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Drivers of air pollution variability during second wave of COVID-19 in Delhi, India

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ABSTRACT

To curb the 2nd wave of COVID-19 disease in April–May 2021, a night curfew followed by full lockdown was imposed over the National Capital Territory, Delhi. We have analyzed the observed variation in pollutants and meteorology, and role of local and transboundary emission sources during night-curfew and lockdown, as compared to pre-lockdown period and identical periods of 2020 lockdown as well as of 2018 and 2019. In 2021, concentration of pollutants (except O₃, SO₂, and toluene) declined by 4–16% during night-curfew as compared to the pre-lockdown period but these changes are not statistically significant. During lockdown in 2021, various pollutants decreased by 1–28% as compared to the night-curfew (except O₃ and PM_{2.5}), but increased by 31–129% compared to the identical period of 2020 lockdown except O₃. Advection of pollutants from the region of moderate lockdown restrictions and an abrupt increase in crop-residue burning activity (120–587%) over Haryana and Punjab increased the air pollution levels over NCT during the lockdown period of 2021 as compared to 2020 in addition to a significant contribution of long-range transport. The increase in PM_{2.5} during the lockdown period of 2021 compared to 2020 might led to 5–29 additional premature mortalities.

1. Introduction

The novel coronavirus disease started in Wuhan, China in late 2019 and quickly spread worldwide within a few months (Singhal, 2020). During February–March 2020, the COVID-19 cases increased sharply at a rate of more than 300% per week in various countries like China, Germany, Italy, UK, USA, and India, etc. (Pai et al., 2020). Therefore, the governments imposed restrictions on human activities to curb the spread of the disease. These restrictions substantially reduced air pollution which was seen in both the ground-based and space-borne observations (Jain and Sharma, 2020; Sharma et al., 2020; Rahaman et al., 2021; Sathe et al., 2021). The first COVID-19 case in India was reported on January 30, 2020, and COVID-19 cases kept gradually increasing up to March 10, 2020. 366

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cases were reported between March 16–23, 2020 (<https://covid19.who.int/region/searo/country/in>). To control the spread of the COVID-19 disease, the Government of India announced a nationwide lockdown of 21 days starting March 24, 2020 and extended the lockdown further for the next 21 days till May 3, 2020 (Sharma et al., 2020). All businesses like industries, transport, markets, shops, construction, hotel, and mining, etc. were shut down. As a result, a large reduction in air pollution levels was reported in Delhi (Jain and Sharma, 2020; Sharma et al., 2020). The Government of India implemented a phased reopening plan starting in June 2020 despite a gradual increase in COVID-19 cases (Table 1). India experienced the first COVID-19 peak in August–September 2020 and active COVID-19 cases steadily declined thereafter until January 2021. Unfortunately, COVID-19 cases started to surge again in late February and early March of 2021 and India experienced the second peak of COVID-19 in late April to early May of 2021. In Delhi, positive COVID-19 cases started rising sharply in the first week of April. The government of Delhi started imposing restrictions and initially implemented a night curfew from April 07 to April 16, but cases continued to rise. Therefore, the Government decided to impose more restrictions and announced a lockdown from April 17, 2021, for one week, which was later extended by one more week till April 30, 2021.

During the second wave, the timing and extent of the restriction on human activities were determined by the state governments and not by the central government as was the case in the first wave. In 2021, the restrictions were not as strict as during phases 1 and 2 of the lockdown in 2020 (from March 24 to May 03 2020). People were allowed to commute for essential goods, medical services, takeout deliveries from restaurants, retail shops, e-commerce, taxi services (e.g., Ola and Uber), metro trains, and public transport with a limited number of buses and trains for migrants as well as the frontline staff who served during the lockdown, and airports were allowed with limited activity. The neighboring states (Uttar Pradesh, Haryana, Punjab, and Rajasthan) of Delhi did not impose a lockdown until April 30, 2021. Therefore, this period provides a unique opportunity to understand the effect of local versus transboundary emission sources on air quality in Delhi.

Most of the studies on the air quality of Delhi target the pollution sources in the late autumn season (October and November) because of the potential influence of large scale crop residue burning in upwind states of Haryana and Punjab (Kumar et al., 2020; Kulkarni et al., 2020) and meteorology which favors the dispersion of pollutants through slower winds, lower temperature, shallower planetary boundary height thereby causing accumulation of pollution not only in Delhi but throughout the Indo-Gangetic plain (Sharma et al., 2020; Beig et al., 2021). During April–May, crop-residue burning activity is similar but the meteorology favors the dispersion of pollutants (Agarwala and Chandel, 2020) in Delhi. We saw a sharp rise of about 2–8 times in springtime fire activity over Haryana and Punjab (of IGP, India) in 2020 as compared to the previous years (Fig. S1), which coincided with a significant increase in the air pollution level in Delhi despite of COVID-19 induced restrictions as discussed later.

Here, we present a comprehensive analysis of the drivers of air pollution variability during the second wave of COVID-19 in Delhi. We also provide information about the potential primary sources and regions affecting air quality in Delhi. Finally, we estimate the health impacts of changes in PM_{2.5} concentration in Delhi from the lockdown of 2020 to 2021.

2. Data and methods

2.1. Different phases of the study period

We have analyzed air pollution variability during March–April of 2018 to 2021 in Delhi in view of the varying COVID-19 restrictions on both the local and transboundary sources relative to the non-COVID-19 affected years. COVID-19 restrictions in 2020 and 2021 led to a unique opportunity that allows the investigation of the relative importance of local and non-local sources in Delhi. The lockdown in 2020 was implemented nationwide from March 24 to May 3, 2020 and thus both the local and non-local sources experienced similar restrictions. In contrast, the lockdown restrictions from April 07 to 30, 2021 were imposed only in Delhi but not in the neighboring states of Delhi.

We classify our observations in pre-lockdown and lockdown periods to investigate the impacts of varying restrictions on Delhi's air

Table 1
Different phases of lockdowns and unlocks during 2020 and 2021 in Delhi.

Year	Phase	Date	Duration	Description
2020	Lockdown 1	25 Mar - 14 Apr 2020	21 days	Complete lockdown. Only emergency services were allowed.
	Lockdown 2	15 Apr - 3 May 2020	19 days	Complete lockdown. Only emergency services were allowed.
	Lockdown 3	4 May - 17 May 2020	14 days	Lockdown with limited public movement for essential goods and mass movement with limited public transport.
	Lockdown 4	18 May - 31 May 2020	14 days	Lockdown with limited public movement for essential goods and mass movement with limited public transport.
	Unlock 1	1 Jun - 30 Jun 2020	30 days	Govt office with limited capacity.
	Unlock 2	1 Jul - 31 Jul 2020	31 days	Industrial activity and corporate office with limited capacity.
	Unlock 3	1 Aug - 31 Aug 2020	31 days	All activity allowed with covid-19 guidelines with limited manpower.
2021	Night curfew	7 Apr - 16 Apr 2021	10 days	Restriction only between 10 PM to 5 AM.
	Lockdown 4	17 Apr - 30 Apr 2021	14 days	Only essential and emergency services allow.

Source: <https://www.mha.gov.in/notifications/circulars-covid-19>

quality. For 2020, we define the pre-lockdown period from March 01 to 23 as Period A, and lock down period from March 24 to April 30 as Period B (Table S1). Period A in 2021 is defined as March 01 to April 06 because the government of Delhi implemented restrictions from April 07 onwards. Period B is divided into two periods, namely B1 (April 07 to 16) and B2 (April 17 to 30) corresponding to the night curfew and full lockdown, respectively. We use 2018 and 2019 as years representative of normal anthropogenic activities and also classify their observations in A, B, B1, and B2 periods.

2.2. Ground station observation and methods

This study focused on the National Capital Territory (NCT) Delhi (Fig. 1), which is the second-largest populous megacity in the world (United Nations, 2018). The NCT is a part of the Indo Gangetic Plain in northern India with a subtropical humid climate. As per the report “Population Projections for India and States (2011–2036)” published by the Government of India (National Commission on Population, 2019), the total projected population of Delhi is about 20.19 million for 2020. The NCT consists of the highest number of automobiles in the country and covers a 1484 km² land area, and a population density of 11,297 people/km² (Census, 2011). It shares land boundaries with two Indian states (Haryana and Uttar Pradesh).

In this study, fine and coarse particulate matter (PM_{2.5}, PM₁₀), oxides of nitrogen (NO_x), ozone (O₃), ammonia (NH₃), sulfur dioxide (SO₂), carbon dioxide (CO), benzene, and toluene, along with meteorological parameters like ambient air temperature (AT), relative humidity (RH), wind speed (WS), planetary boundary layer height (PBLH), and rainfall (RF) were used. The air pollutants and meteorological data were collected from 36 continuous air quality monitoring stations (Fig. 1), which are operated by the central pollution control board (CPCB), Delhi state pollution control committee (DPCC), the Indian Institute of Tropical Meteorology (IITM), and the Indian Meteorological Department (IMD) over the NCT (<https://app.cpcbcr.com/ccr/#/cqm-dashboard/cam-landing/data>). The hourly data for PBLH was obtained from the climate data store(<https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels?tab=form>). All the original data were subjected to quality control as explained in the supplementary material Part-1. The Ventilation Coefficient (VC) is directly related to the dispersion of pollutants with higher VC means a greater vertical mixing of pollutants within the PBLH (Sujatha et al., 2016). The VC is estimated at each site using Eq. (1):

$$VC = PBLH \times WS \quad (1)$$

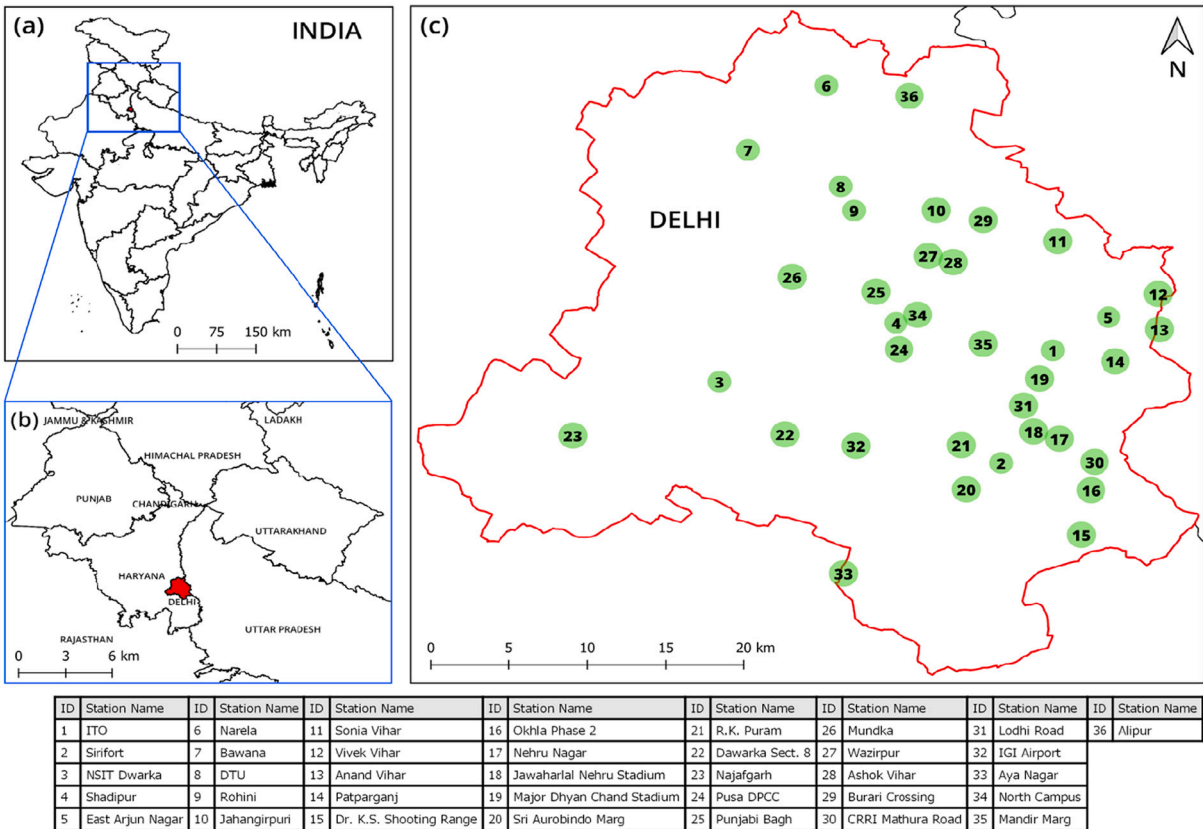


Fig. 1. [a] Map of India showing the location of the study region marked by the blue square. [b] A zoom-in view of the study region with Indian state names. Delhi is filled with red color. [c] Location and names of all the monitoring stations in Delhi. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

We have also evaluated the major sources and source regions of pollutants in Delhi, using the Principal Component Analysis (PCA) and HYSPLIT trajectory analysis. The five days backward trajectories were calculated using the HYSPLIT model to identify the movement of air parcels during each period and year. The backward trajectory only reports the movement of the air parcel but does not tell us about the potential source region. Therefore, we used 3D cluster analysis at a height of 500 m and concentrated weighted trajectory analysis, to identify the potential source region and contribution of a region to the receptor site (Fig. S5a and S5b). The clusters were formed by the angle distance clustering method that follows Yang et al. (2017). PM_{2.5} concentrations were combined with a backward trajectory to identify the potential contributing region by the concentration-weighted trajectory method. The receptor site was National Physical Laboratory (central Delhi) for backward trajectory analysis. The Detailed methodology about PCA and HYSPLIT are explained in supplementary material Part-1(Sections S2 and S3).

2.3. Changes in emission

Accurate quantification of changes in emissions due to COVID-19 induced restrictions deserves a separate study but here we have attempted to provide a first order approximation of COVID-19 induced modification in emissions. Specifically, we have estimated the adjustment factor for three sectors to quantify the reduction in the emission of pollutants namely black carbon (BC), organic carbon (OC), NO_x, SO₂, NH₃, CO, and non-methane volatile hydrocarbon (NMVOC). Anthropogenic emissions data from the Copernicus Atmosphere Monitoring Service (CAMS) global anthropogenic emission inventory (CAMS-GLOB-ANT_v4.2_R1.1; Granier et al., 2019) are used as the base emissions. The adjustment factor is generated for each sector based on the activity data for each sector on each day during April 2021 as given by Doumbia et al. (2021). The activity data are obtained from various sources (Google mobility, Apple, and TomTom) for different timescales, depending on geographical location but only Google mobility data (COVID-19 Community Mobility Reports (google.com)) available on each day for Delhi are used to calculate adjustment factors. The Apple and TomTom datasets were not available for public use. The Google mobility data available in percentage changes from baseline activity for 6 categories namely retail, park, grocery & pharmacy, transition, workplace, and residential. The baseline activity data are defined by Google as median value of activity data of 5 week from January 01 2020 (Doumbia et al., 2021). The percentage changes in daily activity from baseline activity data during March and April 2020–21 are shown in Fig. 2.

This study covers the period from April 1 to April 30, 2021 for emission adjustment and compares it with the same period of 2019. The adjustment factors (AF) are calculated by this formula (Fig. 3).

$$AF = 1 + G/100$$

Where, G represents the daily change in mobility from baseline mobility data for each sector.

AF < 1 represents the reduction in daily activity.

AF > 1 represents an increase in daily activity.

The adjustment factor for the road transportation sector is generated from the estimation of transit usage (i.e., public transportation

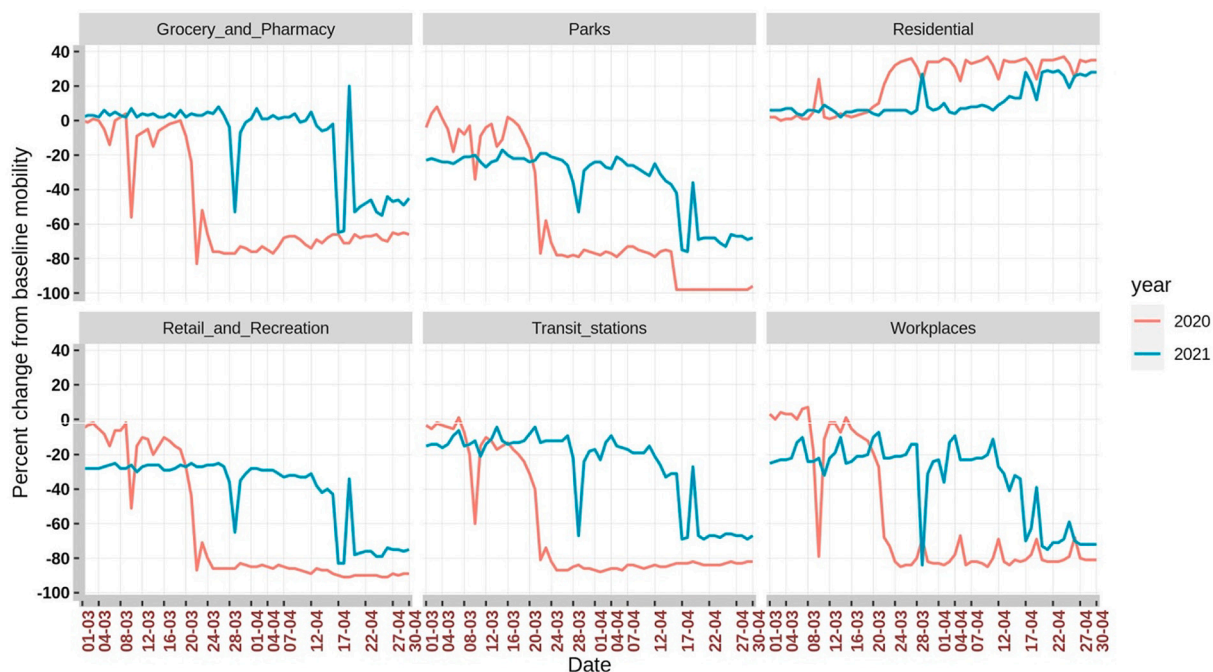


Fig. 2. Percentage changes in google mobility data for 6 categories during March–April of 2020 and 2021 in Delhi.

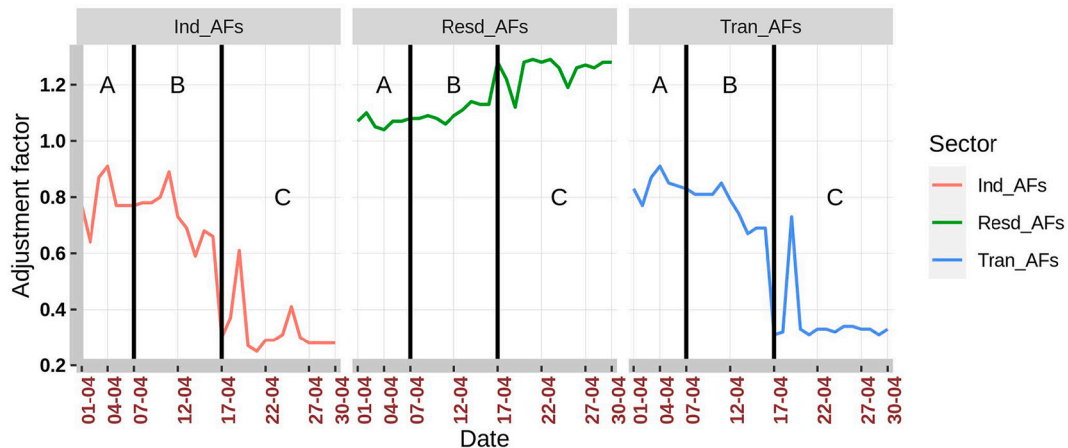


Fig. 3. Daily adjustment factor for road transport, industries, and residential sectors during April 2021. Here A, B, C represent the pre-lockdown (April 1–6), night-curfew (April 7–16) and lockdown (April 17–30) period.

including buses, subways, and train stations) made by Google. The major part of industrial emission comes from the manufacturing activities utilizing fossil fuel. Since daily data about industrial processes was not available, we use Google workplace mobility data to calculate an AF, which represents the percentage of people traveling to or from their workplaces, representative of changes in industrial activities during each day of April. The AF for residential is generated from Google residential category data, which represents the duration during which people are constrained to their home through restrictions applied by the local government. This duration represents the additional time that people spent at home due to the restrictions (Fig. 3). We could not verify the changes at ground level in these activity data. The changes in local emission were calculated using CAMS-GLOB-ATN_v4.2_R1.1 data. We have resampled the data from cell size 0.1 degree to 0.001 degree using nearest neighbor interpolation, this is crucial since clipping the raw data of cell size 0.1 degree (11 km) at the edges can omit/include surrounding areas and add uncertainty in computing total emission from Delhi. Reducing the cell size of data before clipping enables better quantitative estimates of total emission over the study area. The AF is applied to each sector separately on each day and daily emission files for each sector are generated. Then combined emission of all three sectors on each day have been estimated and calculated for each period of study (April 1–6, April 7–16 and April 17–30). This study only have considered changes in emission over Delhi in three sectors, road transport, industries and residential rather actual emission.

2.4. Short-term mortality

The short-term mortality burden of all causes, ages, and both sexes from exposure to $PM_{2.5}$ over the NCT are estimated for each day following Atkinson et al. (2014), who estimated that a $10 \mu g m^{-3}$ increase in daily mean $PM_{2.5}$ (C_d) ambient exposure enhances the risk (γ) of mortality by 1.04% (0.52%–1.56% with 95% Confidence Interval) with a $PM_{2.5}$ reference concentration (C_r) of $0 \mu g m^{-3}$. This study assumes no upper limits for exposure concentration. No India-specific risk functions for $PM_{2.5}$ exposure are available at this time. 1.04% is on a conservative side compared to other studies like Levy et al. (2012) and WHO (2013), who estimated the percentage risks as 1.2% and 1.23%, respectively. Relative Risk (RR_d) of short term exposure is estimated as follows (Chen et al., 2020):

$$RR_d = 1 + [\gamma (C_d - C_r) \times 0.1] \quad (2)$$

The premature mortality for each age and all causes or specific causes (disease) were calculated as a function of population, relative risk, and baseline mortality rate for the study region. We use annual baseline mortality data from the Institute of Health Metrics, and Evaluation (GBD, 2019) for all causes, gender, and age across India due to lack of daily mortality data from India. The annual death rate was divided by 365.25, similar to Chen et al. (2020) and Jat and Gurjar (2021). The daily mortality (M_d) is estimated as:

$$M_d = P \times I_d \times \frac{RR_d - 1}{RR_d} \quad (3)$$

Where, P , RR_d , and I_d are Population, Relative risk, and daily death rate over the study area.

3. Results and discussion

3.1. Daily variations in meteorology

Daily averaged AT, WS, PBLH, and VC increased but RH decreased from period A to period B of all the years. WS decreased from $1.32 m s^{-1}$ to $1.19 m s^{-1}$ (Table 2) in the year of 2021 only. These changes in meteorological parameters occur naturally as we move from a cooler March to hotter and drier April. To understand interannual variability in meteorological parameters during the lockdown

Table 2

Mean value of pollutants, meteorological parameters except RF (sum of rainfall considered during each period) and percentage variation in-between period A, B, B1 and B2 during 2018, 2019, 2020, and 2021.

Variable	Average value of variable										% change in variables value between time period A, B, B1, and B2 for each year				
	Period-A				Period -B			Period-B1		Period-B2					
	1 March to 23 March		1 March to 6 April		24 March to 30 April			7April to 16 April		17 April to 30 April	From A to B		From A to B1		From B1 to B2
	2018	2019	2020	2021	2018	2019	2020	2021	2021	2021	2018	2019	2020	2021	2021
PM ₁₀ ($\mu\text{g m}^{-3}$)	216.8	174.3	140	248.5	244.4	219.7	93.7	233.8		225.9	13	26	−33	−6	−3
PM _{2.5} ($\mu\text{g m}^{-3}$)	100.2	84.4	65.6	94.8	91.6	82.8	42.9	79.9		96.3	−9	−2	−35	−16	21
CO (mg m^{-3})	1.2	1.2	1	1.2	1.4	1.3	0.8	1.1		1.1	10	14	−24	−10	−3
Ozone ($\mu\text{g m}^{-3}$)	50.9	42.1	41.3	36.2	64.5	55.8	45.6	41.7		48	27	33	10	15	15
NOx(ppb)	62.3	52.2	45.4	52.2	58.2	52.9	20.2	49.4		35.8	−7	1	−56	−5	−28
NH ₃ ($\mu\text{g m}^{-3}$)	41	32	33	49	40.1	33.6	27.9	41.1		39	−2	5	−16	−16	−5
SO ₂ ($\mu\text{g m}^{-3}$)	15.9	17.4	14.7	19	15.1	21.4	14.3	19.7		19.5	−5	23	−3	4	−1
Benzene ($\mu\text{g m}^{-3}$)	3	4.2	2.9	3.6	3.7	4.3	1.9	3.5		3.3	25	3	−34	−4	−6
Toluene ($\mu\text{g m}^{-3}$)	NA	32.4	25.6	25	NA	31.8	12.4	26.2		23.1	NA	−2	−52	5	−12
AT (°C)	24.3	19.8	20.3	24.8	29.6	29.3	27.4	29.4		29.2	22	48	35	19	−1
RH (%)	50	52.6	64.1	44.9	37.5	35.6	47.7	30.9		37.4	−25	−32	−26	−31	21
WS (m s^{-1})	1.2	1.4	1.2	1.3	1.4	1.4	1.2	1.2		1.3	18	3	3	−10	5
PBLH (m)	432.8	438.1	392.9	483.7	646.1	639.3	532.2	854		866.1	49	46	35	77	1
VC ($\text{m}^2 \text{s}^{-1}$)	519.4	628.2	464	665.1	902.7	931.2	628.6	1015.6		1067.9	74	48	35	53	5
RF (mm)	0	3	24.105	0.04	0	3.5	11.2	0		1	0	16	−53	−100	

Data not available: (NA), Period-A:- No restriction period; Period-B:- Lockdown period in 2020; Period-B1:- Partial restriction (Night curfew) in 2021, Period-B2:- Lockdown period in 2021.

periods, we compared Period B of 2020 and B2 in 2021 with the identical period of previous years (Table S2). In 2020, a decrease of 7, 16, 17, and 31% was observed in AT, WS, PBLH, and VC, respectively, from the average of identical periods in 2018 and 2019, while the RH increased. This suggests that meteorological conditions were more favorable for the accumulation of pollutants during the lockdown period of 2020 than during 2018–2019. The changes in VC in 2021 shows that meteorology favored less dispersion than 2018 and 2019 but more dispersion than 2020. Rainfall data was not available for 2018, and very few rainfall events were seen in 2019, 2020, and 2021 (Fig. S2). A two-day rainfall event occurred in 2020 during period A and another single event occurred during period B (Fig. S2). No major rainfall events were seen in other years.

3.2. Daily variation of pollutants

Daily mean values of PM_{2.5}, PM₁₀, CO, O₃, NO_x, and SO₂ over Delhi for the period of March 1 to April 30 (2018–2021) are presented in Fig. 4 and other pollutants (NH₃, benzene and toluene) are shown in Supplementary Fig. S2.

During period A (not affected by COVID-19 related restrictions), the daily averaged values of PM₁₀, PM_{2.5}, CO, O₃, and NO_x decreased from 2018 to 2020. The mean concentration of all these species in 2021 was either higher than the last three years or comparable to 2018 except for O₃ which continued to decrease (Table 2). Mean value of NO_x was higher in 2021 compared to 2020 but smaller than in 2018–19. Benzene concentration showed random fluctuations, but toluene decreased consistently from 2018 to 2021 during Period A (Fig. S2). All the pollutants (except O₃) have a significant negative correlation at $p > 0.05$ with AT and WS and a significant positive correlation with RH (Fig. S3). The lowest pollutant concentration in Period A was found during 2020 despite the lowest VC as compared to the earlier three years. This could be due to a possible decrease in emission from different sources as several policies and initiatives e.g., the Pradhan Mantri Ujjwala Yojana to reduce household solid biofuel consumption, nationwide

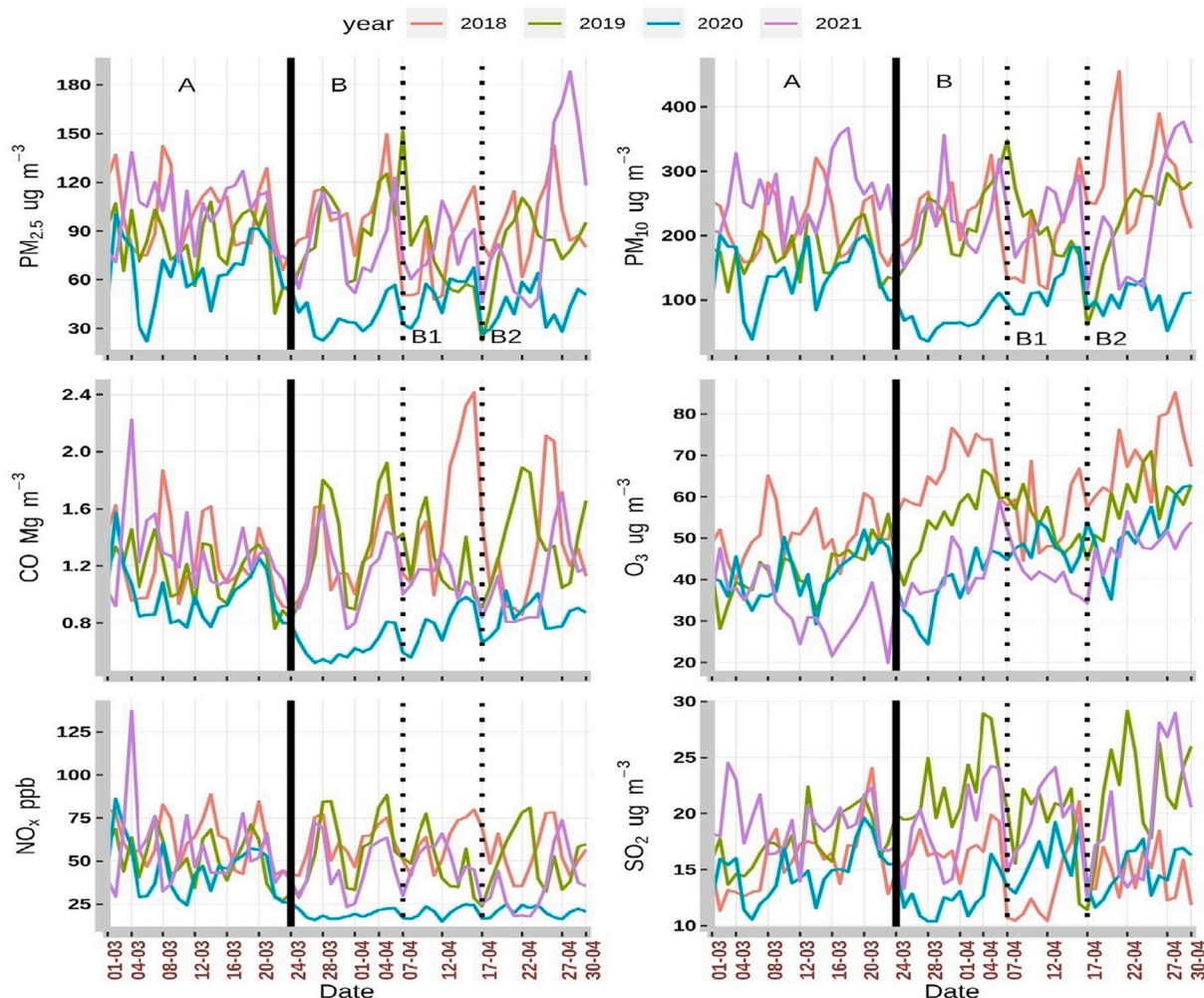


Fig. 4. Daily mean variation of selected pollutants and meteorological parameters in Delhi. All air pollutants data are averaged over monitoring sites of Delhi. The graphs are divided into two parts by thick black lines, where A and B represent pre-lockdown and lockdown periods for 2020, while B1 and B2 between thin dotted lines represent partial and complete restriction periods for 2021.

enforcement of Bharat Stage IV emission norms in April 2017, increase in CNG based public transport, reduced age of diesel-based automobile (both light, and heavy-duty vehicle) in the NCT, and expansion of electric metro facility over the NCT region) initiated by the government in the last decade to tackle the issue of air pollution in Delhi (Singh et al., 2021). Singh et al. (2021) reported the decrease in the mass concentration of PM_{2.5} over Delhi at a rate of $\sim 4 \mu\text{g m}^{-3}$ per year from 2014 to 2019. However, a more systematic analysis of these policies along with observations over a large area is needed. Note that the impact of these policies since 2020 has been negated partially by inflation and the impact of COVID-19 caused economic depression. The lockdown during 2020 severely impacted poor and middle-class families in India, with about 67% job losses reported in this section, and the highest impact was seen on women's jobs (Basole et al., 2021). The price of an LPG cylinder was also raised in 2020. All these factors forced the poor or middle-class families to switch back to using solid biofuel for household energy needs (Ravindra et al., 2021). The results of Principal Component Analysis (PCA) also suggest fossil fuel and solid biomass burning as the major sources during Period A in 2021 (Tables 3a and 3b). This could be a possible reason behind an increase in the level of pollutants, despite the highest dispersion rate in 2021 compared to the past three years.

During Period B (affected by 2020 lockdown restrictions), the daily mean value of pollutants reduced by 3 to 63% as compared to period A except for O₃ (Table 1), and a decrease of 6 to 65% was observed, when compared with an average concentration of identical period of the previous two years (Table S2). The highest decrease in PM₁₀, PM_{2.5}, NO_x, benzene, and toluene may be due to the greater contribution of direct anthropogenic emissions to these species (Klimont et al., 2017; Sharma et al., 2020). The SO₂ concentration slightly decreased compared to period A, likely because no restrictions were imposed on thermal power plants in northern India (Sharma et al., 2020). A 10% increase in O₃ was seen as compared to Period A but it decreased 24% compared to the identical period of the last two-year average. Since near-surface O₃ in urban areas is primarily produced by photochemistry, changes in ozone cannot be related linearly to changes in the emissions of its precursors. The changes in ozone depend on whether ozone production in a given area is controlled by VOCs or NO_x (Sharma et al., 2021). Large increases in ozone along with a decrease in VOCs and NO_x during COVID-19 induced lockdown period was also observed in China and other counties over the world (Shi and Brasseur, 2020; Faridi et al., 2021). The lowest fire counts were observed over Haryana, and Punjab in 2020 as compared to previous years (Fig. S1). The dispersion of pollutants was at the lowest level during Period B as compared to Period A (Table 2) and the identical period of previous two years (Table S2). Due to the absence of conducive dispersion the pollution reduction is likely driven by a reduction in anthropogenic emission sources only. Therefore, this period provides an opportunity to define the baseline achievable concentration of pollutants in Delhi.

During Period B1 (night curfew) in 2021, the concentration of PM₁₀, PM_{2.5}, CO, NO_x, NH₃, and benzene decreased by 6, 16, 10, 5, 16, and 4%, respectively but the value of O₃ and SO₂ increased by 15%, and 4%, as compared to Period A (Table 2). This reduction in PM₁₀, PM_{2.5}, CO, NO_x, and benzene is due to a reduction in fossil fuel burning from vehicles, restaurants, and pubs, etc. (Dantas et al., 2020), as compared to Period-A. An increase in O₃ concentration is usually due to changes in meteorology precursors as explained in the previous paragraph. In addition, due to non-available of fresh NO and NO₂ in the evening, nighttime ozone chemistry i.e., destruction of O₃ by NO₂ for the formation of N₂O₅ is slowed (Brown and Stutz, 2012). Here we saw an increase in SO₂ peak during day time, as compared to period A, which means during day time the industrial or other activities have increased, likely to minimize the impact of nighttime curfew on industrial production that leads to an increase in the concentration of SO₂.

During Period B2 (full lockdown) in 2021, the concentration of PM₁₀, CO, NO_x, SO₂, NH₃, benzene, and toluene decreased by 3, 3, 28, 1, 5, 6, and 12%, respectively but an incremental change in PM_{2.5} and O₃ was observed, as compared to the Period B1 (Night curfew). Furthermore, compared with an average of identical periods of 2018 and 2019, we observe a decrease in the concentration of PM₁₀, CO, O₃, NO_x, and benzene, but an increase in the value of PM_{2.5}, NH₃, SO₂, and toluene. The reduction in the concentration of CO, NO_x, and benzene could be due to a reduction in anthropogenic sources like vehicles, industries, etc., while the gain in PM_{2.5}, CO, and SO₂ could be due to a sharp rise in crop-residue burning in the upwind states of Haryana and Punjab (Fig. 6). The O₃ concentration

Table 3a
PCA results of each period during 2018–19.

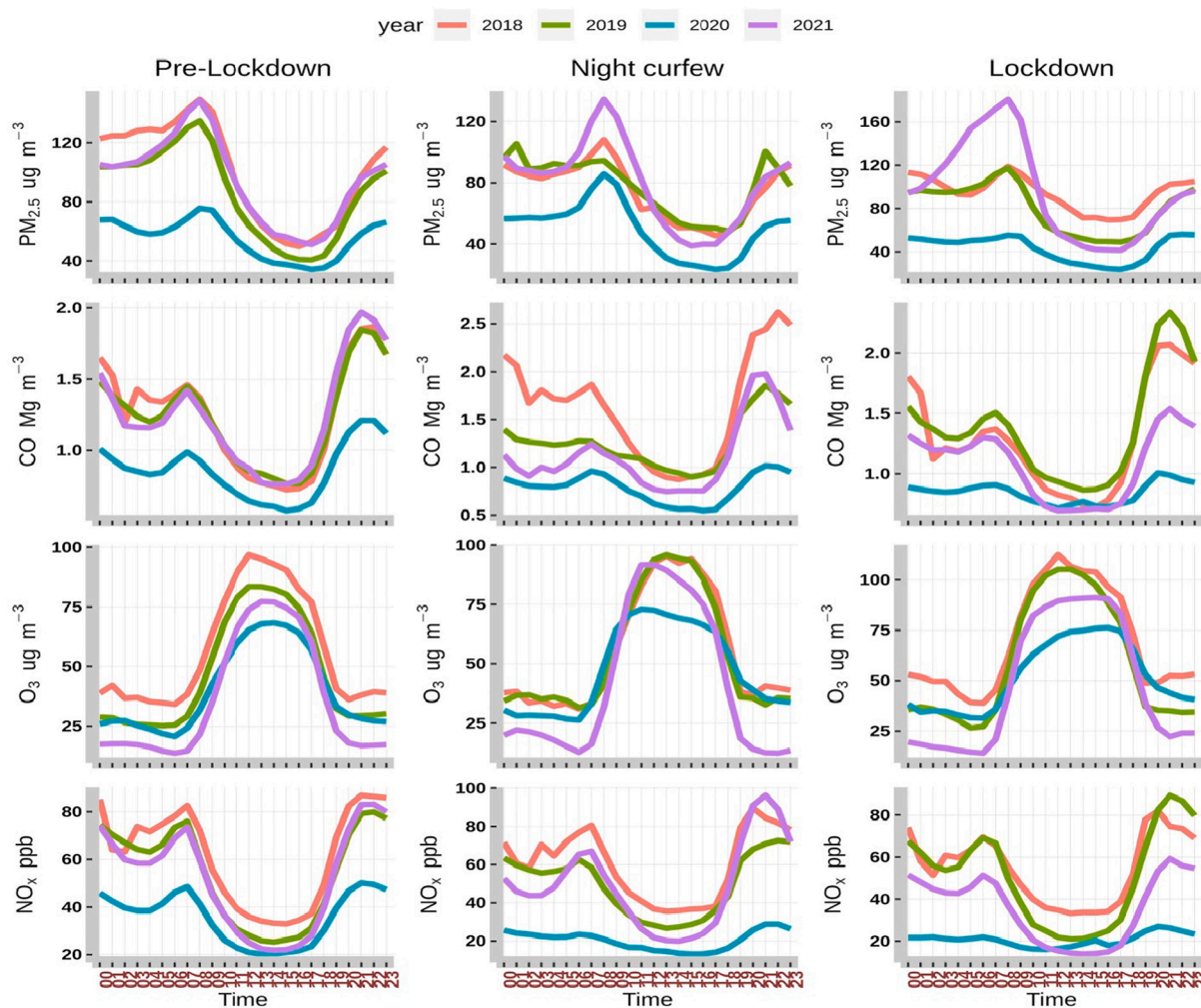
Parameter	2018				2019			
	March-01 to March-23		March-24 to April-30		March-01 to March-23		March-24 to April-30	
	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
PM ₁₀	0.89	0.22	0.62	0.53	0.9	0.26	0.76	0.41
PM _{2.5}	0.88	0.3	0.79	0.49	0.88	0.34	0.83	0.43
CO	0.87	−0.34	0.84	−0.23	0.89	−0.23	0.89	−0.27
Ozone	−0.38	0.37	−0.52	0.43	−0.61	0.35	−0.47	0.51
NO _x	0.86	−0.25	0.88	−0.21	0.89	−0.32	0.91	−0.3
NH ₃	0.75	0.45	0.57	0.32	0.78	0.33	0.57	0.62
SO ₂	0.57	0.68	0.69	0.4	0.53	0.72	0.67	0.26
Benzene	0.71	−0.31	0.7	−0.38	0.95	−0.05	0.94	0.05
Toluene	0.7	−0.43	0.61	−0.5	0.78	−0.45	0.81	−0.34
Xylene	0.29	0.03	NA	NA	0.24	−0.33	0.56	−0.24
Eigen Value	5.19	1.4	4.43	1.46	6.04	1.4	5.74	1.4
% variance	51.9	14	49.25	16.16	60.38	13.99	57.43	14.02
Cumulative %	51.9	65.9	49.25	65.4	60.38	74.38	57.43	71.45

Data not available: (NA).

Table 3b

PCA results of each period during 2020–21.

Parameter	2020				2021					
	March-01 to March-23		March-24 to April-30		March-01 to April-06		April-07 to April-16		April-17 to April-30	
	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
PM ₁₀	0.87	0.33	0.84	0.31	0.67	0.39	0.69	0.45	0.84	0.09
PM _{2.5}	0.86	0.34	0.91	0.2	0.78	0.55	0.73	0.64	0.84	0.21
CO	0.89	−0.26	0.9	−0.04	0.9	−0.23	0.84	−0.38	0.91	−0.16
Ozone	−0.42	0.73	−0.32	0.81	−0.63	0.12	−0.73	0.21	−0.73	−0.07
NO _x	0.88	−0.35	0.75	−0.39	0.9	−0.27	0.89	−0.31	0.9	−0.11
NH ₃	0.73	0.21	0.64	0.15	0.74	0.16	0.59	0.51	0.31	0.79
SO ₂	0.47	0.77	0.75	0.47	0.4	0.72	0.7	0.55	0.73	0.25
Benzene	0.96	0.07	0.93	−0.02	0.93	0	0.92	−0.08	0.87	−0.01
Toluene	0.85	−0.18	0.84	−0.35	0.78	−0.37	0.73	−0.39	0.49	−0.75
Xylene	0.14	−0.11	−0.23	−0.05	0.75	−0.37	0.83	−0.42	0.85	−0.12
EigenValue	5.67	1.63	5.64	1.32	5.8	1.4	5.93	1.81	5.94	1.35
% variance	56.76	16.34	56.38	13.22	58.03	14.18	59.29	18.06	59.44	13.5
Cumulative %	56.76	73.12	56.38	69.6	58.03	72.21	59.29	77.36	59.44	72.94

**Fig. 5.** Diurnal variation in pollutants and meteorological parameters during Pre-lockdown (March 1 to April 06), Night curfew (April 7 to April 16), and Lockdown (April 17 to April 30) in 2021 and comparison with identical periods of the previous three years.

increased, as compared to Period B1, despite a slight reduction in temperature, which could be due to enhancement in precursors concentration, and en route ozone production in the fire plumes traveling from Punjab/Haryana to Delhi (Parrish et al., 1998). The concentration of pollutants increased by 31 to 129%, as compared to the identical period of 2020 except O_3 (Table S2) likely because of the relaxation in 2021 for local sources like restaurants, street vendors, construction, taxis, public transport (buses, train, and airplane), and the transboundary sources as there was no strict restriction on human activities in neighboring states (Haryana, Uttar Pradesh, Rajasthan, Punjab) and a sharp rise in fire counts over Haryana and Punjab.

3.3. Diurnal variation

This section presents a diurnal variation in three phases, Period A, B1, and B2 during 2021, and compares it with identical periods of previous years and among the three phases to understand varying effects of restrictions (Fig. 5 and Fig. S4).

During Period A, most of the pollutants followed the same pattern and peak hours as previous years. The pollutants PM_{10} , $PM_{2.5}$, SO_2 , and NH_3 showed a peak during morning hours (7 to 10 AM) and NO_x , CO, benzene, and toluene peaked during evening hours (8 to 11 PM), but O_3 concentration showed a peak during the afternoon from 11 am to 5 PM, which is typical of urban areas. The concentration of PM_{10} and $PM_{2.5}$ were high during the daytime in 2021 compared to the other years, while WS was slightly less than in 2018 and 2019 during the day. The pollutants followed the same pattern across all the years during Period A indicating that there is not a large change in pollution sources. A decrease in the concentration of the $PM_{2.5}$, CO, NO_x , and benzene was observed during the night in Period B1, as compared to Period A and previous years (Fig. 5 and Fig. S4). During peak hours, the concentration of NO_x and SO_2 increased, as compared to pre-lockdown. NO_x peaked from 8 PM to 10 PM while SO_2 peaked from 7 AM to 9 AM. These hours mark the closing and opening of the offices, industries, market shops, and restaurants, etc. Other activities like traffic, hotels, restaurants, and pubs also increased during the evening before 10 PM. In the morning, the calm winds, low temperatures, and shallower PBLH restrict the dispersion of pollutants and may lead to higher concentrations of SO_2 and NO_x during peak hours. During Period B2, most pollutant concentrations during peak hours as well as throughout the day decreased except $PM_{2.5}$ and SO_2 when compared to identical periods of previous years and night curfew. This could be due to a reduction in emission sources, but an increase in $PM_{2.5}$ and SO_2 could also be due to a rise in fire activity over Haryana and Punjab (Fig. 6).

3.4. Effect of crop residue burning

In Haryana and Punjab, almost the same temporal pattern in the fire activity has been seen during 2018–2021. The number of fire counts sharply rose during the last week of April (Fig. 6). To understand the impact of crop residue burning in Haryana and Punjab on air pollution in Delhi, we calculate the mean value of pollutants and meteorological parameters, and the sum of fire counts during last

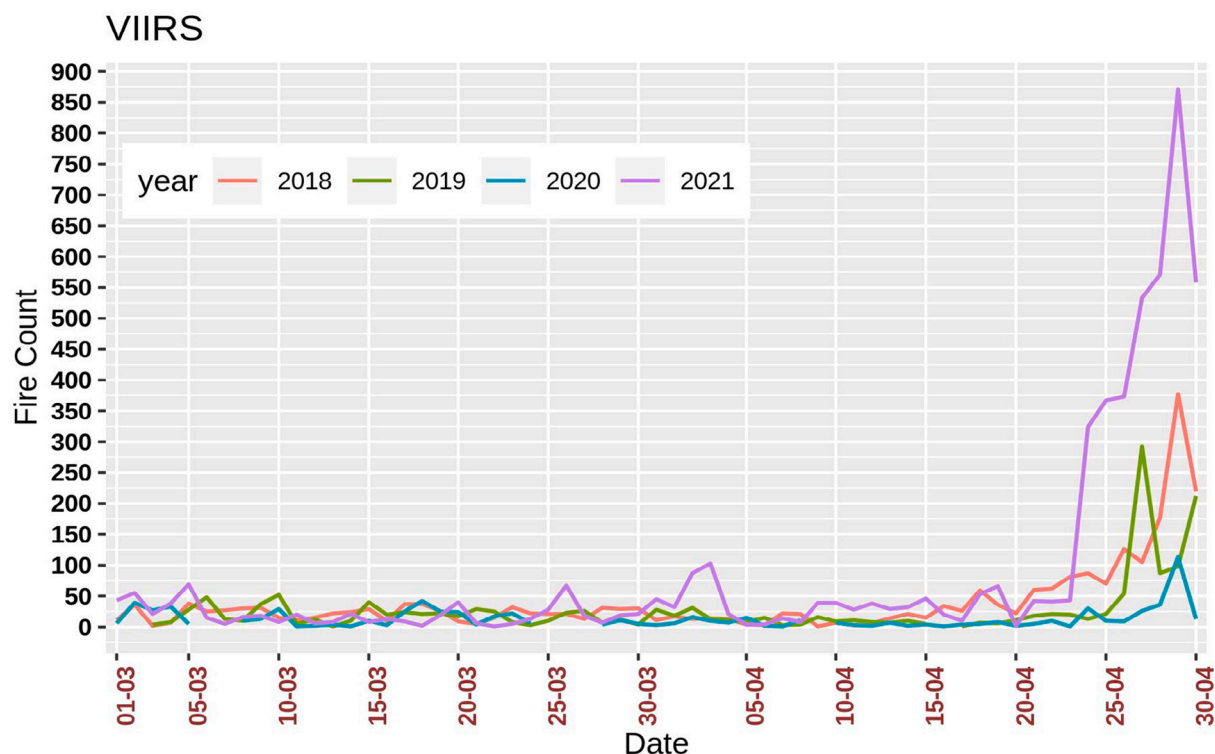


Fig. 6. Daily fire count over Haryana and Punjab from March 01 to April 30 for 2018 to 2021.

week (Period Q) of April, and compared it with last to the first week (Period P) (Table S3). The total fire counts in Period P were 346, 84, 35, and 257 during 2018, 2019, 2020, and 2021, respectively, and the corresponding fire counts in Period Q were 1162, 777, 237, and 3597. The percentage change in fire counts from Period P to Q was 235, 825, 577, and 1300% during 2018, 2019, 2020, and 2021, respectively. The percentage change during 2019 and 2020 was higher than in 2018, but the number of fire counts was much smaller than in 2018. The fire counts increased much more swiftly in Period Q, as compared to Period P in 2021. Therefore, a smaller percentage change in concentration of pollutants was observed during 2019 and 2020 compared to 2018 and 2021. In 2019, the concentration of NO_x, CO, and benzene decreased compared to Period P. This could be due to an increase in AT, WS, PBLH, and VC of 21, 26, 26, and 62%, respectively during Period Q. However, an increased concentration of PM₁₀ could be due to resuspension and transboundary movement of dust by high WS.

In 2020, the nationwide lockdown and lowest fire count over Haryana and Punjab led to the lowest level of pollution in both periods compared to the rest of the years. In 2021, the concentration of PM₁₀, PM_{2.5}, CO, O₃, NO_x, SO₂, benzene, and toluene increased by 82, 124, 43, 8, 79, 44, 22, and 65%, respectively, as compared to Period P, despite both periods facing similar lockdown restrictions in Delhi, which means almost no changes in local emission sources. At that same time, there was no lockdown restriction in neighboring states of Haryana, Punjab, Rajasthan, and Uttar Pradesh. Thus, the probability of fluctuation in regular anthropogenic emission sources in these states during this period was also very low, except for a sharp increase in fire counts of about 1299% (257 to 3597) in Haryana and Punjab, as compared to Period P. During Period Q, the abrupt increase in pollutants concentration is possible due to unfavorable meteorology, and huge crop residue burning in Haryana and Punjab. The meteorological parameters AT, RH, and PBLH were in favor of dispersion, but a 9% decrease in WS caused a 4.5% reduction in VC, which means the atmosphere was less dispersive during Period-Q. It means meteorology played a minor role in this increase. Therefore, the major contribution is most likely from an increase in crop residue burning in Haryana and Punjab.

3.5. Identification of sources

There are many source apportionment techniques used for the identification and quantification of the sources at receptor location. The detailed explanation about each method used for HYSPLIT analysis are available in Hopke (2016). Each technique has its limitations and benefits. The best possible techniques on available data are Principal Component Analysis (Tables 3a and 3b). The backward trajectory analysis (Cluster and concentrated weighted trajectory analysis) are used to track the movement of air parcels (Figs. S5a and S5b) which helped us to identify the potential source regions during each Period and year.

3.5.1. HYSPLIT trajectory analysis

During Period A, 40–80% of air parcels reaching Delhi originated in Northwestern countries of India including Pakistan, Turkmenistan, and Turkey, and the rest of the parcels originated in the Indian states of Haryana, Punjab, Himachal Pradesh, Uttarakhand, and Rajasthan, during 2018 to 2021. In 2020, the majority of trajectories that reached Delhi started from Uttar Pradesh, Madhya Pradesh, and Nepal, and 40% from the countries located northwest of India. The concentration weighted trajectory (CWT) showed that a strong to a moderate source region of PM_{2.5} was located over Punjab, Haryana, Uttar Pradesh, Rajasthan, and Pakistan, as these regions showed the highest CWT value (≥ 100) during Period A.

In Period B, the dominance of parcels reaching Delhi was affected by long-range transport from Iran, Turkey, Saudi Arabia, Turkmenistan, and Pakistan, and short-range transport from the Himalayan region (Himachal, Uttarakhand, and Jammu & Kashmir) as shown in the cluster plot (Fig. S5a). The major source contribution region was located in Haryana, Uttar Pradesh, Punjab, Himachal Pradesh, Rajasthan, Jammu & Kashmir, Northern part of Pakistan, Afghanistan, and south-west part of China during 2018. In 2019, the strong to moderate source region shifted north to the northwestward, as compared to 2018, and the least contribution was seen from the China region (Fig. S5b). During Period B, the potential source regions were located in Iran, Afghanistan, Pakistan, and Indo Gangetic plain states (Haryana, Punjab, Uttar Pradesh, Himachal Pradesh, Rajasthan). The CWT also shows the Arabian Sea also as a potential source during 2020 Period B which is likely related to the transport of fine mode sea-salt aerosols during the rainfall events observed during this period.

In periods B1 and B2, the strong to moderate source regions were located in Haryana, Punjab, Rajasthan, Uttar Pradesh, Himachal Pradesh, Pakistan, and Afghanistan (Fig. S5b). Most of the long-range transport during March–April are from North-Western countries of India, while potential source regions are located in Punjab, Haryana, Uttar Pradesh, Himachal, Jammu & Kashmir, Pakistan, and Afghanistan. Note that the CWT analysis only identifies potential source regions affecting Delhi and chemical transport modeling studies should be conducted in the future to quantify the contribution of each non-local source to air pollution in Delhi particularly during the lockdown periods.

3.5.2. Principal component analysis (PCA)

This is a dimension reduction technique, which transforms the original dataset into multiple factors, and each factor corresponds to a variable of the dataset with different factor loading. The factors having an eigenvalue of more than one are considered as sources (Table 2a and b).

During Period A, the two factors explained about 66, 74, 73, and 72% of the cumulative variance in 2018, 2019, 2020, and 2021, respectively. In 2018, factor-1 explained 51.9% variation with the highest factor loading from PM₁₀, PM_{2.5}, CO, NO_x, NH₃, benzene, and toluene, while factor-2 explained 13.4% variation with top loading from PM_{2.5}, NH₃, SO₂, O₃ and toluene. In urban areas, major factor loading from PM₁₀, PM_{2.5}, CO, NO, NO₂, NO_x, and benzene, with the highest contribution from PM_{2.5}, CO, NO_x, and benzene suggest the burning of fossil fuel, while about the equal contribution from PM_{2.5} and CO represents the addition of solid biofuel burning

for household energy need, and higher contribution from PM_{10} than $PM_{2.5}$ indicates the additional influence of dust sources (Ravindra et al., 2019; Singh et al., 2020), while factor-2 could be secondary aerosols due to the highest contributions from $PM_{2.5}$, NH_3 , and SO_2 (Jain et al., 2021). During 2019–2021, both of these factors explained more than 74% variance, and almost common factor loading as 2018.

During period B, the two-factor solution has been produced. Top factor loading of PM_{10} , $PM_{2.5}$, CO, NO_x , SO_2 , benzene, and toluene in factor-1 represent the burning of fossil fuel from vehicles, industries, etc., and solid biofuel burning for cooking and crop residue burning in an open field (Ravindra et al., 2019), but an equal contribution of CO and $PM_{2.5}$ reflects a larger contribution from the burning of solid biofuel in residential and waste dumping sites rather than vehicles or industries. The household consumption of solid biofuel during 2019 was 0.415Mt/yr and produced 12Gg/y of NMVOC in Delhi (Mondal et al., 2021). The NMVOC (C1–10) are major precursors for photochemical reaction to produced secondary aerosols. During the first and second phase of lockdown in 2020, the government of India prohibited the free movement of residents and only permitted very essential goods (medicine, foods, ambulance, securities vehicle, etc.) supply (Jain and Sharma, 2020). Due to strict norms, the supply of goods like LPG cylinders in rural areas was disturbed and trudge marches by laborers due to the lack of availability of public transport could be possible reasons for increasing household consumption of solid biofuel (Ravindra et al., 2021). In factor 2, the highest contribution from O_3 , NO_x , SO_2 , and toluene presents the photochemical reaction and formation of secondary aerosol (Sbai et al., 2021). The identical period in 2018 and 2019 shows the same factor loading as Period B, but NO_x contribution was high in factor 1 during 2018–19. During Period B1 and B2, the loading of variables was common as found during Period B in previous years in factor-1, which means the sources are fossil fuel, residential sold biofuel, and crop residue burning, but the equal contribution of PM_{10} and $PM_{2.5}$ represents the additional sources as dust (from dust storms and resuspension from the road). Factor 2 with loading from PM_{10} , $PM_{2.5}$, NH_3 , and SO_2 during Period B1 represents the secondary aerosol formation (Jain et al., 2021) and during Period B2 loading from NH_3 and toluene could be from

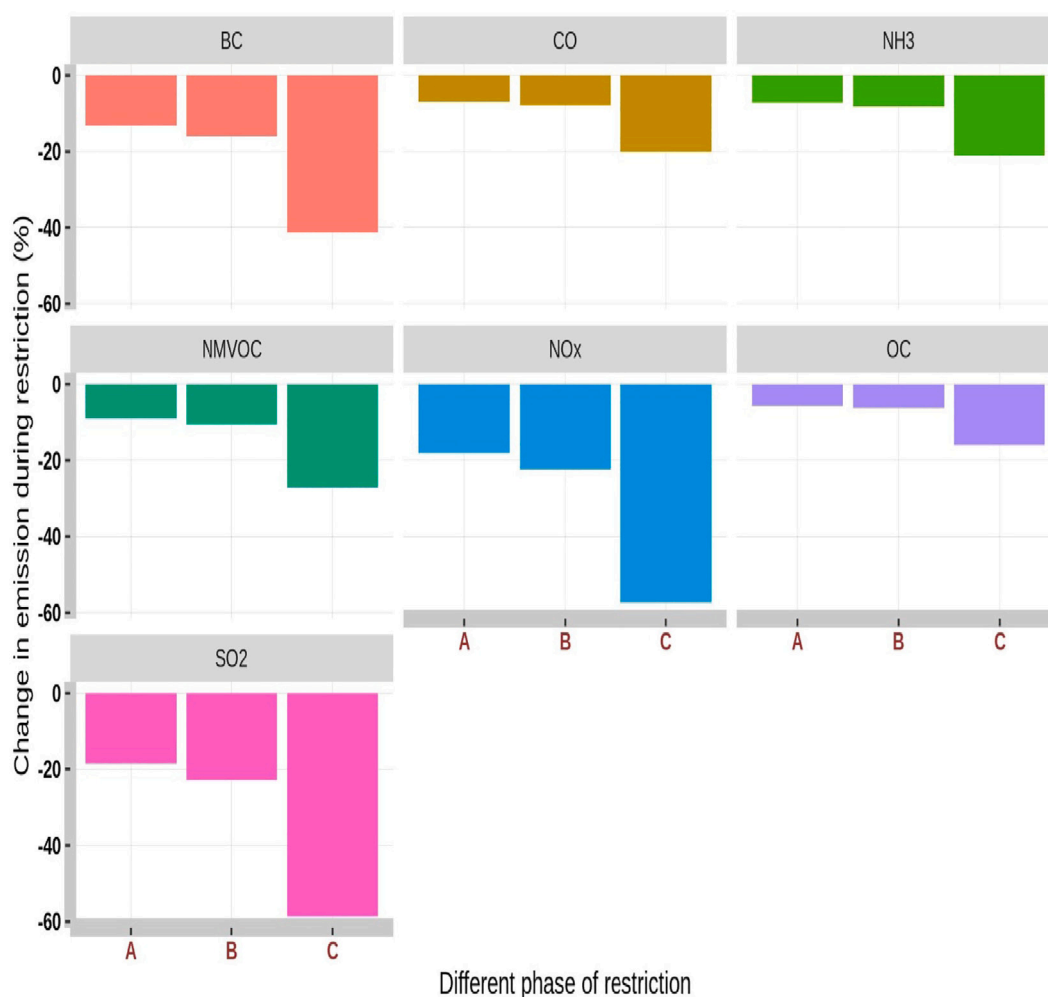


Fig. 7. The percentage change in total emission from the sum of three sectors (road transport, residential and industries) during each phase in April 2021 as compared to the identical period of 2019. Here A, B and C represent the pre-lockdown (April 1–6), night-curfew (April 7–16) and lockdown (April 17–30) period.

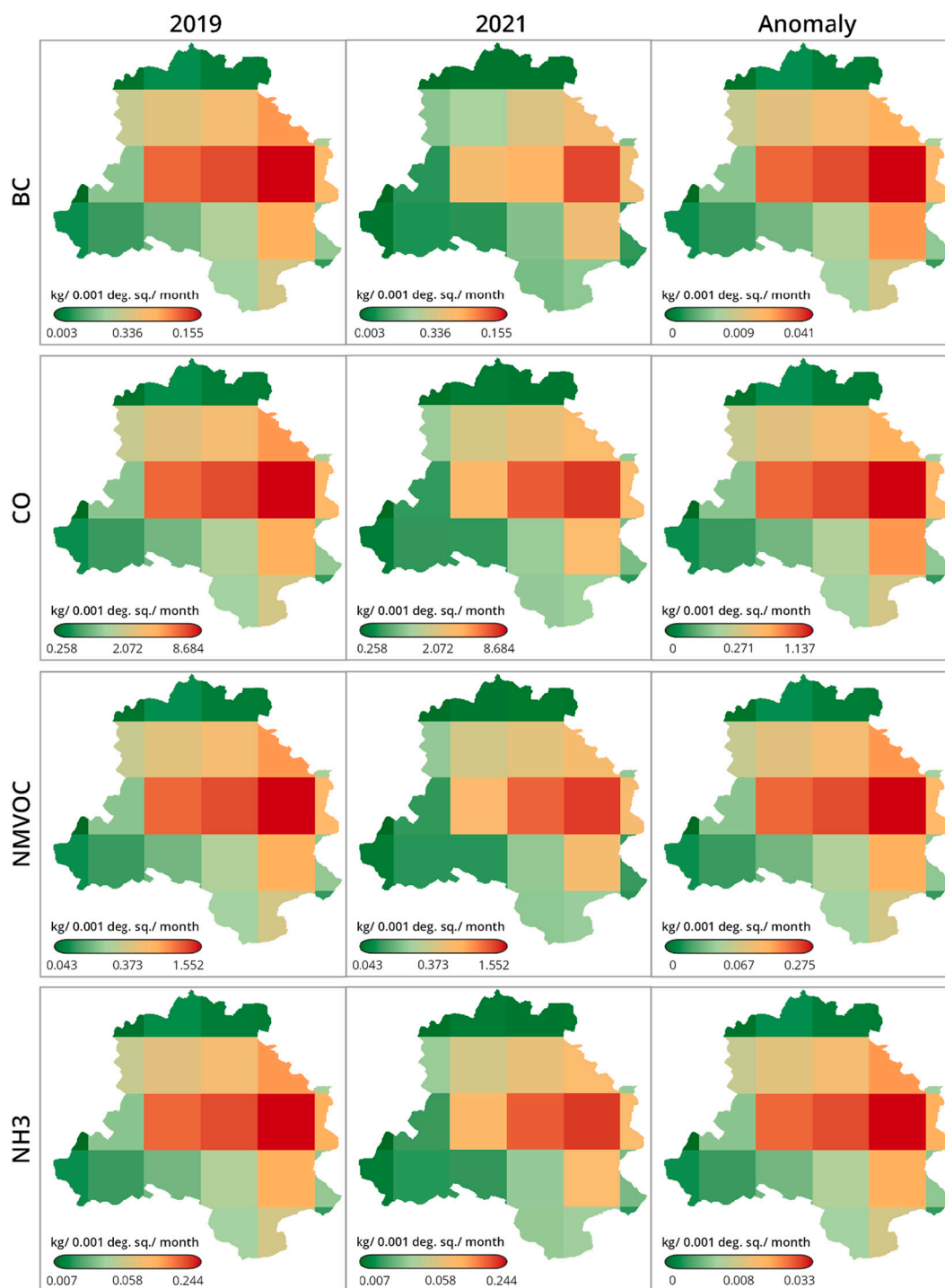


Fig. 8. a. Total gridded emission from road transport, industries and residential sector during April 2019 and 2021, and their anomaly over Delhi. b. Total gridded emission from road transport, industries and residential sector during April 2019 and 2021, and their anomaly over Delhi.

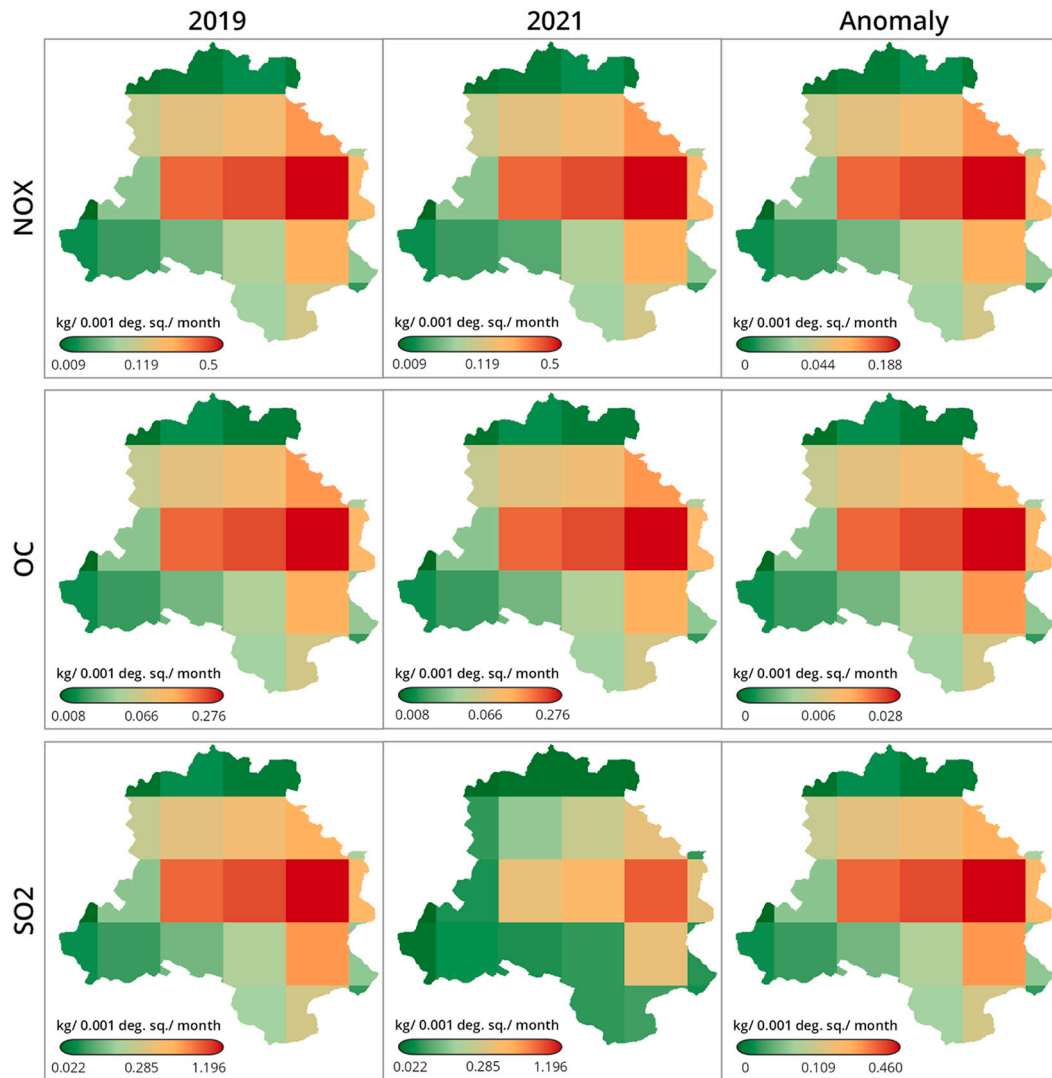


Fig. 8. (continued).

industrial emission sources because no restriction in neighboring states till April 30, 2021. Here we find that the major possible sources during March are burning of fossil fuel (from vehicles, industries, etc.), solid biofuel (household and waste site), road dust, and secondary aerosol formation, while in April the dominant pollution from the burning of fossil fuel, solid biofuel in household and open field (crop residue), and secondary aerosol.

3.6. Quantification of changes in pollutants emission

The AF related to road transport and industries slightly decreased during night curfew compared to pre-lockdown but during lockdown, the reduction was high and consistent except on April 19, 2021 (Fig. 2). This could be due to the Delhi government allowing the movement of industrial workers from Delhi to their native states and arranging special transport facilities for movement of workers that belong to other states with the help of the Government of India and other state governments. The AF for residential sectors increased slightly during night curfew but was consistently high during lockdown as compared to pre-lockdown. The Percentage changes in emission during each phase of restriction and pre-lockdown periods in April 2021 are represented in Fig. 7 and Fig. S6. The total emission of pollutants showed a small decrease during night-curfew and pre-lockdown. But in lockdown, the estimated emissions of pollutants decreased 18–60% as compared to the identical period of 2019 (Fig. 7). We see the highest decrease in NO_x and SO₂ and the lowest decrease in CO and OC during lockdown as compared to the identical period of 2019. The emissions from the residential sector increased by about 25%, while those from the road transport and industrial sector decreased by 50–60% in Delhi during lockdown 2021 as compared to 2019 (Fig. S6). Fig. 8a and b depict that central to eastern Delhi grids experience highest emissions and

consequently estimated emission reduction during April 2021 from the sum of three sectors (road transport, residential and industrial). The largest magnitude change observed in CO while the least changes were seen in OC.

3.7. Short term mortality

This study estimates the health impact as all cause mortality from short-term exposure of PM_{2.5} during March and April in Delhi. Table S4 contained detailed data about short-term health impact during each phase of study. Phase wise mean mortality, in period A, the mean premature mortality was 35 (31–39), 30 (27–33), 24 (21–26), and 33 (29–37) during 2018, 2019, 2020, and 2021, respectively (Table S4). In Period B, the daily mortality from PM_{2.5} was 16 (14–18) in 2020, while in Period B2, during the first and 2nd week of lockdown were 21 (19–24) and 45 (40–50), respectively. The change in short-term mortality is directly proportional to ambient PM_{2.5} concentration (Eq. (2)). The lowest mortality count was calculated during Period B because all anthropogenic sources (like transport sector, Industries, Aviation, hostels, Infrastructure, etc) were closed to control the spread of novel coronavirus in the country. Therefore, we calculate excessive PM_{2.5} during study period by subtracting the mean concentration of PM_{2.5} during Period B from PM_{2.5} daily mean concentration during March–April period.

$$\text{Excessive PM}_{2.5}\text{concentration} = A - B$$

Where

A = PM_{2.5} daily concentration during March–April.

B = Mean concentration of PM_{2.5} during Period B (March 24 to April 30, 2020).

This excessive PM_{2.5} concentration causes additional mortality 13(11–14), 5(5–06) and 29 (26–32) per day during Period B1, the first and second week of Period B2, respectively (Table S4). This could be due to more relaxation in local sources, and no restriction in neighboring states and an additional source of crop-residue burning over Haryana and Punjab in the 2nd week of Period B2. This excessive death count was only calculated for Delhi (population about 201.93 lakh) (Fig. S6).

4. Conclusion

The key findings of this study are listed below.

- During lockdown in 2020, the meteorology did not favor the dispersion of pollutants as much as it did during the previous years (Table 2). However, we still observed the lowest concentrations during this period highlighting the importance of reducing anthropogenic emission sources. We suggest that air pollution levels during this period could be considered as an achievable concentration for Delhi but it will likely take many years to achieve these levels as emission control policies also need to take into account the economic growth and cannot simply suspend all non-essential activities as was done during the 2020 lockdown.
- During Period B1, the mean concentration of PM₁₀, PM_{2.5}, CO, NO_x, NH₃, benzene and toluene decreased by 4 to 16% from Period A. The changes varied as a function of the time of the day depending on whether the COVID-19 related restrictions were imposed during the nighttime or throughout the day, and what activities were limited.
- During Period B2, the concentration of pollutants decreased in the first week of lockdown, but the second week saw an increase in pollutants (Table S3), which means that the effect of controlling local sources was overwhelmed by a rapid increase in crop residue burning activity over Haryana and Punjab and decrease in WS and VC possibly contributed to the accumulation of pollutants in Delhi.
- The sources apportionment and trajectory analysis result show that the possible sources during March and April are burning of fossil fuel, solid biofuel in residential and agriculture field, dust, and secondary aerosol formation, and most probably non-local source regions are located in Haryana, Punjab, Rajasthan, Himachal Pradesh, Pakistan, and Afghanistan.
- The short-term health impact analysis, as all causes of mortality from exposure of ambient PM_{2.5}, finds 5 (5–06) and 29 (26–32) addition mortalities per day during the first and 2nd week of lockdown in 2021 compared to the 2020 lockdown in Delhi. These results are based on a short term reduction in air pollution and should be considered as merely indicative of potential health benefits of improving air quality.

This study provides evidence to policymakers and the scientific community that air pollution control in Delhi should target the burning of fossil fuels, solid biofuel in households, solid waste dumping sites, and agriculture fields not only at the local level in Delhi but also in neighboring states. The possible solutions might be as easy availability of affordable LPG cylinders in rural areas and increase awareness among the public of the health benefits of shifting to clean energy from solid biofuels, shifting of public transport to electric buses and trains in urban areas, by using electricity produced by renewable energy sources, and tax relaxation and subsidies on electric vehicles. Farmers should be provided with affordable alternatives for cleaning their fields and avoid crop-residue burning.

While this study focuses on Delhi, the results presented here should be viewed as representative of the COVID-19 lockdown induced changes in air quality in a developing megacity. In this perspective, this study contributes to rapidly evolving global knowledge of the impact of COVID-19 restrictions on air quality in different environments of the world.

Declaration of Competing Interest

The authors declare that there are no known competing financial and personal interests to influence the results and work reported in the manuscript.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.uclim.2021.101059>.

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