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Impact of COVID-19 lockdown on air quality in Chandigarh, India: Understanding the emission sources during controlled anthropogenic activities

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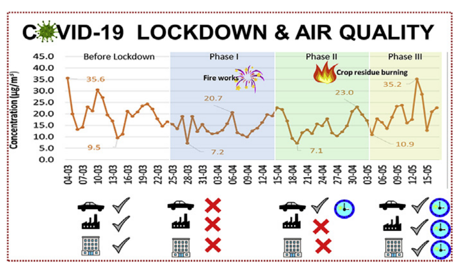
HIGHLIGHTS

- Studied impact of COVID-19 lockdown phases on 14 air quality parameters.
- Reduction in pollutants found to be associated with meteorology and emissions.
- Increase in O_3 concentration was linked with atmospheric reactivity.
- Key emission sources identified were vehicles, stubble burning, and coal power plants.
- VOCs variations shows association with regional emissions e.g., solid biomass burning.

GRAPHICAL ABSTRACT



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ABSTRACT

The variation in ambient air quality during COVID-19 lockdown was studied in Chandigarh, located in the Indo-Gangetic plain of India. Total 14 air pollutants, including particulate matter (PM_{10}, $PM_{2.5}$), trace gases (NO_2, NO, NO_x, SO_2, O_3, NH_3, CO) and VOC's (benzene, toluene, o-xylene, m,p-xylene, ethylbenzene) were examined along with meteorological parameters. The study duration was divided into four parts, i.e., a) 21 days of before lockdown b) 21 days of the first phase of lockdown c) 19 days of the second phase of lockdown d) 14 days of the third phase of lockdown. The results showed significant reductions during the first and second phases for all pollutants. However, concentrations increased during the third phase. The concentrations of SO_2, O_3, and m,p-xylene kept on increasing throughout the study period, except for benzene, which continuously decreased. The percentage decrease in the concentrations during consecutive periods of lockdown were 28.8%, 23.4% and 1.1% for $PM_{2.5}$ and 36.8%, 22.8% and 2.4% for PM_{10} respectively. The Principal Component Analysis (PCA) and characteristic ratios identified vehicular pollution as a primary source during different phases of lockdown. During the lockdown, residential sources showed a significant adverse impact on the air quality of the city. Regional atmospheric transfer of pollutants from coal-burning and stubble burning were identified as secondary sources of air

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pollution. The findings of the study offer the potential to plan air pollution reduction strategies in the extreme pollution episodes such as during crop residue burning period over Indo-Gangetic plain.

1. Introduction

The pandemic of COVID-19 has enforced most of the countries of the world to implement preventive measures to stop its spread. The lockdown was one of the major preventive measures adopted by all the nations. In India, the lockdown was enforced from 25th March to May 17, 2020 in three phases (Lockdown 1, 2, and 3). The lockdown imposed strict regulations on almost all the human activities like restrictions on vehicular movement, shutting down of industries, closure of all government and private commercial offices, closure of all the educational institutes, closure of construction projects and the onset of curfews in all the states and cities of the country.

All these restrictions were predicted to cause a significant decrease in the concentrations of almost all the air pollutants resulting from anthropogenic activities and have been reported in various studies around the globe (Dantas et al., 2020; Abdullah et al., 2020; Li et al., 2020; Bao and Zhang, 2020; Zambrano-Monserrate et al., 2020) and India (Biswal et al., 2020; Chauhan and Singh, 2020; Singh et al., 2020). In India, all three phases of lockdown had relatively few differences in the regulations imposed on human activities. These differences resulted in variation in human activities, having very few activities in the first phase of lockdown, and a consecutive increase in activities thereafter in each succeeding stage of lockdown. In the absence of detailed source apportionment studies for the Chandigarh city, the simultaneous temporal study of air quality and restricted human activities could help to understand the primary sources contributing to the ambient air pollution of the city.

Air pollution is a major health risk to human health and observed to be in the severe category in the cities of low and middle-income countries like India (Balakrishnan et al., 2019). Indian cities are among the most polluted cities in the world (Gurjar et al., 2016; Guttikunda et al., 2014). Also, Fattorini et al. (2020) hypothesized that regions having consistent poor air quality for long-terms could have higher chances of the spread of COVID-19 disease as compared to other areas. Chandigarh is one of the non-attainment cities of India and the primary sources contributing to its declining air quality have been found to be the local sources (Ravindra et al., 2020; Guttikunda et al., 2019; Bhargava et al., 2018) which include the emissions from automobiles and industrial sectors along with emissions originating from residential households, biomass and waste burning, construction activities and an influx of all kind of pollutants into the city due to atmospheric transport from upwind neighboring areas (Ravindra et al., 2020, 2019; Ravindra, 2019). Use of fossil fuel in vehicles and industries have been found to be the major sources of air pollution in urban areas. Hence, assessing the air quality of urban areas while most of these sources are not active, less active, or are active only for fixed timings every day will provide an understanding of all the prominent sources of air pollution.

In the present study, the impact of lockdown, i.e., March 25, 2020 to May 17, 2020 (in three phases) on the ambient air quality of city Chandigarh, has been investigated. The air pollution trends of 54 days lockdown periods were compared within the phases of lockdown and with the air quality trends of 21 days before the lockdown. Additionally, the peak concentrations during the lockdown were examined, including the possible sources using the Principal Component Analysis, pooled trajectory analysis, and characteristic ratios of pollutants. The study provides an opportunity to understand and identify air pollution sources during restricted anthropogenic activities and plan air pollution reduction strategies.

2. Methodology

2.1. Study area

The study area, Chandigarh, is situated in the northern part of India and is one of the eight union territories of the country. It is located at 30° 44′ 14 N and 76° 47′ 14 E and is having a humid subtropical climate. The total area of Chandigarh is around 114 km^2, with a total population of 1.05 million, out of which 97.25% is the urban population. The population density of the Chandigarh is 9258 people per sq km and has the largest per capita number of vehicles in the country (Census, 2011). It also shares its boundaries with states of Haryana and Punjab, which contribute to an enormous flux of vehicles into the city daily.

2.2. Sampling and instrumentation

The air pollutants investigated in the study were particulate matter ($PM_{2.5}$, PM_{10}), nitrogen dioxide (NO_2), nitric oxide (NO), oxides of nitrogen (NO_x), sulfur dioxide (SO_2), ozone (O_3), ammonia (NH_3), carbon dioxide (CO), benzene, toluene, x-xylene, m,p-xylene, ethylbenzene along with meteorological parameters which were air temperature (AT), relative humidity (RH), wind speed (WS)), wind direction (WD), solar insolation, and rainfall. The concentrations of parameters studied were obtained from the continuous ambient air quality monitoring station (CAAQMS), which has been set up by Chandigarh Pollution Control Committee (CPCC) at Panjab University, Chandigarh under National Clean Air Programme (NCAP) and having analyzers of Environnement S.A. India Pvt. Ltd. (a 100% Subsidiary of Environnment S.A. Group, France) $PM_{2.5}$ and PM_{10} were measured by models PM101 M and MP101 M, respectively, which both work on the principles of Beta Attenuation. Nitrogen oxides and NH_3 were analyzed based on Chemiluminescence methods with model AC32e. Carbon monoxide was analyzed using model CO12e, which works on the principle of Non-Dispersive Infrared. SO_2 was analyzed based on the principle of Ultra-Violet Fluorescence with model AF22e. While O_3 was analyzed using model O342e, which works on the UV Photometric principle. All the VOCs were measured with model VOC72 M, which works on the principle of Gas Chromatography. Among the meteorological parameters, temperature and humidity were measured with PT100 and Capacitive methods, respectively (Model DMA875). wind speed and wind direction were measured using Ultrasonic Anemometer (Model DNB105). Solar radiation was measured with Pyranometer (DPA855), and rainfall was measured using Tipping Bucket (DQA 230.1).

Table 1
Average concentrations of pollutants and meteorological parameters before and during lockdown phases.

Study Period	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	NO_2 (μg/m^3)	NO (μg/m^3)	NOx (ppb)	NH_3 (μg/m^3)	SO_2 (μg/m^3)	CO (mg/m^3)	O_3 (μg/m^3)	Benzene (μg/m^3)	Toluene (μg/m^3)	o-xylene (μg/m^3)	m,p-xylene (μg/m^3)	ethylbenzene (μg/m^3)
Average pollutants concentration before lockdown and during all phases of lockdown periods														
Before Lockdown	20.1	56.9	13.9	7.2	13.0	68.0	9.9	0.55	13.8	4.4	1.4	1.0	0.6	3.2
Lockdown 1	14.3	35.9	10.7	1.9	7.0	38.3	10.0	0.46	19.2	3.2	0.4	1.1	0.8	3.1
Lockdown 2	15.4	43.9	11.6	2.4	8.0	32.1	11.4	0.52	26.5	3.0	0.7	1.1	0.9	2.9
Lockdown 3	19.8	55.5	13.0	2.3	8.6	32.5	11.8	0.57	31.7	2.2	1.0	0.9	2.0	2.4

Average values of meteorological parameters before and during all phases of lockdown period

Study Period	**AT (°C)**	**RH (%)**	**WS (m/s)**	**WD (Degrees)**	**SR (W/m^2)**	**RF (mm)**
Before Lockdown	18.0	75.3	1.0	188.6	212.0	0.024
Lockdown 1	22.5	59.7	1.1	244.3	262.6	0.001
Lockdown 2	25.8	52.1	0.8	165.5	256.4	0.018
Lockdown 3	27.4	50.1	1.0	190.3	272.0	0.000

3. Results & discussion

The comparison of the concentrations of various pollutants analyzed in this study showed that the levels of almost all of the pollutants decreased during the lockdown period. However, few pollutants (SO_2 and O_3) showed a continuous increase in their concentrations as the lockdown progressed (Table 1). These results are similar to the other studies across the globe during the lockdown events (Sharma et al., 2020; Mahato et al., 2020; Collivignarelli et al., 2020), but the current study provides a comprehensive view in term of sources, chemistry, and meteorology. The percentage decrease in the concentrations of all pollutants during all three phases of lockdown as compared to the 21-days period before lockdown is tabulated in Table 2. The average levels of all the pollutants and meteorological parameters for the study periodsi.e., before and during lockdown are shown in Table 1. The Fig. 1 depicts the daily average variations of pollutants during the whole study period, i.e., before lockdown, first phase (PH I), second phase (PH II), and third phase (PH III) of lockdown. The comparison with the previous year's pollution levels could not be performed due to the unavailability of real-time air quality data.

3.1. Variation in particulate matter ($PM_{2.5}$, PM_{10}) concentrations

The average $PM_{2.5}$ concentrations for the period of 21 days before the lockdown was 20.1 μg/m^{3,} which were reduced to 14.3 μg/m^3 during the first phase of the lockdown period. The PM_{10} concentration decreased from 56.9 μg/m^3 to 35.9 μg/m^3 during the first phase of lockdown. A decrease of 28.8% and 36.8% were seen for $PM_{2.5}$ and PM_{10} during this phase as compared to the pre-lockdown period. A major fraction of these reductions in PM concentrations can be attributed to the massive decrease in the vehicular traffic, halting of industries and stopping of all construction activities (chdcovid19, 2020) which are the significant sources of the particulate matter in the urban ambient air.

During the second phase, the average $PM_{2.5}$ and PM_{10} concentrations were recorded as 15.4 μg/m^3 and 43.9 μg/m^{3,} respectively. The levels increased by 7.7% and 22.3% for $PM_{2.5}$ and $PM_{10,}$ respectively, as compared to the first phase of lockdown. There was not much difference in the regulations for human activities between the first and second phase of lockdown. But the regulation for movement of residents for essential services was implemented after the completion of few days of total lockdown during the first phase, while in the second phase, this movement occurred daily throughout the stage of lockdown (chdcovid19, 2020). This might be the primary reason for a little increase in emissions, which can be attributed to automobiles and, consequently, which lead to higher concentrations in the ambient air.

The third phase of lockdown was implemented with few relaxations like reopening of government offices, private offices, and few essential industrial manufacturing units (chdcovid19, 2020). The average concentrations of $PM_{2.5}$ and PM_{10} were 19.8 μg/m^3 and 55.5 μg/m^3 respectively, during this phase of the lockdown period. A very slight decrease of 1.1% and 2.4% for $PM_{2.5}$ and PM_{10} was seen as compared to the before lockdown period. This can be attributed to the emissions from increased vehicular traffic and emissions from industrial units (Singh et al., 2020; Lawrence et al., 2013; Sawyer, 2010).

3.2. Variation in trace gases concentrations

3.2.1. Oxides of nitrogen and ammonia

The 21 days average concentrations of NO_2, NO, $NO_{x,}$ and NH_3 before lockdown were 13.9 μg/m^3, 7.2 μg/m^3, 13.0 ppbv, and 68.0 μg/m^3 respectively, which were well under the national ambient air quality standards (NAAQS). During the first phase of lockdown period concentrations of these gases, NO_2, NO, NO_x, NH_3 reduced to 10.7 μg/m^3, 1.9 μg/m^3, 7.0 ppbv, and 38.3 μg/m^3 respectively. NO_2 reduced by 23%, whereas NO, NO_x and NH_3 showed a significant decrease of 74.1%, 46.4%, and 43.7%, respectively. During the second phase of the lockdown, the average concentrations of these gases were recorded as 11.6 μg/m^3, 2.4 μg/m^3, 8.0 ppbv, and 32.1 μg/m^3 for NO_2, NO, NO_x and NH_3 respectively. A decrease of 16.5% for NO_2, 66.5% for NO, 38.6% for $NO_{x,}$ and 52.8% for NH_3 were recorded as compared to the before lockdown period. The third phase of lockdown showed the average concentrations of 13 μg/m^3, 2.3 μg/m^3, 8.6 ppbv, and 32.5 μg/m^3 for NO_2, NO, NO_x and NH_3 respectively. The percentage decrease for these gases was found as 6.1% for NO_2, 67.4% for NO, 33.9% for $NO_{x,}$ and 52.3% for $NH_{3,}$ respectively, as compared to the average concentrations before lockdown period.

The various sources of oxides of nitrogen (NO_2, NO, NO_x) and NH_3 in the atmosphere are agricultural activities, biomass burning, and domestic emissions, along with automobiles and industries. Restrictions on automobile movements and shutdown of industries may be the possible reasons for these reductions in nitrogen oxides. Agricultural, domestic, and biomass burning emissions are considered the major sources of NH_3 emissions. The continuous decrease in the concentration of NH_3 can be due to the formation of secondary aerosols like ammonium sulfate and ammonium nitrate with an increase in the rate of photochemical reactions (Bhanarkar et al., 2005; Asman et al., 1998; Olivier et al., 1998; EPA, 2020).

Table 2
Percentage change in the concentrations of pollutants during all phases of lockdown compared to before the lockdown period.

Study Period	$PM_{2.5}$ (%)	PM_{10} (%)	NO_2 (%)	NO (%)	NOx (%)	NH_3 (%)	SO_2 (%)	CO (%)	O_3 (%)	Benzene (%)	Toluene (%)	o-xylene (%)	m,p-xylene (%)	ethylbenzene (%)
Lockdown 1	−28.8	−36.8	−23.0	−74.1	−46.4	−43.7	+1.4	−17.4	+38.7	−26.8	−69.8	−87.8	+37.8	−3.5
Lockdown 2	−23.4	−22.8	−16.5	−66.5	−38.6	−52.8	+15.9	−6.1	+91.4	−31.2	−51.1	−87.5	+50.9	−11.0
Lockdown 3	−1.1	−2.4	−6.1	−67.4	−33.9	−52.3	+19.1	+2.8	+128.9	−50.3	−24.6	−90.0	+244.2	-24.2

3.2.2. Sulfur dioxide, carbon monoxide, and ozone

The concentration of SO_2 kept on increasing from 9.9 μg/m^3 before lockdown period to 10.0 μg/m^3 during the first phase to 11.4 μg/m^3 during the second phase and to 11.8 μg/m^3 during the third phase of lockdown (Table 1). A continuous increase of 1.4%, 15.9%, and 19.1% was recorded during these consecutive phases of lockdown as compared to the average concentration of before lockdown period. Coal burning is the primary source of SO_2 emissions (EPA, 2020). Therefore, this increase in SO_2 level can be attributed to the atmospheric transportation of SO_2 emissions from coal-burning plants upwind of Chandigarh city, as coal-fired thermal power plants present in neighboring states were working during all the phases of lockdown period.

The average concentration of carbon monoxide before the lockdown was 0.55 mg/m^3 and decreased to 0.46 mg/m^3 during the first phase of lockdown and then increased to 0.52 mg/m^3 during the second phase and 0.57 mg/m^3 during the third phase of lockdown. A percentage decrease of 17.4% was recorded during the first phase, a 6.1% decrease during the second phase, and an increase of 2.8% was recorded during the third phase of lockdown in comparison to the before lockdown period.

O_3 showed a significant increase (38.7%) during the first phase, second phase (91.4%), and third phase (128.9%) of the lockdown period as compared to the 21 days period before the lockdown. The average concentrations of O_3 were found to be 13.8 μg/m^3, 19.2 μg/m^3, 26.5 μg/m^3 and 31.7 μg/m^3 during the study periods of before lockdown, the first phase, second phase and third phase of lockdown respectively. This can be due to the following two reasons: 1) Continuous increase in the intensity of solar radiation as the study period proceeded, which helped to the continuous rise of photochemical reactions and O_3 formation. 2) The decrease in the concentration of NO during the lockdown period as NO helps in the breakdown of O_3 to O_2 and comparatively less decrease in the concentrations of CO and VOCs, which helps in the formation of O_3 in the ambient air.

3.3. Variation in volatile organic compounds concentrations

The average concentrations of benzene, toluene, o-xylene, and ethylbenzene before lockdown were 4.4 μg/m^3, 1.4 μg/m^3, 8.8 μg/m^3 and 3.2 μg/m^3 respectively, which reduced to 3.2 μg/m^3, 0.4 μg/m^3, 1.1 μg/m^3 and 3.1 μg/m^3 respectively during the first phase of lockdown. All the concentrations for these VOCs were under NAAQS prescribed standards except for o-xylene before the lockdown.

Benzene and ethylbenzene went on decreasing during the next phases of lockdown having average concentrations of 3.0 μg/m^3 and 2.2 μg/m^3 for benzene and 2.9 μg/m^3, and 2.4 μg/m^3 for ethylbenzene, respectively. o-xylene had the same concentration in the second phase as it was in the first phase and further reduced to 0.9 μg/m^3 during the third phase of lockdown. Toluene increased from 0.4 μg/m^3 (during the first phase of lockdown) to 0.7 μg/m^3 during the second phase and to 1 μg/m^3 during the third phase of lockdown. This increase can be attributed to the increased vehicular movement and the reopening of industries during the third phase of lockdown in the city (Bruno et al., 2006).

A significant decrease of 26.8%, 69.8%, 87.8%, and 3.5% were seen for all VOCs during the first phase of the lockdown period (Table 2). The primary sources of VOCs in the urban environment are vehicular exhaust, evaporation from fuel tanks, emissions from industries, and coal burning (Chattopadhyay et al., 1997; Kerchich and Kerbachi, 2012; Bretón et al., 2017). This decline during lockdown can be attributed to the restrictions on automobiles movement and shutting down of industries, which are the major sources of VOCs in the urban ambient environment. The m,p-xylene showed a continuous increment in its concentrations throughout the study period. It increased from 0.6 μg/m^3 to 0.8 μg/m^3 during the first phase, to 0.9 μg/m^3 during the second phase and to 2 μg/m^3 during the third phase of lockdown. The slight increase during the early two phases can be attributed to the coal burning in power plants in the vicinity of the city and possible emissions from residential areas (EPA, 1994). The more significant increase during the third phase shows fresh local emissions in the city during the third phase by increased vehicular emissions.

3.4. Variations in the diurnal pattern of pollutants

3.4.1. Particulate matter

Diurnal analysis of PM concentrations before the lockdown period showed that $PM_{2.5}$ had its peak concentrations during the midnight as 24.5 μg/m^3 at 1:00 a.m., while the lowest concentration was found as 16.6 μg/m^3 at 4:00 p.m., The $PM_{2.5}$ concentrations remain high around midnight followed by the peaks during day time between 10 a.m. and 1 p.m.. During the first phase of lockdown, the peak was recorded as 20.7 μg/m^3 at 11:00 a.m., and lowest concentrations was recorded as 11.5 μg/m^3 at 7:00 & 8:00 p.m. During the second phase, the highest peaks for $PM_{2.5}$ were found at midnight time with a peak of 18.8 μg/m^3 at around 12 a.m. followed by a second peak during day time between 12 p.m. and 5 p.m., and the lowest concentrations was recorded as 12.2 μg/m^3 at 9 a.m. The highest peaks for all the parts of the study period were found during midnight except for the first phase of lockdown. The reason for the accumulation of $PM_{2.5}$ during night time seems to associated with poor dispersion and accumulation of pollutants due to lower mixing height, lower wind speed, and favorable relative humidity, as also reported by (Lou et al., 2017). The peak of $PM_{2.5}$ during the first phase of lockdown was observed around 11 p.m., whereas for subsequent lockdown phases the $PM_{2.5}$ peaks were observed during evening time. These peaks during day time can be attributed to the increase in automobile movement as relaxations were given to the residents of the city for movement between 10 a.m. and 3 p.m. (chdcovid19, 2020). During the third phase, again, the highest concentrations was 23.8 μg/m^3 at 12 a.m., followed by lower peaks at 12 p.m. and 1 p.m. (Fig. 2).

In the case of PM_{10}, the peak was 71.7 μg/m^3 at 11:00 p.m., and the lowest levels were recorded (43.2 μg/m^3) at 6:00 a.m. before the lockdown, and the second lowest peaks were observed between 10 a.m. and 2 p.m. During the first phase of lockdown, the highest and lowest concentrations were found as 44.4 μg/m^3 at 11:00 a.m. and 28.7 μg/m^3 6:00 a.m., respectively. During the second phase of lockdown, the maximum concentration was found to be 53.5 μg/m^3 at 12 p.m., and the minimum was 31.2 μg/m^3 at 7 a.m.

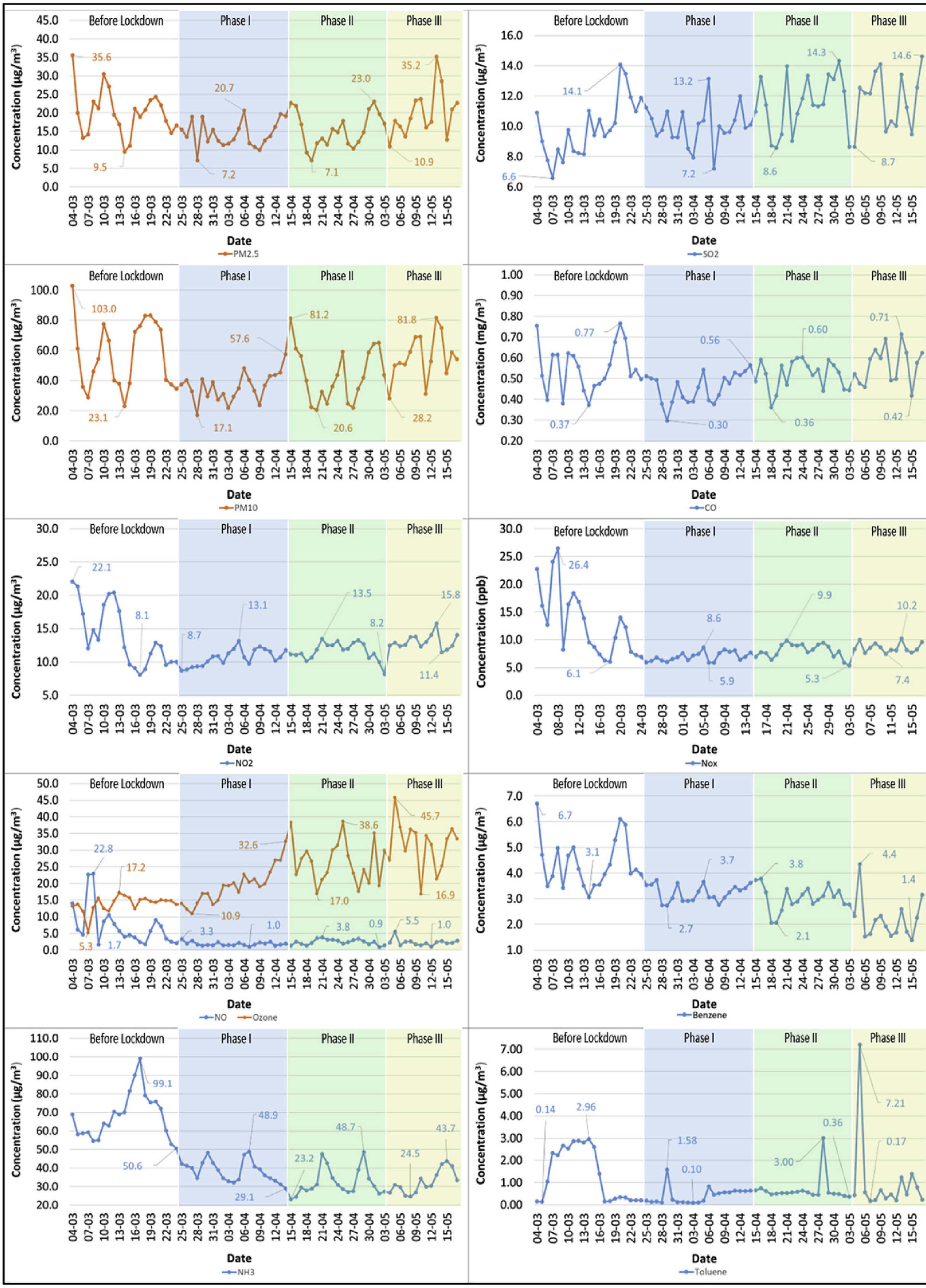

Fig. 1. Daily average variations of pollutants during the whole study period: before lockdown, first phase (PH I), second phase (PH II) and third phase (PH III) of lockdown.

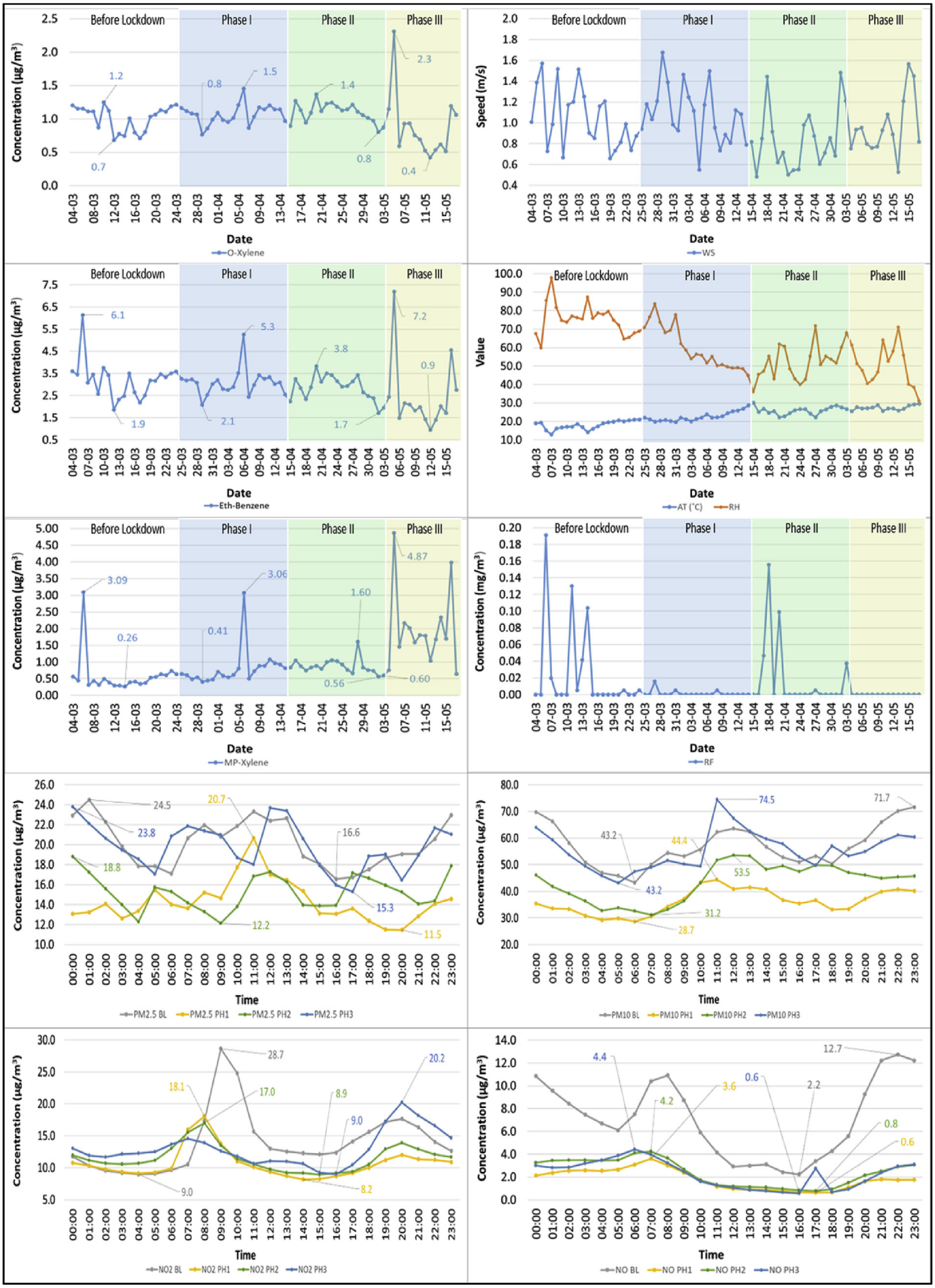

Fig. 2. Daily average and diurnal variations of pollutants and meteorological parameters for the whole study period: before lockdown (BL), first phase (PH I), second phase (PH II) and third phase (PH III) of lockdown.

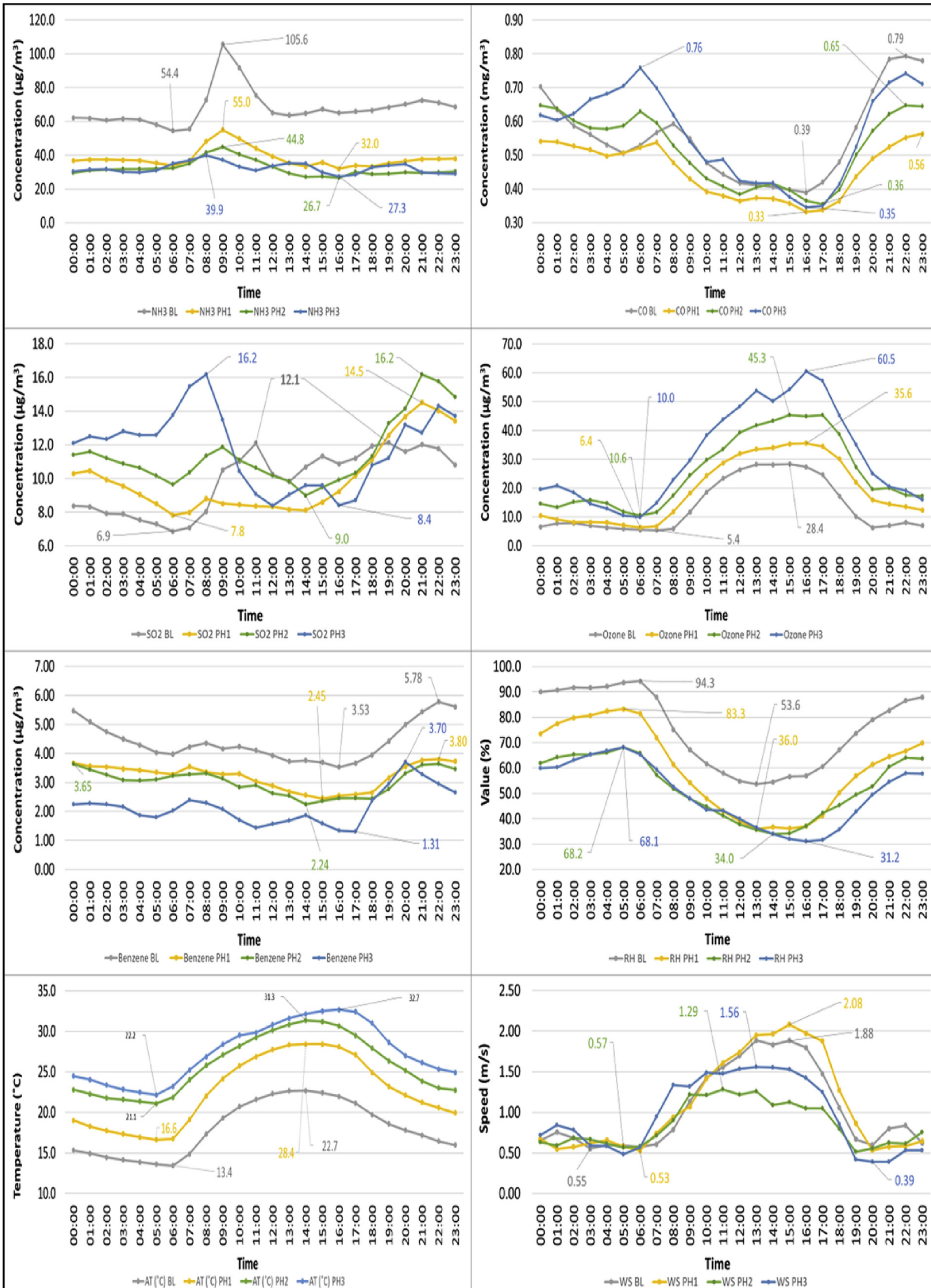

Fig. 3. Diurnal variations of pollutants and meteorological parameters for the whole study period: before lockdown (BL), first phase (PH I), second phase (PH II), third phase (PH III) of lockdown.

In the third phase, the highest concentration was 74.5 μg/m^3 at 12 p.m., and the lowest level was 43.2 μg/m^3 at 5 a.m. (Fig. 3). The peak concentration of PM_{10} during the day time (11 p.m. and 12 p.m.) could be due to the favorable meteorological conditions for its accumulation in ambient air, i.e., the relative humidity of 35–40% (Fig. 3), higher emissions (Lou et al., 2017) and turbulence of PM_{10} in ambient air due to greater wind speed. The high pollution peak during the lockdown period or before the lockdown at night can be attributed to the lower dispersion of PM_{10}.

3.4.2. Diurnal variations in trace gases and VOCs

The peak NO_2 concentration was recorded as 28.7 μg/m^3 at 9 a.m. before lockdown, and the peaks during phases 1 and 2 of lockdown were found at 8:00 a.m. with concentrations of 18.1 μg/m^3 and 17 μg/m^3. During the third phase of the lockdown, the highest concentration was recorded as 20.2 μg/m^3 at 8 p.m. The minimum concentrations were recorded as 9 μg/m^3 at 4 a.m. in before lockdown study period, 8.2 μg/m^3 at 2 p.m. during the second phase, 8.9 μg/m^3 at 3 p.m. during the second phase and 9 μg/m^3 at 4 p.m. during the third phase of lockdown (Fig. 2).

NH_3 showed almost similar trends in its diurnal average concentrations throughout the study period with maximum concentrations of 105.6 μg/m^3, 55 μg/m^3, 44.8 μg/m^3 at 9 p.m. for periods of before lockdown, the first phase, the second phase of lockdown, respectively and 39.9 μg/m^3 at 8 a.m. for the third phase of lockdown. The least concentrations were found to be 32 μg/m^3, 26.7 μg/m^3 and 27.3 μg/m^3 at 4 p.m. for all the consecutive phases of lockdown while for the study period of before lockdown it was found as 54.4 μg/m^3 at 6 a.m. (Fig. 3).

The diurnal average of SO_2 for the study period before the lockdown showed two peaks of concentration 12.1 μg/m^3 at 11:00 a.m. in the morning and 7:00 p.m. in the evening. The least concentration for the same period was recorded at 6:00 a.m. in the morning as 6.9 μg/m^3. The analysis showed the maximum and minimum concentration during the first phase of the lockdown period was 14.5 μg/m^3 at 9 p.m. and 7.8 μg/m^3 at 6:00 a.m., respectively. During the second phase, the maximum and minimum concentrations were 16.2 μg/m^3 at 9 p.m. and 9 μg/m^3 at 2 p.m. During the third phase, the maximum and minimum concentrations were recorded as 16.2 μg/m^3 at 8 a.m. and 8.4 μg/m^3 at 12 p.m. & 4 p.m., respectively (Fig. 3).

Diurnal averages of carbon monoxide didn't show significant differences in the time of highest and lowest concentrations throughout the study period except for maximum concentration during the third phase of lockdown which was found to be 0.76 mg/m^3 at 6 a.m. In contrast, the maximum concentrations for the study period (before lockdown, first phase and the second phase of lockdown) were found to be 0.79 mg/m^3 at 10 p.m. 0.56 mg/m^3 at 11 p.m. and 0.65 mg/m^3 at 10 p.m., respectively. The lowest average concentrations were 0.39 mg/m^3 at 4 p.m., 0.33 mg/m^3 at 4 p.m., 0.36 mg/m^3 at 5 p.m. and 0.35 mg/m^3 at 4 p.m. for the study periods of before lockdown, the first phase, second phase and third phase of lockdown respectively (Fig. 3).

The diurnal trend of average O_3 concentrations showed similar patterns throughout the whole study period with peaks during mid-day at around 3 p.m. & 4 p.m. and lows at 6 a.m. & 7 a.m. The peak concentrations were 28.4 μg/m^3 at 3 p.m., 35.6 μg/m^3 at 4 p.m., 45.3 μg/m^3 at 3 p.m. and 60.5 μg/m^3 at 4 p.m. for the serialized parts of the whole study period as the study progressed. The primary reason for increased concentrations is the continuous increase in solar radiation, which provides a better environment for the enhancement of photochemical reactions in the atmosphere and results in enhanced formation of O_3. The least concentrations were 5.4 μg/m^3 at 7 a.m., 6.4 μg/m^3 at 6 a.m., 10.6 μg/m^3 at 6 a.m. and 10 μg/m^3 at 6 a.m. for study periods of before lockdown, the first phase, second phase and third phase of lockdown respectively.

Benzene also showed similar diurnal patterns across all phases of the study period with the peak concentration during the midnight time and lower concentration during day time. This pattern could be due to the higher dispersion of benzene during the daytime and accumulation in the ambient air during midnight. The maximum average concentrations were found as 5.78 μg/m^3 at 10 p.m., 3.8 μg/m^3 at 10 p.m., 3.65 μg/m^3 at 12 a.m., and 3.7 at 8 p.m. for all the consecutive parts of the study as the phases of lockdown progressed. The minimum concentrations were 3.53 μg/m^3 at 4 p.m., 2.45 μg/m^3 at 3 p.m., 2.24 μg/m^3 at 2 p.m. and 1.31 μg/m^3 at 5 p.m. for the periods of before lockdown, the first phase, second phase and third phase of lockdown respectively (Fig. 3).

3.5. Impact of meteorological parameters

Rain scavenging is a very crucial phenomenon for the removal of several air pollutants (Ravindra et al., 2006; Shukla et al., 2008). Yoo et al. (2014) found that the PM_{10} has higher removal rates followed by SO_2, NO_2, CO, and O_3, and the removal rates are directly proportional to the intensity of rain. The comparison of PM (PM_{10} and $PM_{2.5}$), SO_2, NO_2, CO, and O_3 daily average variation graphs with rainfall graph (Figs. 1 and 2) showed the rain scavenging of these pollutants. Throughout the whole study period, very few events of rainfall were recorded, and subsequently, the effect of rain on the concentrations of pollutants was limited to these very few events. The study periods of first and third phases of lockdown received negligible or no rainfall.

Air temperature and wind speed are important parameters that affect the dispersion of air pollutants. The increase in air temperature due to the onset of summer season directly decreases the stability of the atmosphere and correspondingly increases the mixing height of pollutants and therefore increases the vertical mixing of pollutants in the troposphere (Ravindra et al., 2019; Cichowicz et al., 2017; Akpinar et al., 2008). The increase in air temperature due to the rise in solar radiation also enhances the intensity of photochemical reactions in the atmosphere. The average temperature during the first phase, second phase, and third phase of lockdown increased by 4.5 °C, 3.3 °C, and 1.6 °C respectively, as compared to the pre-lockdown period due to the onset of the summer season. Hence, a slight fraction of the total decrease in the concentrations of pollutants during the lockdown period can be attributed to the increase in temperature. The effect of increasing air temperature during the day time on the O_3 concentration is also observed in the graph of diurnal variation of O_3. This increase is due to the availability of intense solar radiation for the photochemical reactions to occur, which results in the formation of ground-level O_3.

The increase in wind speed is favorable for the dispersion of pollutants and local suspension of the geological sources of PM_{10}. The average wind speed during the consecutive phases of lockdown slightly increased from 1.0 m/s before the lockdown to 1.1 m/s during the first phase, then decreased to 0.8 m/s during the second phase and then again increased to 1 m/s during the third phase of lockdown. The comparison of daily average wind speed and the pollutant variation shows a decrease in the concentrations of NO_2, SO_2, CO, PM and VOCs during the peaks of wind speed during the whole study period (Figs. 1 and 2). Among the particulate matter, PM_{10} showed less decline, which might be due to the resuspension of dust particles and higher wind speeds (Akpinar et al., 2008; Lawrence et al., 2013, 2016; Singh et al., 2020).

Relative humidity affects the accumulation rates of particulate matter and other pollutants. Lou et al. (2017) reported that accumulation rates of $PM_{2.5}$ are favored by very dry (<45%), dry (45–60%), and low (60–70%) relative humidity in the ambient air, whereas the PM_{10}, SO_2, and NO_2 accumulations are peaked at 40 ± 5% relative humidity. Munir et al. (2017) also found somewhat similar relationships of relative humidity with particulate matter. During the study period, relative humidity went on decreasing except few peaks, which were accompanied by rainfall events (Fig. 2). The average value of 75.3% before the lockdown study period decreased to the average value of 59.7% during the first phase, 52.1% during the second phase, and 50.1% during the third phase of lockdown. The relative humidity remained in the range favorable to the $PM_{2.5}$ accumulations throughout the study period except for few peaks during the before lockdown period. This also partly explains the lower variations in the $PM_{2.5}$ concentrations during the study period. By comparing the daily average relative humidity and daily average PM_{10}, SO_2, and NO_2 concentration, it was found that these pollutants showed a lot of fluctuation throughout the study period, which signifies rapid reductions and rapid accumulations due to addition and reduction of pollutant loads from sources.

Along with the meteorological parameters, daily backward trajectories of air masses were also studied for all the different phases of lockdown using Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) as shown in Fig. 4. The backward trajectories showed that prevailing backward air masses originate from North-west direction during the study period, i.e., Punjab and Haryana. These states are adjacent and upwind to the study area, reflecting the regional transport of pollutants to the Chandigarh (Singh et al., 2020a). There are several thermal power plants in the region, and some of them are situated within 30 km of aerial distance from Chandigarh.

3.6. Source identification

To identify and quantify the pollution sources, there are many robust source apportionment approaches, as detailed by Hopke (2016). However, due to the limitation of detailed speciation and specific source markers, best possible approaches (Ravindra et al., 2008, 2016), i.e., Principal Component Analysis - PCA (Table 4), Characteristic Ratios (Table 3), Pearson's Correlations and HYSPLIT backwind trajectories (Fig. 4) were applied to identify the possible sources of pollutants before and during the lockdown period in Chandigarh.

3.6.1. Principal component analysis

To identify the sources of air pollutants, PCA was applied to the dataset using SPSS software. The technique of PCA reduces the multidimensionality of large dataset having more number of variables and converts it to a more interpretable small dataset with a new set of factors or components which represent the variables of an old dataset having linear combinations between them (Ravindra et al., 2007; 2008). These new factors are arranged according to the computed percentage variance. This technique is widely used to identify the pollution sources of soil, water, and air pollutants (Kong et al., 2015; Lu et al., 2010; Ravindra et al., 2008, Guo et al., 2004). Varimax Rotation and Kaiser Normalization methods were used for

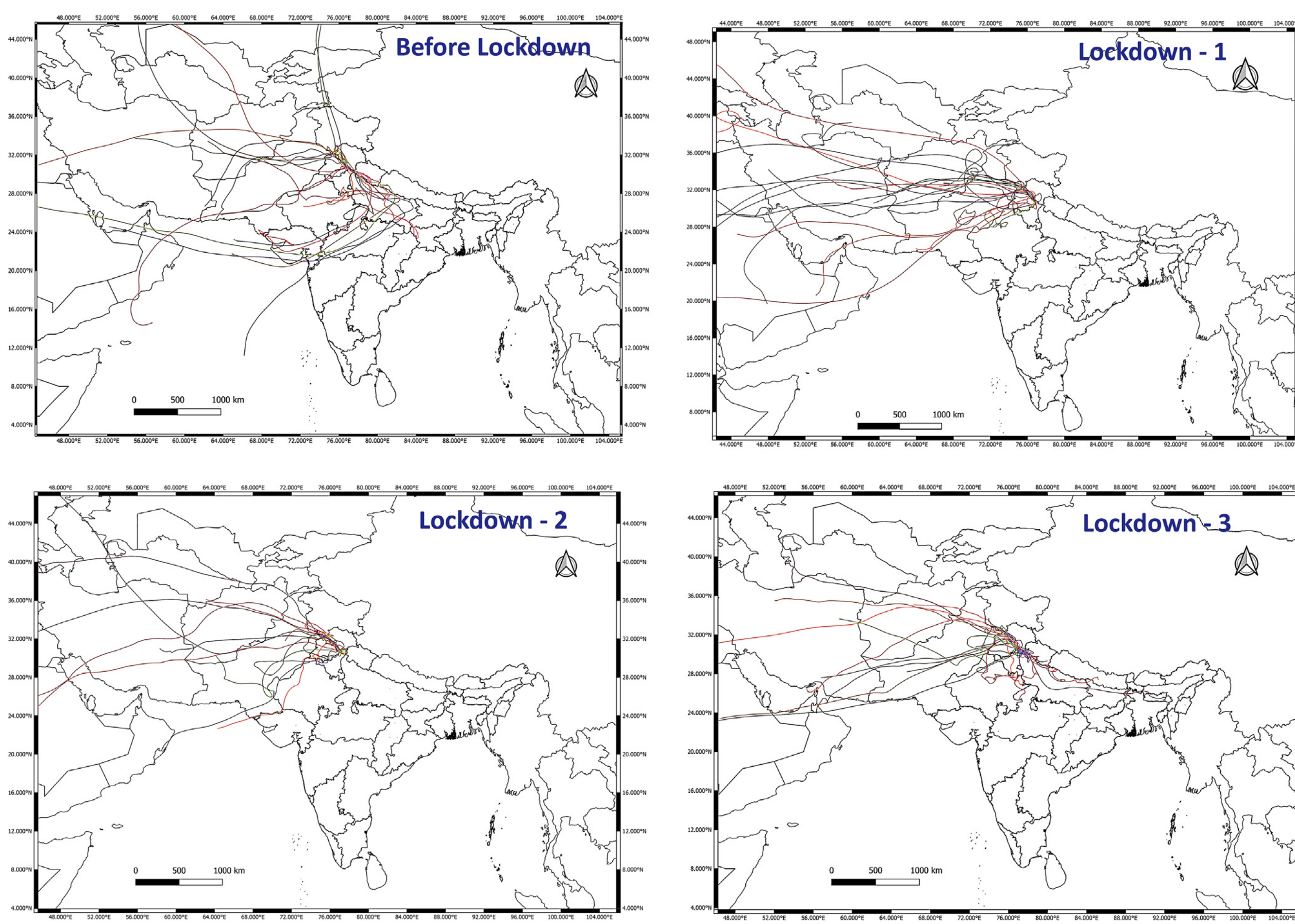

Fig. 4. HYSPLIT backward trajectories of air masses during different phases of the study period.

Table 3
Characteristics ratios of various pollutants.

Study Period	$PM_{2.5}/PM_{10}$	T/B	EB/B	m,p-X/B	o-X/B	o-X/EB	m,p-X/EB
Before Lockdown	0.35	0.31	0.73	0.13	0.23	0.32	0.18
Lockdown Phase 1	0.40	0.13	0.96	0.25	0.33	0.35	0.26
Lockdown Phase 2	0.35	0.22	0.94	0.29	0.36	0.38	0.31
Lockdown Phase 3	0.36	0.47	1.11	0.91	0.40	0.36	0.82

where, T/B is toluene to benzene ratio; EB/B is ethylbenzene to benzene ratio; mp-X/B is m,p-xylene to benzene ratio; o-X/B is o-xylene to benzene ratio; o-X/EB is o-xylene to ethylbenzene ratio and m,p-X/EB is m,p-xylene/ethylbenzene.

Table 4
PCA results for the whole study period.

Parameter	Before Lockdown			Lockdown 1			Lockdown 2			Lockdown 3		
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3
$PM_{2.5}$	0.91	0.08	−0.04	0.23	0.79	−0.17	−0.26	0.84	−0.34	0.00	0.95	0.18
PM_{10}	0.88	−0.09	0.03	0.12	0.91	−0.01	−0.47	0.75	−0.41	−0.02	0.93	0.08
NO_2	0.41	0.07	0.14	0.17	−0.02	0.91	0.74	0.17	0.43	−0.10	0.32	0.83
NO	0.23	0.88	−0.13	0.26	−0.26	−0.04	0.80	−0.07	0.47	0.91	−0.03	0.17
NOx	0.34	0.72	−0.04	0.04	−0.18	0.88	0.83	0.06	0.45	0.52	0.28	0.73
NH3	0.17	−0.14	−0.20	0.16	−0.08	−0.71	0.26	0.11	0.84	0.12	0.47	−0.67
SO_2	0.41	−0.35	−0.09	0.71	0.37	−0.12	0.09	0.87	0.27	0.29	0.38	0.66
CO	0.83	0.39	−0.11	0.13	0.19	0.47	0.74	0.48	−0.08	−0.11	0.82	0.28
Ozone	0.02	−0.89	−0.28	0.04	0.68	0.58	−0.08	0.25	−0.65	0.59	−0.54	0.20
Benzene	0.94	0.17	0.00	0.01	0.25	0.10	0.19	0.92	−0.01	0.82	0.12	0.43
Toluene	−0.41	0.19	−0.33	0.11	0.72	−0.04	0.03	0.01	0.20	0.91	−0.09	0.00
o-xylene	0.33	0.31	0.48	0.90	−0.02	0.18	0.95	−0.17	0.02	0.92	−0.17	0.15
m,p-Xylene	−0.14	−0.03	0.95	0.90	0.33	0.03	0.42	0.04	0.05	0.85	0.12	−0.25
ethylbenzene	0.04	0.15	0.91	0.97	−0.05	0.01	0.91	−0.22	0.14	0.97	−0.05	−0.04
Eigen Value	4.60	3.38	2.51	4.38	2.93	2.33	6.14	3.29	1.64	5.82	3.78	1.89
% Variance	32.88	24.16	17.90	31.25	20.96	16.63	43.88	23.49	11.70	41.56	26.97	13.51
Cumulative %	32.88	57.05	74.95	31.25	52.21	68.84	43.88	67.36	79.06	41.56	68.53	82.04

the factor analysis, and all the factors showing the eigenvalues greater than one were included.

Three factors showed the maximum variance during the study period before lockdown and represented the cumulative variance of 74.9%. Factor 1 showed a 32.9% variance with significant factor loadings for $PM_{2.5}$, PM_{10}, benzene, and carbon monoxide. Factor 2 showed a 24.2% variance with considerable factor loadings for NO and NO_x. The primary anthropogenic source of particulate matter, carbon monoxide, benzene, NO, and NO_x in urban air is the burning of fossil fuels in vehicles (Buzcu and Fraser, 2006; Fernandes et al., 2002). Hence, vehicular emissions can be the primary source of these emissions. Factor 3 showed 17.9% variance with significant factor loadings for m,p-xylene and ethylbenzene. These VOCs emissions can be associated with industrial and residential sources.

During the first phase of lockdown, three factors showed the maximum variance with a cumulative variance of 68.8%. Factor 1 showed a 31.3% variance with significant factor loadings for SO_2, o-xylene, m, p-xylene, and ethylbenzene. The main source of these emissions could be the regional transport of these pollutants from coal-burning thermal power plants working in the upwind regions of the city, as also observed by Wang (2013). The backward trajectories of air masses for the study area are depicted in Fig. 4. Factor 2 explained 20.9% variance with substantial factor loadings for PM_{10}, $PM_{2.5}$, O_3, and toluene. Factor 3 showed a 16.6% variance with higher factor loadings for NO_2, NO_x, and O_3. Factors 2 and 3 indicate that vehicular pollution could be the major source of toluene, NO_x, and PM.

During the second phase of lockdown, Factor 1 had a 43.9% variance with significant factor loadings for o-xylene, ethylbenzene, NO_x, NO, NO_2, and CO. The source of these emissions can either be fuel burning in vehicles (Gallego et al., 2008) or regional transport of these pollutants occurring due to stubble burning from upwind areas or can be the combination of both these sources (The Hindu 2020; Ravindra et al., 2020; Sembhi et al., 2020). Factor 2 showed 23.5% variance with higher factor loadings for benzene, SO_2, $PM_{2.5}$, and PM_{10}, which can be attributed to the emissions from power plants burning coal (Wang et al., 2013). Factor 3 showed an 11.7% variance with a significant factor loading for NH_3. The source of NH_3 can be the agricultural and rural residential sources in the vicinity of the city.

During the third phase, Factor 1 showed a 41.6% variance with higher factor loadings for ethylbenzene, NO, o-xylene, toluene, benzene, m,p-xylene, and NO_x. Again, the source of these emissions can either be fuel burning in vehicles or regional transport of these pollutants occurring due to stubble burning from upwind areas or can be the combination of both sources. Factor 2 explained a 27% variance with significant factor loadings for $PM_{2.5}$, PM_{10}, and CO, which are mainly emitted by vehicular exhausts (Lawrence et al., 2013). Factor 3 showed 13.9% variance with significant factor loadings for NO_2 and SO_2, which indicates pollution originating from coal-powered thermal power plants.

3.6.2. *Characteristic ratios of $PM_{2.5}/PM_{10}$, $PM_{2.5}$/CO, and VOCs ratios*

The $PM_{2.5}/PM_{10}$ ratio was used to identify the possible sources because different types of sources emit different sized proportions of particulate matter (i.e., particles <2.5 μm and 2.5 μm < particles<10 μm). The combustion sources such as internal combustion engines, coal combustion, and biomass combustion emit a large proportion of $PM_{2.5}$. Whereas, particles larger than $PM_{2.5}$ in PM_{10} mostly contain geological matter, pollens, etc. (Querol et al., 2001; Das et al., 2006). During the study period, the ratio increased from 0.35 before lockdown to 0.4 during the first phase, decreased to 0.35 during the second phase, and again slightly increased to 0.36 during the third phase of lockdown. There was no significant difference observed in the ratios among all the parts of the study period. The $PM_{2.5}/PM_{10}$ ratio generally increases

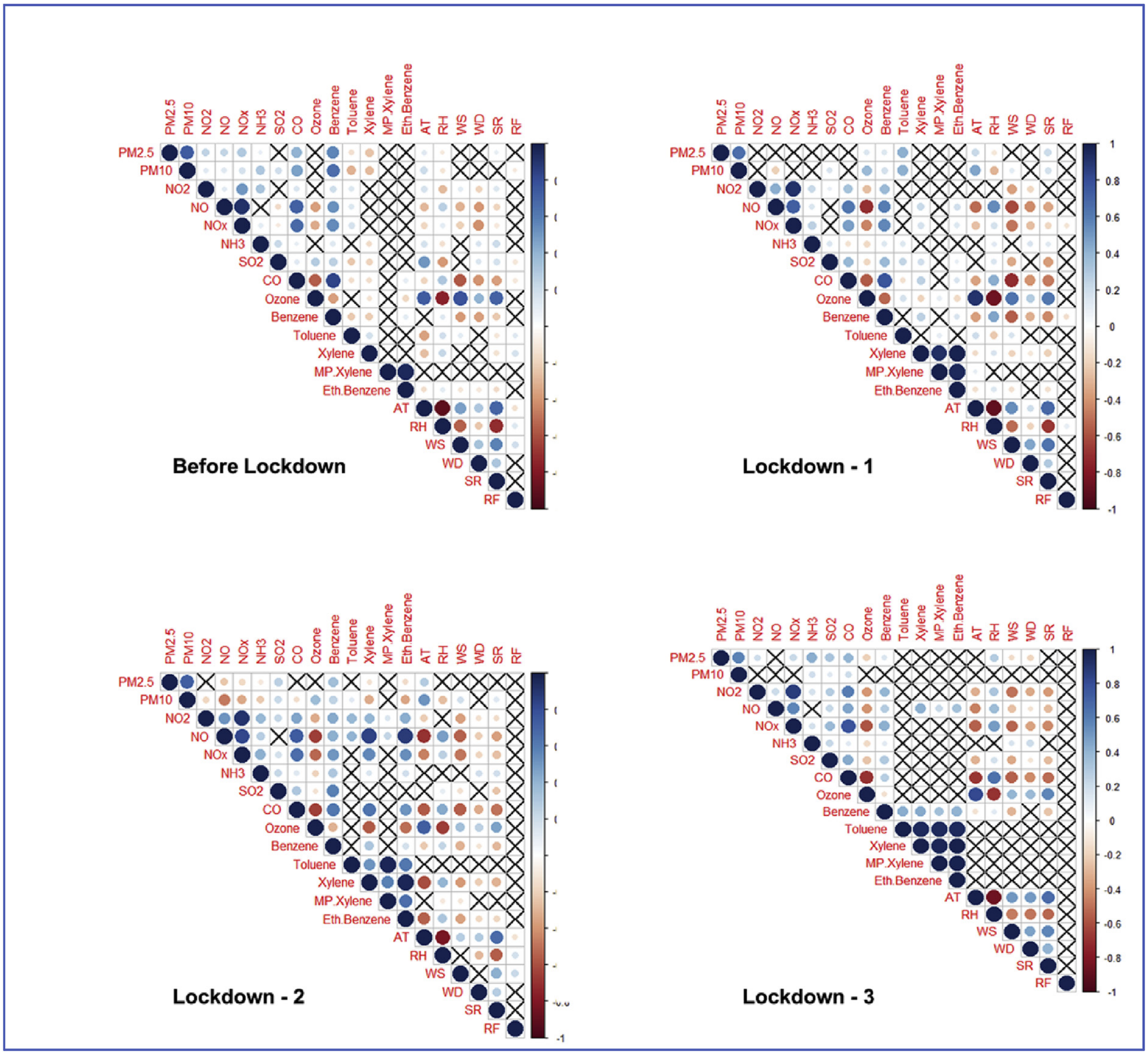

Fig. 5. Pearson correlation between various pollutants during different phases of the study period.

during the winter seasons and decreases during summer seasons (Brook et al., 1997; Xu et al., 2016). As the lockdown announced, the summer season also started resulting in a slight increase in temperature. Further, there were some dust events, which affected the $PM_{2.5}/PM_{10}$ ratio as coarser particles from dust introduced in the atmosphere in summers. The lockdown minimized the significant sources of $PM_{2.5}$ in urban air, i.e., combustion emissions from vehicles and industries activities. Hence, there was no considerable variation observed in $PM_{2.5}/PM_{10}$ ratio during the lockdown period. The small variations could be linked to the crop residue burning in the agricultural sector, including the increased uses of solid biomass fuel due to restricted supply of liquefied petroleum gases in rural areas of neighboring states (Fig. 4, The Hindu 2020; Ravindra et al., 2020). In the third phase of lockdown, there was further addition of local vehicular $PM_{2.5}$ emissions in the city, resulting in a slight increase in $PM_{2.5}/PM_{10}$ ratio.

The toluene/benzene (T/B) ratio was 0.31 during the period before lockdown, then decreased to 0.13 in the first phase, 0.22 in second, and increased to 0.47 in the third phase. T/B ratios less than 2 signifies emissions from vehicular sources, whereas greater than 2 signifies that there can be additional pollutant load from industrial sources (Hui et al., 2018. Bruno et al., 2006). This indicates that vehicular emissions are the prominent source for toluene in Chandigarh. The decrease in the first two phases of lockdown can be due to the less emissions of toluene because of less vehicle movement. Whereas the ratio shows greater toluene concentrations during the third phase, which can be due to the increased vehicular movement (Monod et al., 2001). The proportionally slight decrease of benzene concentration during lockdown indicates its other sources, which could be linked with the uses of coal in thermal power plants, including the smoldering of crop residues in agriculture fields (Singh et al., 2020b; Ravindra et al., 2006).

All other ratios (ethylbenzene/benzene, m,p-xylene/benzene, o-xylene/benzene, o-xylene/ethylbenzene, m,p-xylene/ethylbenzene) showed a similar pattern during the whole study period. A continuous increase in the concentrations of all isomers of xylene and less reduction of ethylbenzene concentration was seen during the lockdown as compared to the before lockdown period. The primary source of VOCs in urban air is emissions from vehicular exhaust. During the lockdown, when the vehicular emissions were very less as compared to before, therefore higher concentrations of xylene isomers despite their shorter lifetimes (especially during summer season) indicate that other sources contributed to their concentration. A comparatively more significant decrease in the concentration of ethylbenzene can be due to the halting of industries, which is its major source as ethylbenzene is used as a resin in various industrial processes.

Also, the ratios of xylene isomers with benzene and ethylbenzene help to determine the photochemical age of the air masses (Ravindra et al., 2019; Roukos et al., 2009). These ratios were recorded very less in the study (Table 3), which also indicates the regional transport of VOCs and aged air masses. This shows regional transport of VOCs from coal-burning thermal power plants and biomass burning in the nearby upwind states were also contributing to the city's VOC pollution load (Fig. 4, Ravindra et al., 2020).

3.6.3. Restricted anthropogenic activities and air pollution peaks during COVID-19 lockdown

During the whole lockdown period, the lowest concentrations were observed during the first phase of lockdown, which can be due to the strict restrictions on all human activities. During this first phase of lockdown, NO_2, NO_X, benzene showed their highest peaks on 5th April. While $PM_{2.5}$, SO_2, ethylbenzene, o-Xylene, m,p-xylene showed their highest peaks on 6th April. PM_{10}, toluene, and CO also showed their second-highest peaks on 6th and 5th April. NH_3 and NO showed their highest peaks on 7th April. All these peak concentrations can be attributed to the event of excessive fireworks and open burning of oil in traditional diyas (lamps) done by the residents of city Chandigarh on the night of 5th April (Business Standard 2020; Greven et al., 2019; Zu et al., 2018; Zhao and Zhao, 2018; Kong et al., 2015; Wang et al., 2007; Ravindra et al., 2003). This shows that household-related pollution also affects the air quality of Chandigarh.

The second phase of lockdown showed almost similar concentrations but with little increase as compared to the first phase of lockdown. The restrictions and relaxations on human activities were identical to the first phase. The reason for aggravated concentrations can also be the regional transport of pollutants to the city from stubble burning, and coal-fired power plants in the upwind neighboring states (Singh et al., 2020, Ravindra et al., 2020). The PCA analysis and Pearson's correlations for this phase (Table 4, Fig. 5) also showed linear correlations among the pollutants which are released from stubble burning, and coal-fired power plants (Ravindra et al., 2019a; Xue et al., 2016). The characteristic ratios of VOCs (Table 3) also indicated the regional transport of aged air parcels to the study area.

The third phase of lockdown introduced many relaxations in the city, like reopening of the government, commercial offices, and essential manufacturing industrial units. The concentrations during this phase increased as compared to the other stages of lockdown. They were slightly less than the before lockdown period for almost all pollutants except for nitrogen oxides and NH_3. The reason for lower concentrations of trace gases seems to be the enhanced photochemical reactions with increased solar radiation and the formation of secondary compounds like O_3, ammonium sulfate, and ammonium nitrate.

4. Conclusions

The comparison of air quality 21 days period before the lockdown and during the different phases of lockdown showed that there was a significant reduction in the concentrations of almost all pollutants except for sulfur dioxide, ozone, and m,p-xylene. The maximum decreases were found to be associated with the restriction on automobiles and the shutting of industries during the first two phases of lockdown. When these activities were partly started during the third phase of lockdown, higher concentrations were recorded for almost all the pollutants as compared to the first two phases. The pattern of meteorological parameters during the lockdown also elaborated certain variations in the concentrations of PM, SO_2, NO_2, and other pollutants. The principal component analysis, characteristic ratios, and Pearson's correlations identified vehicular emissions (exhaust and non-exhaust) as the primary source of local air pollution. These approaches, along with backward trajectories of air masses, recognized the contribution of pollutants originating from coal-burning and stubble burning in the neighboring upwind areas. The analysis of daily average showed that the local emissions coming from residential areas also adversely affect the air quality of the city. This finding gives vital evidence that local household air pollution also affects the overall air quality of Chandigarh. The study helps to identify and understand the possible sources of air pollution during controlled anthropogenic activities. Though, to assign specific sources contribution, a proper source apportionment study is recommended. However, in the absence of a particular source apportionment study, the finding of the current study will be useful to plan proper policy interventions to reduce the air pollution originating from local and regional sources.

Credit author statement

Suman Mor: Conceptualization; Formal analysis; Investigation; Supervision; Methodology; Validation; Visualization; Writing - original draft; Writing - review & editing. Sahil Kumar: Data curation; Formal analysis; Methodology; Visualization; Writing - original draft Tanbir Singh: Data curation; Methodology; Visualization; Writing - original draft Sushil Dogra: Formal analysis; Methodology; Validation; Visualization; Writing - review & editing. Vivek Pandey: Validation; Visualization; Writing - review & editing. Khaiwal Ravindra: Conceptualization; Formal analysis; Investigation; Supervision; Methodology; Validation; Visualization; Writing - original draft; Writing - review & editing.

Declaration of competing interest

Authors confirm that there's no financial/personal interest or belief that could affect the objectivity. All authors read the paper and approved it.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.127978.

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