

1 Importance of meteorology and chemistry 2 in determining air pollutant levels during 3 COVID-19 lockdown in Indian cities

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7 Abstract

8 Indian cities can experience severe air pollution, and the reduction in activity during the COVID-19
9 lockdown offered a natural experiment to study the contribution of local sources. The current work
10 aimed to quantify the changes due to the lockdown in NO_x, O₃ and PM_{2.5} in two contrasting cities in India
11 (Delhi and Hyderabad) using a boosted regression tree model to account for the influence of
12 meteorology. The median NO_x and PM_{2.5} concentrations were observed to decrease after lockdown in
13 both cities, up to 57% and 75% for PM_{2.5} and NO_x, respectively when compared to previous years. After
14 normalization due to meteorology the calculated reduction after lockdown for PM_{2.5} was small (<8%) in
15 both cities, and was likely less attributable to changes local emissions, but rather due changes in
16 background levels (i.e. regional source(s)). The reduction of NO_x due to lockdown varied by site (on
17 average 5-30%), likely reflecting differences in relative proximity of local sources to the monitoring site,
18 demonstrating the key influence of meteorology on ambient levels post-lockdown. Ozone was observed
19 to increase after lockdown at both sites in Delhi, likely due to changes in relative amounts of precursor
20 concentrations promoting ozone production, suggesting a volatile organic compound (VOC)-limited
21 regime in Delhi. Thus, the calculated reduction in air pollutants due to lockdown in the current work
22 cannot be extrapolated to be solely from a reduction in emissions and instead reflects the overall
23 change in ambient levels, as meteorology and atmospheric chemical process also contributed.

24 Environmental Significance Statement

25 To accurately quantify impact of short-term interventions (such as COVID-19 lockdown) on air pollutant
26 levels, meteorology and atmospheric chemistry need to be considered in addition to emission changes.
27 We demonstrate that regional sources have a significant influence on PM_{2.5} levels in Delhi and
28 Hyderabad due to the small reduction calculated post-lockdown after weather-normalization, indicating
29 that future PM_{2.5} mitigation strategies should focus on national-scale, as well as local sources.
30 Furthermore, we demonstrate with field measurements that ozone production in Delhi is likely volatile
31 organic compound (VOC)-limited, in agreement with previous modelling predictions, indicating that
32 ozone mitigation should focus on dominant VOC sources. This work highlights the complexity in
33 developing mitigation strategies for air pollution due to its non-linear relationships with emissions,
34 chemistry and meteorology.

35 1.0 Introduction

36 Exposure to air pollution is a well-established public health risk, with PM_{2.5} (defined as airborne particles
37 with an aerodynamic diameter less than 2.5 µm) of particular concern ¹. India experiences some of the
38 worst air pollution globally ², with nine Indian cities among the top ten most polluted cities in the world
39 for air pollution, as reported by the World Health Organization in 2016 ³. Of most concern across India is
40 ambient particle pollution, with the four largest cities in India (Delhi, Chennai, Hyderabad and Mumbai)
41 having population-weighted annual-mean PM_{2.5} concentrations (72 µg m⁻³) almost double Indian
42 regulatory limits ⁴. As a result, India has a disproportionately high mortality and disease burden due to air
43 pollution, with Balakrishnan et al.⁵ estimating that air pollution exposure, specifically PM_{2.5}, resulted in
44 1.27 million deaths across India in 2017, with the burden higher in the northern states. With PM_{2.5} levels
45 across India forecast to rise owing in part to increased urbanization, the disease burden due to air
46 pollution exposure is expected to worsen ⁶.

47 Like many countries, to mitigate the spread of COVID-19 through social distancing, India implemented a
48 nationwide lockdown, restricting citizens' activities and movements as well as the closures of non-
49 essential businesses and workplaces. The resultant reduction in major urban sources such as traffic,
50 industry and construction activity would lead to decreased emissions, and hence potentially
51 improvements in air quality. Therefore, the COVID-19 lockdown acts a natural experiment to study the
52 contribution of local sources of air pollutants in cities for an extended period under normal
53 meteorological conditions. Consequently, there is a fast-growing number of studies that have aimed to
54 quantify the reduction in air pollutants during lockdown, with Kumar et al.⁷ and Kroll et al.⁸ providing a
55 summary of recent work. Overall, the consensus within the literature is that levels of air pollution have
56 typically decreased after the implementation of lockdown globally (See e.g. ⁹⁻¹²). Recent work in China
57 demonstrated reductions of up to 90% in emissions, primarily from vehicles and manufacturing, during
58 lockdown but also unexpectedly PM_{2.5} mass concentrations across Northern China ¹³. Le and co-authors
59 proposed that unusually high humidity led to increased secondary aerosol formation, which combined
60 with stagnant air patterns, led to severe haze events, highlighting the non-linear nature of atmospheric
61 chemistry as well as the importance of meteorology.

62 Owing to its severe air pollution, there have been a number of studies already published analyzing the
63 effect of lockdown on air quality in India and, overall, notable reductions after lockdown have been
64 reported across the country ^{7,14,15}. Pathakoti et al. ¹⁶ estimated using satellite data that, compared to 5-
65 year mean levels, aerosol and NO₂ levels decreased by 24% and 17%, respectively. The effect of
66 lockdown on air quality varied across India, with reported reductions in PM_{2.5} of 19-43%, 41-53%, 26-
67 54%, 23-36% and 10-30% for Chennai, Delhi, Hyderabad, Kolkata and Mumbai, respectively, with the
68 level of reduction seemingly related to traffic volume ⁷. For NO₂, similar levels of reductions after
69 lockdown have been reported across Indian cities, with one study estimating it ranged between 50-70%
70 for these cities ¹⁷. However, much of this work in India has been observationally based, that is comparing
71 measurements before and after the implementation of lockdown to determine the change in observed
72 levels (e.g. ^{7,17}). The implicit assumption of this approach is that observed changes are driven exclusively
73 by changing emissions, yet meteorology and atmospheric chemistry play a significant role in
74 determining ambient pollutant levels ⁸. When assessing changes in air pollutants due to short-term
75 interventions (such as lockdown), these two factors need to be considered, with previous work
76 demonstrating the importance of accounting for changes in meteorology ^{11,13,15,18,19}.

77 The current work aims to quantify the changes in regulated air pollutants levels (i.e. NO_x, O₃ and PM_{2.5})
78 due to the lockdown in two contrasting cities in India. We used two methods to achieve this aim, firstly
79 we simply compared observed concentrations during the lockdown period to the corresponding dates in
80 the last three years. Secondly, we employed a boosted regression tree (BRT) model to predict
81 concentrations post-lockdown and compared these predicted values to the measured concentrations, in
82 order to account for the effect of meteorology on pollutant levels. Therefore, we primarily focus on
83 normalizing the effect of meteorology on observed levels in order to estimate change in levels after
84 lockdown. The results of these two approaches to quantify the change in air pollutant levels due to
85 lockdown are compared to explore the importance of meteorology. Finally, the changes in pollutant
86 levels after lockdown allowed us to explore the chemical regime for local O₃ production in Delhi.

87 2.0 Method

88 2.1 Description of study sites

89 We chose Delhi and Hyderabad for this study, as they are both large inland cities but located in the
90 north and south of India, respectively. Delhi is the largest city in India and experiences the highest levels
91 of air pollution⁴. This is partly due to Delhi being located on the Indo-Gangetic plain, which experiences
92 significant regional air pollution due to seasonal agricultural burning²⁰. Hyderabad is the fourth largest
93 city in India and is outside the regional air pollution that affects much of northern India. Consequently,
94 Hyderabad experiences typically lower levels of PM_{2.5} compared to Delhi; however severe pollution
95 events are common in Hyderabad⁴.

96 2.2 Data analysis

97 The data used in the study was obtained from the publicly available database on the Central Pollution
98 Control board (CPCB) of India website (<https://cpcb.nic.in/>), using data collected by the CPCB, Delhi
99 Pollution Control Committee (DPCC), Haryana State Pollution Control Board (HSPCB) and Telangana
100 State Pollution Control Board (TSPCB). As the national lockdown was introduced on 23 March 2020, we
101 collected hourly data between 1 March 2017 and 24 April 2020 for the following species: PM_{2.5}, NO_x,
102 NO₂, NO, and O₃. The lockdown across India did not end on 24 April, but we have chosen to focus on the
103 initial month after its implementation (known as phase-I) as this period encompasses the most severe
104 restrictions on citizens' activity⁷. We focused on urban background sites and these were chosen
105 primarily based upon data availability for the above dates, with 2 sites selected in Delhi: RK Puram
106 (28.563212, 77.186954) and ITO (928.628527, 77.240851). For Hyderabad, 2 sites were selected: Zoo
107 Park (17.349707, 78.451440) and Santhanagar (17.455925, 78.433330). All collected data underwent
108 quality control checks prior to analysis as recommended in Pant et al.²¹. All data processing was
109 performed in R (v3.6.2) primarily utilizing the openair package²². Hourly meteorological and visibility
110 data for 2017-2020 was obtained for Safdarjung (Delhi) and Begumpet (Hyderabad) airports as these
111 were closest to the monitoring sites of interest using the worldmet package in R.

112 2.3 BRT model description

113 To predict pollutant concentrations based on meteorology, a boosted regression tree (BRT) model was
114 developed using the deweather package in R. We chose to focus on PM_{2.5} and NO_x as these have direct
115 emission sources in urban areas. To build the BRT model at each site we followed the procedure
116 outlined in Carslaw et al.¹⁹ and we used meteorological data, measured and background concentrations
117 of the pollutant of interest along with time-based co-variates (e.g. time of day, day of week).

118 Meteorological measurements were obtained from the two airport sites, which included relative
119 humidity, atmospheric pressure, dew point, wind speed, wind direction and temperature.

120 Background levels were included as previous work has shown this can significantly improve the
121 predicting power of the BRT model^{19,23}. The background sites were selected to be upwind of the sites
122 of interest, based on the prevailing wind direction (NW at both cities) as well as data availability over the
123 time period required. Only one site was suitable for Hyderabad (ICRISAT Patancheru), as there were few
124 sites to the NW, and therefore the measurements from this site were taken as representative of
125 background levels. In comparison, at Delhi five background sites were available upwind with enough
126 data coverage and included Vikas Sadan, Gurugram, Alipur, Bawana and Najafargh. The median of the
127 measured concentration at five sites were used as the background concentrations for the model, to limit
128 any local sources emissions overly affecting the analysis.

129 While BRT models are typically robust against including too many variables, in order to minimize the
130 associated error in the predictions, there is typically an optimum number of co-variates. Therefore, we
131 built a series of test models using a multi-year dataset of the parameters above (2016-2020) by
132 systematically changing the number of co-variates to determine the optimal suite. The optimal model
133 was chosen based on its predictive capability (using a randomly selected 25% of the dataset). An
134 advantage of the BRT approach is that the model outputs are physically and chemically meaningful and
135 can be explored by partial dependencies of the co-variates, which is the relationship between the
136 pollutant of interest and the covariates used in the model. To further aid the selection of the optimal
137 model, we examined the model partial dependences to ensure if they made chemical or physical sense,
138 for example if NO_x decreased with increasing wind speed as would be expected if local emissions
139 dominate.

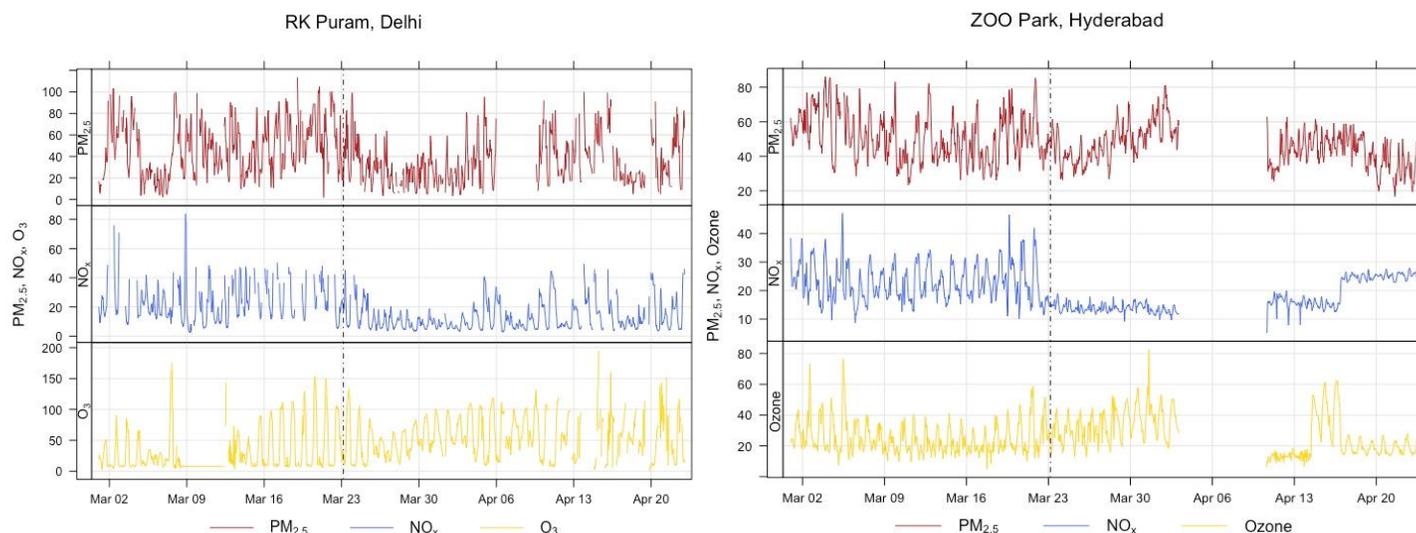
140 The optimized BRT model was used to independently predict pollutant concentrations based only on the
141 meteorology, for 2 months pre-lockdown (1 Feb to 22 Mar 2020) and for 1-month during phase-I
142 lockdown (23 Mar to 23 Apr 2020). In order to optimize the predictive power for each model, the
143 number of iterations/trees were set independently to equate the number of data points. This approach
144 minimizes the variance of the model and improves the prediction capability of the concentrations of
145 pollutants, based on the chosen co-variates using a randomized approach^{24,25}. We built separate BRT
146 models for PM_{2.5} and NO_x for each site in Delhi and Hyderabad, because the source influence would be
147 expected to vary (e.g. the direction of important sources relative to the site). Owing to poor data
148 coverage post-lockdown, we did not employ this analysis for NO_x at Zoo Park and PM_{2.5} at Santhanagar.
149 For NO_x and PM_{2.5} at all sites, the background concentration and temporal trends were the most
150 important co-variates for the model, followed by meteorology; dew point, air temperature, wind speed,
151 RH and wind direction. The partial dependencies of each meteorological co-variate differed between the
152 two cities and for NO_x and PM_{2.5}, as would be expected. The optimized BRT model for PM_{2.5} at both sites
153 in Delhi (RK Puram and ITO), were found to have similar co-variate influence. This was likely due to
154 strong influence of background concentrations on the BRT models at both sites, and this is discussed
155 later in the manuscript. For NO_x, the BRT models were notably different in terms of co-variate influence
156 at RK Puram and ITO.

157 3.0 Results and Discussion

158 3.1 Time series of pollutants pre- and post-lockdown

159 Fig 1 presents a time series of selected species levels (PM_{2.5}, NO_x and O₃) for March and April 2020 at RK
160 Puram, Delhi and Zoo Park, Hyderabad. The corresponding time series for other sites are presented in
161 Figs S1 and S2, Supporting Information. In India, a nationwide lockdown came into force on the 23
162 March 2020 and a clear and rapid decrease in NO_x was observed after this date at all sites (Figs 1, S1 and
163 S2). As NO_x is a primary pollutant (i.e. directly emitted into the atmosphere from primarily
164 anthropogenic sources such as vehicles), this reduction would be expected if there was significant
165 reduction in human activity during lockdown and is consistent with studies elsewhere^{10,11}.

166 Contrasting trends in PM_{2.5} and ozone after lockdown were observed at Delhi and Hyderabad (Fig 1). At
167 Hyderabad, the levels of PM_{2.5} and ozone were not observed to change after lockdown (23 March, Fig 1),
168 as would be expected if regional sources were dominant for these two species. At the two Delhi sites,
169 there is an apparent reduction in the levels of PM_{2.5} immediately after lockdown compared to the before
170 (Fig 1), with levels appearing to recover towards pre-lockdown levels after 2 weeks (i.e. on the 6th April).
171 While this may point to a reduction in primary emissions for PM_{2.5}, a significant rain event that coincided
172 with the implementation of lockdown, may also explain these observed changes. Therefore, this would
173 suggest that meteorology influenced the observed levels during the lockdown period.



174 Fig 1: A time series of PM_{2.5} (µg/m³), NO_x (ppbv) and O₃ (µg/m³) levels pre-lockdown and during phase-I
175 lockdown at RK Puram, Delhi and Zoo Park, Hyderabad. The marker placed on 23 March denotes the
176 start of phase-I lockdown.

177 3.2 Comparison of concentrations during lockdown period to previous years

178 We first compared the levels of PM_{2.5}, NO_x and O₃ during the phase-I lockdown period (24 March to 24
179 April 2020) to the corresponding dates in previous three years (24 March to 24 April 2017-2019, referred
180 to as L3Y) to evaluate the changes due to lockdown. This comparison is shown in Figs 2 and 3 for PM_{2.5}
181 and NO_x, respectively. Generally, the median levels during phase-I lockdown were lower than the L3Y
182 in both cities (Table 1). Overall, we observed a greater reduction in median NO_x compared to PM_{2.5} levels
183 at each site (Table 1). The exception was ITO, where the median NO_x levels were comparable during

184 phase-I lockdown compared to L3Y (Fig 3), while at the other sites the reduction in median NOx levels
 185 ranged from 34-75%. From Fig 3, the inter-quartile ranges were notably different at most sites,
 186 suggesting the observed reduction during phase-I lockdown was significant and likely reflects that NOx
 187 has more local emissions sources affected by the lockdown (e.g., vehicle emissions).

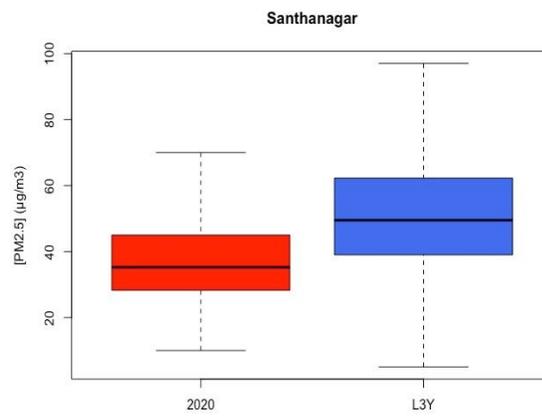
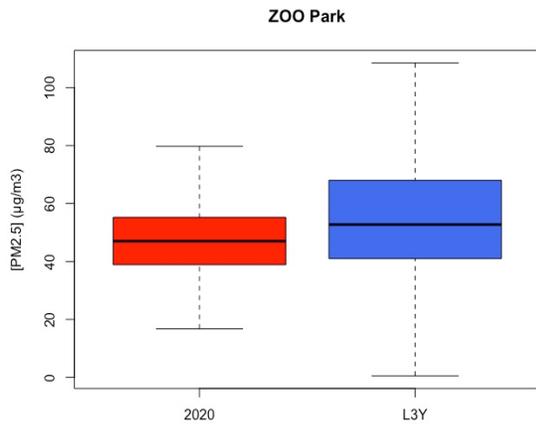
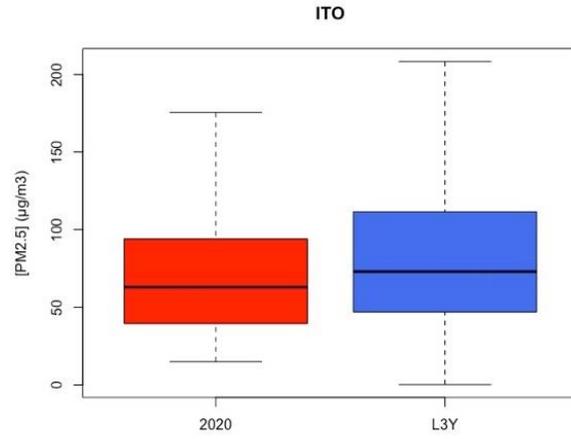
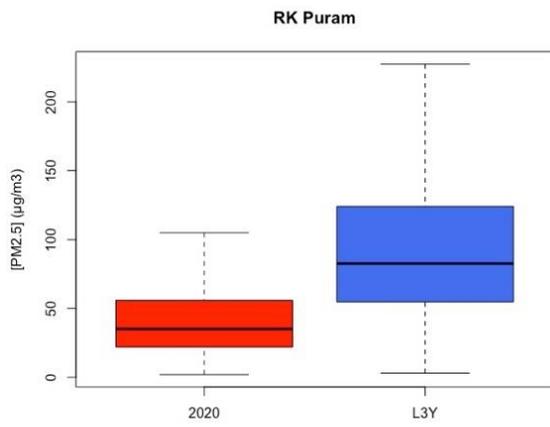
188 The largest change for PM_{2.5} during phase-I lockdown compared to L3Y was observed at RK Puram,
 189 where the median PM_{2.5} mass concentration decreased by 57%. The reduction in PM_{2.5} during phase-I
 190 lockdown was similar at the other sites (11-36%, Table 1), and is lower than previously reported. Sharma
 191 et al.¹⁵ reported a decrease of 34% in mean PM_{2.5} mass concentrations across North India during
 192 lockdown compared to previous years, while Kumar et al.⁷ calculated a decrease of 41-53% and 26-54%
 193 for Delhi and Hyderabad, respectively. The differences in calculated reduction between studies may
 194 reflect differences in lockdown time periods chosen for analysis, as well as monitoring site locations
 195 within the cities. However, while the median levels for both PM_{2.5} were lower during phase-I lockdown
 196 compared to L3Y, we observed similar inter-quartile ranges (Fig 2) for most sites (with exception of RK
 197 Puram), suggesting the changes may not be significant. This suggests that regional sources of PM_{2.5} may
 198 be dominant at both cities, and that meteorology needs to be considered in order to properly account
 199 for regional source influence. Understanding changes in regional or background levels is critical to
 200 properly assess changes in local emissions, and in the next section we employed a BRT model to predict
 201 the influence of meteorology on PM_{2.5} and NOx levels during phase-I lockdown.

202 Table 1: Median levels of PM_{2.5} and NOx during the phase-I lockdown period (24 March – 24 April 2020)
 203 and corresponding dates for 2017-19 (L3Y).

			<i>PM_{2.5} (µg m⁻³)</i>		<i>NOx (ppbv)</i>	
<i>Delhi</i>			2020	L3Y	2020	L3Y
	RK Puram	Median	36.3	83.3	14.9	60.6
		% decrease in phase-I	57%		75%	
	ITO	Median	63.0	73.0	51.2	49.3
		% decrease in phase-I	14%		-4%	
<i>Hyderabad</i>	Zoo park	Median	47.0	52.8	18.2	34.1
		% decrease in phase-I	11%		47%	
	Santhanagar	Median	35.3	49.5	9.8	28.4
		% decrease in phase-I	29%		66%	

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208 Fig 2: Box plots of hourly measured PM_{2.5} mass loadings at the sites of interest in Delhi and Hyderabad
 209 comparing them during phase-I lockdown (24 March to 24 April 2020) to the corresponding dates in
 210 2017-2019 (L3Y). Note the different y-axis for each plot.

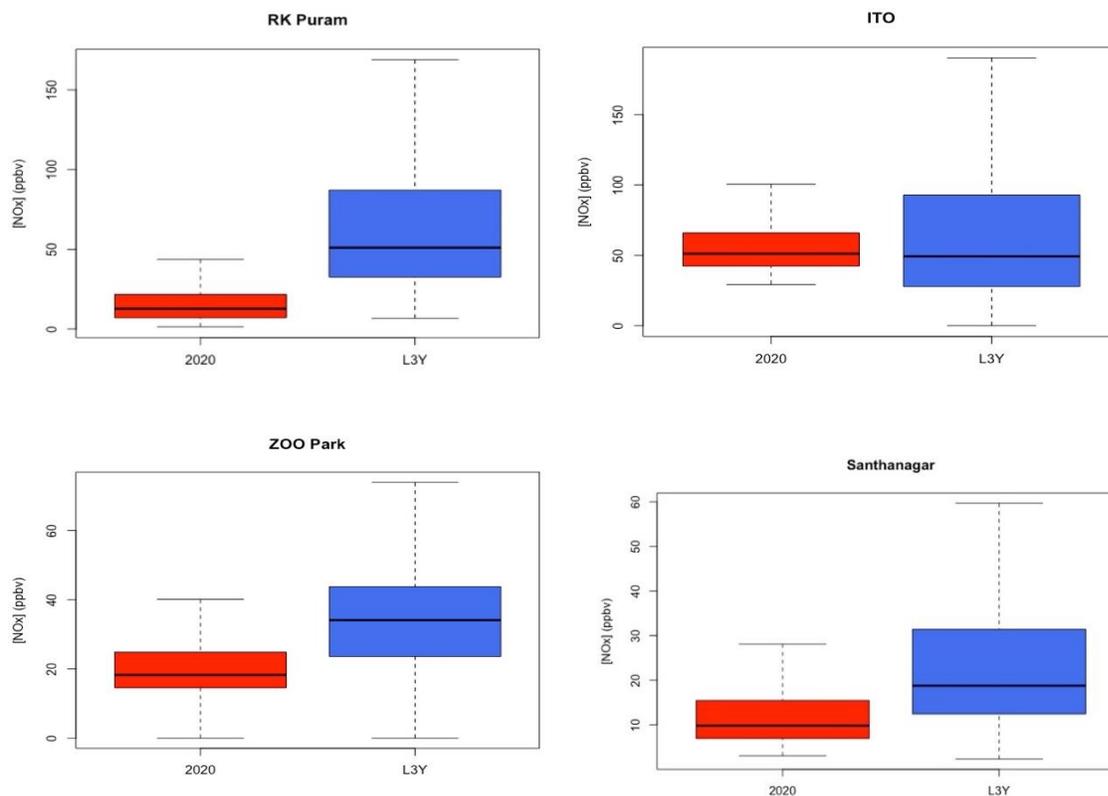
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218 Fig 3: Box plots of measured NOx mixing ratios at the sites of interest in Delhi and Hyderabad comparing
 219 them during phase-I lockdown (24 March – 24 April 2020) to the corresponding dates in 2017-2019
 220 (L3Y). Note the different y-axis for each plot.

221 3.4 Predicted levels from BRT model compared to measured concentrations during phase-I
 222 lockdown

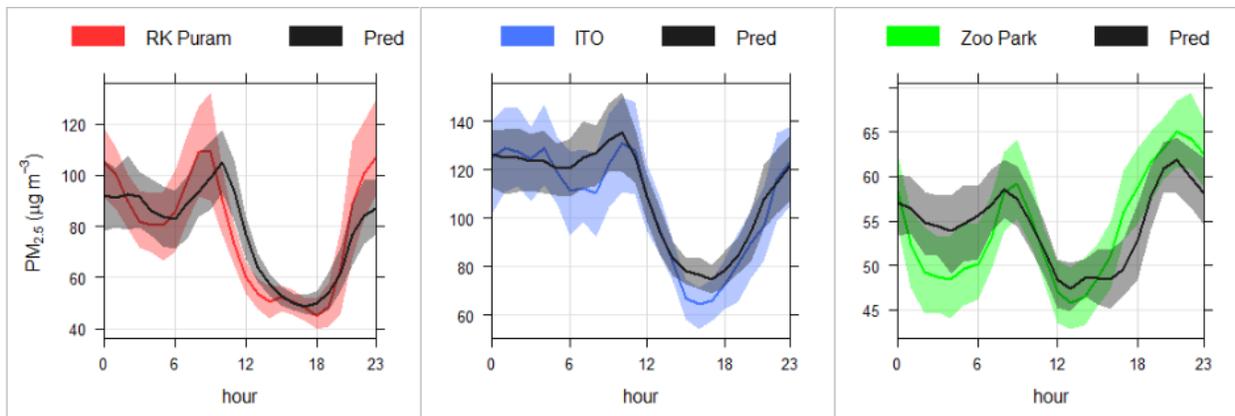
223 Using the BRT model, we predicted levels of NOx and PM_{2.5} based primarily on the meteorology at each
 224 site for Feb-Apr 2020, in order to capture the changes between pre-lockdown and phase-I lockdown
 225 periods. The predicted values from the model can be considered as representative of the expected
 226 levels during phase-I lockdown, and any difference between predicted and measured may be attributed
 227 to changes in local emissions¹⁹. We compared the predicted and measured levels at each site, and as a
 228 summary we present the mean diurnal trends in Figs 4-5. The time series of predicted and measured
 229 levels pre- lockdown and phase-I (Feb-Apr 2020) are shown in the Supporting Information (Figs S3 and
 230 S4). From Figs 4 and 5, the model performed well in predicting measured levels and capturing the
 231 temporal trends prior to lockdown at each site for PM_{2.5} (r^2 of 0.61-0.93) and NOx (r^2 of 0.76-0.9).

232 The predicted PM_{2.5} mass concentration diurnal trends in phase-I lockdown are similar to those
 233 measured at all sites (Figs 4). The difference in PM_{2.5} levels after weather normalization during phase-I
 234 lockdown compared to pre-lockdown was 8%, -0.6% and 3% for RK Puram, ITO and Zoo Park,
 235 respectively. Thus, the changes in PM_{2.5} mass concentrations observed at these sites during phase-I
 236 lockdown was likely less attributable to local emissions, but rather due changes meteorology. The
 237 importance of meteorology on PM_{2.5} levels suggests regional sources may play a significant role.

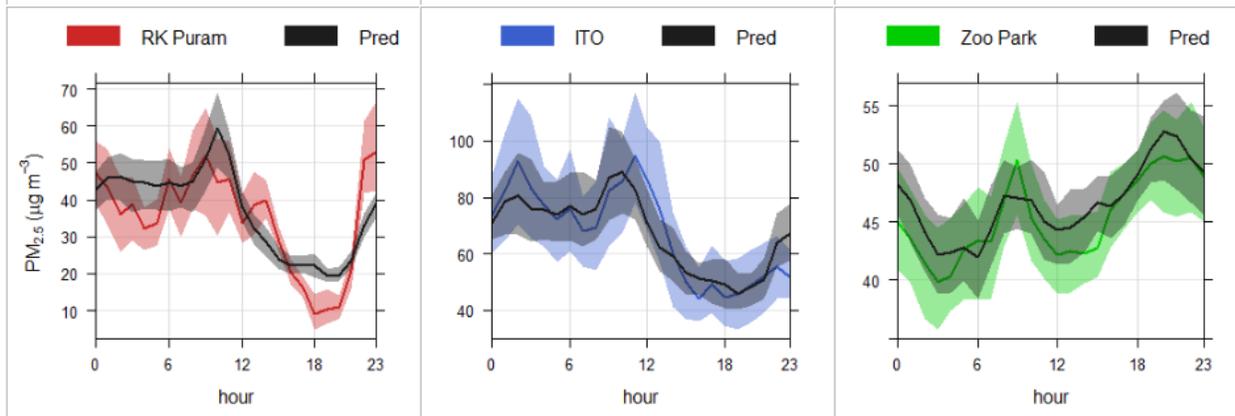
238 Differences in NO_x mixing ratios were observed between mean diurnal predicted and measured levels
239 during phase-I lockdown at two sites (Figs 5). The difference in mean diurnal trends pre-lockdown
240 compared to phase-I lockdown for NO_x was on average was 20%, 5% and 30% at RK Puram, ITO and
241 Santhanagar, respectively. At RK Puram, the measured NO_x levels were notably lower than predicted
242 during the afternoon (12-6pm), with average reductions of 50%, while at ITO and Santhanagar, the
243 reduction was 5.5% and 33%, respectively, similar to the daily mean difference. There was a notable
244 difference in predicted reduction in NO_x during phase-I lockdown between the two sites in Delhi. At RK
245 Puram, there was a large difference between predicted and measured NO_x, while predicted and
246 measured NO_x levels at ITO were similar. Both sites are near major roads and the reason for this
247 difference was likely the prevailing wind direction with respect to this major source. During phase-I
248 lockdown the prevailing wind direction was west/northwest (Fig S5, Supporting Information). Under
249 these conditions, the RK Puram site was downwind and the ITO site was upwind from major roads.
250 Therefore, while lower NO_x levels are observed during phase-I lockdown at ITO, this is predicted by the
251 model based on the meteorology, and this is perhaps best illustrated by the flat diurnal cycle in
252 observed NO_x during phase-I lockdown (Fig 5), atypical if vehicle emissions were dominant. In the pre-
253 lockdown period, when there was significant portion of wind from east (placing the site downwind from
254 major roads, Fig S5), the model performed well at capturing the NO_x diurnal trends that are more typical
255 for vehicle emissions at ITO. However, without detailed traffic activity data for these two locations it is
256 difficult to ascertain the true cause of the differences, but what is clear is that the changes in
257 meteorology affected the observed levels.

258 Overall, we observed a greater difference between predicted and measured levels with NO_x compared
259 to PM_{2.5}, and this likely reflects that NO_x is a primarily emitted by sources most affected by phase-I
260 lockdown (i.e. vehicle exhaust emissions). The reduction in NO_x emissions would also affect chemical
261 processes, notably ozone production, which is discussed in more detail in the next section.

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264 Figure 4. Diurnal trends of measured PM_{2.5} mass concentrations and predicted levels pre-lockdown (top)
265 and during phase-I lockdown (bottom) at the three sites.

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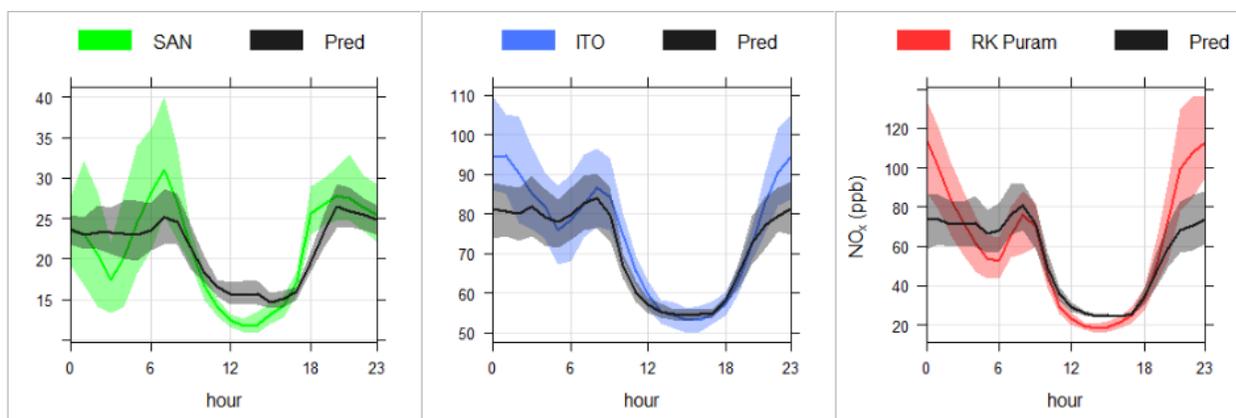
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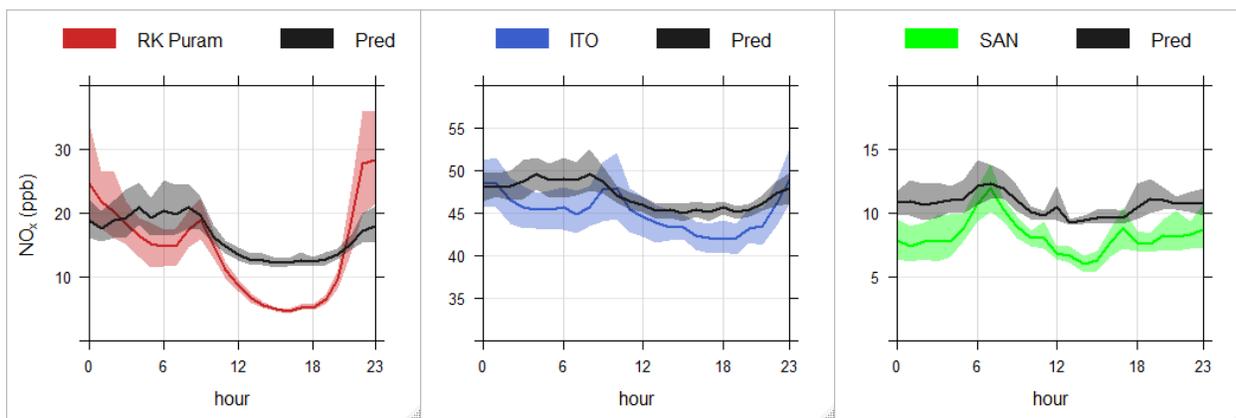
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274 Fig 5: Diurnal trends of measured NO_x mixing ratios and predicted levels pre-lockdown (top) and phase-I
275 lockdown(bottom) at the three sites.

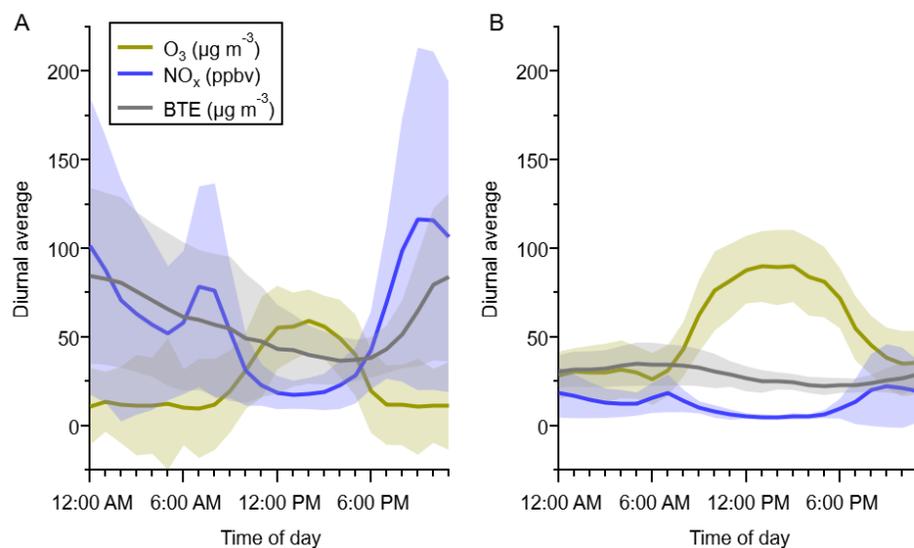
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277 3.5 Effect of lockdown on atmospheric chemistry: Evidence for local ozone production in Delhi 278 being VOC-limited

279 The phase-I lockdown provided an opportunity to observe the impacts of a different chemical regime on
280 local ozone production. At all sites, O₃ levels increased following the lockdown (Figure S6). However, at
281 the RK Puram site, O₃ mixing ratios increased significantly following for all daytime hours in the 17 days
282 during phase-I lockdown compared to the 51 days prior to lockdown (paired t-test for hourly data,
283 Figure 6, Figure S7, Figure S8), corresponding to a maximum average increase of 62% at 15:00. For this
284 site, we examined the potential impacts of NO_x, VOCs, and light availability on the chemical formation
285 of O₃. As described above, NO_x mixing ratios decreased following lockdown (Figure 6). Mixing ratios
286 were statistically lower for all hours of the day (paired t-test for hourly data). Measurements of VOCs
287 were not publicly available from RK Puram, so we collected benzene, toluene and ethylbenzene
288 (collectively referred to as BTE) measurements from the nearby Sirifort site (3 km away). Although this
289 does not represent the full spectrum of VOCs in Delhi, recent work has shown that aromatic compounds
290 comprise the largest fraction of VOCs in Delhi and are predominately from traffic and solid fuel burning
291 emissions²⁶. In addition, the O₃ formation potential of BTE is high²⁷, thus BTE can be a useful proxy for
292 the impact of VOCs on local O₃ formation. Reported levels of BTE were statistically lower during phase-I
293 lockdown for all daylight hours (paired t-test for hourly data). Daytime visibility (Safdarjung airport, 3.3

294 km from RK Puram) was generally higher during phase-I lockdown, although data reporting frequency
295 was insufficient to compare to pre-lockdown period statistically (Figure S9). This is generally consistent
296 with a decrease in PM_{2.5} during phase-I lockdown.

297 Decreased levels of NO_x and VOCs during phase-I lockdown, along with increased light, led to increased
298 O₃ at the RK Puram site in Delhi. These results agree with recent modelling work that predicted O₃
299 formation in Delhi is in the traditionally defined VOC limited regime²⁸. Although VOC limited is the most
300 common descriptor for this regime, it can also occur in areas with high VOC levels²⁹. A VOC limited
301 regime may be described as NO_x saturated or radical limited with respect to O₃ production, in which
302 emissions of NO_x exceed radical production (from VOC oxidation and other sources). Chen et al. (2020)
303 also noted the impact of visibility on O₃ formation, suggesting the chemistry was light limited, which is
304 consistent with a radical-limited regime. A decrease in NO_x leads to increased O₃ formation in a radical-
305 limited regime because of a reduction in the loss pathway for radicals through reaction with NO₂. Similar
306 observations of increased O₃ resulting from pandemic-related decreased NO_x were made in urban areas
307 in China¹³ and the UK³⁰.



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309 Fig. 6: Diurnal averages of O₃, NO_x, and the sum of benzene, toluene, and ethylbenzene (BTE) in RK
310 Puram in the (A) 51 days preceding lockdown and (B) 17 days following start of phase-I lockdown.
311 Shaded areas represent the standard deviation of the measurements.

312 4.0 Conclusions

313 Indian cities can experience severe air pollution from a complex mixture of sources, and the reduction in
314 activity during the COVID-19 lockdown offered a natural experiment to study the contribution of local
315 sources in urban areas. The concentrations of NO_x and PM_{2.5} were observed to decrease during phase-I
316 lockdown in both Delhi and Hyderabad at all selected sites, as would be expected if local emissions were
317 driving ambient levels at these sites. Compared to previous years, the calculated reduction in median
318 concentrations during phase-I lockdown period was generally large, up to 57% and 75% for PM_{2.5} and
319 NO_x, respectively. This calculation assumes that the local emission solely controls ambient levels, yet
320 meteorology also impacts air quality. To normalize for its effect, we employed a BRT model to predict
321 concentrations based on meteorology and compared this to measured values during phase-I lockdown.

322 The calculated reduction in PM_{2.5} and NO_x levels during phase-I lockdown using a BRT model to account
323 for effect of meteorology (Figs 4 and 5) were notably different to those calculated by comparing median
324 concentration during phase-I lockdown to the same time period in previous years (Table 1). While the
325 relative trends were similar, with both methods suggesting smaller change during phase-I lockdown at
326 ITO compared to RK Puram and the Hyderabad sites (Table 1 and Figs 4 and 5) the absolute magnitude
327 differed. Overall, a higher percentage reduction was calculated in comparing median concentrations
328 (Table 1) than when the effects of meteorology were normalized by the BRT model. This would suggest
329 that, despite lower concentrations being observed at all sites for PM_{2.5} and NO_x during phase-I lockdown
330 compared to pre-lockdown (Fig 1), much of the observed decrease after lockdown was, at least in part,
331 driven by changes in meteorology.

332 For PM_{2.5}, after normalization due to meteorology the calculated reduction during phase-I lockdown was
333 small (Fig 4). Thus, the changes in PM_{2.5} mass concentrations observed in Delhi and Hyderabad during
334 phase-I lockdown (at least for the sites studied) were likely less attributable to local emissions, but
335 rather due changes in background levels (i.e. regional source(s)). This result stands in contrast to
336 previous work based on solely on observational data in Delhi, which concluded significant reductions
337 due to lockdown (<60%,⁷ and references therein). But this result is perhaps not surprising when
338 considering the significant influence of regional sources on PM_{2.5} levels across northern India²⁰. These
339 sources include rural/agriculturally based emissions, that were possibly less affected by lockdown²⁶.

340 While for NO_x the reduction during phase-I lockdown varied by site (on average 5-30%, Fig 5), likely
341 reflecting differences in local source emissions and its relative proximity to the monitoring station,
342 highlighting the importance of meteorology (i.e. wind direction) on the observed levels. Overall, we
343 observed a greater difference between predicted and measured levels with NO_x compared to PM_{2.5}, and
344 this likely reflects that NO_x is a primarily emitted by sources most affected by lockdown (e.g. vehicle
345 exhaust emissions). Changes in relative amounts of precursor concentrations led to observed increased
346 in O₃ post-lockdown at both sites in Delhi. Consistent with previous modelling work, O₃ in Delhi was
347 shown to be in the VOC limited regime (also known as radical limited). Decreased levels of NO_x and
348 increased light led to increased O₃ in the phase-I lockdown. This emphasizes the need for clear
349 consideration of chemistry when targeting emissions reductions. Reductions in NO_x and PM_{2.5} can lead
350 to increased O₃ formation in Delhi, which indicates potential trade-offs in emissions reduction.

351 The presented changes in air pollutant levels during phase-I lockdown in the current work cannot be
352 extrapolated to be solely from reduction in emissions activity and instead reflects the complex
353 interactions between emissions, meteorology and chemistry. This work highlights that the impacts of all
354 three must be considered when assessing the effects of a short-term intervention on air pollutants.

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358 [Author contributions](#)

359 LRC conceptualized the study. YEI performed the modelling. All authors contributed to data analysis,
360 manuscript writing, and editing.

361 **Conflicts of Interest**

362 There are no conflicts of interest to declare.

363

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453 *Supporting information for*

454 Importance of meteorology and chemistry
455 in determining air pollutant levels during
456 COVID-19 lockdown in Indian cities

457 Leigh R. Crilley, Yashar E. Iranpour and Cora J. Young

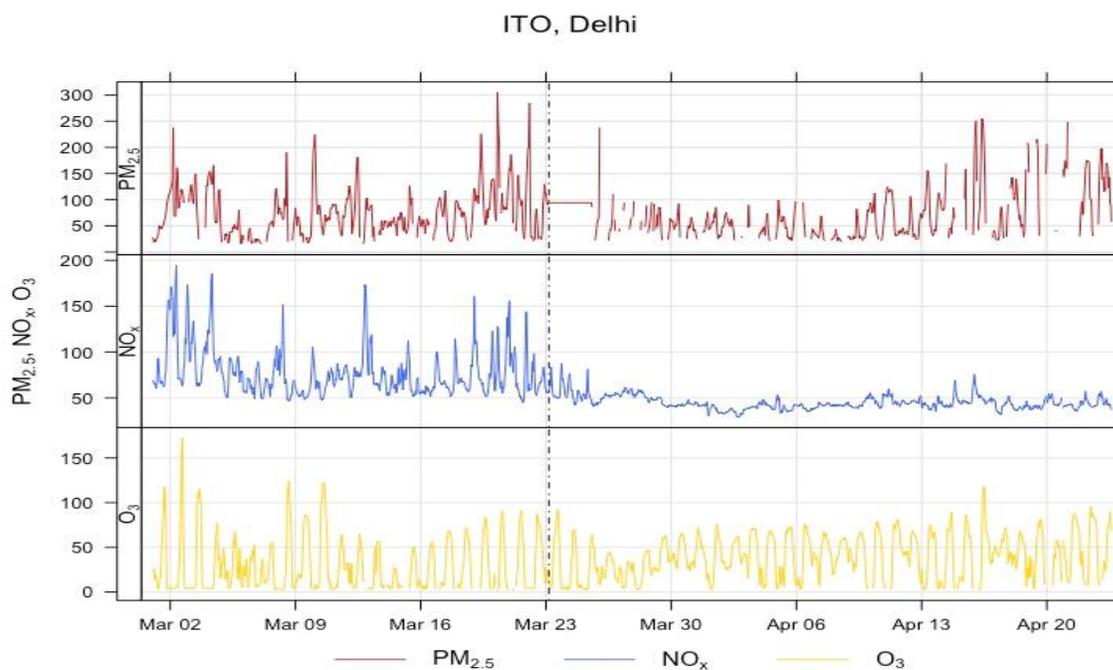
458 *Department of Chemistry, York University, Toronto, ON, Canada*

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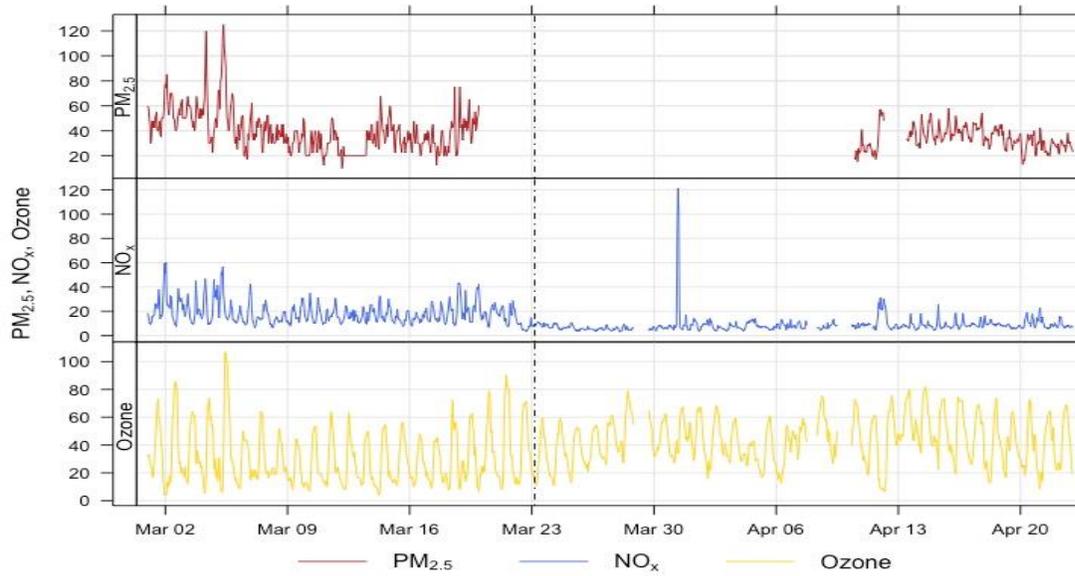
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464 Figure S1: A time series of PM_{2.5} (µg/m³), NO_x (ppbv) and O₃ (µg/m³) levels pre-lockdown and during
465 phase-I lockdown at ITO, Delhi. The marker placed on 23 March denotes the start of phase-I lockdown.

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Santhanagar, Hyderabad



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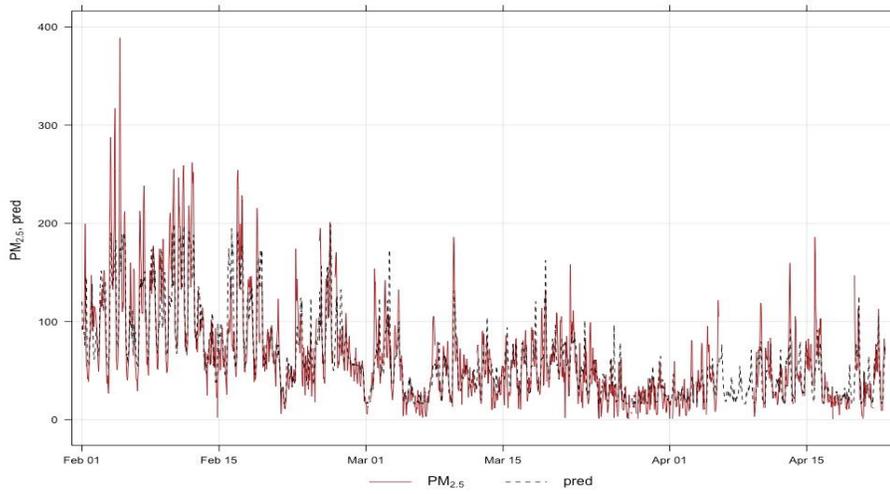
468 Figure S2: A time series of $PM_{2.5}$ ($\mu g/m^3$), NO_x (ppbv) and O_3 ($\mu g/m^3$) levels pre-lockdown and during
469 phase-I lockdown at Santhanagar, Hyderabad. The marker placed on 23 March denotes the start of
470 phase-I lockdown.

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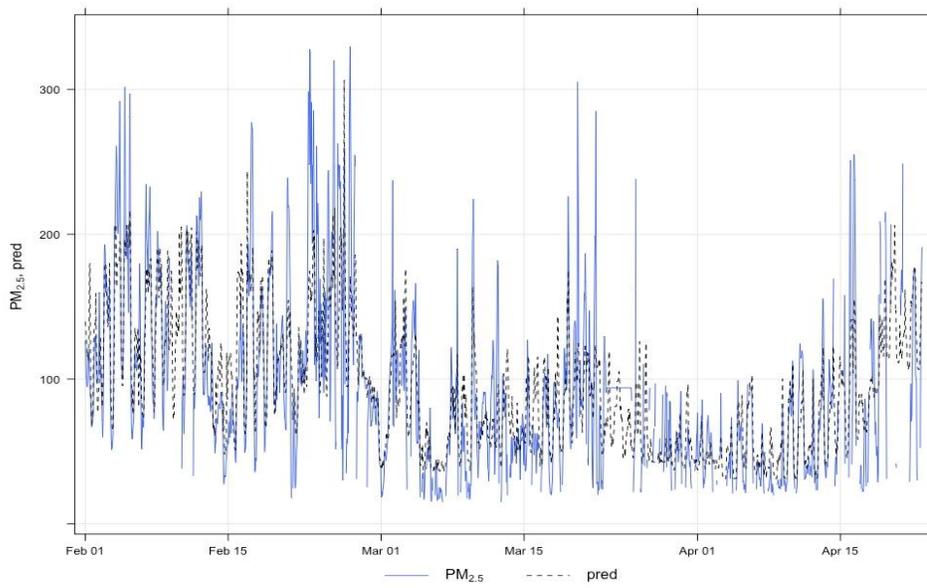
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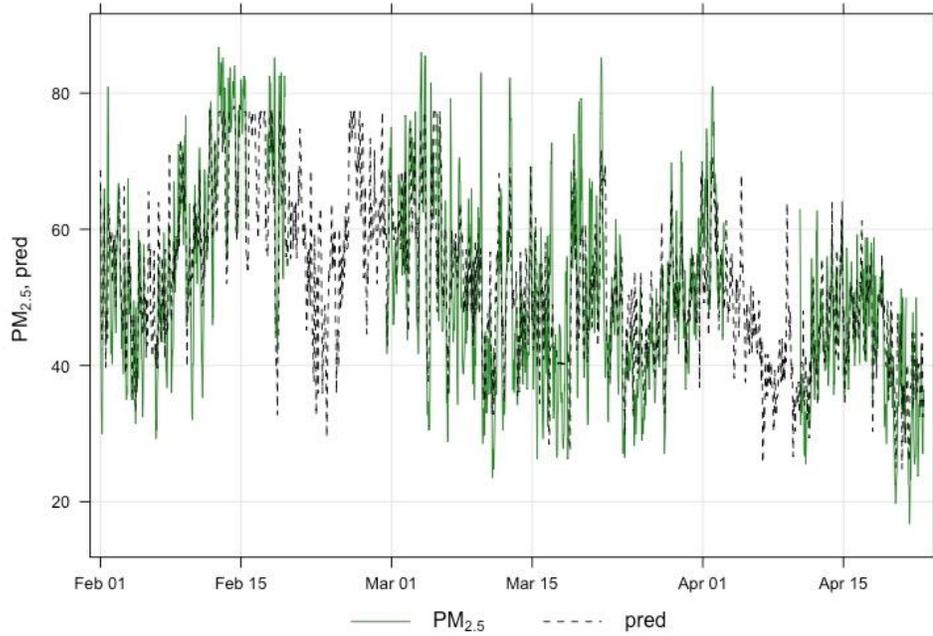
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ZOO Park, Hyderabad

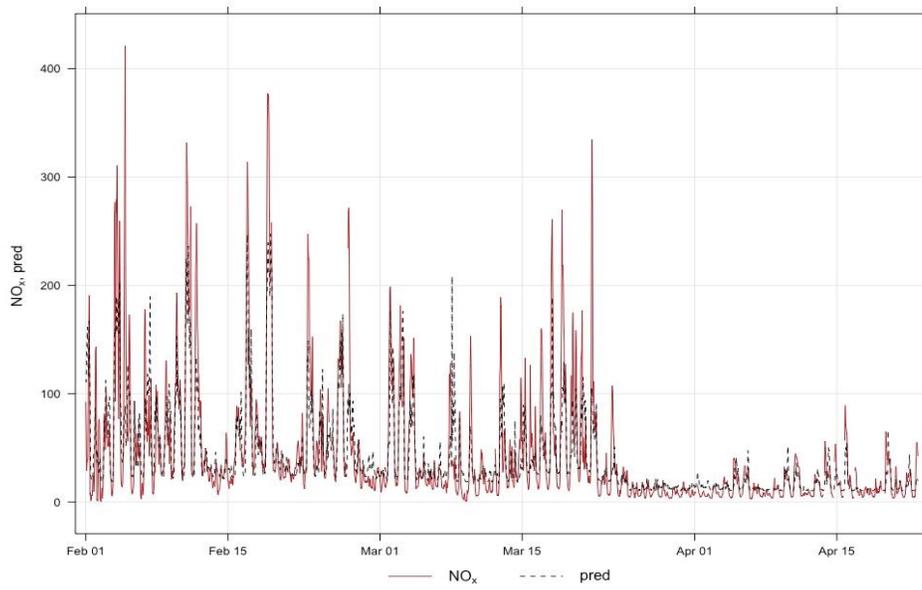


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479 Figure S3: Time series of measured and predicted PM_{2.5} mass concentrations pre-lockdown and during
480 phase-I lockdown at RK Puram (Delhi), ITO (Delhi) and Zoo Park (Hyderabad).

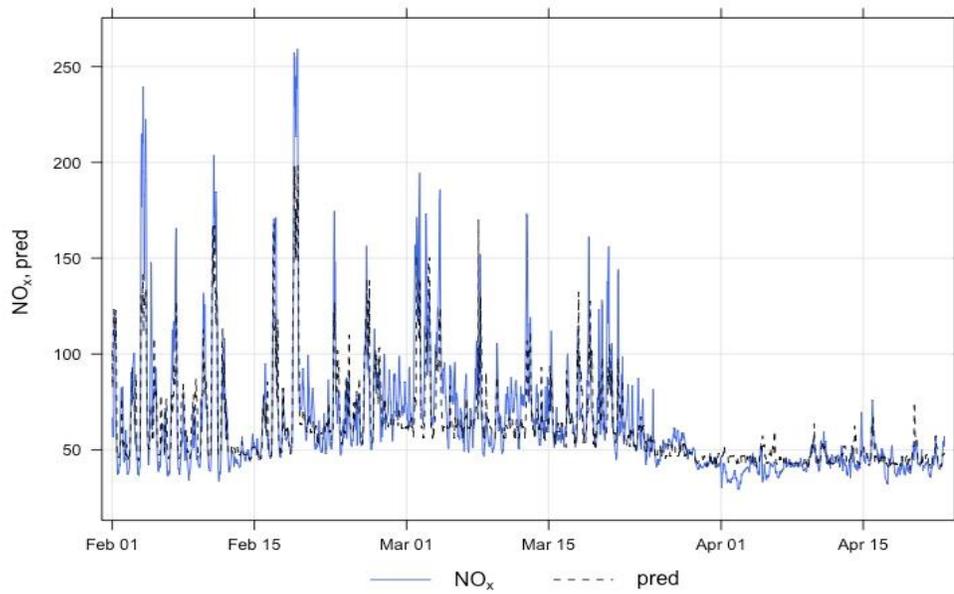
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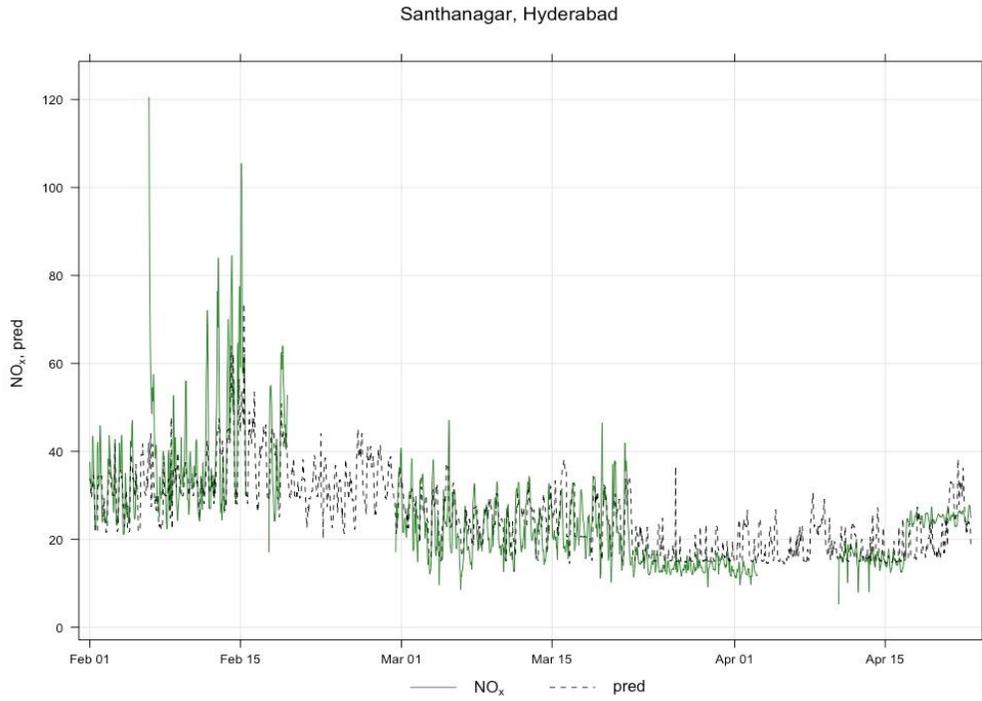
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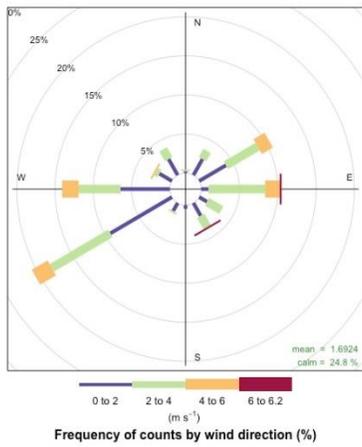
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490 Figure S4: Time series of measured and predicted NOx mass concentrations pre-lockdown and during
 491 phase-I lockdown at RK Puram (Delhi), ITO (Delhi) and Santhanagar (Hyderabad).

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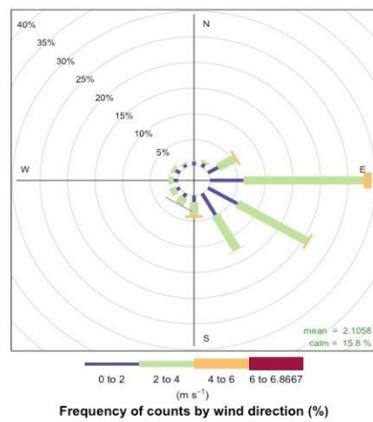
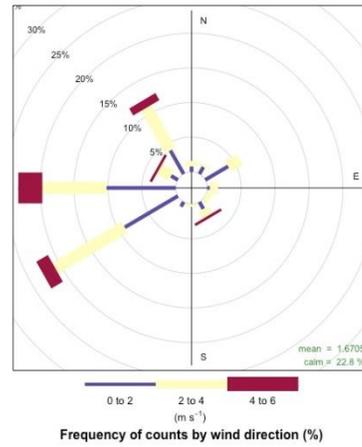
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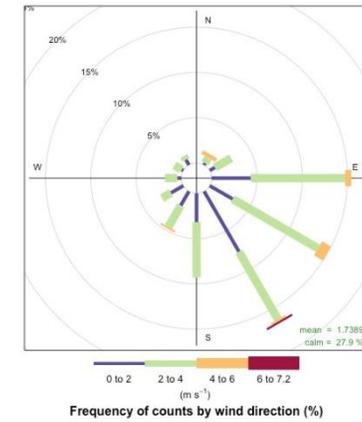
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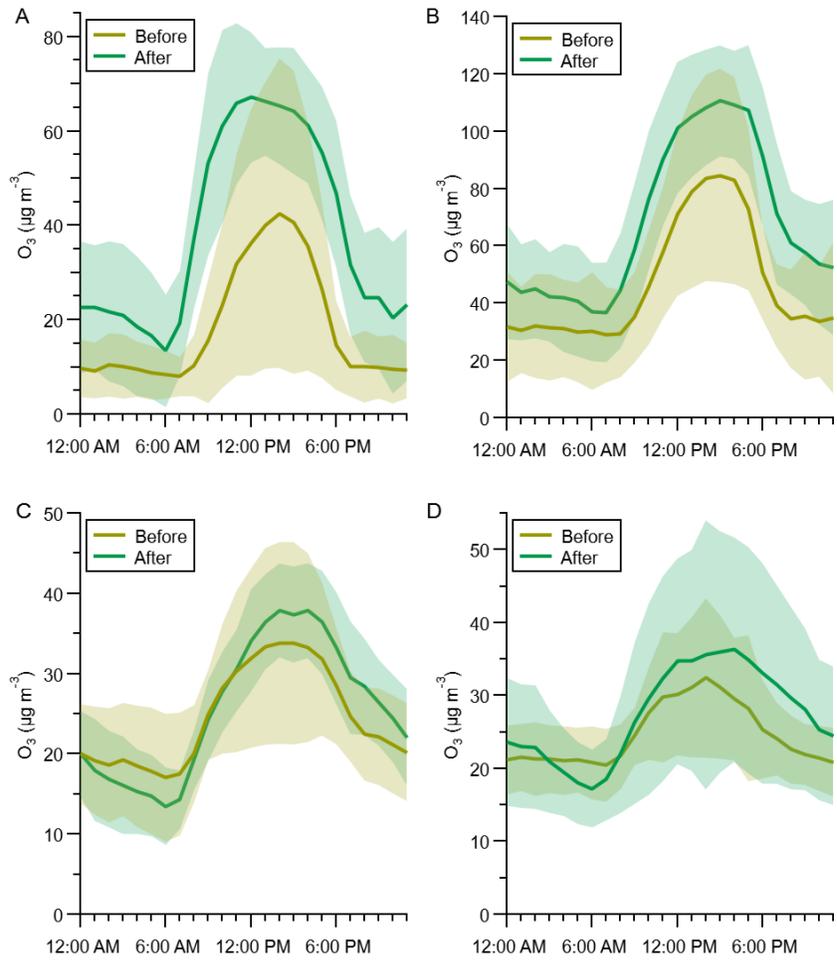
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499 Figure S5: Wind rose plot for pre-lockdown and during phase-I lockdown period at Safdarjung airport
500 (Delhi – top) and Begumpet airport (Hyderabad – bottom) (Feb-Apr 2020).

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504 Fig S6: Comparison of diurnally-averaged O₃ pre-lockdown and during phase-I lockdown at: (A) ITO, (B)
 505 DTU, (C) ICRISAT, and (D) Zoo Park sites.

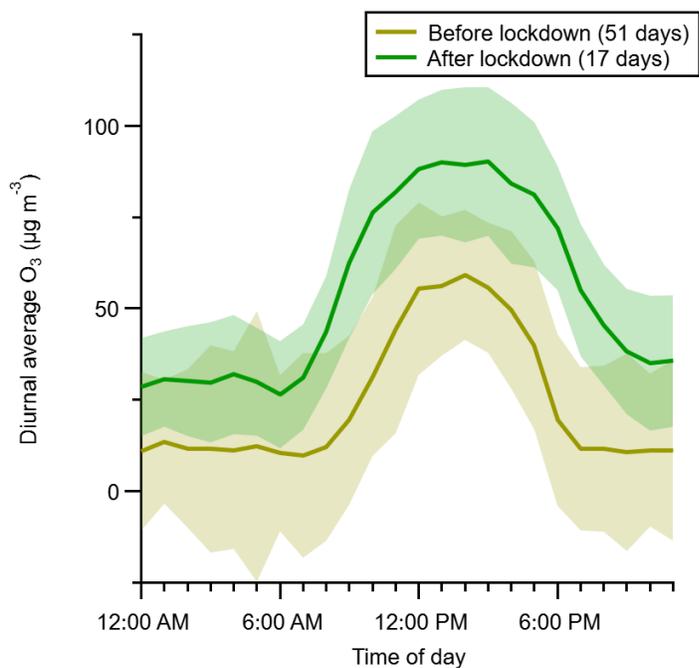
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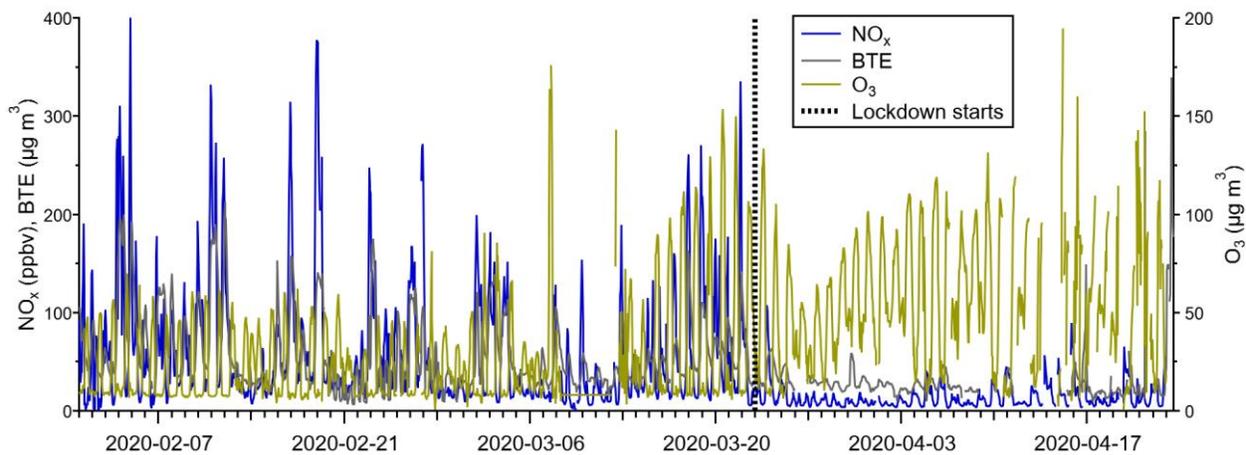
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512 Fig S7: Comparison of diurnally-averaged O₃ pre-lockdown and during phase-I lockdown at RK Puram.
513 Shaded areas represent the standard deviation.

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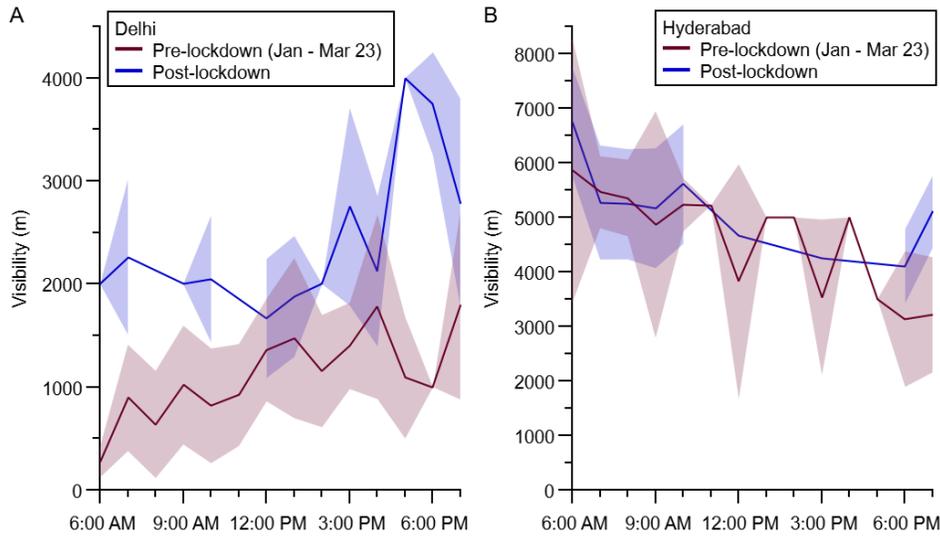
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516 Fig S8: Time series of O₃, NO_x, and the sum of benzene, toluene, and ethylbenzene (BTE) from RK
517 Puram. The vertical line indicates the start of the lockdown.

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522 Fig S9: Comparison of visibility measurements before and after the lockdown at: (A) Safdarjung Airport,
523 Delhi, and (B) Begumpet Airport, Hyderabad.