall meteorological influence on the spatial difference

in pollutant levels. The overall workflow from data acquisition to further analysis in this study has been shown in the supplementary Figure S1.

Validation of satellite observations

Satellite data may contain some known unknowns whose removal before the study poses analytical challenges (Loew et al., 2017). Since relative changes have been compared in this study, it is obvious that the systemic error cancels out and results are unaffected. However, it is important to check whether the satellite measurements are sensitive enough to capture the changes observed in the surface level by the ground stations. This has been achieved by validating the satellite using surface measurements. The correlation between the hourly average of satellite and surface data has been determined for a few representative locations where a significant increase or decrease in pollutant levels has been observed. Only a few stations had more than 60% valid data during the BAU and LDN periods as shown in Fig. 1a (I Delhi, II Pali, III Asansol, IV Chandrapur, V Solapur, VI Gadanki). Table 1 and supplementary Figure S2 indicate Pearson’s correlation value (*r*-value) for pollutants in sites with valid datasets. According to Student’s *t*-test, for *p*=0.05 confidence level, *r*-value greater than or equal to 0.13 has a significant correlation between the two datasets (*N*=135 to 200) (Weathington et al., 2012). O₃ and NO₂ have sufficient correlation (*r*>0.13), but for CO, a good association is not evident. The concentration of CO and NO₂ have been dependent on spatial variation and anthropogenic activities in an area (Shmool et al., 2014). The surface data considers the

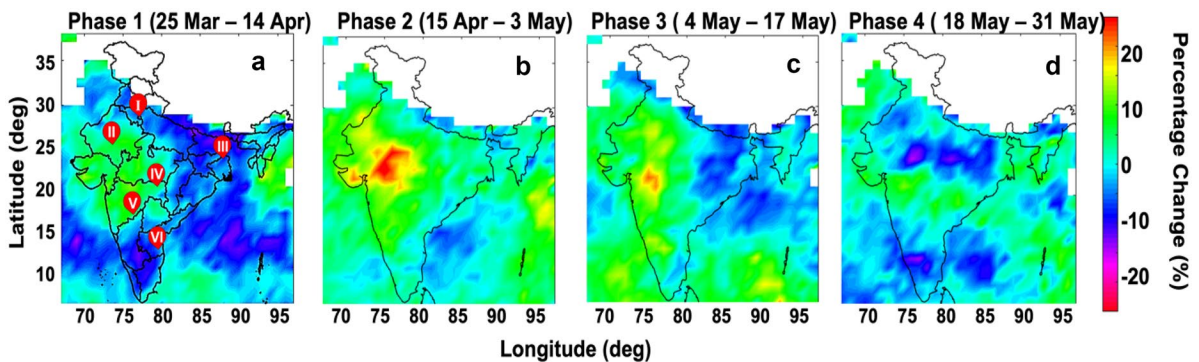


Fig. 1 Phase-wise variation of CO over India during LDN-1 (2020). Roman letters in (a) indicate surface stations selected for validation of satellite data, (I) Delhi, (II) Pali, (III) Asansol, (IV) Chandrapur, (V) Solapur, (VI) Gadanki

Table 1 Pearson's correlation analysis results between surface and satellite measurements for few representative locations

Zone	State	Station name	Correlation coefficient (<i>r</i> -value)					
			CO		NO ₂		O ₃	
			BAU	LDN	BAU	LDN	BAU	LDN
North	Delhi	Anand Vihar	0.14	0.13	0.15	0.35	0.23	0.2
West	Rajasthan	Pali*	0.24	-0.11	0.51	0.08	0.42	0.2
East	West Bengal	Asansol*	0.01	0.04	0.15	0.59	0.21	0.58
Peninsular	Maharashtra	Chandrapur	0.11	0.2	0.13	0.2	0.22	0.25
	Andhra Pradesh	Gadanki**	-	0.29	0.27	0.36	-	0.56
	Kerala	Plamoodu, TVM*	-	-	0.32	0.17	0.37	0.34
	Maharashtra	Solapur	0.22	-0.01	0.25	0.46	0.1	0.27

*BAU period includes only 2018 and 2019, **BAU period includes only 2019
Correlation values in bold italics are statistically significant at 95% confidence level ($p=0.05$)

measurements from a specific point area which may be industrial, forest, urban, rural, etc. In urban and industrial regions, CO and NO₂ concentrations are high during peak office hours (Cichowicz & Stelegowski, 2019). AIRS data takes an average of 10×10 spatial resolutions that cover a large area. Therefore, considerable variation in values ought to happen when the area is diverse with different intensities of activities. NO₂ shows a much better *r*-value when compared to CO because the OMI instrument measures data with a spatial resolution of 0.25°×0.25°, which gives much more accurate data. The atmospheric lifetime of O₃ is several weeks (Venter et al., 2020), and its concentration is mainly dependent on seasonal variation, temporal changes, solar radiation and also on the concentration of precursor molecules rather than spatial changes; hence, a good correlation has been evident in the case of ozone measurement. In general, it can be inferred that satellite measurements have been able to qualitatively pick up the changes observed from the surface, and both datasets go hand in hand. Therefore, the satellite data can be confidently used for the spatial study of relative pollutant changes after validating with the surface measurements.

Results

Spatial variation in air pollutant's level

Carbon monoxide (CO)

The variation of CO in Fig. 1 shows a substantial increase in the levels in the LDN-1 period when compared with the same period in BAU for the majority of the Indian regions. The increase in

concentration has been higher in the central-west part of India, with a percentage increase ranging from 10 to 25% for all four phases of the lockdown. Maharashtra, Madhya Pradesh and Gujarat are among the top states having the greatest number of highly polluting industries; it has been one of the main industrial hotspots in India. South and north-eastern parts of India witnessed a slight decrease in concentration during this period. During LDN-2 phase 1 (Fig. 2), the concentration of CO has been less compared to the BAU period in most parts of the Indo-Gangetic plain and South India. In Central India, an increase of up to 10 to 25% has been observed, which changes to a 5 to 15% decrease compared to BAU when it transitions to later phases of LDN-2. When the lockdown periods have been compared for both 2020 and 2021 (Fig. 7a), it has been observed that there is an overall decrease in CO concentration in 2021. Except in Jharkhand, West Bengal, Chhattisgarh and a few north-eastern states where an increase in concentration up to 5% has been observed, as the second lockdown has not been strictly imposed in those regions (Bagwaiya, 2021). However, during the phase 1 period, the decrease in levels in 2021 has been observed only in north-west regions and the rest of the country witnessed an increase of 10 to 20%.

Previous studies have been carried out in different parts of the country by (Jain & Sharma, 2020) and in rural areas of Gadanki by (Jain et al., 2021) show an overall decrease of ~28 to ~10% in the CO levels during the 2020 lockdown period. A rise in CO concentrations (~20%) has been reported by Rawat and Naja (2022) and Alladi et al. (2021) over the central and western parts of India during the 2020 lockdown. Similar results have been observed

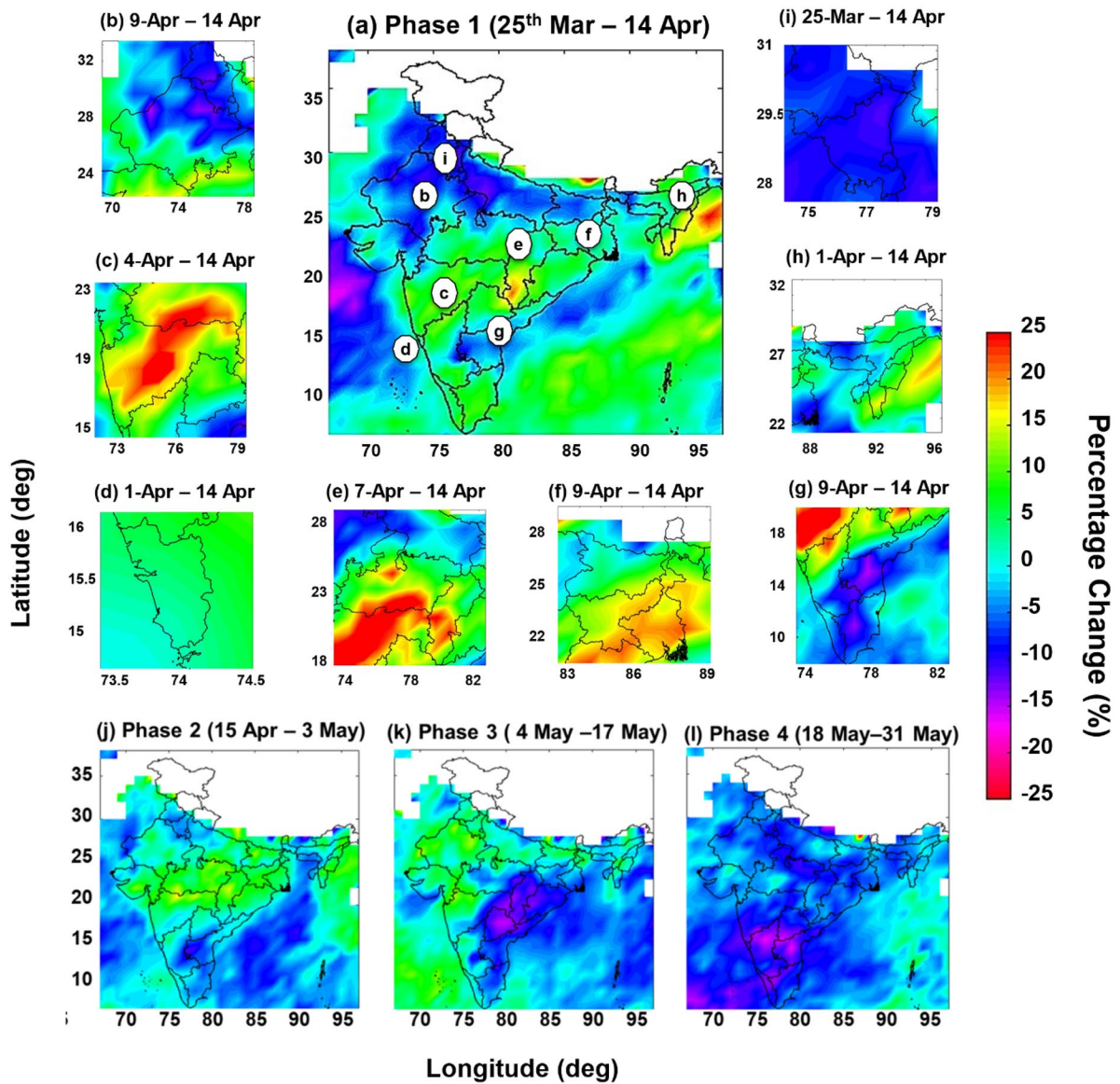


Fig. 2 CO percentage change for (a–i) LDN-2 (2021) phase 1 vs corresponding BAU period for states which announced curfew, (a) Indian subcontinent, (b) Rajasthan, Delhi, (c) Maharashtra, (d) Goa, (e) Madhya Pradesh, Chhattisgarh, (f) Bihar,

West Bengal, (g) Tamil Nadu, Telangana, Andhra Pradesh, (h) north-east states, (i) Chandigarh, (j) LDN-2 phase 2 (k)LDN-2 phase 3, (l) LDN-2 phase 4

in our studies, where an increase of 10 to 20% in the central-west region and a decrease of ~10% in the southern part of India has been observed.

Nitrogen dioxide (NO₂)

There has been an overall decrease in NO₂ levels during LDN-1 phases when compared with BAU

(Fig. 3). The degree of variation however varies over different regions; in Central-West India negative changes of about 5% have been observed, while a 10 to 20% decrease has been seen in most of the Indian regions. A few small areas of Jharkhand, West Bengal, Delhi and Mizoram witnessed a change of about –30 to –40%. The decrease in NO₂ levels witnessed in phase 1 has been in the range of –30 to

–40%. A slight increase or no change in NO_2 concentration has been observed during the first three phases of LDN-2 when compared with the BAU period (Fig. 4), with most of the Indian region having up to a 10% increase in concentration. As the LDN-2 period progressed from phase 1 to phase 4 significant decreases in NO_2 levels have been witnessed and the decrease is prominent over the southern, central-east and eastern part of the country. The LDN-2 period when compared with LDN-1 (Fig. 7b) showed an increase in the NO_2 levels over the entire Indian subcontinent. A 30 to 40% increase can be seen in the central and north-eastern parts of India for all the lockdown phases. While phase 4 witnessed a slight decrease of 5 to 10% in South India.

A satellite study by Biswal et al. (2020) reported a decrease of ~30% in the urban agglomeration, and up to ~50% lower levels in megacities such as New Delhi, Bangalore, Chennai and Mumbai, and 10–24% decrease have been observed over forestland, which is in good correlation with the results we have observed for 2020 lockdown. A study comparing the changes in the 2020 and 2021 lockdown period by Gopikrishnan et al. (2022) shows NO_2 level rise in the range of 24, 21, 17, 31, 8 and 5.5% in Indo-Gangetic Plain, Central, Hilly, North-East, North-West and Peninsular India respectively during 2021 lockdown, which has been similar to the 10 to 25% increase observed in IGP, central, peninsular and north-east part of the country in this study.

Ozone (O_3)

The O_3 concentration during LDN-1 has been less compared to the BAU period in almost all regions

of the Indian subcontinent (Fig. 5). In central-west and North-East India, there has been a slight increase in the O_3 concentration up to 5% in phase 1. The rest of the country witnessed an overall decrease of up to 10%. During phase 3 and phase 4, a slight increase in levels in some regions has been observed. The LDN-2 showed an overall increase in levels during phase 1 and phase 2, and phase 3 and phase 4 witnessed an overall decrease (Fig. 6). The O_3 levels during LDN-2 phase 1 and phase 2 have been higher by 10 to 15% when compared to LDN-1 in all regions of India, while a decrease has been seen during phase 3 and phase 4 of LDN-2 with respect to LDN-1 (Fig. 7c).

O_3 levels varied spatially over different parts of the country, based on the level of activity in that region. The rural areas witnessed a decrease in the levels up to 7% (Jain et al., 2021) during LDN-1. Based on a study by Selvam et al. (2020), an increase in the range of 16–48% has been observed in the urban areas of Gujarat, similar to that observed trend in our study. A decrease in levels has been witnessed over the southern part of India, which is in line with the studies by Singh et al. (2020).

Effect of meteorology

Apart from anthropogenic influence, natural factors like meteorological parameters (temperature, relative humidity), planetary boundary layer (PBL) height and long-range transport also have a significant role in maintaining pollutant concentrations (Ratnam et al., 2021). A rise in humidity may impact the chemical processes in the atmosphere by involving the formation of oxidant species such as OH (Seinfeld & Pandis, 2016). An increase in RH may also influence the

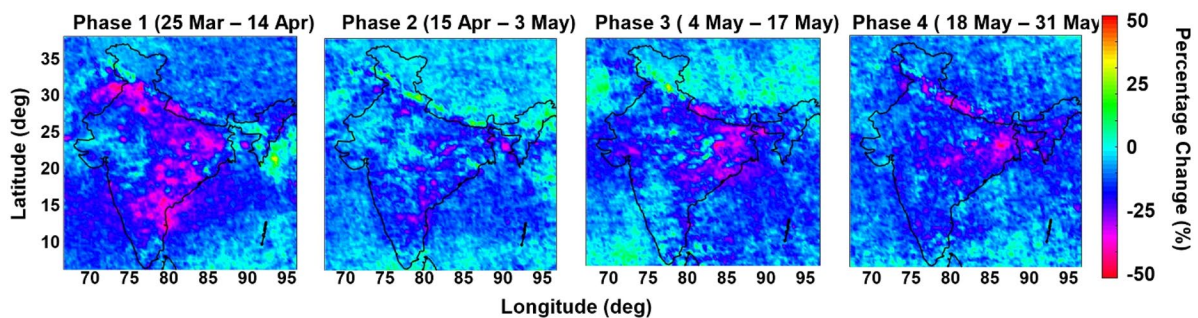


Fig. 3 Phase-wise variation of NO_2 over India during LDN-1 (2020)

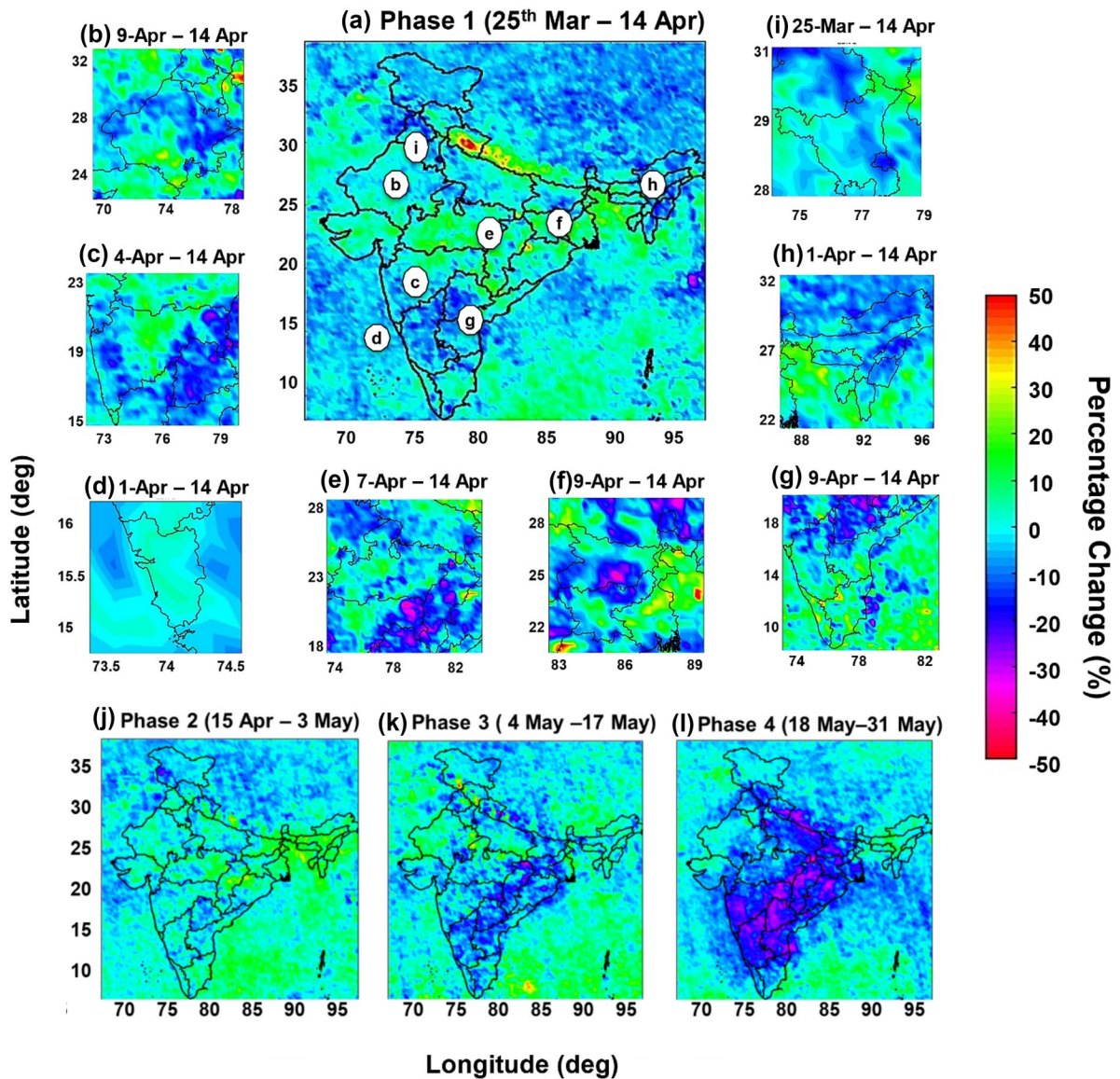


Fig. 4 NO₂ percentage change for (a–i) LDN-2 (2021) phase 1 vs corresponding BAU period for states which announced curfew (a) Indian subcontinent, (b) Rajasthan, Delhi, (c) Maharashtra, (d) Goa, (e) Madhya Pradesh, Chhattisgarh, (f) Bihar,

West Bengal, (g) Tamil Nadu, Telangana, Andhra Pradesh, (h) north-east states, (i) Chandigarh, (j) LDN-2 phase 2 (k) LDN-2 phase 3, (l) LDN-2 phase 4

optical and chemical properties of aerosols which may indirectly influence the photochemical oxidation of different chemical species in the atmosphere. Changes in the vertical temperature structure within the troposphere may have an impact on the evolution and dynamics of the boundary layer, as well as the mixing processes that influence the dispersion and transport of air pollutants (Ratnam et al., 2021),

and it can also accelerate the rate of reaction involved in the production of tropospheric ozone. Changes in PBL height directly affect the distribution of pollutants within the boundary layer and their volume mixing ratio. Analysis of the meteorological changes modelled over India during the 2020 phase 1 lockdown period showed a considerable increase in RH (10–40%), a reduction in surface temperature (17%)

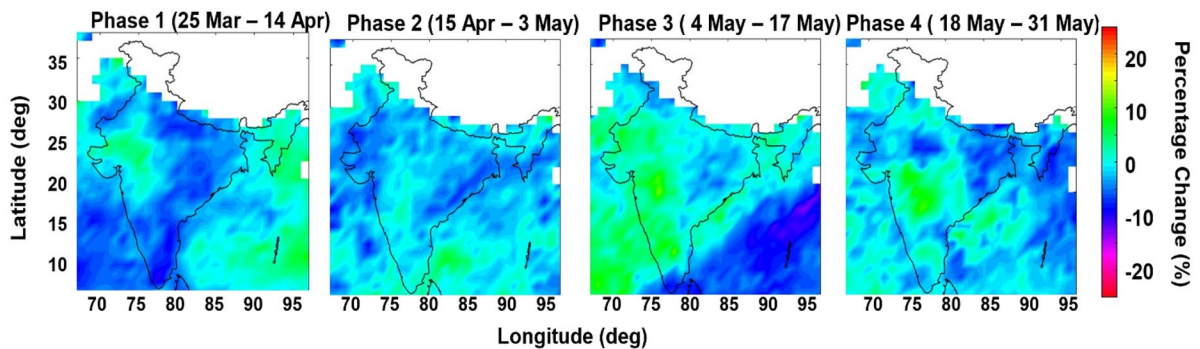


Fig. 5 Phase-wise variation in O_3 over India during LDN-1 (2020)

and a significant increase in PBL height (40–80%) when compared with non-lockdown years (Basha et al., 2022; Ratnam et al., 2021). Therefore, it is very important to judiciously analyse the meteorological entities while interpreting the data during the lockdown periods.

Phase-wise variation in the PBL height (Fig. 8), temperature (supplementary Figure S3) and relative humidity (RH) (supplementary Figure S4) for LDN-1 and LDN-2 with respect to BAU over India has been compared. High solar insolation and maximum peak in temperature observed during afternoon hours result in the highest peak of PBL evolution during that period of the day. In most of the regions, the temperature was lower during the LDN-1 and LDN-2 periods when compared to the BAU periods. The effect of PBL variation can be witnessed in the pollutant levels over that region (supplementary Figure S6(c) and Fig. 9c) and the regions having low PBL height show increased pollutant levels and vice versa during both the lockdown period. A similar trend in PBL spatial variation by Basha et al. (2022) has been observed while considering the ERA data sets as well. Changes in RH have been consistent with the variations observed in temperature. During LDN-1, the effect of RH has not been much evident in the reduction of pollutants over a region (supplementary Figure S6(d)). However, during LDN-2, RH plays a significant role in exhibiting changes in pollutant levels, especially from phase 2 onwards, where the economic activities have not been restricted at all (Fig. 9d). Increased RH levels during phase 3 and phase 4 have been due to the transition to the monsoon season and rainfall over India which led to the washout of pollutants. Overall

rainfall has been higher during LDN-2 compared to that of LDN-1 (supplementary Figure S5).

Discussion

The changes in trace gas levels have been influenced by both reductions in anthropogenic emissions as well as natural factors like meteorology and long-range transport. Temperature, RH and PBL height have been varying during LDN compared to BAU years. The high levels of pollutants observed in the central part of India may be due to the effect of long-range transport of pollutants and aerosols from the Middle East and Africa due to changes in wind direction and further getting constrained in the anti-cyclonic circulation over Central India (Ratnam et al., 2021). In LDN-1, the variations in pollutant concentration have been mainly due to change in anthropogenic emission intensity rather than the changes in meteorology over an area. However, in the case of LDN-2, the anthropogenic emissions levels have been comparable with BAU and the variations in pollutant levels observed have been mainly due to the natural factors like meteorology and long-range transport.

Since major sectors such as mining, thermal power plants and steel industries have been exempted from being closed down during LDN-1 and LDN-2, they continued to contribute to the pollution (Menon & Kohli, 2020). An overall decrease in the concentration of pollutants has been observed over different zones of India (Biswal et al., 2020; Gopikrishnan et al., 2022; Jain & Sharma, 2020; Jain et al., 2021; Kumar et al., 2020; Ratnam et al., 2020; Rawat & Naja, 2022) during both the lockdown years

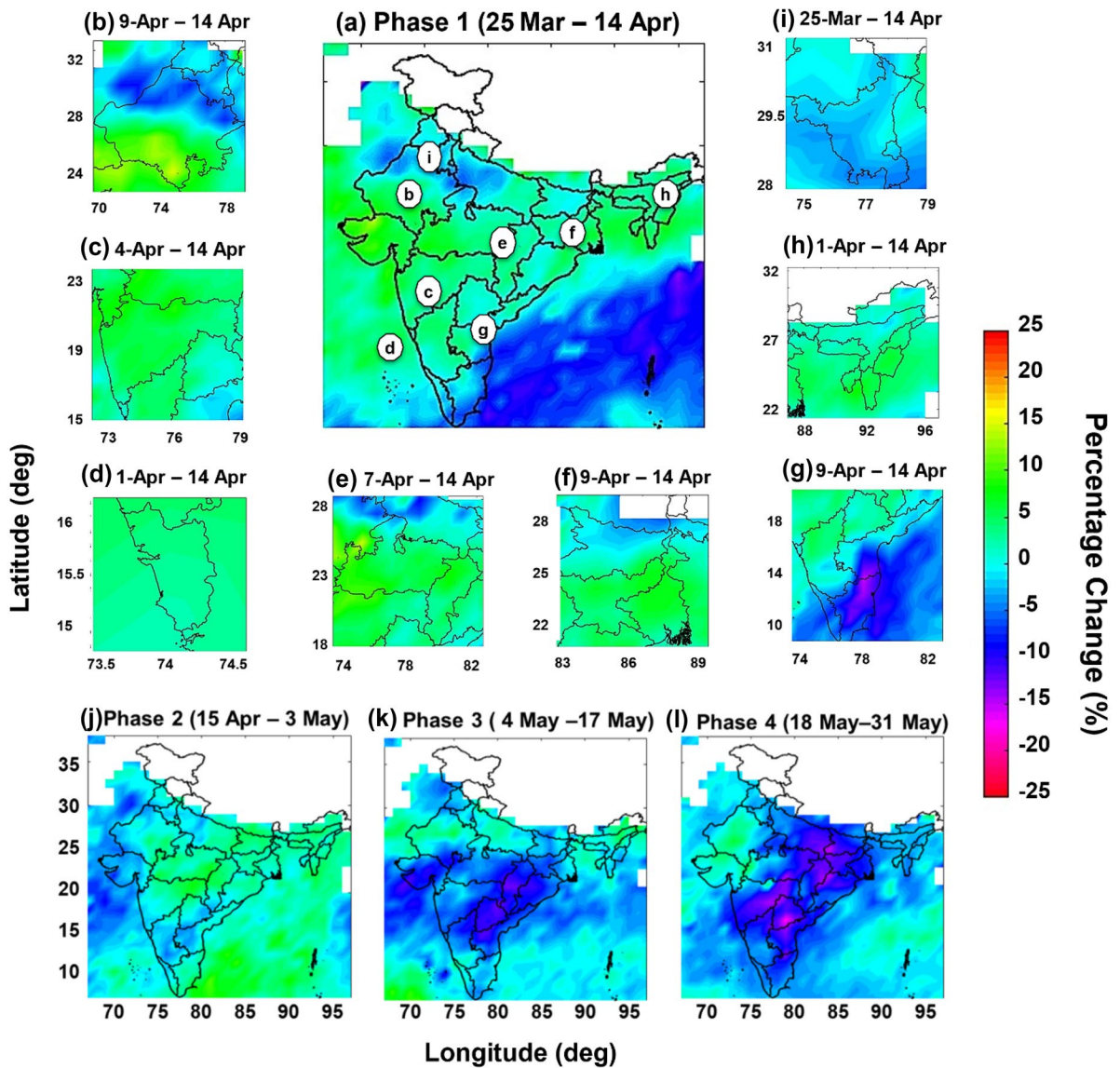


Fig. 6 O₃ percentage change for (a–i) LDN-2 (2021) phase 1 vs corresponding BAU period for states who announced curfew (a) Indian subcontinent, (b) Rajasthan, Delhi, (c) Maharashtra, (d) Goa, (e) Madhya Pradesh, Chhattisgarh, (f) Bihar,

West Bengal, (g) Tamil Nadu, Telangana, Andhra Pradesh, (h) north-east states, (i) Chandigarh, (j) LDN-2 phase 2 (k) LDN-2 phase 3, (l) LDN-2 phase 4

compared to BAU period depending on the pollutant characteristics and the region of observation. Changes in CO have been more complex because of the interference of forest fires and biomass-burning activities along with the anthropogenic sources. Biomass burning and forest fires have been higher in the western part of Central India and North India during the 2020 lockdown when compared to the 2016–2020

average (Biswal et al., 2020), which would have resulted in an abnormal increase in CO levels. Fire count analysis using MODIS data for the LDN-1 and LDN-2 showed that, for the LDN2 phase 1 fire counts have been higher than that of LDN-1 phase 1 levels in central and north-east regions, and directly coincide the CO levels (supplementary Figure S6(a) and Fig. 9a). Higher CO levels have been observed in

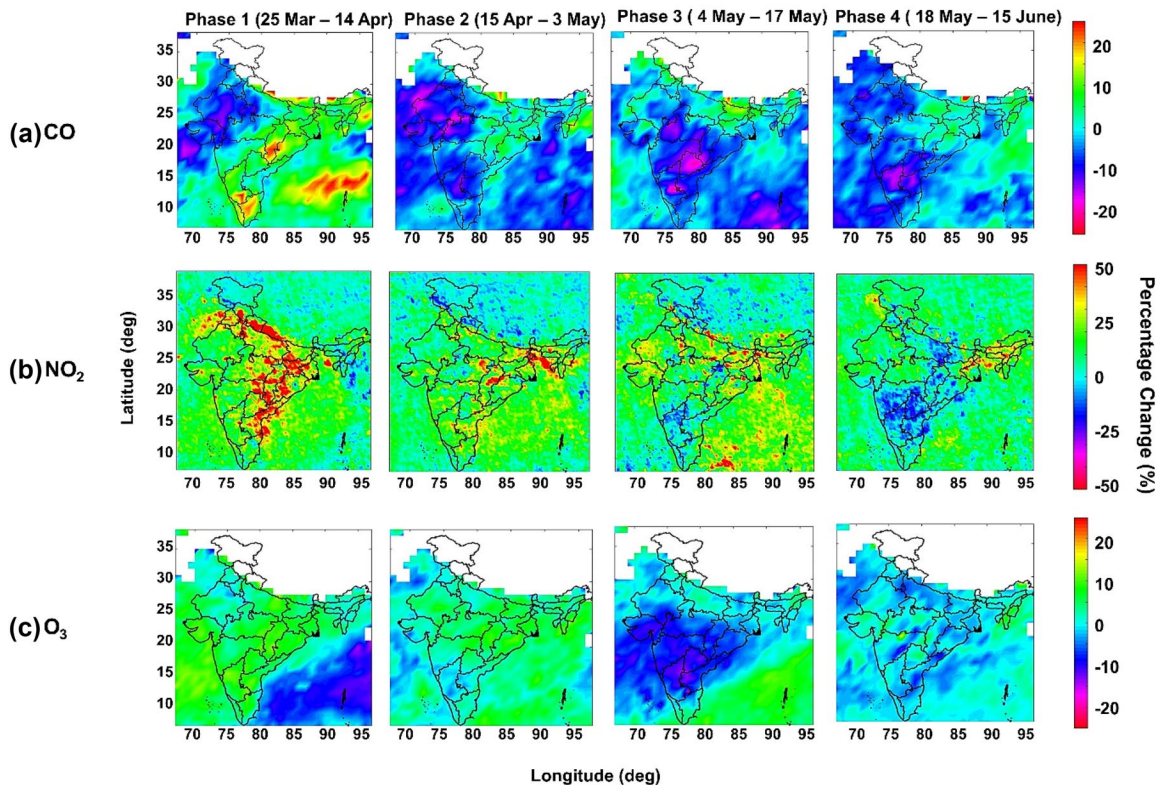


Fig. 7 Phase-wise variation of (a) CO, (b) NO₂ and (c) O₃ in LDN-2 (2021) compared to LDN-1 (2020) levels

those locations where more fire activity was detected. Vehicular and industrial emissions have a direct impact on the NO₂ levels in the atmosphere over a region and hence faced an overall reduction during the LDN-1 period. The largest reductions in levels have been seen in urban areas and in most of the megacities. Post-LDN-1 witnessed the regeneration of NO₂ to its normal level as the strict government lockdown norms have been released. During LDN-2, the lockdown rules have not been strictly implemented, and hence the economic activities have been similar to that during BAU; therefore, the changes in concentration of NO₂ in 2021 have been comparable to that during the BAU period. The decrease in pollutant levels in LDN-2 phase 3 and phase 4 has been mostly associated with the incoming south-east monsoon during that period. Rainfall causes washout of the pollutants from the atmosphere, thereby reducing its level (Yoo et al., 2014). Rainfall has a considerable

effect on the pollutant levels over a region. In LDN-1, the role of rainfall in reducing the pollutant level is minimal, as the pollutant levels have been already low due to reduced anthropogenic actions and the changes observed due to washout has been insignificant (supplementary Figure S6(b)). However, from LDN-2 phase 2 onwards washout effects have been clearly witnessed (Fig. 9b), where regions having less rainfall compared to BAU period observed an increase in pollutant levels and vice versa. O₃ formation is dependent on the precursor NO_x and VOC molecules (Seinfeld & Pandis, 2016); during LDN-1, an overall decrease in O₃ concentration can be seen throughout the country except in the central region, where an increase in concentration is observed; this can be due to the less concentration of NO₂ in urban areas during this period. O₃ concentration increased during LDN-2 in the initial two phases compared to the levels during LDN-1; this can be drawn from the fact that NO₂

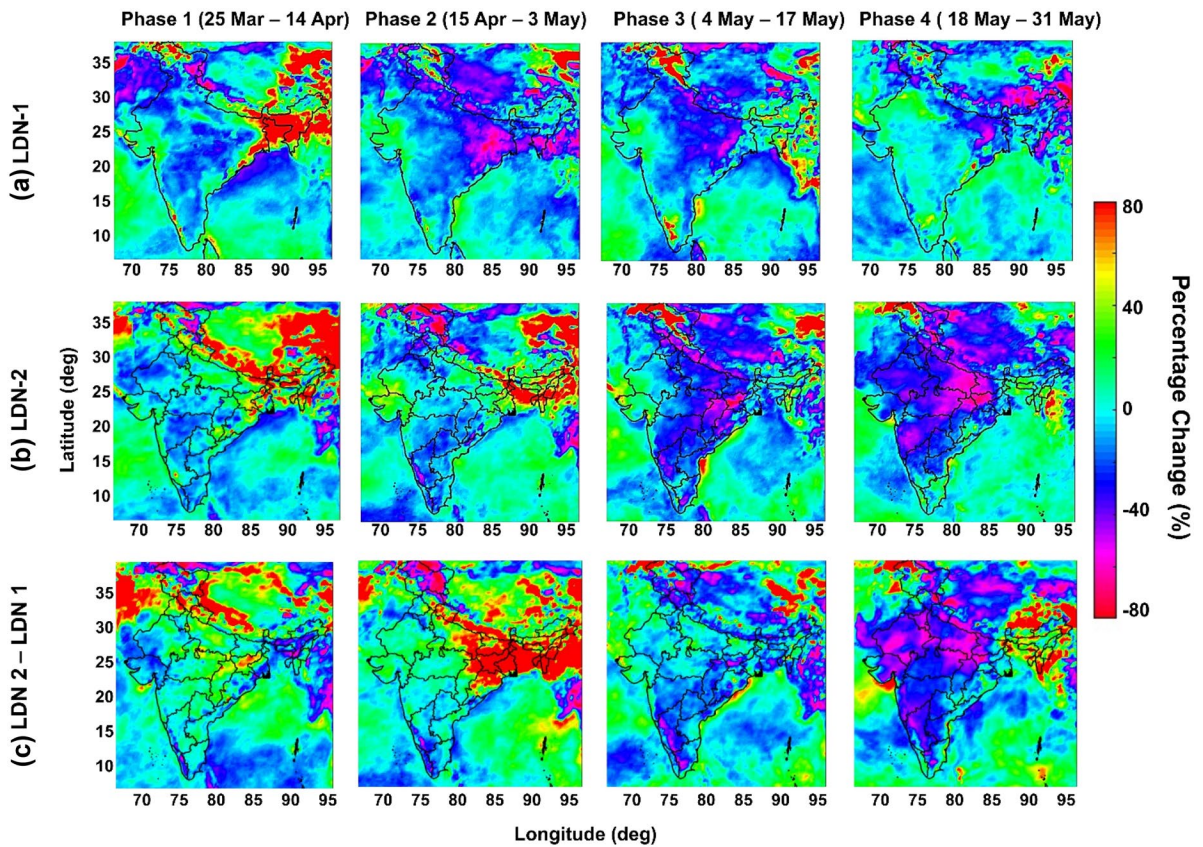


Fig. 8 Phase-wise PBL differences during (a) LDN-1 (2020) compared to BAU, (b) LDN-2 (2021) compared to BAU and (c) LDN-2 (2021) compared to LDN-1 (2020)

concentration increased much more in 2021 when compared to 2020, and hence optimum levels of NO_2 and VOCs enhanced the production of ozone in that region. The decrease in ozone during LDN-2 phase 3 and phase 4 are again mainly due to the cleansing effect due to higher rainfall events.

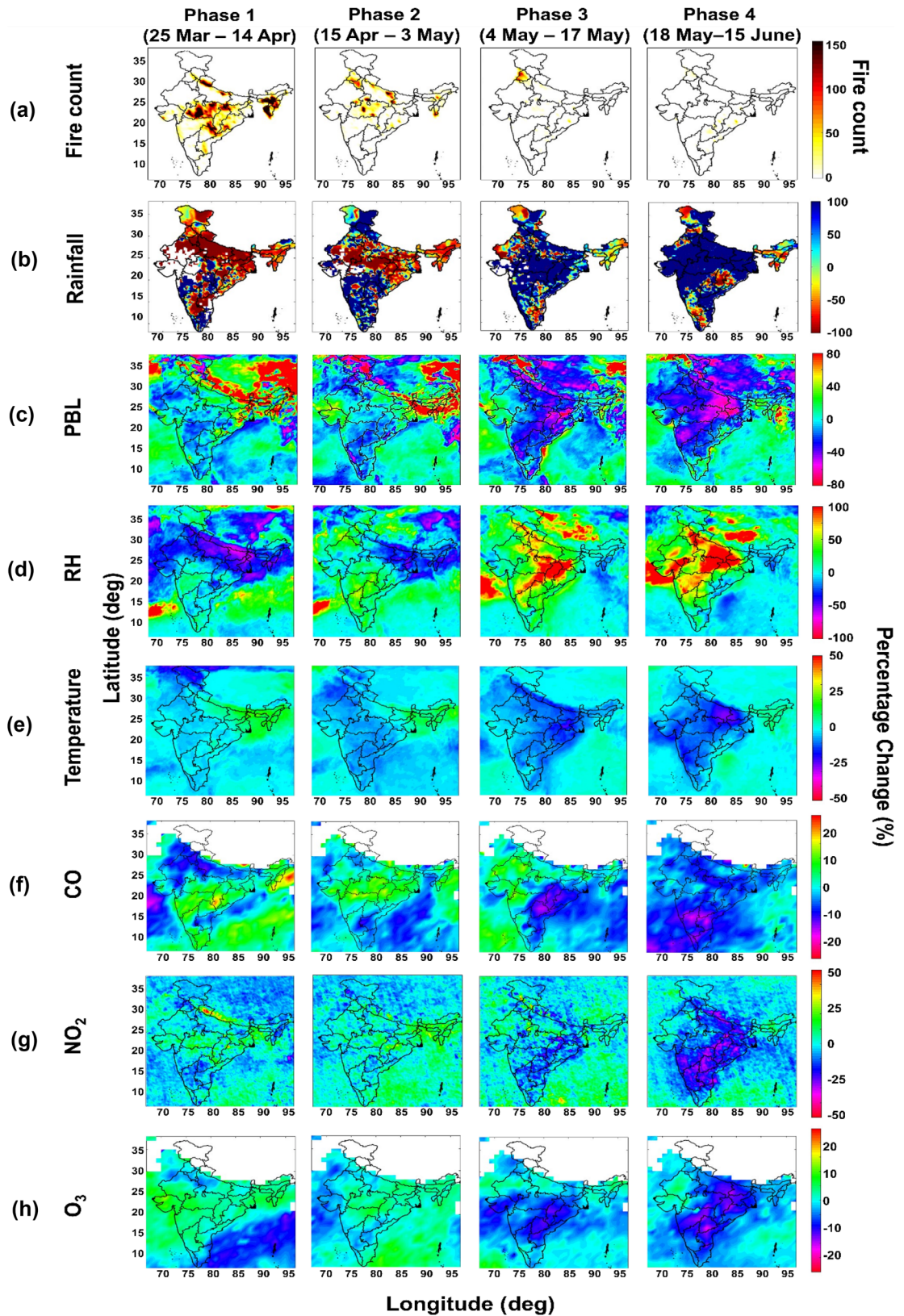
Conclusions

The lockdown scenario enabled us to get more significant insights into the variation of trace gas levels with the changing emission intensities and other natural processes during the lockdown period in 2020 (LDN-1) and 2021 (LDN-2) over the Indian subcontinent. The pollutants having direct influence from anthropogenic activities such as CO and NO_2 witnessed a sufficient decrease in their levels during LDN-1 and LDN-2. However, the degree of change varied for

both years according to the level of economic activities prevalent in that region. Ozone formation chemistry is influenced by the levels of precursor molecules, which has been reflected in the results obtained. The main findings are summarised in the following:

1. CO had an overall increase of 10–25% during LDN-1, and a mixed variation in the levels during LDN-2 due to the influence from biomass burning and other meteorological factors such as rainfall and PBL. The NO_2 concentration reduced drastically by 20 to 40% throughout India in LDN-1, and the reduction has been less during LDN-2. O_3 levels have reduced by 5 to 10% in LDN-1 and witnessed a rise by 10% during LDN-2 in most parts of the Indian subcontinent as optimum levels of precursor NO_x and VOCs have been favouring O_3 formation. O_3 levels vary with the changes in NO_2 levels.

2.



◀**Fig. 9** Phase-wise variation of (a) fire counts, (b) rainfall, (c) PBL, (d) RH, (e) temperature, (f) CO, (g) NO₂, and (h) O₃ during LDN-2 (2021) when compared to BAU

Satellite and surface data correlate with each other significantly in most cases (with Pearson's correlation of $r \geq 0.13$ showing a significant correlation between the datasets). Low Pearson's coefficient observed in some cases can be due to the coarser data resolution and increased spatial emission sensitivity of the pollutant.

3. The impact of meteorological factors on the observed trace gas level is pronounced in LDN-2 than LDN-1 period. In contrast with LDN-1, the anthropogenic emissions remained unchanged during partial lockdown in 2021 and most of the variations observed in pollutant levels can be attributed to the varying PBL height, RH and rainfall pattern.
4. This study showed that partial lockdowns at the local level do not have much impact on emission and pollutant levels. The overall changes observed have been mainly due to meteorological influence and other natural factors unlike in the LDN-1 where it has been mostly due to a reduction in anthropogenic activity.

These findings need to be considered while implementing any policy for the reduction of air pollution in India.

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Author contribution V. G. Vignesh: data curation and analysis, methodology, plotting and writing—original draft preparation; Chaithanya D. Jain: conceptualisation, methodology, visualisation, data curation and writing—original draft preparation; K. Saikranthi: methodology, data curation and writing—reviewing and editing; M. Venkat Ratnam: methodology, visualisation, resources, supervision and writing—reviewing and editing.

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Data availability The data used in this study can be obtained on request.

Declarations

Competing interests The authors declare no competing interests.

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