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Letter

Persistence of Primary and Secondary Pollutants in Delhi: Concentrations and Composition from 2017 through the COVID Pandemic

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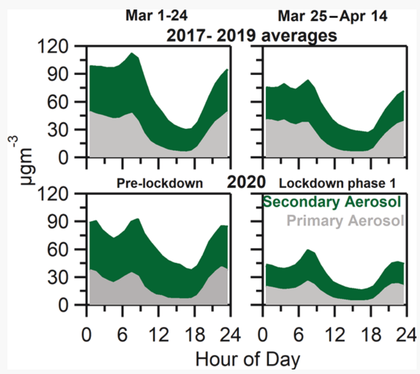
ABSTRACT: We assess impacts of the 2020 COVID-19 lockdown on ambient air quality in Delhi, building on over three years of real-time measurements of black carbon (BC) and nonrefractory submicrometer aerosol (NR-PM_1) composition from the Delhi Aerosol Supersite and public data from the regulatory monitoring network. We performed source apportionment of organic aerosol (OA) and robust statistical analyses to differentiate lockdown-related impacts from baseline seasonal and interannual variability. The primary pollutants NO_x, CO, and BC were most reduced, primarily due to lower transportation emissions. Local and regional emissions such as agricultural burning decreased during the lockdown. $PM_{2.5}$ declined but remained well above WHO guidelines. Despite the lockdown, NR-PM_1 changed only moderately compared to prior years. Differences in the trends of hydrocarbon-like OA and BC suggest that some sources of primary aerosol may have increased. Despite notable reductions in some primary pollutants, the lockdown restrictions led to rather small perturbations in the primary fraction of NR-PM_1, with secondary aerosol continuing to dominate. Overall, our results demonstrate the impact of secondary and primary pollution on Delhi's air quality and show that large changes in emissions within Delhi alone are insufficient to bring about needed improvements in air quality.



■ INTRODUCTION

Delhi, India, routinely experiences particulate matter (PM) concentrations that rank among the very highest in the world.[1] However, the sources and atmospheric dynamics of PM in Delhi remain incompletely understood. One highly policy-relevant knowledge gap is the interplay between local emissions in this megacity region of ∼30 million people and the regional atmospheric chemistry of the broader Indo-Gangetic Plain.[2,3] Numerous major primary sources of PM in Delhi include traffic, biomass burning from cooking and heating, waste burning, construction, emissions from both formal and informal industries (e.g., brick kilns), and electric power generation.[4,5] The fraction of primary submicrometer aerosol (PM_1) in Delhi is high (∼30%−40%) relative to other megacities.[6] However, the generally dominant contribution of secondary PM remains both a matter of surprise in public discourse about air pollution and a vexing challenge for Delhi-centric air pollution control efforts that target primary emissions.

Restrictions in cities around the world aimed to control the COVID-19 pandemic have provided a novel opportunity to study how changes in human activities and their associated emissions affect atmospheric chemistry and air quality.[7−18] In India, lockdowns in 2020 were implemented in four phases, phase 1 (March 25−April 14), phase 2 (April 15−May 03), phase 3 (May 04−17), and phase 4 (May 18−31), with restrictions easing with time (Figures S1 and S2).[19−23] During phases 1 and 2, activities such as transportation and construction were severely restricted, and several factories and businesses were shut down.[24] Power plants in several parts in India operated at a lower capacity or were shut down owing to lower demand.[25−27] These changes offered insights on how future emission reductions might affect air quality in Delhi.

Recent analyses of the COVID-19-related lockdown in Delhi and other Indian cities have been generally limited to a small number of pollutants (e.g., $PM_{2.5}$, O_3, NO_x, CO) that are influenced by a large number of local and regional sources, with one study focused on the $PM_{2.5}$ composition during the lockdown.[23−25,28−37] Here, we utilize a unique long-term (∼3.5 years) measurement data set of submicrometer aerosol (PM_1) composition measured using an aerosol chemical speciation monitor and a multichannel aethalometer as part

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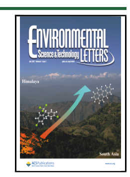

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of the Delhi Aerosol Supersite (DAS) study,[6,38−40] coupled with receptor modeling and pollutant data from the regulatory monitoring sites, to investigate the impacts of the lockdown on both primary and secondary pollutants in the Delhi megacity.

■ MATERIALS AND METHODS

DAS Instrumentation. We have been measuring non-refractory PM_1 (NR-PM_1; submicrometer aerosol that flash vaporizes at 600 °C) using an aerosol chemical speciation monitor (ACSM, Aerodyne Research, Billerica MA)[41] and black carbon using a multichannel aethalometer (Magee Scientific Model AE33, Berkeley, CA)[42] almost continuously since Jan 2017. The site is located at the Indian Institute of Technology, Delhi campus, in New Delhi.[6,38−40] Details on ACSM calibration and data processing are presented in Section S1 of the Supporting Information (SI). For the analysis presented in this article, we used data from March−May across four years, 2017−2020.

Positive Matrix Factorization (PMF). We conducted positive matrix factorization (PMF) analysis for two periods, "before lock down" (March 01−24) and "during lockdown" (March 25−May 31 and March 25−April 25 for 2018) across three years (2018−2020) using the ACSM organic aerosol (OA) spectrum. We determined a three-factor solution (hydrocarbon-like OA, "HOA"; oxidized biomass burning OA, "OBBOA"; and oxidized OA, "OOA") to best represent the data set for all periods/years analyzed (Figures S3 and S4).[43−46] Further details on the PMF runs and factor identification are presented in Section S2 and Figures S3−S7.

Other Data. We retrieved CO, NO_x, SO_2, O_3, PM_{10} ,and $PM_{2.5}$ data from the Central Pollution Control Board (CPCB) with additional processing steps summarized in Section S3. Details on the supporting meteorological, fire count, and electricity production data are also presented in Section S3.

Data Analysis. In core analyses, we used the method of "robust differences" (D-value, eq 1) to compare the concentrations in 2020 to recent trends (2017−2019 average, henceforth referred to as historical concentrations).[47]

$$D_i = (C_{2020,i} - C_{h,i})/I_{h,i} \quad (1)$$

where $C_{2020,i}$ is the weekly median concentration of the daily averaged data in week i, $C_{h,i}$ is the historical median concentration (based on years 2017−2019; 2018−2019 for PMF factors) for $i \pm 2$ weeks, and $I_{h,i}$ is the historical interquartile range (IQR) for $i \pm 2$ weeks. The use of median concentration makes it robust to the presence of outliers, and the use of ±2 weeks (i.e., ∼28 days) of data allows for the smoothing of unusual weeks in the observational record. The D-value metric compares conditions at one time of year over multiple years, thereby assessing the degree to which concentrations are typical or atypical for a given period. For example, a D-value of −2 indicates that concentrations in week i in 2020 were lower than historical concentrations by a factor of 2 IQR, indicating an influence of reduced emissions during the lockdown.

As a supplemental and complementary analysis technique ("percent difference", Figure S8), we computed the percentage difference between concentrations in each phase of the lockdown and the baseline pre-lockdown period (March 01−24). We compared this seasonal evolution in air pollutants for 2020 with that for prior years (Figure S8). Further, we conducted statistical tests to determine significance of the difference in concentrations relative to historical concentrations and concentrations before the lockdown (Figure S9). Overall, the test results compare well with our interpretation of D-values and percentage differences. Thus, while year-to-year differences in meteorology and emissions do provide a degree of irreducible uncertainty in our analyses, the consistency of our results among multiple analytical techniques suggest that they are robust. We focus on D-values in the Results and Discussion and use percent difference data and associated p-values as Supporting Information to complement the D-value method.

■ RESULTS AND DISCUSSION

As shown in Figure 1a and b, with the onset of the lockdown, ambient $PM_{2.5}$, NR-PM_1, CO, NO_x, and BC reduced. SO_2 did not change much, and O_3 increased (see average concentrations in Table S2 and Figure S10a). Notably, most species concentrations were lowest during the first phase (P1) of the lockdown (relative to other phases). While the lockdown restrictions likely had an impact, it is important to note that Delhi experienced rains during that period (Figure 1d) which also contributed to the improvement in air quality. Further, the ventilation and temperature increased during the lockdown (Figure 1d), which can partly explain the observed trend. Concentrations of all particle species are expected to decrease with increased ventilation, and the concentrations of volatile particle-phase species, especially ammonium nitrate and

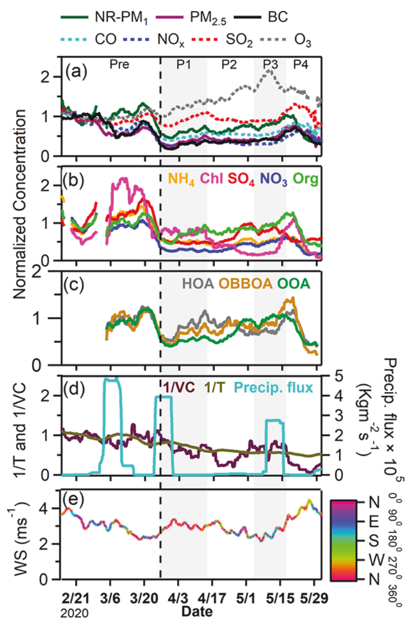

Figure 1. (a) Weekly moving averages of pollutants NR-PM_1, $PM_{2.5}$, BC, CO, NO_x, SO_2, and O_3 before and during the lockdown. P1−P4 correspond to the four phases of lockdown. Weekly moving averages of (b) NR-PM_1 species, (c) OA PMF factors, and (d) meteorological parameters: 1/VC (VC = ventilation coefficient), 1/T (T in °C). The parameters have been normalized to 02/16−02/28 averages; the PMF factors have been normalized to average concentrations during the pre-lockdown period. Weekly moving averages help smoothen out the day-to-day variability and capture the longer-term trends. Normalization helps understand the relative deviation from early spring concentrations. Precipitation is shown in terms of precipitation flux ($Kgm^{-2}\ s^{-1}$). (e) Wind speed in ms^{-1}. Colors represents the wind direction.

ammonium chloride, are also expected to further decrease with increasing temperature (see average NR-PM_1 species concentrations in Table S3 and Figure S10b).[48] Figure S11 shows the evolution of PM_1 (= NR − PM_1 + BC) species during the lockdown in 2020 and compares it to the average historical trends during those periods. The trend of the PM_1 concentration was different in 2020. It was lowest during P1 in 2020, while its decrease was more gradual in the previous years, reaching lowest values during P2, before increasing in P3. Concentrations of all PM_1 species decreased during the initial phases of the lockdown, and the ratio of primary to secondary aerosols did not decrease due to lockdown restrictions (Figure S12d), which mostly impacted emissions of primary pollutants. This is a further testament to the rapid photochemical processing of primary pollutants and importance of secondary aerosol in Delhi.[6,39] Below, we discuss and present explanations for the variability of PM_1 species and other pollutants.

The concentrations of NO_x, CO, and BC were significantly lower than historical concentrations during the initial phases (P1−P3) of the lockdown (Figure 2a−c; *D*-values ∼ −2 or

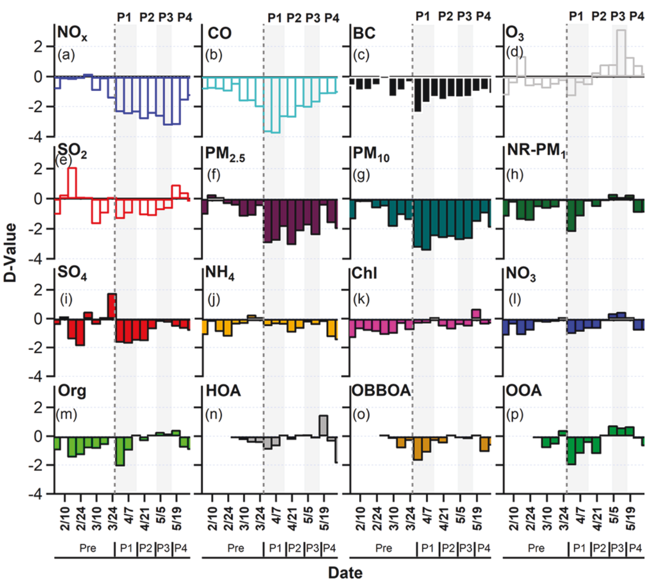

Figure 2. Weekly robust difference *D*-value of pollutants from 02/01 to 05/31 2020, reflecting the changes in air pollutants in 2020 relative to multiyear observational baselines. Data for panels (a) and (b) and (d)−(g) reflect pollutants monitored by the official regulatory network in Delhi, while other panels result from our observations at the Delhi Aerosol Supersite. P1−P4 and associated shaded regions correspond to the four phases of lockdown.

lower; low *p*-values in tests (1) in Figure S9), likely due to reduced vehicular emissions, which is their dominant contributor. In fact, their *D*-values (and those of PM_{10} and $PM_{2.5}$) became negative a few weeks before the lockdown (Figure 2), indicating some reduction in activity before March 25, consistent with the mobility trends in Figure S13 and the nationwide curfew on March 22.[23] While the *D*-values of NO_x remained low through P1−P3, those of CO and BC increased gradually after P1, possibly due to contributions from residential activities such as cooking and biomass burning which contribute relatively less to NO_x, as also indicated by Delhi emission inventories.[49] The ratios of BC/CO and BC/$PM_{2.5}$ (Figure S12) were lowest during P1 and increased gradually, while NO_x/CO kept decreasing until P3, consistent with a change in source mix during the lockdown and increased contribution from residential activities. Historically, NO_x and CO concentrations increased during P3/P4 (Figure S8a, b), despite increased ventilation, likely due to contribution from agricultural burning (the lockdown coincided with the rabi crop burning season from mid April−May[50]), which was significantly reduced this year (see fire counts in Figure S14). Thus, in addition to reduced vehicular emissions, reduced crop burning likely also impacted the concentrations of NO_x and CO in 2020. By P4, the *D*-values of NO_x, CO, and BC returned to near baseline values observed before the lockdown, consistent with high *p*-values in test (2) (Figure S9) and low percentage differences (Figure S8).

During the lockdown, the decrease in NO_x concentration was accompanied by an increase in O_3 concentration (Figure 2a, d). However, in P1 and P2, the O_3 concentration was not substantially higher than in previous years (*D*-value < 1) (Figure 2d). Further, the increase in O_3 concentration going from March to April was also observed in previous years (see temporal trend in Figure S15), In P3, O_3 was higher relative to prior years (*D*-value ∼ 3), and the weekly moving averages (Figure S15) indicate that it continued to increase despite the relatively small temporal changes in NO_x during that period. While this is consistent with VOC-limited conditions,[51] it can also be due to enhanced photochemical processing, given the increase in photochemical activity during that time of the year and the concurrent increases in O_3, organic aerosol (Org), and sulfate (SO_4^{2-}) during the period (Figure 2 and Figure S8). Future VOC measurements are needed to understand ozone formation in the city.

$PM_{2.5}$ and PM_{10} concentrations during the lockdown were lower relative to previous years (*D*-values ∼ −4 to −2) (Figure 2 f, g). While daily averaged $PM_{2.5}$ decreased to less than 60 μgm^{-3} (Indian National Ambient Air Quality standard for 24-h average[52]), on many days it remained higher than the World Health Organization guideline of 25 μgm^{-3} (Figure S16).[53] The relative increase in *D*-value in P4 (consistent with increased *p*-value in Figure S9) was likely due to the influence of crop burning and photochemical processing during that period since the temperature/ventilation favored lower concentrations (Figure 1d). Historically, $PM_{2.5}$ did not change significantly, while PM_{10} increased during the summer (April−May) period (Figure S8f, g), despite increased ventilation and temperature. This may be due to agricultural burning, photochemical processing, and the influence of dust events in summer.[54] Overall, the reduced $PM_{2.5}$ concentrations during the lockdown in 2020 were likely due to decreases in local and regional sources during the period. NR-PM_1 during the lockdown was lower than the historical concentrations only during P1 (*D*-value ∼ −2 to −1 in Figure 2h; high *p*-values in tests (1) in Figure S9 after P1). The different influences of the lockdown on $PM_{2.5}$ and NR-PM_1 could be due to an increased influence of regional transport of crop burning and dust events on larger particles[54,55] and the different locations where these were measured (NR-PM_1 inside the IIT-D campus, $PM_{2.5}$ averaged across several sites in the city).

Among the NR-PM_1 species, ammonium (NH_4^+) and chloride (Cl^-) concentrations during the lockdown (Figure 2j, k) did not differ significantly from historical concentrations (*D*-value ∼ −1 to 1; high *p*-values in tests (1) in Figure S9). This is likely due to the relatively high vapor pressure of ammonium chloride making temperature the most important

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factor controlling its concentrations (Figure S8k). Further, potential chloride sources such as steel processing plants and trash burning[38] likely did not "turn off" during the lockdown period. Nitrate (NO_3^-) was slightly lower than historical concentrations (D-value < −1) during the first week of P1 but not after that (Figure 2l; consistent with the lower p-value in P1 in tests (1) in Figure S9). The lower reduction in nitrate compared to NO_x is indicative of the complex nitrate chemistry in Delhi and is similar to the observations by Sun et al.[56] in Beijing, China, where particulate nitrate decreased less than NO_x during the lockdown. In Delhi, the temperatures during the latter phases of the lockdown were higher (summer) than the temperatures observed during the lockdown in Beijing (winter). At the higher summer temperatures, little inorganic nitrate is present in the particle phase, which may explain the similarity to historical trends for nitrate in Delhi. Sulfate was lower than previous years during P1 and P2 (D-value ∼ −2 in Figure 2i; low p-values in P1 and P2 in test (1) in Figure S9) and relative to the March 01−24 average (by ∼50%−60%; Figure S8i). This could partly be due to reduced coal-based electricity generation in India during the lockdown period (see temporal trend in Figure S17). However, SO_2 was not significantly impacted, indicated by the low percent changes during the lockdown (Figure S8e; consistent with high p-values in test (2) Figure S9) and comparably low D-values before and during the lockdown (Figure 2e). The differences between the trends of SO_2 and sulfate could be due to the fact that that SO_2 hotspots (e.g., coal-fired power plants) are outside of Delhi,[27] and sulfate is more regional than SO_2.[51,57] This contrasts with Beijing, which observed increases in the concentrations of particulate sulfate during the initial period of the lockdown despite reduced SO_2, due to higher relative humidity during this period.[56] The larger excursions in sulfate and nitrate relative to ammonium during the initial phases of the lockdown in Delhi are consistent with higher particle pH during these phases.

Organic aerosol concentrations were lower than previous years during P1 but not after that (Figure 2m; consistent with low p-value in P1 in test (1) in Figure S9). PMF factors offer more detailed insights. The reductions in HOA were lower than those in BC (lower drops in D-value in Figure 2c, n; higher p-values during P2 onward in Figure S9), pointing to the influence of sources other than traffic on HOA, such as diesel-based electricity generators[58] and/or residential cooking which likely increased during the lockdown, as also indicated by the MS of HOA (enhanced m/z 55/57, Section S2). The OBBOA D-value decreased during P1 (Figure 2o) but increased during P2 and P3 due to the influence of agricultural burning. OOA experienced a greater reduction (more negative D-values) than HOA and OBBOA (Figure 2n−p; lower p-values for more phases than HOA/OBBOA in Figure S9), consistent with its fast photochemical production from local primary sources, which were reduced during the lockdown, and regional transport.[6,39] This is also consistent with recent VOC measurements in Delhi, which demonstrated a role of local oxidation of primary emissions on secondary OA formation.[59] The increase in the OOA D-value in P3 was likely due to photochemical processing of emissions from agricultural burning. Thus, unlike Beijing, where the overall OOA contribution increased during the lockdown,[56] in Delhi, it decreased during the initial phases of the lockdown and increased during the latter phases.

Overall, strict activity restrictions helped reduce the emissions from some major sources: transportation, construction, some industries, power plants, and agricultural burning. However, even these strict restrictions did not reduce $PM_{2.5}$ levels to the WHO 24-h air quality guideline of 25 $\mu g\ m^{-3}$. The lower reductions in HOA relative to BC suggest that certain primary aerosol sources such as residential cooking and diesel generators may have increased. The ratio of primary to secondary aerosol did not decrease due to lockdown restrictions, and secondary aerosol continued to dominate. While it is true that emissions and concentrations of primary pollutants including primary aerosol are high in Delhi, rapid photochemical processing as well as regional transport results in even higher concentrations of secondary aerosol. To better understand the formation and processing of secondary PM[60] and O_3, it is important for future studies to include gas phase measurements to characterize VOC emissions.

To address the major air pollution challenge in Delhi, there is an urgent need for integrative, multiscale, and multisectoral policies, which include tackling multiple sources simultaneously (such as local and regional biomass burning, transportation, power plants, diesel generator sets, industrial emissions) and formulating a regional action plan, including but not limited to Delhi.[3]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00211.

> Details on ACSM calibration and data processing (section S1), details on PMF runs (section S2), and details on other supporting data including regulatory meteorological data (section S3), and 17 figures and three tables (PDF)

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Notes

The authors declare no competing financial interest.

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