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NOVEL CORONA VIRUS (COVID-19) IN ENVIRONMENTAL ENGINEERING PERSPECTIVE

Frequency distribution of pollutant concentrations over Indian megacities impacted by the COVID-19 lockdown

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Abstract

The megacities experience poor air quality frequently due to stronger anthropogenic emissions. India had one of the longest lockdowns in 2020 to curb the spread of COVID-19, leading to reductions in the emissions from anthropogenic activities. In this article, the frequency distributions of different pollutants have been analysed over two densely populated megacities: Delhi (28.70° N; 77.10° E) and Kolkata (22.57° N; 88.36° E). In Delhi, the percentage of days with $PM_{2.5}$ levels exceeding the National Ambient Air Quality Standards (NAAQS) between 25 March and 17 June dropped from 98% in 2019 to 61% in 2020. The lockdown phase 1 brought down the PM_{10} (particulate matter having an aerodynamic diameter ≤ 10 μm) levels below the daily NAAQS limit over Delhi and Kolkata. However, PM_{10} exceeded the limit of 100 μgm^{-3} during phases 2–5 of lockdown over Delhi due to lower temperature, weaker winds, increased relative humidity and commencement of limited traffic movement. The $PM_{2.5}$ levels exhibit a regressive trend in the highest range from the year 2019 to 2020 in Delhi. The daily mean value for $PM_{2.5}$ concentrations dropped from 85–90 μgm^{-3} to 40–45 μgm^{-3} bin, whereas the PM_{10} levels witnessed a reduction from 160–180 μgm^{-3} to 100–120 μgm^{-3} bin due to the lockdown. Kolkata also experienced a shift in the peak of PM_{10} distribution from 80–100 μgm^{-3} in 2019 to 20–40 μgm^{-3} during the lockdown. The $PM_{2.5}$ levels in peak frequency distribution were recorded in the 35–40 μgm^{-3} bin in 2019 which dropped to 15–20 μgm^{-3} in 2020. In line with particulate matter, other primary gaseous pollutants (NO_x, CO, SO_2, NH_3) also showed decline. However, changes in O_3 showed mixed trends with enhancements in some of the phases and reductions in other phases. In contrast to daily mean O_3, 8-h maximum O_3 showed a reduction over Delhi during lockdown phases except for phase 3. Interestingly, the time of daily maximum was observed to be delayed by ~ 2 h over Delhi (from 1300 to 1500 h) and ~ 1 h over Kolkata (from 1300 to 1400 h) almost coinciding with the time of maximum temperature, highlighting the role of meteorology versus precursors. Emission reductions weakened the chemical sink of O_3 leading to enhancement (120%; 11 ppbv) in night-time O_3 over Delhi during phases 1–3.

Keywords Frequency distribution · COVID-19 · Lockdown · NAAQS · Anthropogenic emissions · Megacities

Introduction

The air pollution levels are often observed to be in exceedance of the National Ambient Air Quality Standards (NAAQS) limits (Table ST1) over the megacities of India (Gurjar et al. 2016; Sen et al. 2017; Singh et al. 2021). Considering the adverse effects of air pollution on health and agricultural productivity in the region, poor air quality is of profound scientific and policy interest (Chowdhury et al. 2019; Ghude et al. 2016; Lelieveld et al. 2015; Sharma et al. 2019). Besides atmospheric dynamics and meteorology, the air pollution levels over Indian cities are also enhanced by strong local anthropogenic emissions and photochemistry (Ansari et al. 2016; Dhaka et al. 2020; Kumar et al. 2015;

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Ojha et al. 2012, 2020). Additionally, biomass-burning emissions, natural emissions and chemistry play important roles in photochemical ozone buildup (Kumar et al. 2016, 2018). Several studies have highlighted the complex interplay between meteorological conditions and aforementioned diverse emissions in affecting air quality over the Indian region (Dhaka et al. 2020; Ojha et al. 2020; Singh et al. 2020). However, it remains unclear how the reductions in regional emissions could help in improving the air quality in the Indian megacities. Studies based on chemical transport modelling have provided first-hand information; however, considerable biases are seen when model simulations are compared with in situ measurements over India (Chutia et al. 2019; Ojha et al. 2012, 2020; Sharma et al. 2017).

Stringent lockdowns to contain the spread of COVID-19 resulted in considerable reductions in the anthropogenic emissions allowing researchers to assess the impacts of local and regional emissions on the air quality over different environments. In this regard, numerous studies have recently evaluated the effects of lockdown on the air pollution by analysing satellite-based and ground-based observations globally as well as in the Indian region (Dhaka et al. 2020; Le et al. 2020; Singh & Chauhan 2020; Singh et al. 2020). A few studies also reported the declining trend in the air quality index (AQI) (Nigam et al. 2021; Srivastava et al. 2020) and reduction in aerosol optical depth over the Indo-Gangetic basin (Srivastava et al. 2021). These studies, along with several other efforts, established general reductions in the concentrations of primary air pollutants; however, enhancements in surface O_3 were observed in some urban and rural environments during the lockdown.

India experienced one of the most comprehensive lockdowns of the world which led to unprecedented reductions in the emissions from the traffic, industries, as well as from other sectors (Dhaka et al. 2020; Singh et al. 2020). Table 1 lists the different lockdown phases in India with the duration, restriction and relaxation guidelines during the different phases (Mondal et al. 2021). The phase 5 lockdown was lifted in a controlled manner unlike the earlier phases (phases 1–4). Analysis of satellite-based observations revealed that mean NO_2 levels over India were lowered by 17–18% as compared to the pre-lockdown period and 5-year average (Pathakoti et al. 2020). Analysis of ground-based measurements across the Indian region revealed sharp reductions by ~40–60% in $PM_{2.5}$ and PM_{10}; ~30–70% in NO_2 and ~20–40% in CO (Singh et al. 2020). Large reductions in the $PM_{2.5}$ levels (by 40–70%) during the first week of the lockdown were also reported from the Delhi region; however, some haze was observed due to interplay of baseline pollution and meteorological conditions (Dhaka et al. 2020). Mixed changes were observed in cases of O_3 and SO_2 over Indian environments (Dhaka et al. 2020; Girach et al. 2021; Singh et al. 2020; Soni et al. 2021). Nevertheless, it remains unclear how the frequency distributions of air pollutants were impacted by emission reductions during lockdown. The pollutant concentrations typically have skewed distributions, and the changes only in mean and median are insufficient to assess the change in air quality; therefore, the analyses of frequency distributions are very useful (Miles et al. 1991; Sarangi et al. 2014). Additionally, such an analysis would provide a quantitative picture of the likely range of concentrations in a possible mitigation scenario, such as that during the COVID-19 lockdown. A particularly important part of the distribution is the higher side of the pollution concentrations which are extremely sensitive to the emissions and stagnant meteorological conditions. A frequency distribution can also be used for the prediction of pollutant levels using Lognormal, Weibull and type V Pearson distributions. Lu (2003) concluded in a study that the lognormal distribution can closely predict the particulate matter concentrations as compared to type V Pearson and Weibull distributions which over-estimated and under-estimated the actual values respectively. Further, concentrations of air pollutants and the frequency distributions are important factors in assessments

Table 1 Lockdown phases, duration and imposed restrictions/relaxations over India

Lockdown phases	From	To	Duration	Restrictions and relaxations
Phase 1	25th March 2020	14th April 2020	21 days	Complete lockdown with relaxations to healthcare workers and daily need item suppliers only
Phase 2	15th April 2020	3rd May 2020	19 days	Complete lockdown and relaxations limited to 'green zones' (where there were no reported cases of CoVID-19) only
Phase 3	4th May 2020	17th May 2020	14 days	Complete lockdown and relaxations to green zones only. The transport buses were allowed to operate with 50% capacity in green zones only
Phase 4	18th May 2020	31st May 2020	14 days	Lockdown for red zones only (where there were active CoVID-19 cases) and relaxations given to green zones only. Indian railways and airlines resumed operations with low capacity
Phase 5 (a.k.a. Unlock 1)	1st June 2020	30th June 2020	30 days	Lockdown restrictions limited to 'containment zones' (with high number of active CoVID-19 cases). For all other zones, restrictions removed except large public gathering and opening of theatres, gyms, hotels, swimming pools, parks, bars, auditorium etc

of human health risks (Saltzman 1997). This is of paramount significance for potential policy-making for mitigating pollution episodes over the Indian megacities. In this direction, the present study is aimed to analyse the frequency distributions of key air pollutants over two Indian megacities.

Data and methodology

This study analyses the ground-based observations of the ambient air quality over a network of monitoring stations in two megacities of India: Delhi (28.6° N; 77.2° E, 200–250 m amsl) and Kolkata (22.6° N; 88.4° E, ~ 9 m amsl). Delhi—the national capital of India and Kolkata—the cultural capital of India have about 16 and 14 million population, respectively (Census of India, 2011; https://censusindia.gov.in/2011). These megacities experience poor air quality frequently exceeding the NAAQS limits of various pollutants (Firdaus and Ahmad 2011; Gupta et al. 2007; Molina and Molina 2004). The observations were taken from 38 stations for Delhi and 2 stations for Kolkata (as described in the supplementary Table ST2) operated and maintained by Central Pollution Control Board (CPCB) along with Delhi Pollution Control Committee (DPCC), West Bengal Pollution Control Board (WBPCB) and India Meteorological Department (IMD). Two years (2019 and 2020) of observations on daily basis were analysed to estimate the impact of lockdown on the levels and frequency distribution of air pollutants. The stations which had observations of all the species—$PM_{2.5}$, PM_{10}, NO_2, SO_2, O_3, CO and NH_3 between 25 March and 17 June for both 2019 and 2020 (lockdown period) were considered for the analyses. There are 7 monitoring stations installed throughout Kolkata out of which just 2 stations recorded all the parameters ($PM_{2.5}$, PM_{10}, NO_2, SO_2, O_3, CO and NH_3) for both the years 2019 and 2020. We suggest that more denser monitoring network for air quality parameter measurements are required, especially in Kolkata, which can provide further insights in the air quality data. Daily mean of particulate matter ($PM_{2.5}$ and PM_{10}) and pollutant gas (NO_2, SO_2 and NH_3) concentrations were used in the analyses. In addition to daily average, 1-h and 8-h averages are also used in cases of O3 and CO since considering the NAAQS limits. Daily data were screened for values beyond 3-sigma standard deviations for removal of spikes/abnormal values prior the analyses.

Results and discussions

The effects of lockdown on air quality are investigated by comparing the ground-based observations of $PM_{2.5}$, PM_{10}, O_3, CO, NO_2, SO_2 and NH_3 over the Indian megacities during the lockdown and the same period of the year 2019. Besides the changes in mean concentrations given in the Table 2, here the emphasis is on the frequency distributions of pollution concentrations to provide a reference for potential future policies to improve regional air quality. The frequency distribution of the pollutant's concentrations provides the statistical characteristics of the air quality variations (Seinfeld and Pandis 1998). In addition to unravel the spread in the pollution levels and most frequent ranges of variation, the skewness indicates on the source characteristics (Seinfeld and Pandis 1998). The frequency distributions of the pollution concentrations exhibit large variations and also depend on meteorological conditions, besides emissions (Lu and Fang 2002).

$PM_{2.5}$ and PM_{10}

The NAAQS safe limit for $PM_{2.5}$ annual and daily average concentrations is 40 μgm^{-3} and 60 μgm^{-3} respectively. The average $PM_{2.5}$ and PM_{10} concentrations over Delhi were found to be 82.2 ± 27.7 μgm^{-3} and 236.5 ± 75.2 μgm^{-3} respectively during the year 2019 (25th March–30th June), which dropped significantly to 50.5 ± 6.9 μgm^{-3} and 120.05 ± 26.2 μgm^{-3} due to lockdown in 2020 (Table 2). The reduction in $PM_{2.5}$ concentrations by 11.6–53.8 μgm^{-3} during the 1st phase of lockdown brought it below NAAQS limit over Delhi (Figure S2). The strong shift in the frequency distribution towards the lower range of the $PM_{2.5}$ concentrations is clearly visible (Fig. 1b). In Delhi, 77% of the total days experienced $PM_{2.5}$ levels above the NAAQS during 25 March to 17 June 2019 which dropped to 21% in the year 2020 due to the lockdown. However, following relaxations towards the phases 4–5 of lockdown $PM_{2.5}$ again showed enhancements (Fig. 1a). Kolkata also experienced drop of

Table 2 Mean concentration levels of pollutants over the Indian megacities during lockdown period (25 March–17 June) of 2020 and same period in the preceding year. The mean values are calculated from daily averages of pollutants concentration

Megacity	Year	$PM_{2.5}$ (μgm^{-3})	PM_{10} (μgm^{-3})	NO_2 (μgm^{-3})	O_3 (μgm^{-3})	SO_2 (μgm^{-3})	CO (mgm^{-3})	NH_3 (μgm^{-3})
Delhi	2019	82.22 ± 27.70	236.57 ± 75.26	48.15 ± 10.98	48.54 ± 8.30	19.28 ± 4.76	1.33 ± 0.28	33.79 ± 5.74
	2020	50.57 ± 6.97	120.05 ± 26.25	23.70 ± 4.56	49.58 ± 9.66	14.71 ± 1.46	0.85 ± 0.17	30.23 ± 2.87
Kolkata	2019	42.29 ± 14.77	88.38 ± 33.18	24.72 ± 15.34	29.23 ± 9.09	4.27 ± 3.77	0.503 ± 0.09	13.32 ± 8.86
	2020	19.87 ± 10.06	41.76 ± 13.27	9.37 ± 2.34	44.07 ± 13.07	6.96 ± 1.58	0.505 ± 0.18	23.57 ± 4.92

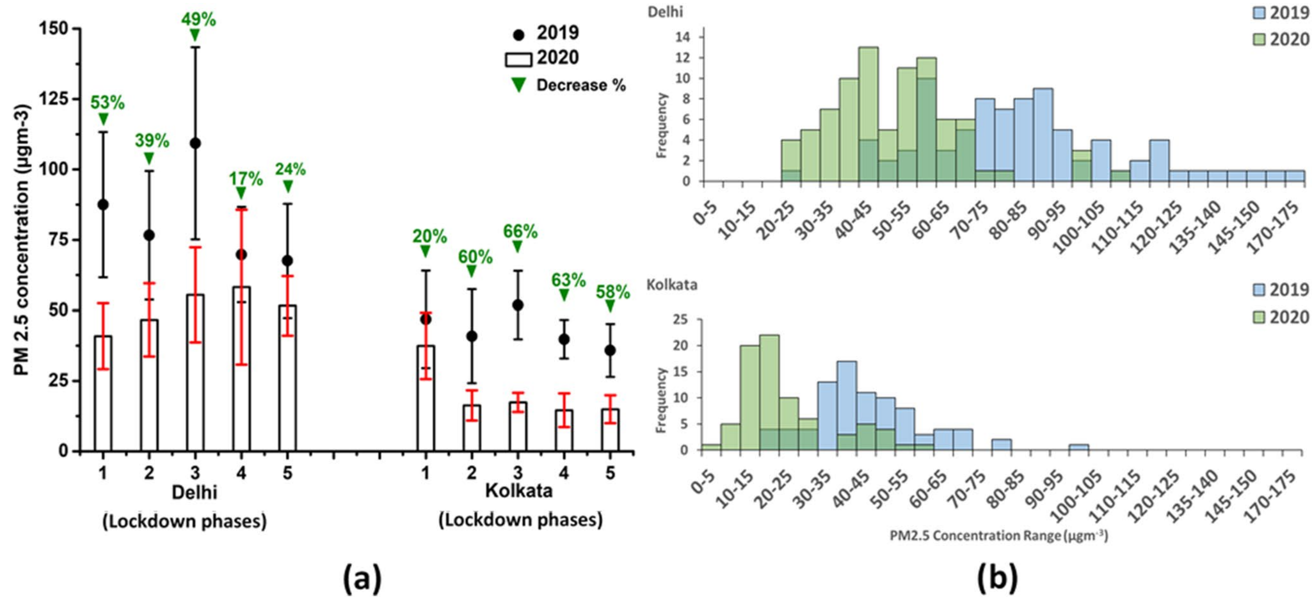

Fig. 1 (**a**) Change in daily $PM_{2.5}$ concentrations; (**b**) frequency distribution of $PM_{2.5}$ concentrations during the lockdown phases of 2020 and the same period of 2019

about 13% of the days with higher levels of ambient $PM_{2.5}$ during 2020 than the preceding year. The mean concentration levels of $PM_{2.5}$ in Kolkata during 2019 dropped from 42.2 ± 14.7 μgm^{-3} to 19.8 ± 10.06 μgm^{-3}, while the PM_{10} level dropped from 88.3 ± 33.1 μgm^{-3} to 41.7 ± 13.2 μgm^{-3} due to the lockdown.

PM_{10} levels over Delhi were mostly above the daily NAAQS limit (100 μgm^{-3}) during 2019 (Figure S1). The reduction of PM_{10} to 120.1 ± 26.3 μgm^{-3} concerning 2019 levels (236.2 ± 39.8 μgm^{-3}) reveals strong decline by about 50%. The lockdown phase 1 brought down the PM_{10} levels below the daily NAAQS limit over Delhi. However, PM_{10} increased again exceeding 100 μgm^{-3} during the phases 2–5 of lockdown over Delhi (Fig. 2a). The increase in particulate matter loading during the lockdown phase 2 in Delhi could have resulted due to sudden change in weather plus

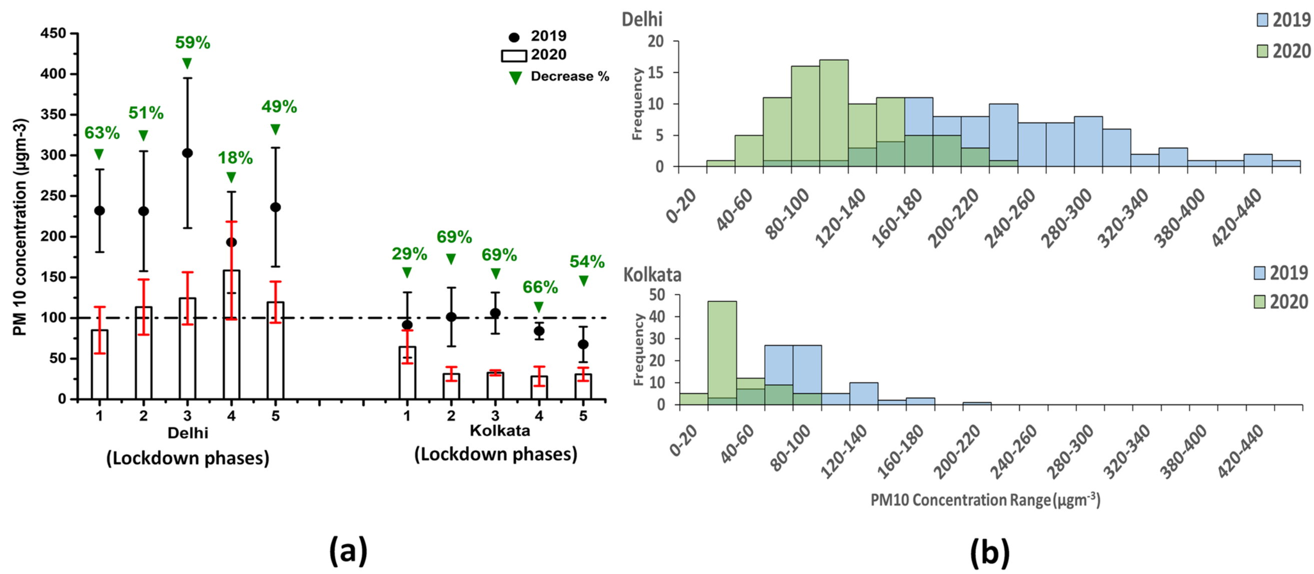

Fig. 2 (**a**) Change in daily PM_{10} concentrations; (**b**) frequency distribution of PM_{10} concentrations during lockdown phases of 2020 and the same period of 2019

gusty winds (18th April 2020) in the northern India region. Relaxation allowing some market places and the transport from phase 3 (from 4th May, 2020) onwards in a controlled manner also contributed to the observed recovery in the pollution loading. The reductions were estimated in the range of 33.7–177.1 μgm^{-3} (18–59%) during other phases. The reduction over Kolkata were smaller (26.2 μgm^{-3}) during phase 1, whereas, larger reductions by 36.7–67 μgm^{-3} (52–66%) were observed during phases 2–5. In contrast with Delhi, lower PM_{10} concentrations during phase 4 over Kolkata were due to state-level restrictions on vehicular movements. Delhi and Kolkata recorded about 98% and 24.7% of the total days (lockdown period) with pollution exceeding 100 μgm^{-3} during 2019 which dropped to 61% and 0%, respectively due to lockdown in 2020. The frequency distribution for Delhi (Fig. 2b) shows that the number of days with pollution levels above the NAAQS safe limit are less and PM10 peak shifted to lower range (20–240 μgm^{-3}) during entire lockdown period as compared to the peak variations over 20–460 μgm^{-3} during 2019. PM_{10} variations over Kolkata were also distributed over a broader range 0–220 μgm^{-3} during 2019, whereas, these were squeezed to lower range (0–100 μgm^{-3}) during the lockdown period.

O_3, CO, NOx

Figure 3a shows that the daily mean O_3 levels were decreased by 12% and 15% during the phases 1 and 5 respectively as compared to the preceding year, whereas an increase by 15.2 μgm^{-3} (33%) was seen at surface O_3 levels during the phase 3. Few studies have reported a decrease in O_3 over Delhi during the initial stage (phase 1) of lockdown earlier (Sathe et al. 2021; Saxena and Raj 2021). However, Kolkata witnessed a significant rise in daily O_3 concentrations during phase 1 with 28 μgm^{-3} (91%), while it fluctuated between 3.8 and11.3 μgm^{-3} (11–42%) during phases 2–5. The 8-h maximum O_3 remained just below the NAAQS limit (100 μgm^{-3}) during 2019 over Delhi (Figure S3). The highest 8-h O_3 levels were lower during the lockdown as compared to 2019. As shown in Fig. 3b, O3 shows reduction by 28 μgm^{-3} (32%) over Delhi during phase 1 with respect to the levels during 2019. However, the surface O_3 levels were seen to be within the variabilities (standard deviation) during subsequent phases. O_3 levels over Kolkata showed an enhancement by 49.2 μgm^{-3} (83%) during phase 1 with respect to the levels during 2019. This highlights the nonlinear ozone chemistry over these regions. Large reduction in NO_2 (discussed subsequently), a major O_3 precursor, could have caused O_3 reduction over Delhi in phase 1. However, enhancement or reduction depends upon changes in NO_2 with respect to NO as well as Volatile Organic Compounds (VOCs). The NO_2 reduction was relatively less over Kolkata and the changes in NO as well as VOCs would have contributed to O_3 enhancement during phase 1. O_3 levels remained comparable during phases 2–5 concerning the values during 2019 over Delhi as well as Kolkata. The daily mean O_3 over Delhi and Kolkata were consistently below 80 μgm^{-3} during 2019 as well as 2020. Figure 3c shows the frequency distribution of 8-h maximum O_3 during entire lockdown period with many occasions of O_3 exceeding the NAAQS limits. While the broad O_3 distribution shifted to lower values over Delhi, it has squeezed with greater frequencies over 40–60 μgm^{-3} over Kolkata.

The frequency distribution of 1-h maximum O_3 (Fig. 4) shows skewed distribution. The maximum 1-h O_3 values were reported for 13:00–14:00 for the year 2019 which got shifted by 2 h during the pandemic. Time of maximum O_3 depends upon the availability of precursor gases, solar radiation, ambient temperature and evolution of boundary layer. With reduction in precursor concentrations, influences of temperature (38.7 ℃; 32–48 ℃) maximum in the afternoon hours (14–16 h) could have contributed to O_3 maximum around 15 h during the lockdown. The shift in peak O_3 time is about 2 h over Delhi. However, such effect was less pronounced over Kolkata which experienced daily maximum O_3 most frequently at 14 h as compared to 13 h seen during 2019. Interestingly, there are higher number of occurrences of daily maximum at night-time over Kolkata. The 8-h maximum O_3 frequency distribution is shown in Figure S4.

NO_2 levels over Delhi and Kolkata were below daily NAAQS limits (80 μgm^{-3}) during lockdown period as well as the same period of 2019 (Figure S5). Large reductions by 35–58% (14.6–29.9 μgm^{-3}) were observed over Delhi (Fig. 5a) with a shift in the peak of frequency distribution to 15–20 μgm^{-3} bin from 40–45 μgm^{-3} in 2019 (Fig. 5b). The NO_2 reductions over Delhi during phases 1–3 are seen to be beyond 2-sigma (standard deviation), which are not observed for other pollutants in both the locations. The reduction was about 50% over Kolkata during phases 1–2, whereas during the subsequent phases NO_2 levels were within the variabilities. Also, there was a slight shift in frequency distribution, and the peak frequency was observed over the 10–15 μgm^{-3} bin.

Night-time O_3 (averaged over 2100–0500 h local time) shows a prominent enhancement (by ~ 11 ppbv or 124%) over Delhi (Fig. 6a) coinciding with the reduction in NO by 32 ppbv (90%; Fig. 6b). This shows that the night-time O_3 titration by NO has been weakened, whereas, the noontime O_3 buildup has sustained resulting into higher night-time O_3. Typically, night-time O_3 levels are low in such urban environments (~ 9 ppbv; Fig. 6a). These changes suggest profound impacts of emission reductions on the urban air chemistry, which are also anticipated to affect particle-phase processes. Similar O_3 enhancement is reported over cities in Europe (17%) and Wuhan, China (36%) recently (Sicard et al. 2020).

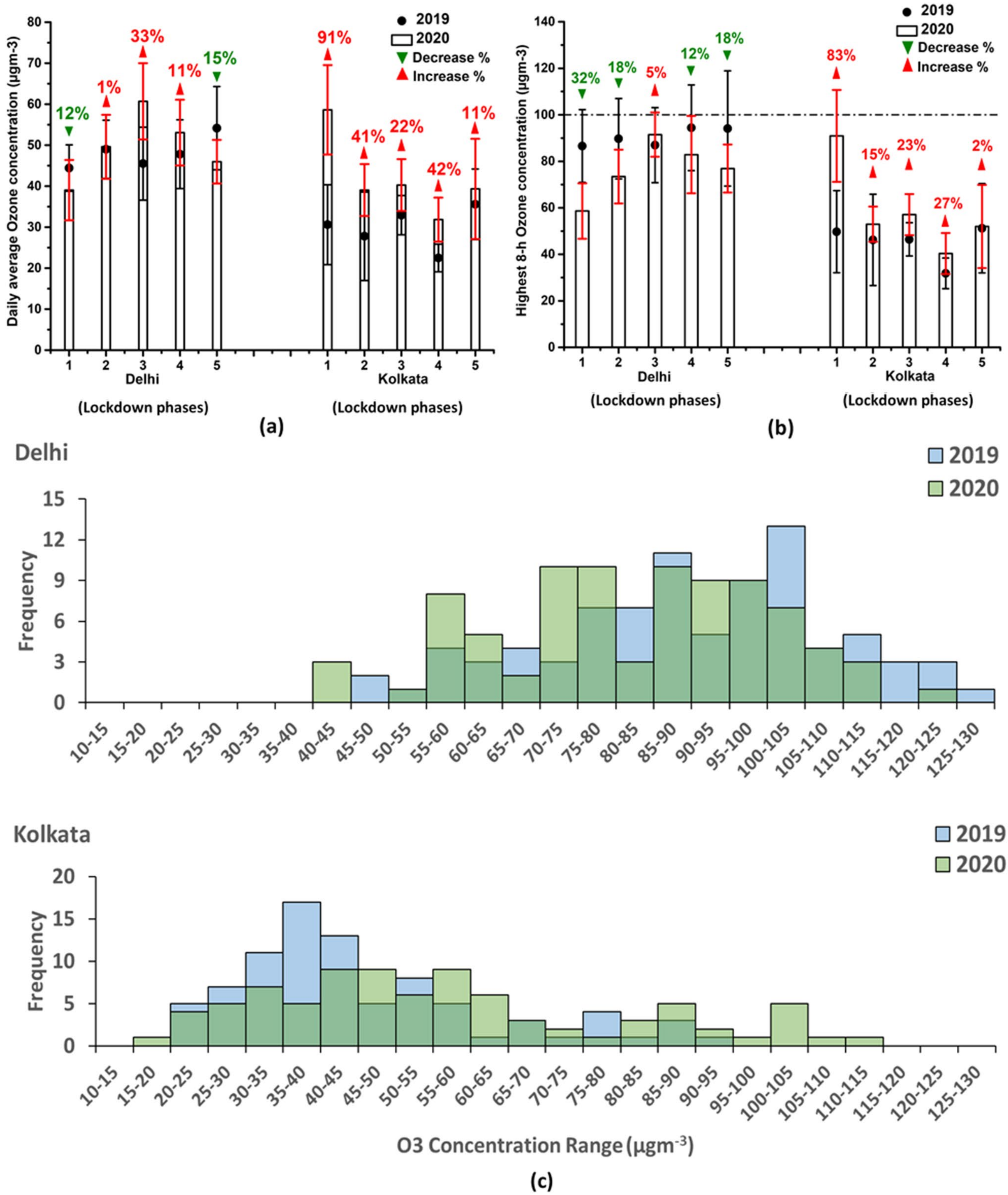

Fig. 3 Change in (a) daily O_3; (b) maximum 8-h O_3; (c) frequency distribution of daily O_3 during lockdown phases of 2020 and the same period of 2019

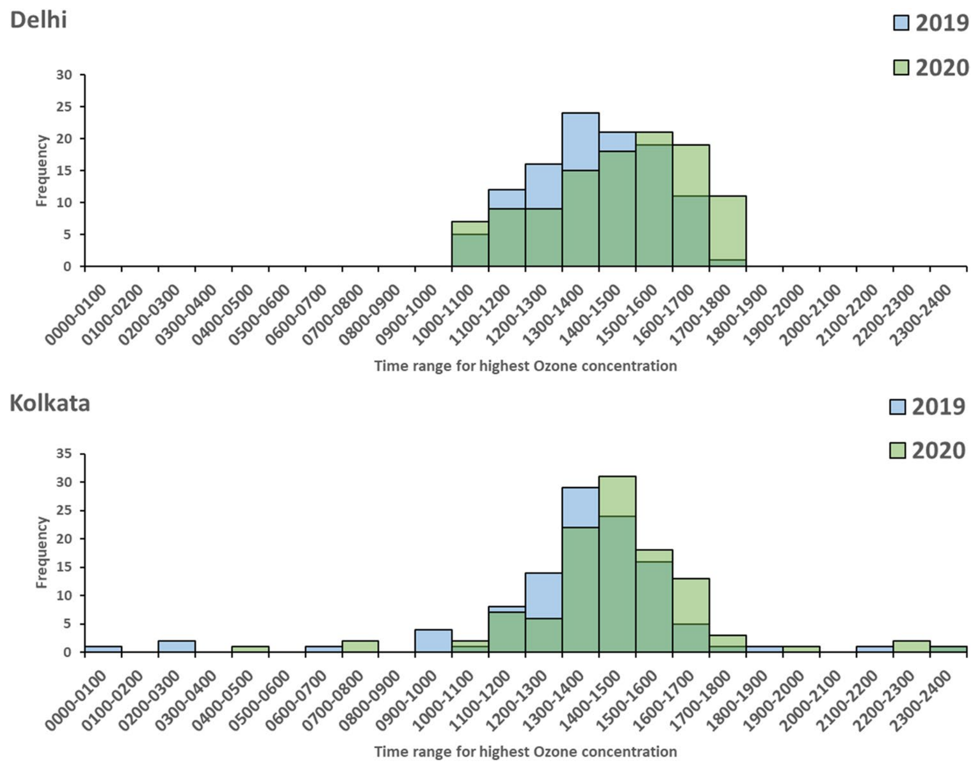

Fig. 4 Frequency distribution of 1-h maximum O_3 concentration during lockdown phases of 2020 and the same period of 2019

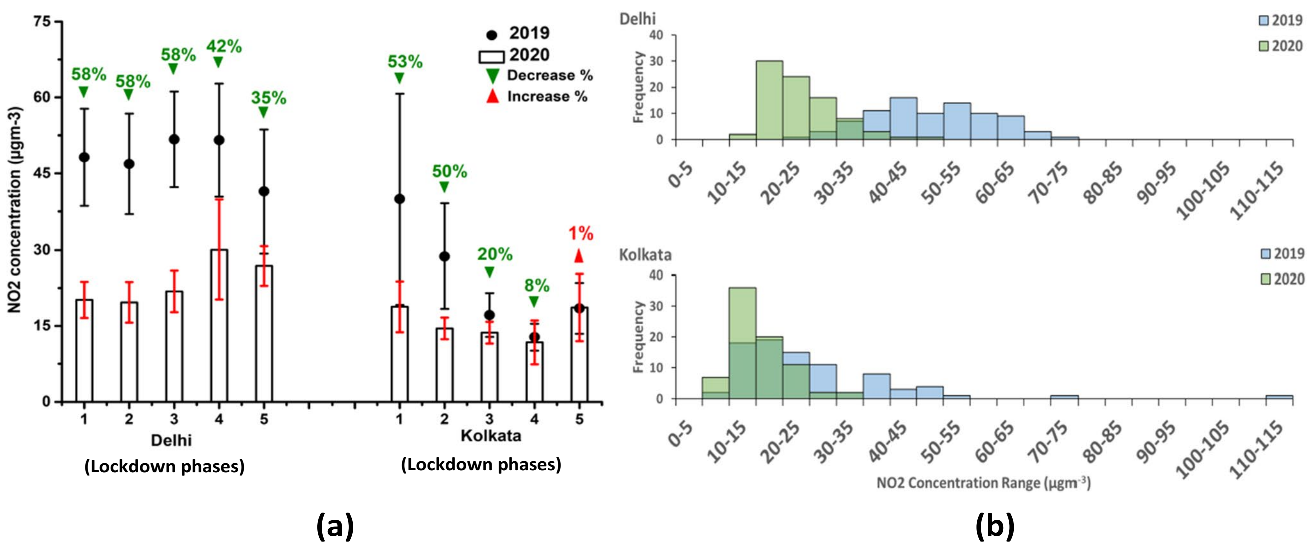

Fig. 5 (**a**) Change in daily NO_2 concentrations; (**b**) frequency distribution of NO_2 concentrations during lockdown phases of 2020 and the same period of 2019

SO_2 and NH_3

Similar to NO_2, SO_2 remained within the daily NAAQS limit during lockdown and the same period of 2019 (Figure S6). The SO_2 levels were 14.4–21.5 µgm^{-3} during 2019 over Delhi which reduced to 13.6–17 µgm^{-3}. The SO_2 levels in Kolkata were enhanced to 13.6–17 µgm^{-3} during the lockdown from 5.1–12.5 µgm^{-3} during 2019. The increase in SO_2 levels observed in the Kolkata region is not clear, but speculation could be a slower reduction in the local activities towards the lockdown (Chakraborty et al. 2021). SO_2 reduction by 7.9 µgm^{-3} (37%) was larger during phase 1 over Delhi (Fig. 7a), and reductions were lower (6–30%) during other phases and within typical variabilities. Peak

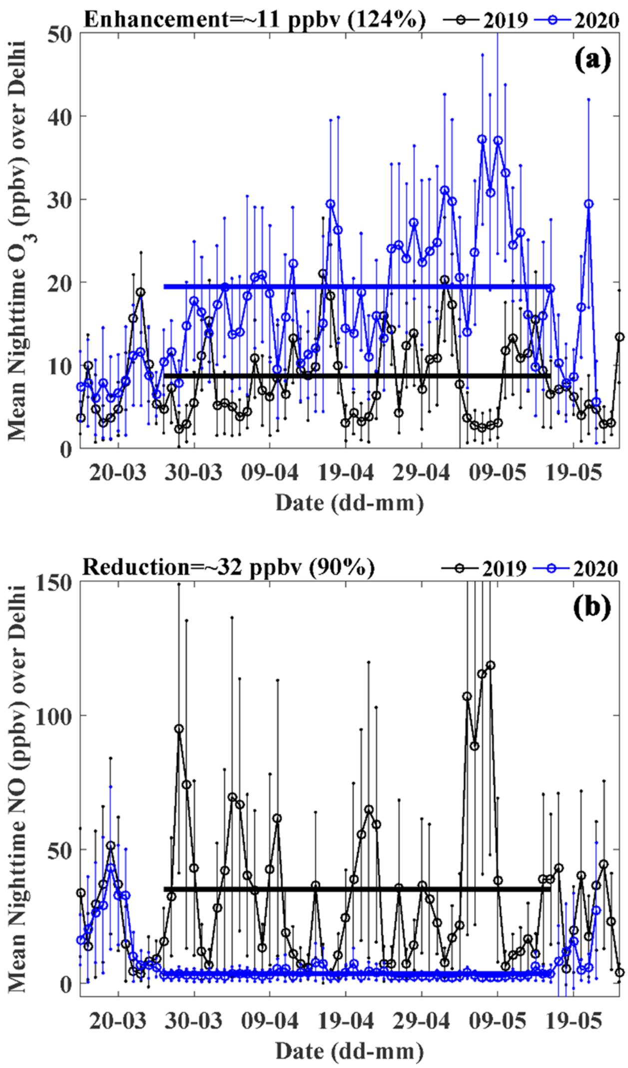

Fig. 6 Night-time surface O_3 and NO variations over Delhi during March–May 2019 (black) and 2020 (blue) covering the lockdown period. Error bars represent standard deviations corresponding to spatial variability over Delhi. Horizontal lines represent the mean

of the frequency distribution shifted to 12–15 μgm^{-3} during the lockdown from 20–22.5 μgm^{-3} in the preceding year (Fig. 7b). As expected, the frequency distribution was shifted to lower range and became skewed for Delhi. SO_2 levels remained well within variability (1-sigma standard deviation) over Kolkata during the lockdown phases except during phases 1 and 3.

The mean concentrations of CO were 0.9 mg m^{-3} (0.7–1.0 mg m^{-3}) and 0.5 mg m^{-3} (0.4–0.7 mg m^{-3}) over Delhi and Kolkata, respectively, during lockdown phases (Figure S7). The reduction was estimated to be in the range of 0.3–0.6 mg m^{-3} (23–49%) during lockdown phases over Delhi (Fig. 8a), with shift in peak of distribution to lower range (0.8–1 mg m^{-3}) (Fig. 8b). The reduction was beyond 1-sigma standard deviation during phases 1–3 over Delhi. Similar to the case of SO_2, CO concentrations showed insignificant changes over Kolkata within the variabilities.

NH_3 also showed a significant reduction by 25% over Delhi during phase 1, whereas it remained within the variabilities during other phases (Fig. 9a). This is reflected in a marginal shift towards a lower range of values in the frequency distribution (Fig. 9b). In contrast to Delhi, NH_3 showed some enhancement during phases 3–5 over Kolkata (by 12.3–18.7 μgm^{-3}). The increase in NH_3 levels could be due to various factors such as increase in emission from dumping grounds and cattle rearing in the surrounding region (Gupta et al. 2008). Cyclone 'Amphan' in Kolkata during the lockdown period brought marine airmass that could have higher background levels due to the chemistry of nitrogen-containing compounds and excretion of zooplanktons near the sea surface (Quinn et al. 1996). Increase in vitalisation (ammonium to ammonia) under moist conditions due to cyclone-related rain could also have also a contribution. The peak of frequency distribution shifted towards 20–25 μgm^{-3} during lockdown period from 5–10 μgm^{-3} in Kolkata (Fig. 9b). Nevertheless, the NH_3 levels over these megacities remained below the NAAQS limits during 2020 as well as 2019 (Figure S8).

Comparison with NAAQS standard and implications

Delhi witnessed 98% of its days (25th March–17th June) with PM_{10} concentration exceeding the safe limits in 2019, which dropped significantly to 61% in 2020 during the lockdown (Fig. 10). Kolkata had observed 25% of the days exceeding the NAAQS limits for PM_{10} in 2019, but no days were observed with concentrations in exceedance of NAAQS during lockdown. The number of days with $PM_{2.5}$ levels higher than the prescribed safe limits fell from 77% in 2019 to 21% in 2020 over Delhi, whereas Kolkata did not experience exceedances during 2020 as compared to 13% in the preceding year. Concentration of NO_2 and SO_2 levels during the years 2019 and 2020 was seen to be within the safe limits. The ambient NH_3 levels also did not exceed the safe limits during both the years in both the megacities. Delhi witnessed 32% of its days which recorded higher 8-h CO levels in ambient air during 2019, but such instances were not observed during the lockdown. The 8-h ozone levels higher than the NAAQS safe limits were recorded in 34% of total days of study period during the year 2019 which dropped to 18% due to lockdown. Meanwhile Kolkata experienced rise in by 8% in number of days exceeding safe limits of surface ozone as compared to 2019.

Summary and conclusions

To limit the spread of COVID-19 infections, the Government of India imposed lockdown during 25 March–31 May 2020 in a phased manner, which caused cessation in

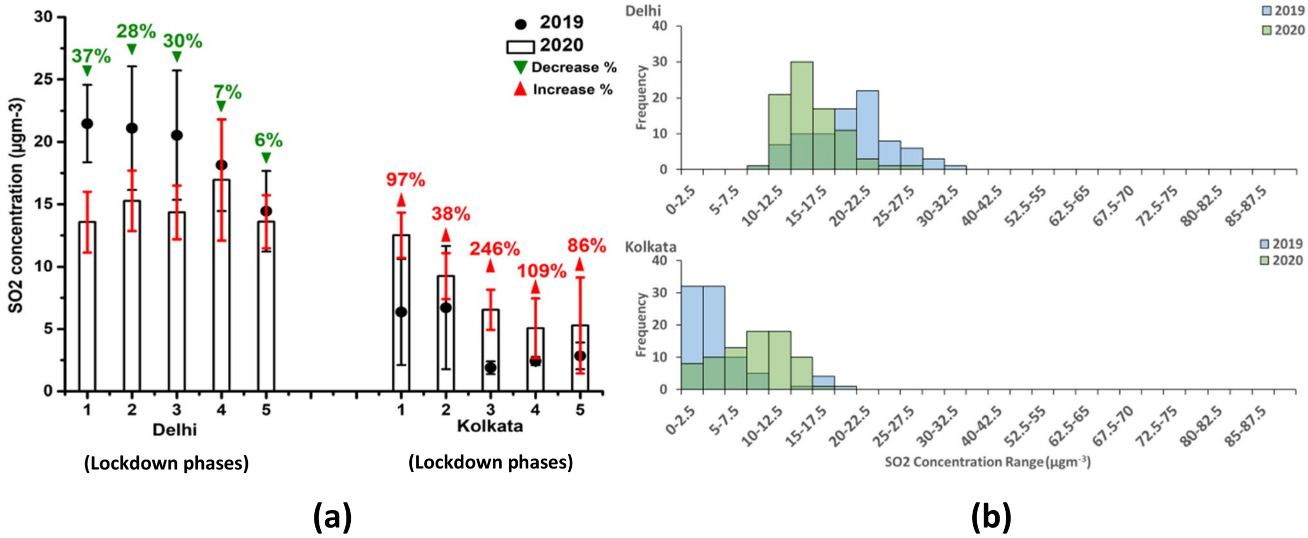

Fig. 7 (**a**) Change in daily SO_2 concentrations; (**b**) frequency distribution of SO_2 concentrations during lockdown phases of 2020 and the same period of 2019

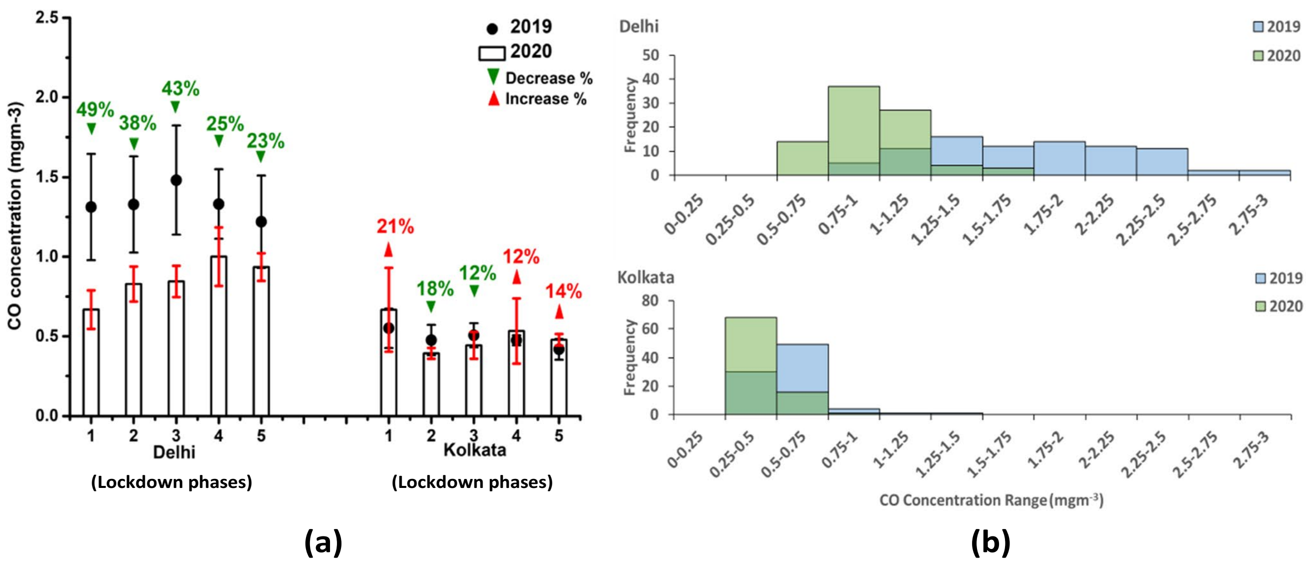

Fig. 8 (**a**) Change in daily SO_2 concentrations; (**b**) frequency distribution of SO_2 concentrations during lockdown phases of 2020 and the same period of 2019

anthropogenic activities. The strict actions taken to curtail the traffic movement and activities in the industrials sectors drastically reduced the pollution levels resulting in air quality levels under the safe limits over Delhi and Kolkata. Many studies have reported decline in pollution levels in Indian cities as a result of the lockdown (Bera et al. 2021; Jain and Sharma 2020; Kumari et al. 2020).The air quality during the lockdown in the Indian megacities exhibited significant reductions in NO_2 (Gautam et al., 2020) and particulate matter concentrations while the daily mean O_3 concentration showed some enhancements due to non-linear photochemistry, lower concentration of O_3 titrating trace gases (e.g. nitric oxide) and more intense solar radiation (Bedi et al. 2020; Mor et al. 2021; Sharma et al. 2020; Singh et al. 2020). The key findings of the present study are as follows:

- $PM_{2.5}$ levels were reduced by 17–66% over the Delhi and Kolkata. The number of NAAQS exceedance days for $PM_{2.5}$ were 65 and 11 days over Delhi and Kolkata in 2019 which were reduced to 18 and *zero* days respectively due to the lockdown.

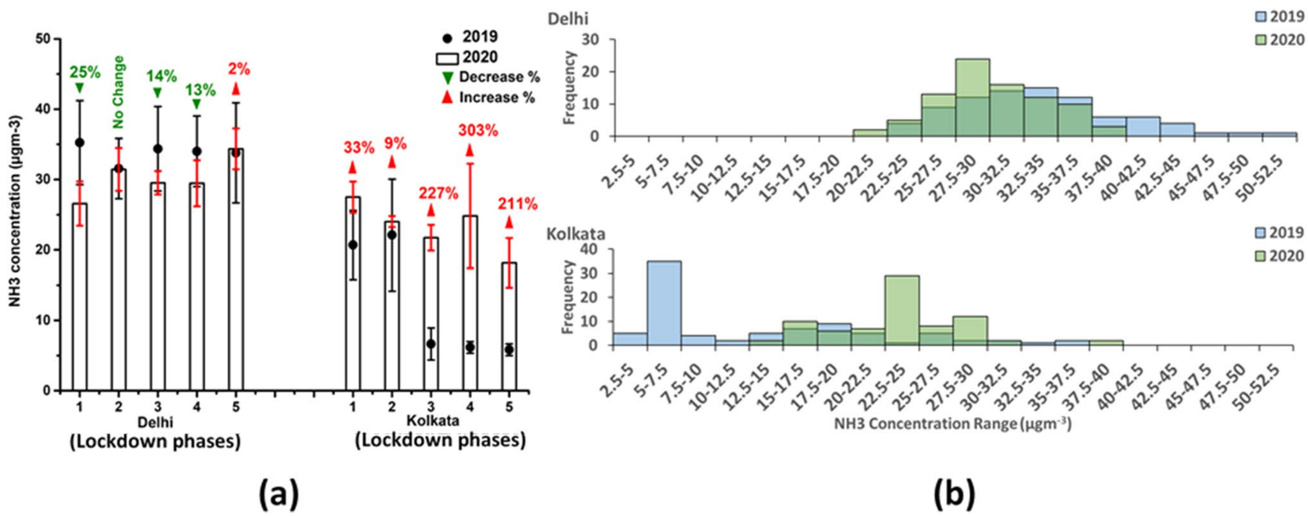

Fig. 9 (**a**) Change in NH_3 concentrations; (**b**) frequency distribution of NH_3 concentrations during lockdown phases of 2020 and the same period of 2019

- PM_{10} concentration levels reduced in the range of 18–69% over Delhi and Kolkata combined. The number of days with PM_{10} above the NAAQS safe limits reduced from 83 to 52 days in Delhi and from 21 days to *zero* days in Kolkata due to the lockdown.
- 1-h maximum O_3 peak in Delhi, which was seen at 13:00–14:00 h during 2019, was delayed by ~ 2 h during lockdown in 2020. Whereas, the O_3 peak in Kolkata shifted by 1 h to 14:00–15:00. This shift in O_3 peak is attributed to interplay between meteorology and reduced levels of O_3 precursors.
- Besides the large reductions in mean concentrations, the peak of frequency distribution for all trace gases was shifted to lower range of concentrations (NO_2: 10–15 µgm^{-3}; CO: 0.25–0.50 µgm^{-3}), except for SO_2 and NH_3 in Kolkata.
- In general, O_3 showed higher levels as compared to 2019 over both Delhi and Kolkata. However, there was some decrease in maximum 8-h and daily mean O_3 over Delhi during phases 1 and 5. A prominent night-time enhancement in ozone (120%; 11 ppbv) over Delhi was due to weaker chemical sink of O_3 during phases 1–3.

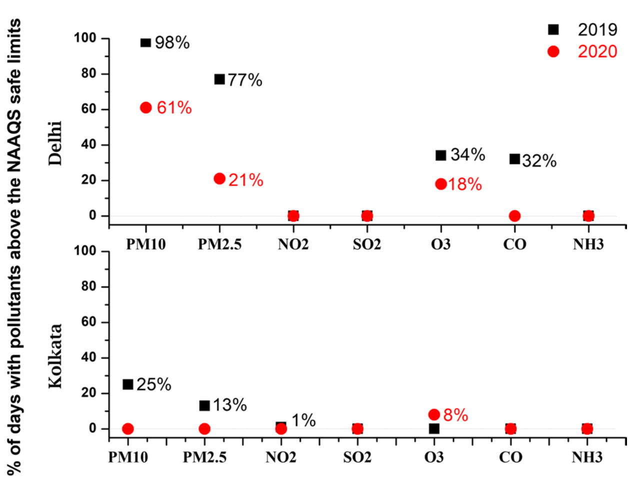

Fig. 10 Percentage of days with pollutant levels exceeding the NAAQS safe limits for the lockdown phases of 2020 and the same period of 2019. There are no exceedances in NAAQS levels except for O_3 during lockdown period over Kolkata

The quantitative analyses on the changes in air quality with varying strength of emissions during COVID-19 lockdown provide valuable insights for designing mitigation strategies also in the normal conditions. Air quality variations and slight ozone enhancements in some phases of the lockdown unravel air chemistry in background conditions over this part of the world. The study based on actual conditions performed here would help evaluating modelling studies and emission inventories to explore the mitigation pathways.

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Author contribution AM had conducted the data analyses and wrote the draft. SKS and TKM conceptualised the idea and contributed in shaping the manuscript. IG and NO contributed to the design and provided inputs on the analysis.

Availability of data and materials All data generated or analysed during this study are included in this published article and its supplementary information files.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

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