



Research article

Anomalous behaviour of ozone under COVID-19 and explicit diagnosis of O₃-NO_x-VOCs mechanismA. Rathod^a, S.K. Sahu^b, S. Singh^c, G. Beig^{a,*}^a Indian Institute of Tropical Meteorology, Pune 411008, India^b Utkal University, Bhubaneswar, Orissa, India^c India Meteorological Department, New Delhi, India

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ABSTRACT

Air pollution is linked to higher rates of human mortality especially those infected with COVID 19. Ozone is a harmful pollutant and is responsible for many health issues. However, some reports suggest that ozone is a strong disinfectant, and can kill the viruses. We hereby, report on the vulnerability of ozone due to COVID-19 lockdown whose levels flutter from surging to saturation in a highly polluted Indian capital, due to significant decline in anthropogenic emissions of ozone precursors. Average observed levels stabilized at 30 ppb, 12 ppb, 740 ppb, and 900 ppb for ozone (O₃), nitrogen dioxide (NO₂), carbon monoxide (CO) and volatile organic compounds (VOCs) respectively during lockdown period from 27th March to 10th April 2020. The NO₂, CO and VOC declined by 50 %, 37 %, 38 % respectively during the lockdown period of 2020 as compared to similar period in 2019. The anomalous response of ozone during the lockdown is explained by resolving the poorly known complex O₃-NO_x-VOCs mechanism with the help of data from air monitoring stations in Delhi, India. The data obtained from this study advances the fundamental understanding of ozone chemistry that may lead to improved ozone parameterization in chemical transport models and better planning of ozone risk management strategies for any global mega cities.

1. Introduction

Corona Virus Disease (COVID-19) pandemic is the greatest challenge that confronts humanity since World War-II [1]. As of October 2020, the virus has infected around 39 million people globally and has an infection fatality ratio of 2.79% [2]. According to the study at Harvard University, long term exposure to fine particulate matter such as PM_{2.5} aggravates the adverse health effects from COVID-19 infections [3]. Economically, the COVID-19 pandemic led to a decline in industrial production, revenue by air couriers and the tourism industry [4]. Global manufacturing sector industries such as automobile, food, metal, electronics etc. suffered a decrease in FDI (Foreign Direct Investment). As estimated by United Nations Conference on Trade And Development (UNCTAD) manufacturing sector FDI has decreased by 5–15 % due to pandemic coupled with shutdown [5]. Travel restrictions imposed by government resulted in loss of \$200 billion globally and a loss of around \$113 billion by the airlines has been forecasted by The International Air Transportation Association (IATA) [6, 7]. COVID-19 lockdown led to a decline in mean concentration of nitrogen dioxide (NO₂) by 28 %, 57 % 46 %, 50

% and 62 % in Beijing, Wuhan, Guangzhou, Barcelona and Madrid respectively [8, 9]. In São Paulo, Brazil, 13 stations reported 34 %–68 % reduction in NO_x levels when compared with business as usual weekdays prior to lockdown [10]. Observations reveal a drastic drop in gaseous pollutants and particle pollution over major cities of the world like Delhi, London, Los Angeles, Milan, Mumbai, New York, Rome, São Paulo, Seoul and Wuhan, ranging between –9 % and –60 % compared to 2019 data, and between +2 % and –55 % compared with the prior four-year average [11, 12]. COVID-19 related reduction in NO₂ was up to 30 % as compared to pre-lockdown period based on satellite data released by National Aeronautics and Space Administration (NASA) and European Space Agency (ESA) over areas of Europe, China and North America.

Breathing ozone can trigger many health conditions, such as chest pain, coughing, throat irritation, and airway inflammation in humans [13]. It can also reduce lung function and harm lung tissues, symptoms almost similar to COVID-19 ailment [14]. However, a contemporary school of thought advocates that ozone effectively destroys bacteria and viruses, due to its disinfectant and oxidizing properties [15]. Thus, an increase in ozone concentration in the atmosphere may assist to reduce

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the spread of COVID-19 infections among the human population [16]. Current study, reports on the level of ozone in the atmosphere during the lockdown in Delhi, India. Ozone pollution has also been studied from radical chemistry to O₃-VOC-NO_x sensitivity in Chengdu, south-western China [17]. Ambient ozone trends at urban, suburban, and rural monitoring sites across the United States over a period of decreasing NO_x and VOC emissions (1998–2013) has also been reported earlier [18]. Studies on NO_x and VOCs, a major component of O₃ has been reported earlier from photochemical air quality models [19, 20]. However, studies indicate that high uncertainties are associated with the input parameters used to develop the photochemical models. Subsequently, leading to poor estimation of O₃, VOCs and NO_x levels in the atmosphere. The split between NO_x-sensitive and VOC-sensitive chemistry is a major source of uncertainty in most forecasting models. NO_x-VOC sensitivity is difficult to predict, and there are no straight forward explanation for distinguishing NO_x-sensitive from VOC-sensitive conditions. The COVID-19 lockdown period (23rd March 2020–10th April 2020) offers a unique opportunity to understand the distribution pattern of O₃, VOCs and NO_x in the atmosphere over Delhi, India. In addition, this study highlights O₃ production and the sensitivity to NO_x, CO and VOCs based on data obtained from air monitoring stations in Delhi, India. In this study, we have represented methane and non-methane hydrocarbons as VOCs (includes variety of chemically different compounds such as benzene, ethanol, etc.) whose levels are measured collectively by the analyser is adopted in this work. This work is carried under System of Air Quality and Weather

Forecasting and Research (SAFAR), a research project of Ministry of Earth Science, Government of India which is also recognized as pilot study of World Meteorological Organization (WMO) [21] established in Indian capital city Delhi (Figure 1).

1.1. Experimental design and methodology

This study is focused on Indian capital Delhi as shown in Figure 1. Delhi is a highly urbanized landlocked city and with an elevation of 216 m above sea level. The city covers an area of 1483 sq km and has a population of about 17 million [22]. It is over populated and has many establishments compared to other major cities in India. The study design is based on the data obtained from the project SAFAR, which consists of 8–10 Air Quality Monitoring Stations (AQMS) in Delhi, India. The air monitoring analyzers such as O₃, NO_x, CO, and VOCs at the 10 stations are approved by the United States Environmental Protection Agency (US EPA) and certified by Bureau Veritas Certification (ISO9001) for quality control. The micro environments considered under this study are viz. downtown area, background, industrialized area, residential area, traffic areas etc. of Delhi and the average concentration of pollutant is considered to be representative of the city as per WMO guidelines [21]. The data is time resolved and have frequency of 5 min and then binned for one-hour interval for further analysis. The NO_x analyser (Model 42i, Thermo Fisher Scientific, US), works on the principle of chemiluminescence and measures nitric oxide and total oxides of nitrogen

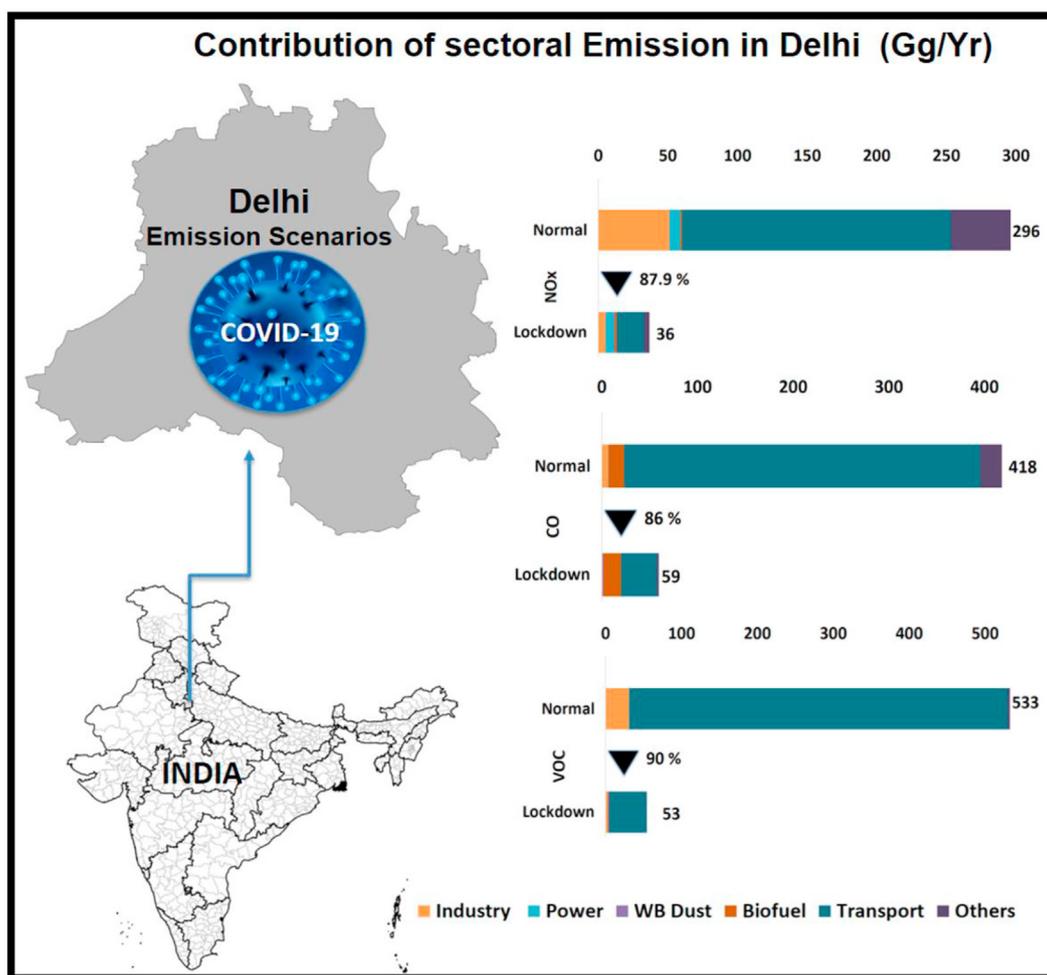


Figure 1. Location of Mega city Delhi in India. The standard emission (Gg/Yr) of ozone precursors, namely NO_x, CO and VOCs from six broad categories of sources in Delhi region and their relative share is depicted by different colors by the horizontal panel in the right side of maps. The downward arrows indicate the total reduction (%) in the magnitude of the emissions of these pollutants under COVID-19 lockdown scenario. The lower horizontal panel depicts the reduced magnitude of emissions for lockdown scenario. The total emissions (Gg/Yr) in standard as well as lockdown scenario of each pollutant are written in extreme left side of top panel.

concentration within the gaseous sample. The CO was measured using Non-Dispersive Infra-Red detectors using Gas Filter Correlation Carbon Monoxide Analyser (Model: 48i, Thermo Fisher Scientific, US). Ozone is monitored by Ozone analyser (Model: 49i, Thermo Fisher Scientific, US) which uses mercury vapor lamp as light source that emits wavelength of 254nm. Ozone concentration is determined by the UV light absorbed by the air sample. Flame ionization detector was used to determine Methane/Non-Methane hydrocarbons by using Model: HC51M-LCD (Environment SA, France). The instruments were calibrated and maintained according to the US EPA standard operating procedure [21]. Further detailed information about calibration procedure can be found by referring to title 40 of the Code of Federal Regulations (CFR) part 50 [23].

1.2. Emission inventory

The pollution level in a city is largely dependent on the sources of emissions [21, 24, 25]. The sources of NO_x , CO and VOC emissions are classified in 6 different categories by SAFAR over Delhi [24, 25]. These are fossil fuel (vehicular transport), bio-fuel (residential combustion), industry, power, windblown dust (re-suspended dust) and rest others (like brick kiln, open trash burning and solid waste). These sources have been found as potential sources for the emissions into the atmosphere in Delhi [24]. The relative distribution of various sources of emissions for NO_x , CO and VOCs are shown in Figure 1. The emission inventory of ozone precursors for the lockdown scenario is constructed based on the combination of standard emissions of Delhi [24] and the local field survey report on activity data during the lockdown period. Building an inventory for air pollutants is a challenging task as it requires qualitative and quantitative data and source apportionment of various sources of emission. The emission inventory used in this work is based on the latest report released by Indian government [24] for Delhi, which is based on the bottom up approach, and a Geographical Information System (GIS) based statistical model [25]. The bottom up approach consists of repository of activity data from various sources on a finer resolution and the information about emission factors. Emissions from various sources have been classified broadly in six sectors namely transport, bio-fuel, power, industrial, re-suspended dust and others (like brick kiln, open trash burning and solid waste). Development of such complex inventory, involves the recognition of each source and distribution of emission geographically. The estimations are based on primary and secondary activity data collected at grid level. The total emission is expressed by following equations for all pollutants:-

$$TE = \sum_a \sum_b FU_{a,b} \left[\sum_c EF_{a,b,c} U_{a,b,c} \right] \quad (1)$$

Where,

- a, b, c = sector, fuel type, technology
- TE = Total Emission
- FU = Sector and fuel specific amount
- EF = Emission Factors (Technology specific)
- U = Fraction of fuel for a sector with particular technology, where $\sum U = 1$ for each fuel and sector.

Specific vehicular emission factors for transport sector for India are developed by Automotive Research Association of India (ARAI). The emission from transport sector has been calculated as per the equation below:

$$E_t = \sum (veh_t \times D_t) \times EF_{t,km} \quad (2)$$

Where,

- E_t = Total Emission of compound

veh_t = Number of Vehicle per type

D_t = Distance travelled in a year per different vehicle type

$EF_{t,km}$ = emission of compound, vehicle type per driven kilometre

In the present work, the reduction in emission in various sectors has been done as per the local statistics for each species NO_x , CO and VOCs as shown in Figure 1.

2. Results

Figure 2 shows the time series of NO_2 , CO and VOC and O_3 for the period 20th February 2020 to 10th April 2020, covering 3 regimes- (a) *Normal*: Business as usual scenario during Feb 20 to 5th March; (b) *Partial Lockdown Scenario*: Fear and panic escalated due to COVID-19 related deaths in India led to partial lockdown voluntary and due to government induced selected restrictions during the period 5th March till about 23rd March; (c) *Full Lockdown*: It is considered from 23rd March 2020 onwards. The data of 2020 is compared with the data of identical period of 2019 in Figure 2. We compared data obtained during the full lockdown period between 2019 and 2020 for Figure 2. The levels of all pollutants declined significantly except ozone immediately after the lockdown period of 2020 imposed, as evident from Figure 2. The distribution of these parameters in 2019 continues to follow the normal pattern and do not show any visible declining trend. Table 1 shown in volume mixing ratio of O_3 , NO_2 , CO and VOCs averaged over the full lockdown period for 2020 and 2019. The statistical error associated with observational data is also shown. The percentage reduction of these pollutants in 2020 with respect to 2019 is also provided. There was a decline of 25 %, 50 %, 37 % and 38 %, in concentration of O_3 , NO_2 , CO and VOCs respectively. During this lockdown period of 2020, as per the survey and statistics of internal reports, transport activities almost came to a halt and only essential duty and supply vehicles were on road. Similarly rest of other sectors (like brick Kiln, solid waste, open trash burning, etc) were affected except bio-fuel emissions in fringe areas of Delhi where people use coal and wood for residential cooking [24]. Lockdown resulted in reduction of emission of ozone precursors by ~85–90 % as discussed in the previous section. The reduced emission scenario is provided in Figure 1 for NO_x , CO and VOCs (see Table 2).

The variability in partial and full lockdown periods with respect to normal period in 2020 is provided in Table-2. A reduction of 40 %, 34 % and 46 % is observed in NO_2 , CO and VOCs respectively during full lockdown period w.r.t. partial lockdown period. However, a nominal increase of 3 % in ozone level is observed for the same scenario. The reduction during full lockdown period as compared to normal period is found to be 42 %, 44 %, 31 % and 7 % for NO_2 , CO, VOCs and O_3 respectively.

Figure 3 (a) shows, the time series of the volume mixing ratios of O_3 , NO_2 , CO and VOCs for the period 20th February 2020 to 10th April 2020 covering all 3 periods. Figure 3 (b) shows the relationship between the ratio $(\text{NO}/\text{NO}_2) \times 100$ and ratio VOC/NO_x for the same period as Figure 3 (a). As discussed earlier, a significant declining trend in NO_2 , CO and VOC is observed during the full lockdown period. After about a week of lockdown, levels got stabilized and further no more decline was observed. During this period, the weather was fairly uniform and consistent as confirmed by the automatic weather station co-located with air quality analysers. Smaller fluctuations during this period are mainly due to minor variability in weather parameters. Ozone levels declined in the initial few days after the full lockdown while it was still under the NO_x saturation regime which will be discussed in the next section. However, ozone level started increasing from 26th March onwards until around 2nd April. Thereafter, it tends to stabilize around 36 ppb. The variability in the ratio of NO to NO_2 and that of VOC to NO_x was relatively quite high before full lockdown as compared to lockdown. However, while $(\text{NO}/\text{NO}_2) \times 100$ ratio became constant after the full lockdown, the VOC/NO_x ratio increased and touched a peak value coinciding with the minimum in ozone at around 2nd April. Thereafter, VOC/NO_x ratio also declined rapidly. The variability of ozone and its

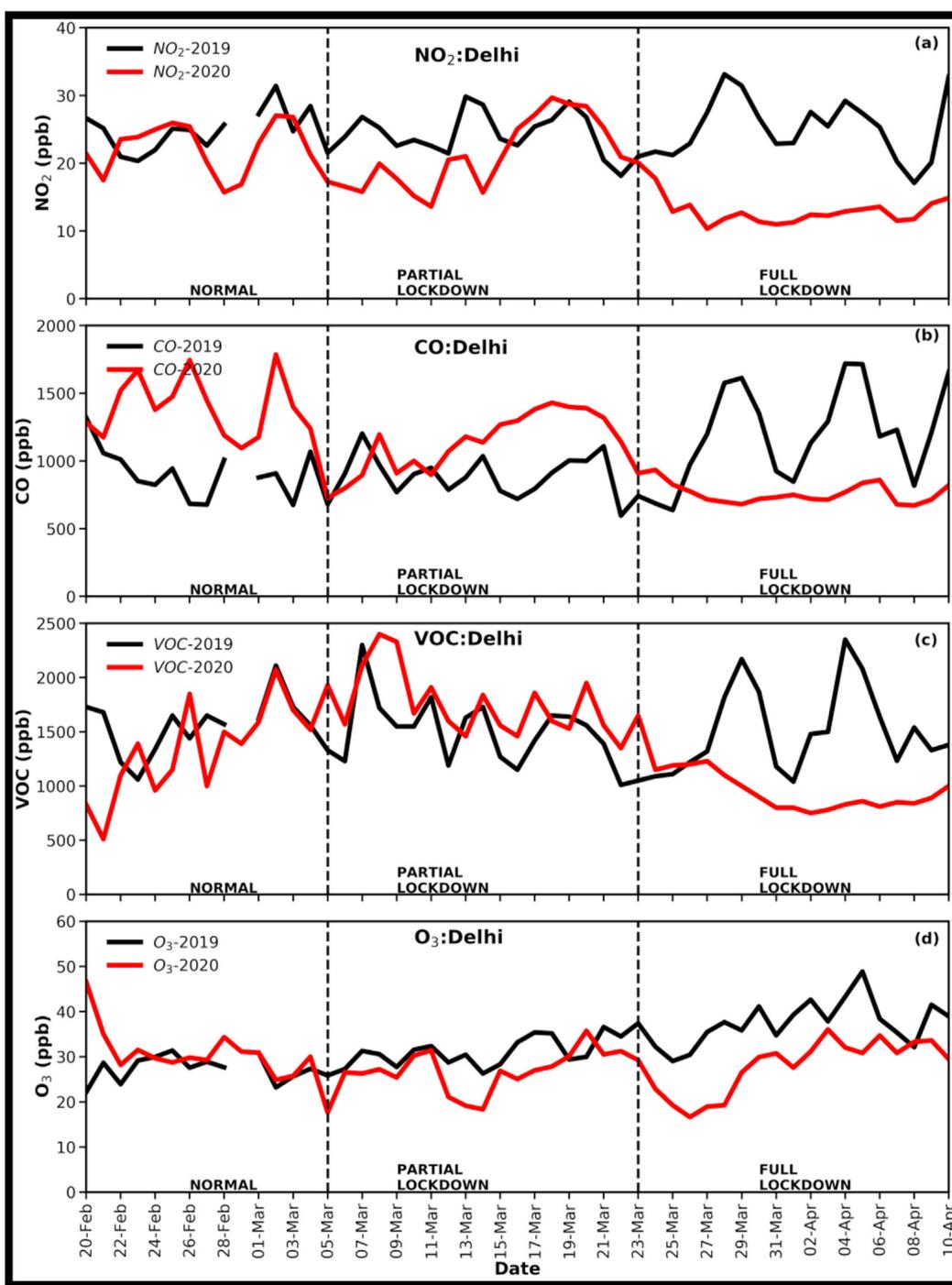


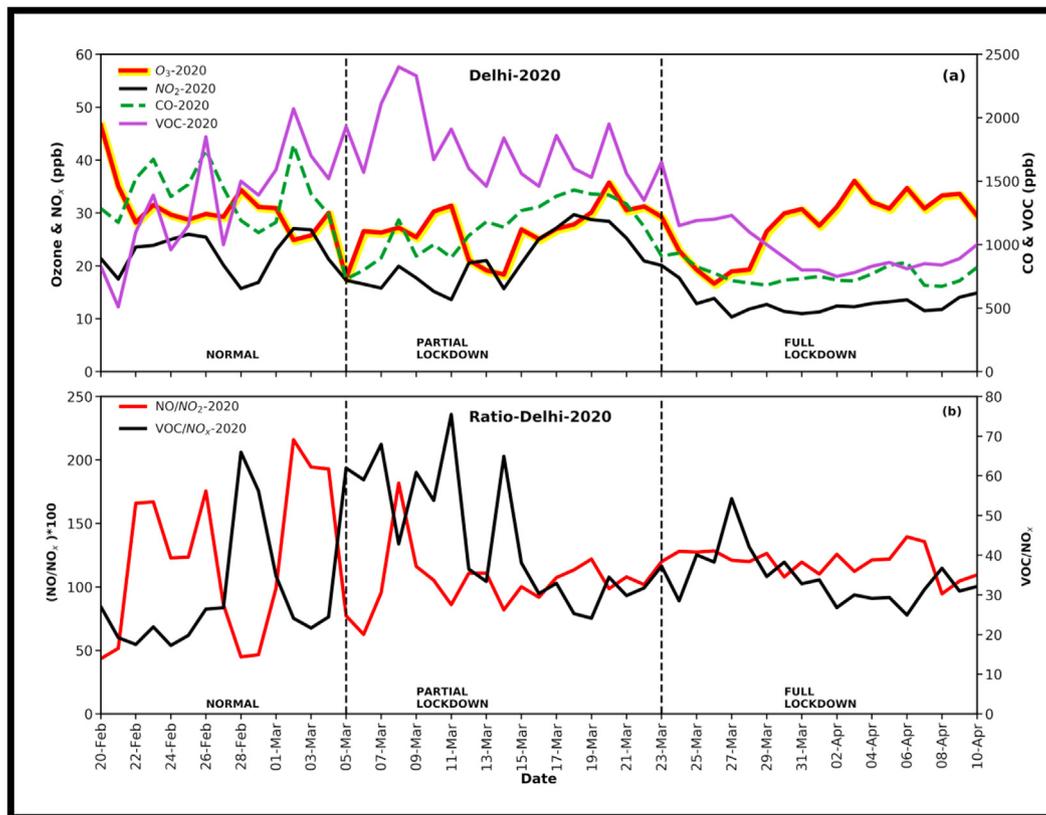
Figure 2. Time series of (a) NO₂, (b) CO, (c) VOCs and (d) O₃ for period 20th February to 10th April 2020 compared with identical period data of 2019.

Table 1. Average concentration of O₃, CO, VOC's and NO₂ during the full lockdown period (23 March to 10th April) of 2020 compared with identical period in 2019. The percentage changes in 2020 lockdown period as compared to 2019 levels are also given.

Pollutants	Full Lockdown Period (23 March – 10 th April)		Reduction in 2020 w.r.t 2019 (%)
	Volume Mixing Ratio (ppb)		
	2019	2020	
O ₃	37 ± 5	28 ± 4	-25 %
NO ₂	25 ± 5	13 ± 2	-50 %
CO	1210 ± 170	757 ± 110	-37 %
VOC	1520 ± 240	943 ± 150	-38 %

Table 2. Percentage change in O₃, NO₂, CO, and VOCs during Partial Lockdown and Full Lockdown as compared to normal period.

Percentage Change/Pollutant	O ₃	NO ₂	CO	VOC
After w.r.t Before (Full Lockdown)	-3 %	-41 %	-39 %	-39 %
Full lockdown w.r.t Partial Lockdown	3 %	-40 %	-34 %	-46 %
Full Lockdown w.r.t to Normal	-7 %	-42 %	-44 %	-31 %

**Figure 3.** Distribution of ground level O₃, NO₂, CO and VOC in Delhi for the 2020 during the period 20th February to 10th April, depicting period before and after COVID-19 lockdown in 2020. (b) shows the ratio (%) of NO by NO₂ and that of VOC/NO_x before and after COVID-19 lockdown scenario as marked.

relationship with VOC/NO_x and (NO/NO₂)*100 are indicated in Figure 3 (b) and explained in the next section.

3. Discussion

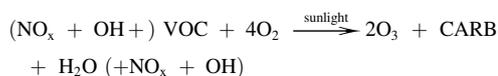
This section discusses the processes responsible for the anomalous variability in the volume mixing ratio of ozone during the lockdown period. Surface O₃ is formed through photochemical reactions involving NO_x (NO + NO₂), volatile organic compounds (VOCs), and CO in presence of solar radiation. Surface ozone formation initiates by the photolysis reaction of NO₂ in presence of sunlight to produce NO and atomic oxygen (O). This atomic oxygen reacts with oxygen molecule to form ozone (O₃) as shown in reaction R1 and R2. The O₃ produced rapidly reacts with NO in the atmosphere to form NO₂. Therefore, dissociation of O₃ takes place as per reaction R3. It implies that an increased (NO/NO₂)*100 ratio reduces the ozone concentration [26]. In a mega city like Delhi, NO is unusually quite high that increases (NO/NO₂)*100 ratio leading to significantly low value of surface ozone (Figure 3a). The effect of reaction R1 is the reverse of reaction R3. In the month of March–April under fair weather conditions temperature remains in the range of 13.5–24.5 °C the reaction R3 dominates:



The mechanism for the formation of ozone by reactions of various VOC with the OH radical is written below (R4). However, the presence of VOC and NO_x allows OH to be regenerated and promote formation of O₃ through NO₂ photolysis. NO₂ is photolyzed to generate atomic oxygen which then combines with oxygen to produce ozone (R5). The rate of ozone formation is controlled primarily by the rate of the initial reaction of VOC with OH. The following equation indicates the sequence of chemical reactions of VOC and NO_x in the formation of ozone [27].



Net:



Here, RO₂ represents any organics with an oxygen molecule attached and M is neutral number density acting as third body. CARB stands for carbonyl compounds, which play the role of hydrocarbons in further oxidation steps. OH and HO₂ are short-lived radicals which play an important role in the ozone formation process. Wang group [28] developed the O₃ attribution technique for O₃ source apportionment in regional chemical transport models to divide the entire range of VOC-NO_x-O₃ formation sensitivity to VOC-limited, transition, and NO_x-limited regimes based on the value of a regime indicator R which provides much needed insight into VOC-NO_x-O₃ mechanism. However all these work are based on modelling. Ozone production in Delhi under normal condition (before lockdown) follows the NO_x-saturated or VOC-limited [29] regime because of high NO_x level. As evident from Figure 3, that level of ozone and its precursors follow usual normal trend in the Normal scenario, which continued, into partial lockdown period as reaction R3 remained much stronger. However, immediately after the full lockdown (23rd March), the levels of NO₂, CO and HC decreased sharply. However, reaction R3 was still stronger which resulted in the fall of ozone amount from 31 ppb (23 March) to 16.5 ppb (26th March) as value of (NO/NO₂)*100 ratio (Figure 3b) steadily increased from 100 % to 130 % and NO₂ value touched 14 ppb. Thereafter, ozone started to increase because it entered in NO_x sensitive regime (11–14 ppb) when production of ozone by reaction R1 started to dominate as compared to losses through reaction R3 and the ratio of (NO/NO₂)*100 declined (Figure 3b). At this point, NO_x level reached saturation level remain constant at 11 ppb from 29th March onwards where production of ozone by R1 and loss of ozone by R3 balanced each other. At the same time, levels of CO and VOC also became saturated and constant. Hence, it can be stated that 11 ppb is the new threshold saturation level for NO₂ and 700 ppb for CO in Delhi. Thereafter, ozone levels continued to surge and became insensitive to NO_x. It appeared that a change over from NO_x sensitive to VOC-sensitive regime has taken place when ozone increased. However, that was not the case as Figure 2. VOC Figure 2(c) was found to decline contrary to our basic understanding of VOC sensitive regime. A study carried out in Chengdu, China to explore the ozone pollution also concluded that VOC sensitive regime is most dominating in major Chengdu stations and hence its control is important for ozone control [17]. A decrease in VOC/NO_x ratio (Figure 3(b)) of 55 from (27th March) to 30 (2nd April) was found to be associated with increasing ozone trend, which was a result of lowering of NO_x value rather than a high VOC level. Ratio of VOC/NO_x became stagnant at 30 from 3rd April onwards when ozone touched a peak level of 36 ppb and became stagnant between 31–36 ppb. Hence, the ozone variability is controlled by VOC/NO_x ratio rather than VOC alone. This suggests that during the last part of full lockdown, when levels of NO₂, CO and VOCs became almost constant at lower level, the basic characteristics of NO_x-sensitive and VOC-sensitive regimes is found to be displaced in controlling the ozone variability and a complex O₃-NO_x-VOCs chemistry indicated the stronger role of VOC/NO_x ratio and levels of CO. In this study, summer baseline level of VOC is found to be 900 ppb as per Figure 3(a). However, defining baseline level of ozone is complex and hence avoided in this work.

4. Conclusions

This study concludes that the concentrations of NO₂, CO and VOC declined by 35–50 % during COVID-19 induced lockdown in Delhi, a Mega-city in India. The variability in the Ozone levels is complex and is characterized by a decline then, an increase even after near negligible emissions of its precursors. This study provides observation-based insight on the processes leading to complex relationship of O₃-NO_x-VOC mechanism, which not necessarily follows the known NO_x-sensitive versus VOC-sensitive regimes but tends to suggest that generalizations about

these regime conditions are subject to exceptions. Full lockdown opportunity has given us a chance of discover markers of summer baseline level concentration that can be used in atmospheric chemistry transport modellers for better accounting the processes defining O₃-NO_x-VOC mechanism. The advantage of ozone as disinfectant and its harmful effect when directly exposed to human being should be understood in proper perspective.

Declarations

Author contribution statement

G. Beig, A. Rathod, S. Singh and S.K. Sahu: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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