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Nitrogen Oxides Concentration and Emission change detection during COVID-19 restrictions in North India

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Abstract: COVID-19 induced restrictions resulted in a lowered fine aerosol particle and trace gas concentration across several urban places around the world, providing a natural opportunity to study effects of anthropogenic activities on emissions of air pollutants. Impact of sudden suspension of human activities on air pollution was analyzed by studying the change in satellite retrieved NO₂ concentrations and top-down NO_x emission over the urban and rural areas around Delhi. NO₂ was chosen for being the most indicative of emission intensity due to its short lifetime of the order of a few hours in the planetary boundary layer. We present a robust temporal comparison of Ozone Monitoring Instrument (OMI) retrieved NO₂ column density during the lockdown with the counterfactual baseline concentrations, extrapolated from the long-term trend and seasonal cycle components of NO₂ using observations during 2015 to 2019. NO₂ columns in the urban area of Delhi experienced an anomalous relative change ranging from 60.0% decline during the Phase 1 of lockdown (March 25–April 13, 2020) to 3.4% during the post-lockdown Phase 5. In contrast, we find no substantial reduction in NO₂ concentrations over the rural areas. To clarify the quantitative impact of the lockdown measures, weekly top-down NO_x emissions were estimated from Tropospheric Monitoring Instrument (TROPOMI) retrieved NO₂ by accounting for horizontal advection derived from the steady state continuity equation. NO_x emissions from urban areas and power plants exhibited a mean decline of 72.19% and 53.4% respectively in Phase 1 compared to the pre-lockdown business-as-usual phase. Due to absence of confounding emission source activity during lockdown, emission estimates over urban areas and power-plants were validated with respective electricity generation reports and Google's mobility reports. A higher anomaly in emission estimates suggests that comparison of only concentration change, without accounting for the dynamical and photochemical conditions, may mislead evaluation of lockdown impact. Our results are also useful for optimizing bottom-up emission inventories.

Keywords: NO_x, lockdown; top-down emission; TROPOMI; Emission inventory; power plant

1. Introduction

Movement restrictions or lockdowns have emerged as the most popular social policy to prevent COVID-19 transmission. This policy has induced natural experiment on a global scale with spillover social¹, economic^{1,2} and environmental impacts³⁻⁵. For environmental impacts, natural experiments like volcanic eruptions and unintended anthropogenic modifications like ship tracks have been utilized for constraining unknowns in understanding the role of meteorological conditions, emissions and atmospheric chemistry on air pollutant concentrations^{4,6}.

Early lockdown in January and February in China resulted in decreased economic activities, revealing a decline in regional pollutants, specially the nitrogen oxides ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$). NO_x forms a key short-lived (2-7 hours) tropospheric air pollutant in urban areas that is produced primarily from fossil fuel combustion. Primary emission mostly takes place in the form of NO which then gets oxidized through chemical and photolytic pathways to NO_2 , collectively referred to as NO_x . Investigating NO_x concentrations gained an additional importance during COVID-19 related lockdowns because it allowed clear estimation of the impact of local policy actions as it is less prone to long-range transport owing to its short life-time. NO_2 concentrations in Chinese cities decreased by up to 50% to 60% as measured by ground monitors^{7,8}, and Ozone Monitoring Instrument (OMI) and TROPospheric Monitoring Instrument (TROPOMI) satellite retrieved columns^{4,6}. Strong declines in NO_2 were also observed in Europe, South Korea, and the United States^{5,9}. Investigating sources of NO_x is important due to their direct and indirect health impacts¹⁰. Indian cities went under varying intensities of 'lockdown' severity since March 25, 2020. About 60% to 90% decline in surface NO_x concentrations was observed at a road-site in Delhi during the first week of lockdown compared to the pre-lockdown week^{11,12}. Similar large reductions were seen across other large cities in Northern India^{13,14}. A review of change in pollutant concentration across Indian cities as measured by ground monitors is summarized elsewhere^{13,15}.

However, short-term NO_x concentrations are regulated not only through the emissions but also non-linearly through meteorology and atmospheric chemistry. Therefore, it remains hard to conclude how much reductions can be attributed to the lockdown restrictions^{4,9,13,16}. NO_2 concentrations have an inverse linear relationship with boundary layer height and wind speed within the boundary layer due to upliftment of air mass¹⁷. These meteorological effects are manifested through seasonality of NO_x concentrations. Long-term trends and seasonality in NO_x concentrations have been extensively studied over India using ground monitoring stations and satellite retrieved datasets^{18,19}. These trends are associated with rapid economic growth translating to rapid urbanization, increased vehicle population and density, increase in industries and industrial output. At the same time policies measures have also been directed to reduce emissions by implementing stricter emission norms for vehicles and industries.

We hypothesize that considering such seasonality and long-term changes, lockdown impacts towards changing the NO_x concentrations can be ascertained by assessing both the change in concentration as well as emissions. So far, a handful of studies have systematically discussed the impact of lockdown on NO_x emissions by considering the effect of wind fields on pollutant transport²⁰⁻²², for example by simulating wind trajectories using for example, NOAA's HYSPLIT model. To assess the impact of lockdown in northern India, we adopt a novel methodology to estimate the impact of lockdown on emissions, whereby top-down emissions were calculated by incorporating spatial changes in NO_2 flux due to wind vector fields. We assessed weekly trends in concentration in the context of 2015-2020 satellite records as well as top-down NO_x emissions. We perform this by spatially analyzing satellite retrieved NO_2 datasets. Our work thus contributes to untangling the emission change from concentration change.

The objective of this research is to use a time-series of satellite retrieved NO_2 columns to investigate the temporal and spatial impact of COVID-19 induced restriction measures on NO_2 concentration and NO_x emission over a polluted region in Indo-Gangetic Plains (shown in Figure 1). Our novelty lies in quantifying the impact of lockdown robustly through satellite derived top-down NO_x emissions which were calculated by applying a steady state continuity model to NO_2 column imagery. Estimating the impact of reduced human activities on nitrogen oxide emissions and its

concentrations can help in constraining the emission inventories and segregating the contribution of the regional emission sources.

2. Methodology

The analysis flowchart of the study is summarized in Figure S1.

2.1. Study Location and policy restrictions

The study domain is defined over a region (74°E, 27°N - 80°E, 30°N) in the Indo-Gangetic Plains (IGP) as shown in Figure 1. The domain is notable for its high air pollution, dense settlements and polluting industries. It includes diverse area and point emission sources such as densely populated urban Delhi (urban center: 77.21°E, 28.60°N), smaller cities such as Meerut (urban center: 77.70°E, 28.98°N), Mathura (urban center: 77.67°E, 27.49°N), Aligarh (urban center: 78.07°E, 27.87°N), strongly emitting power-plants at Dadri (location: 77.61°E, 28.60°N; capacity: 2600 MW), Harduaganj (location: 78.13°E, 28.01°N; capacity 610 MW) and Jhajjar (location: 76.35°E, 28.49°N; capacity: 1320 MW), an expressway network radiating outward from the center, as well as rural background regions not lying downwind of emission sources (sample region bounded by 75.38°E, 29.38°N - 75.13°E, 29.90°N). In the domain, climatic conditions vary from a mean low temperature of 3°C during winter (December to February) to a high of 45°C during the pre-monsoon summer (March - June).

In India business-as-usual (BAU) prevailed prior to March 22, 2020 as there was no movement restriction pertaining to COVID-19. The lockdown was implemented over India in several phases starting from March 25 (week 13) and lasting till May 31. Phase 1 of the lockdown consisted of the strictest restrictions followed by relaxation of restriction intensities in subsequent phases. From June 1, 2020 the lockdown was gradually rolled back (Phase 5). The timeline of the lockdown phases with regards to restrictions on the common emission sources is shown in Table S1.

Figure 1. (top) Dominant land cover types in the study domain (74°E, 27°N - 80°E, 30°N) are urban areas, croplands and desert shrublands. (bottom) Mean NO2 column density (TROPOMI) in the week before the enactment of Phase 1 of lockdown March 16-20 (week 12), 2020. Location of representative

urban area (Delhi) and rural background (Fatehabad) are shown in box, in addition to other prominent neighboring emission sources (power-plant at Dadri and Harduaganj).

2.2. Datasets

2.2.1. OMI (2015-2020) and TROPOMI (2019-2020) retrieved tropospheric NO₂ column

Tropospheric NO₂ column retrievals were obtained from the OMI on-board Aura satellite²³ and the TROPOMI on-board Copernicus Sentinel-5 Precursor satellite²⁴. NO₂ from OMI (henceforth referred to as OMI NO₂) is available since 2004 at a pixel size of approximately 13 × 24 km². While offline and near-real time NO₂ from TROPOMI (henceforth referred to as TROPOMI NO₂) has become available only recently (since mid-2018) but at a higher spatial resolution of 6 × 7 km². OMI NO₂'s 30% cloud-screened daily level 3 product was downloaded from NASA's Giovanni portal²⁵. Level 3 TROPOMI NO₂ product was cloud screened (by considering pixels with effective cloud fraction less than 20%) and analyzed at Google Earth Engine cloud platform²⁶. Both OMI NO₂ and TROPOMI NO₂ follow a similar NO₂ column retrieval algorithm. First slant columnar densities (SCD) are retrieved by differential optical absorption spectroscopy (DOAS) fitting of the backscattered radiance to lab-measured absorption spectra of atmospheric gases such as NO₂, O₃, water vapor and others. From the SCD, tropospheric vertical column densities (VCD) are isolated by removing the stratospheric contribution and applying air mass fractions²⁷. Typically, OMI and TROPOMI retrieve NO₂ at approximately 13:30 hours (local time). Aggregation of the daily concentration (from Monday to Sunday) in each week to their weekly mean concentration was done to overcome the periodic weekly cycle of increase (on weekdays) and decrease (on weekends) in NO₂ concentrations³².

2.2.2. Reanalysis meteorological data

Wind field vectors (w) at 80 meter altitude were used for estimating NO₂ flux. The meteorological fields were obtained from National Center for Environmental Prediction (NCEP) FNL (Final) Operational Global Analysis and Forecast gridded datasets²⁸ and are available at 0.25° × 0.25° spatial resolution every 6 hours. To ensure compatibility with satellite datasets, analysis was performed with forecasts for 13:30 hours (local time). The time series of meteorological parameters over Delhi is shown in Figure S2.

2.2.3. Activity data

Although updated or real-time emission estimation is seldom available in most countries, human activity in the form of energy production and travel behavior change reports serve as reliable proxies for changes in emission activities²⁹. We used daily total electricity production tables available for each power-plant, from the Power System Operation Corporation Limited³⁰ to infer the emission from coal-based thermal power plants situated around Delhi, Harduaganj and Dadri. Since February 2020, percentage reduction in daily mobility patterns from the baseline levels, aggregated for administrative state are available from Google's Community Mobility Reports (Google n.d.). The reports show the movement trends by region, across different categories of places. The defined baseline period represents median value from the 5-week period January 3 to February 6, 2020. Out of the available patterns 'workplace related travel' behavior reduction was used based on the assumption it represents the most common vehicle usage in the study domain.

2.3. Concentration change estimation

NO₂ concentrations over Indian cities in Indo Gangetic Plain (IGP) are known to exhibit annual seasonality and long term trends¹⁸. The seasonality can be explained by the seasonal variation in emission activity and source strengths, NO₂ lifetimes due to chemistry of hydroxyl radical (OH) and photolysis under changing solar zenith angle, and meteorological effects like dispersal, wet and dry deposition processes. Whereas long term trends are in response due to gradual changes in

anthropogenic emissions from industries and vehicles owing to technological, economical and policy related changes.

Owing to the limited temporal record of TROPOMI NO₂, it was used only for comparing the spatial changes in NO₂ column densities between 2019 and 2020. Mean NO₂ concentrations of 2019 for corresponding weeks of the year in 2020 were used as reference concentrations for TROPOMI's spatial anomaly calculation. Although data from just one year is insufficient to represent the weekly baseline concentrations, it still can account for broader seasonality caused due to the changing solar zenith angle to reveal spatial patterns in NO₂ anomaly.

The OMI NO₂ with its coarser spatial resolution but longer temporal record, was used for obtaining the expected baseline concentration in 2020, C_B if business-as-usual conditions had continued. $C_B(t)$ was extrapolated from the low frequency annual trends and high frequency seasonal cycle decomposed from the mean weekly satellite retrieved NO₂ columns C_o during 2015 to 2019. The decomposition was performed using the 'seasonal and trend decomposition' (STL), while extrapolation was performed by exponential trend smoothing (ETS) functions available in R language. To ensure that STL decomposition was not affected by outlying pollution episodes, concentrations higher than 99.5 percentiles were discarded. Box-cox transformation was performed on the concentration prior to STL decomposition to ensure they are normally distributed. Pollution episodes that are non-periodic or random interventions could not be decomposed statistically and were identified as residues. ETS extrapolation was repeated using bootstrap sampling with 1000 draws to obtain the confidence interval of the predictions. Due to computation requirements for STL decomposition and the ETS extrapolation, expected C_B were calculated only over urban Delhi and the background rural region (location bounds are as specified in the previous Section).

2.4. Top-down emission change retrieval

Several methods exist for deriving NO_x emissions using satellite retrieved columns: optimizing the priori available emission inventories with satellite derived top-down constraints on NO_x emissions³³, inversely fitting the dispersion model on the concentrations over the affected region to derive emissions from the source region^{34,35} or applying mass conservation based continuity equation³⁶. Constraining emission inventories present a compromise between the high spatial resolution of TROPOMI and the coarser model simulation, especially close to strong emission sources³⁴. Dispersion models require a careful definition of the region affected by plume advection, by choosing the product of wind-speed and the NO_x lifetime³⁴. Emissions thus calculated are underestimated if the region is not large enough to contain most of the emitted NO_x, or overestimated should the NO_x be influenced by other strong sources outside or inside the region³⁴. In practice such conditions are hard to meet over linear or area-based emission sources. As seen in the mean NO₂ column in Figure 1, advected plumes overlap with downwind emission sources.

With such challenges, continuity equation models are better placed to estimate emissions from multiple point-sources if wind vectors are adequately available. They are based on the energy conservation and mass balance for a fluid in steady state at each location. Furthermore, such models do not require defining a region around each emission plume and allow emission estimation over each pixel unlike dispersion models. Our approach is inspired from Beirle et al.,³⁶, in which using a continuity equation the divergence (D) of NO_x emission flux can be related with the sink (S) and the emission (E) as $D = E + S$. The daytime NO_x sink is constituted by reaction of NO₂ with OH to form HNO₃³⁷. The total emission flux along meridional and zonal wind fields, w , is given as LCw , where C is the daily TROPOMI NO₂ column, and L is the ratio of NO_x and NO₂. Assuming a first order reaction of NO₂ with a time constant τ , S is given as LC/τ . E from using the mass balance is shown in Equation 1.

$$E = \frac{LC}{\tau} + \nabla(LCw) \quad 1$$

τ corresponds to the average lifetime of NO_x in the boundary layer, depending on factors like the photolysis rate, length of night, OH concentrations and meteorological conditions such as wind,

relative humidity and temperature³². It is of the order of one day but varies by about one order of magnitude with time and place. In tropical regions τ varies from 7 hours in January to about 4 hours in July³³. Since the domain lies in a subtropical and semi-arid region, τ was regressed linearly between 7 to 4 hours depending on the day of the year. L accounts for the partitioning of NO_x into NO and NO₂ in the polluted layer by serving as a surrogate for NO to NO₂ chemistry³⁸. It depends on ozone concentration, NO₂ photolysis rate and temperature. Close to the surface, NO_x is present as NO₂ except when close to strongly emitting sources where conversion of NO to NO₂ is limited by ozone concentration. A typical value of L as 1.32 was assumed for the urban noon-time pollution conditions^{36,37}.

Theoretically the divergence term, $D(= \nabla(LCw))$ should be positive over emission sources and negative elsewhere due to the chemical sink. Following Equation 3, an adequately specified chemical loss sink $S(= LC/\tau)$ numerically compensates the negative D to result in a net zero emission rate E over non-emission source field. However, if the S is underestimated such as by assuming τ larger than actual lifetime, we would incorrectly obtain negative E .

Daily meridional and zonal fluxes were stacked and mean averaged for each week. Pixels masked with cloud masks were not considered. The sum of divergence of mean weekly flux and the sink of mean weekly NO₂ column was used to estimate weekly emission rate over each TROPOMI pixel. As the τ of 4 hours was derived for pollution on top of the tropospheric background concentrations³⁹, the sink term S would be biased high in absence of the background correction. A background correction was applied for the NO₂ columns by removing the 5th percentile of all columns in the domain³⁶.

Emission rates over urban Delhi and power-plants at Dadri and Harduaganj were estimated for comparison with activity reports. A region of spanning 50×50 km² (16×16 TROPOMI pixels) around urban Delhi, and 20×20km² (6×6 TROPOMI pixels) regions around Dadri and Harduaganj power-plants were selected. The region for calculating emission rate was chosen to avoid overlapping influence from any other strong source. As the continuity model performs mass balance for each pixel, while selecting the region no special consideration was taken regarding the distance to which the plume was advected. Since the region around the power-plants is devoid of other emission sources, background emissions were assumed to be not more than 5 percentiles of the region's emission.

2.5. Anomaly metric

An anomaly metric, ΔC_t , for a week t , was defined as per Equation 2, to quantify the relative change between the mean weekly concentrations (or emissions) in 2020, $C_{2020,t}$, and the corresponding mean reference values, $C_{reference}$.

$$anomaly = \frac{C_{2020,t} - C_{reference,t}}{C_{reference,t}} \quad 2$$

A high absolute anomaly implies a large deviation in C_{2020} with respect to the reference $C_{reference}$ in the week t . Estimating anomaly as per the Equation 2 also offers the benefit of removing systematic additive or multiplicative biases in satellite retrievals. Anomalies in OMI NO₂ concentration in 2020 were calculated with respect to extrapolated concentration C_B (defined in Section 2.3) as reference. Anomalies in top-down NO_x emissions in 2020 were calculated both with emission in 2019 as reference as well as with mean BAU emission as reference. During the lockdown weeks, anomaly was expected to be negative with large absolute values.

3. Results and discussion

3.1. Concentration change during lockdown

The comparison of the NO₂ column in 2020 with that in 2019 suggests a sharp decline in NO₂ densities in urban areas with Phase 1 and a subsequent slow recovery. Mean anomaly of TROPOMI NO₂ column in 2020 with respect to 2019 for each lockdown phase is shown in Figure S3. Anomaly during BAU was 0.01% ($\pm 11.0\%$), reflecting that prior to lockdown overall densities were not

different from the baseline, except at Rajiv Gandhi power-plant that showed a high positive anomaly (37.4%). During Phase 1 lockdown, overall densities had decreased substantially with a large negative mean anomaly ($-33.7\% \pm 12.1\%$), going as low as -76.8% over central urban Delhi. Apart from urban areas relatively large negative anomalies were also found above power-plants and industries suggesting reduced emissions compared to 2019. During Phase 2, overall anomaly was reduced to $-2.1\% \pm 17.6\%$ on account of a sharp difference in anomaly between the rural western region (28.6%) falling in Rajasthan and the populated central (-58.4%) and eastern regions (-15.3%). A similar spatial dichotomy in anomalies was also found during Phase 3 with urban Delhi showing a 45.6% decline while rural areas showing a 36.5% increase. High positive anomalies observed in the western desert region can be attributed to the small NO_2 columns values wherein; slight changes in concentrations appear as large anomalies. Overall anomaly during Phase 3 was $-7.2\% \pm 31.3\%$. During Phase 4, when most restrictions had been relaxed, NO_2 density anomaly in Delhi reduced to -37.7% from the -76.8% during Phase 1. For the first time since the Phase 1 onset, concentration increased over the Panipat region, which has a power-plant and an oil refinery closely located to each other, showed a positive anomaly of 37.3% . Furthermore, Phase 4 coincided with the large-scale open wheat crop-residue burning in the northern regions of the domain, resulting in positive NO_2 anomalies. The overall anomaly for Phase 4 was $-4.5\% \pm 15.4\%$. During Phase 5, which saw no restrictions except in containment zones, density anomalies remained lower than that during BAU, suggesting that concentrations had still not recovered to the same levels as those in 2019 with overall anomaly as $-17.5\% \pm 12.3\%$. Lower anomalies may also have been partly contributed by unusual precipitation that took place during the latter half of June 2020⁴⁰. The eastern-western spatial dichotomy was not apparent during Phase 5. Interestingly, negative anomalies were persistent over most of the urban region of Delhi and downwind regions of strong emitters such as power plants and refineries during all the phases while fluctuating in other regions.

However, the reduction in concentration cannot be conclusively calculated without adjusting for long-term trends. Reductions in concentrations were further analyzed by calculating anomaly with respect to the expected concentration. Furthermore, spatial differences in anomaly in the western and eastern portion of the domain suggests possible varying influence of wind speeds and direction in dispersing away the pollutants. This confounding by wind was addressed by estimating the impact on emissions (Section 3.2).

(a) Observed, expected NO_2 over urban region

(b) NO_2 anomaly over urban region

(c) Observed, expected NO₂ over rural(d) NO₂ anomaly over rural region

region

Figure 2. Mean weekly observed and predicted NO₂ columns (left) and their anomaly between the observed and predicted columns (right) for Delhi (top) and rural Fatehabad (bottom). The anomaly range corresponds to 95% prediction intervals. Anomaly metric was calculated following Equation 1.

Figure 2 shows the retrieved OMI NO₂ column as well as the expected OMI NO₂ column over urban and the rural location and their corresponding anomalies for both the locations. Based on the STL decomposition, a small but significantly decreasing NO₂ linear trend from 2016 to 2019 was found over the urban Delhi ($-0.142 \times 10^{15} \text{ yr}^{-1}$) as well as rural Fatehabad area. Seasonality of NO₂ after STL decomposition shows a high NO₂ in the winter months (January and February) over Delhi and Fatehabad that gradually decreases with pre-monsoon summer. The seasonality corresponding to the photolysis rate, life-time and change in emissions. The photolysis rate of NO₂ is proportionally related with actinic flux, which increases with decreasing solar zenith angle (as well as depending on direct, diffused and reflected radiation governed by amount and type of the aerosols, absorbing gases, air molecules and the surface albedo¹⁸. A lower actinic flux during winter results in a lower NO₂ photolysis frequency and OH radical production rate, thereby reducing the photochemical loss of NO₂ and increasing its lifetime³². As the pre-monsoon dry summer with its high temperature starts approaching, actinic flux increases, thereby decreasing the lifetime of NO_x and lowering NO₂ column densities. Further with onset of monsoon, wet deposition reduces the NO₂. As seen in Figure 2 (a) and (c), observed and expected NO₂ column densities during the BAU phase were similar, as evidenced by their small mean NO₂ anomalies existing over Delhi (15.7%) and Fatehabad (20.3%) respectively. However, when the lockdown commenced, urban NO₂ showed a sharp deviation from the expected densities. During the Phase 1 of lockdown, similar NO₂ column values were observed in urban Delhi and rural Fatehabad (Figure 2 (a) and (c)). The greatest negative anomaly over Delhi was observed during the first week of the Phase 1 as -64.51% (-56.9% to -70.2%). However, for the rural Fatehabad a positive anomaly was observed during the first week of Phase 1 as 11.2% (-4.7% to 31.8%) which during the second week of Phase 1 dipped to -62.1% (-68.0% to -54.3%).

By comparing mean anomalies across lockdown phase and locations (Table 1), it becomes clear that urban Delhi showed larger anomalies than rural Fatehabad and its reduced concentrations persisted for a longer duration than Fatehabad. Mean anomalies during Phase 2 to Phase 4 in Delhi stayed between -31% to -27%, suggesting slight recovery in emissions compared to Phase 1 reductions. During the same duration Fatehabad had lower mean anomalies between -8% to -15% suggesting that impact of the policy restrictions on human activity and consequently NO₂ was not uniform in urban and rural areas. In urban areas, industrial and transportation activities did not resume to pre-COVID levels even during the unlock Phase 5 resulting in overall negative anomalies. However, in rural areas consistent smaller anomalies could be related to their permission to conduct agricultural activities (such as harvesting and transport) as usual as the inclusion of agricultural activities within 'essential services' excluded them from lockdown restrictions. Another reason could be that contribution of anthropogenic emissions from rural household biomass fuel burning as well as biogenic emission from agricultural soil¹⁸ kept on contributing to rural NO₂ columns despite the lockdown. Sporadic thunderstorms reported in the region between March and May have increased tropospheric NO₂ as lightning can contribute up to 0.4×10^{15} molecules cm^{-2} in tropical continents⁴¹.

Table 1. Mean phase-wise anomaly between the observed and predicted NO₂ concentrations in 2020. The range corresponds to 95% prediction intervals.

Phase	Urban (Delhi)	Rural (Fatehabad)
BAU	15.7% (0.0% to 36.3%)	20.3% (5.6% to 44.0%)
Phase 1	-61.0% (-67.0% to -53.0%)	-33.5% (-43.3% to 20.7%)
Phase 2	-31.1% (-42.0% to -19.3%)	-7.7% (-22.0% to 10.8%)

Phase 3	-30.4% (-40.0% to -17.4%)	-15.0% (-28.6% to 1.8%)
Phase 4	-27.0% (-37.0% to -13.4%)	-10.5% (-25.0% to 6.6%)
Phase 5	-3.4% (-18.4% to 16.2%)	6.2% (10.4% to 27.3%)

3.2. NOx emission change during lockdown

Top-down emissions change during each phase in Figure 3 depicts a more accurate policy impact on NO₂ compared to change in concentrations by overcoming the effect of wind on plume advection. Compared to BAU, substantial emission reduction was observed over urban Delhi as well as surrounding areas. As investigated earlier^{42–44} (also discussed in supplementary file), NO_x emission in Delhi is mostly contributed by traffic and industries, amounting to 58% to 80% of total emissions. Apart from the urban areas, emissions from Harduaganj and Dadri power-plant as well as the oil refinery in Panipat were reduced substantially. Emission rates were similar in Phase 1, 2 and 3. During Phase 4, when most restrictions had been removed, emissions increased in areas within and surrounding Delhi as well as at the Panipat oil refinery. Such resumption in oil refining activities was also been reported in newspapers⁴⁵. Also, several scattered weakly emitting point sources in northern Haryana, possibly corresponding to crop residue fires are visible. During the unlock Phase 5, emissions remained mostly similar to Phase 4, except at Panipat oil refinery where they had decreased compared to Phase 4 but higher than Phase 1.

Figure 3. Phase-wise mean top-down NO_x emission in 2020 during (a) BAU (business-as-usual), and subsequent lockdown phases (b)-(f).

A time-series comparison of top-down emission with activity reports for Delhi (Dadri power-plant and Harduaganj power-plant) is shown in Figure 4 (Figure S4). Interestingly, weekly emission from urban areas showed a slight decreasing trend between January and prior to lockdown in March. It is possible that relatively higher emission estimation in January was due to biomass burning for

heating. It is also possible that emission is slightly underestimated in March due to the assumption of a higher than actual τ , which may be unlikely due to an increased photolysis rate during March. Over these three locations (urban Delhi, Harduaganj power-plant and Dadri power-plant), correlation between the reported activity and top-down emission is higher than the correlation between the activity and concentration (Table S2). This confirms that changes in top-down emissions assess the lockdown impacts better than the concentration since the emission overcome confounding from wind-field related meteorological effects. However for power-plants, a small difference between activity-concentration correlation and the activity-emission correlation suggests that over strong point sources lockdown impacts can be equally informed by changes in either emissions or concentrations. A high correlation between the emission and the mobility activity over the urban area (0.96) re-confirms that most NO₂ emissions are related with people's movement activities from transportation as was also suggested in previous bottom-up emission inventories studies.

Figure 4. Mean relative activity, NO_x emission and NO₂ concentration standardized with respect to mean values in the first week of January 2020 (6 January – 12 January, 2020) at Delhi

Emission anomalies were calculated by comparing the emission during lockdown phases with the mean emission in the three weeks prior to the lockdown. Emission during Phase 1 decreased by 72.19% over the urban area which by Phase 5 had recovered to 49.5% decrease. At the same time, compared to the baseline, workplace mobility had decreased by average 80.6% in the initial Phase 1 and by 45.5% in Phase 5. At the Harduaganj and Dadri power-plants, lower demands led to a power production decrease by 20% and 55% respectively. At the corresponding locations mean emission decreased by 53.4% and 48.5% during Phase 1 and Phase 2 compared to BAU.

Emission anomalies in 2020 were also calculated with respect to the corresponding emissions of 2019 as reference. Emissions anomalies so calculated are visualized spatially for each phase in Figure S3 and summarized for Delhi in Table 2. As transportation and factories came to standstill during the Phase1, for Delhi about 72% NO_x emissions can be attributed solely to the traffic and factories.

Table 2. Mean top-down NO_x emission ($\pm$ standard deviation) in 2020 and its anomaly with respect to 2019 emissions over a 60 $\times$ 60 km² region around central Delhi (77.21 E, 28.60 N).

Phase	mean emission (kg/sec)	anomaly compared to 2019
BAU	2.78 $\pm$ 0.43	-17.3%
Phase 1	0.61 $\pm$ 0.13	-72.0%
Phase 2	0.62 $\pm$ 0.09	-63.2%
Phase 3	0.85 $\pm$ 0.29	-50.3%
Phase 4	1.30 $\pm$ 0.72	-29.4%
Phase 5	1.05 $\pm$ 0.18	-40.6%

3.3. Uncertainty in satellite retrieved tropospheric NO₂ and top-down NO_x emissions

Tropospheric NO₂ retrievals can suffer uncertainty in slant column density (due to measurement noise and spectral fitting errors), stratospheric slant column (due to error in separating stratospheric and tropospheric NO₂) and tropospheric AMF (due to model parameter errors such as assumed profile shape⁷). Trace gas profile is needed to derive VCD by separating AMF from SCD. Column

uncertainty due to AMF is about 30%⁷. If the satellite retrieval assumed NO₂ height profile has a smaller aerosol fraction close to surface compared to the true profile, then tropospheric AMF will be overestimated and correspondingly the retrieved tropospheric NO₂ VCD will be underestimated⁷. Shaiganfar et al., (2011) compared OMI NO₂ VCD with MAX-DOAS observations and found that OMI underestimates high values over Delhi. Over highly polluted regions tropospheric NO₂ VCDs are partially underestimated due to shielding of emitted NO₂ by aerosols¹. The random errors are reduced by temporal and spatial averaging, while a major part of systematic errors is expected to cancel out through the difference and ratio in the defined anomaly metric.

Estimation of total emission is complicated mainly by variation of wind field and chemical transformation^{2,8}. As long as wind has a constant speed and direction and these parameters are known with low uncertainty. However slow winds which change directions with time and space complicate emission estimation as such scenarios have high uncertainty (relative to wind speed) and the sudden change in wind direction breaks the steady-state assumption. However, the error due to non-stationary state is smaller near point sources and that further diminishes by a multi-temporal mean. Another problem is that while the wind speeds and directions change with altitude, the vertical profile of the trace gas itself is not well-known. For example, the near-surface emissions (from vehicles) and chimney-stack emissions are injected at different altitude and their dispersion is subjected to different wind speeds. Chemical reactions may also add uncertainties in the assumed lifetime and the constant Leighton ratio. As stated earlier NO₂/NO_x ratio is lower when close to the freshly emitting source in space and time, which increases in aged plumes after chemical conversion⁹. Near strong emission sources emitted NO may not be quickly converted to NO₂ if the NO mixing ratios locally exceed those of O₃. The NO/NO₂ steady state will be completely achieved only after ambient air has mixed with the emitted plume¹. Turbulence in wind speed, may enhance mixing and conversion of NO to NO₂ near the source or it may disperse NO to downwind before it gets converted to NO₂⁹. We assumed a constant ratio of NO/NO₂, neglecting possible changes by day of the week. We also assumed that chemical loss of NO_x is first-order given by a constant lifetime τ and neglecting possible change in τ due to seasonality in actinic flux, different ozone concentrations on weekends¹⁰ or during the lockdown¹¹ and dilution by wind.

4. Conclusions

The natural experiment induced by COVID-19 lockdown was used to assess the impact of lockdown on NO_x concentration and anthropogenic emissions in a region in Northern India around Delhi. To estimate the decline in NO₂ during the lockdown, seasonality and trend decomposition was used to calculate the expected NO₂ in 2020 by extrapolating it from previous years. Compared to the expected concentrations, NO₂ concentration in urban areas reduced by 60% in Phase 1 of lockdown to 3% in Phase 5 of lockdown. In contrast, the rural areas exhibited much smaller and short-lived reductions suggesting a lower impact of the movement restriction policies. To estimate the detailed impact of the lockdown top-down emissions were estimated spatially using steady state continuity equation by considering the transport of pollutants. The calculations suggest compared to the business-as-usual phase of 2020, emission over the urban area and power-plants decreased by 72.19% and 53.4%, respectively during the Phase 1 of lockdown. By Phase 5, emissions over urban areas had recovered, showing a decrease of 49.5% compared to business-as-usual phase of 2020. Higher correlation of activity reports with emission compared to concentrations, suggested that top-down emissions are representative for estimating the impact of lockdown specially over urban areas. This analysis can be extended to link the varying impacts of diverse policy regulations on emission sources and consequently on NO_x emissions

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Data availability: Publicly available datasets were. TROPOMI was processed and downloaded from Google Earth Engine, OMI was downloaded from NASA's Giovanni platform and meteorological data obtained from NCAR's Research Data Archive. Original code was developed by the authors for top-down emission estimation in Python language and it is available from the corresponding author upon request.

Figures

Figure 1

(top) Dominant land cover types in the study domain (74°E, 27°N - 80°E, 30°N) are urban areas, croplands and desert shrublands. (bottom) Mean NO₂ column density (TROPOMI) in the week before the enactment of Phase 1 of lockdown March 16-20 (week 12), 2020. Location of representative urban area (Delhi) and rural background (Fatehabad) are shown in box, in addition to other prominent neighboring emission sources (power-plant at Dadri and Harduaganj). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

(a) Observed, expected NO₂ over urban region

(b) NO₂ anomaly over urban region

(c) Observed, expected NO₂ over rural region

(d) NO₂ anomaly over rural region

Figure 2

Mean weekly observed and predicted NO₂ columns (left) and their anomaly between the observed and predicted columns (right) for Delhi (top) and rural Fatehabad (bottom). The anomaly range corresponds to 95% prediction intervals. Anomaly metric was calculated following Equation 1.

Figure 3

Phase-wise mean top-down NO_x emission in 2020 during (a) BAU (business-as-usual), and subsequent lockdown phases (b)-(f).

Figure 4

Mean relative activity, NO_x emission and NO₂ concentration standardized with respect to mean values in the first week of January 2020 (6 January – 12 January, 2020) at Delhi

Supplementary Files

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- [20201125NO2DELHISupplementaryPM.pdf](#)