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Impact of reduced anthropogenic emissions during COVID-19 on air quality in India

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Abstract. To mitigate the impacts of the pandemic of coronavirus disease 2019 (COVID-19), the Indian government implemented lockdown measures on March 24, 2020, which prohibit unnecessary anthropogenic activities and thus leading to a significant reduction in emissions. To investigate the impacts of this lockdown measures on air quality in India, we used the Community Multi-Scale Air Quality (CMAQ) model to estimate the changes of key air pollutants. From pre-lockdown to lockdown periods, improved air quality is observed in India, indicated by the lower key pollutant levels such as $PM_{2.5}$ (-26%), maximum daily 8-h average ozone (MDA8 O_3) (-11%), NO_2 (-50%), and SO_2 (-14%). In addition, changes in these pollutants show distinct spatial variations with the more important decrease in northern and western India. During the lockdown, our results illustrate that such emission reductions play a positive role in the improvement of air quality. Significant reductions of $PM_{2.5}$ and its major components are observed especially for secondary inorganic aerosols with the decreasing rates up to 92%, 57%, and 79% for nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium (NH_4^+), respectively. On average, the MDA8 O_3 also decreases 15% during the lockdown period although it increases sparsely in some urban locations, which is mainly due to the lower NO_x and VOCs emissions. More aggressive and localized emissions control strategies should be implemented in India to mitigate air pollutions in the future.

1 Introduction

India, the second most populous country in the world, has been suffered from severe air pollution along with rapid urbanization and industrialization in recent decades (Karambelas et al., 2018), and 13 Indian cities were among the world's top 20 most polluted cities according to the World Health Organization (WHO) (WHO, 2018). High-level pollution leads to health risks and ecosystem damages, which caused 1.24 million deaths in India in 2017 (Balakrishnan et al., 2019) and a great loss of crops (Oksanen et al., 2013; Lal et al., 2017). To mitigate air pollution, the Indian government has been promoting effective emission control strategies such as the conversion of fossil fuels to clean fuels in the nationwide campaign Clean India Mission (CIM).

However, such long-term or short-term reduction strategies seem to show insufficiency in the restoration of ambient air quality (Beig et al., 2013; Purohit et al., 2019; Banerjee et al., 2017).

Due to the pandemic of coronavirus disease 2019 (COVID-19), nationwide or partial lockdown measures have been implemented in many countries (Chintalapudi et al., 2020; Dantas et al., 2020; Ehrlich et al., 2020). India government declared corresponding bans since the detection of the first confirmed case on January 30, 2020. Then, to counter the fast contagion of COVID-19, a 3-week nationwide lockdown was imposed in India on March 24, which was expended till June 30. The lockdown measures mitigate the impact of COVID-19 on Indian health infrastructure and it also helped in curbing the rate of the spread of this infectious disease among people (Pai et al., 2020; Anderson et al., 2020). Because of the prohibition of industrial activities and mass transportation, anthropogenic emissions showed a tremendous reduction. Besides, several studies showed that dramatic emission reductions had an enormous impact on the formation of air pollution and positively influence air quality (Isaifan, 2020; Bao and Zhang, 2020; Gautam, 2020). Thus, the lockdown also provides a valuable opportunity to assess the changes in air pollutants with significantly reduced anthropogenic emissions in a short time.

Conspicuous reductions in concentrations of pollutants were also claimed in different regions (Otmani et al., 2020; Dantas et al., 2020; Nakada and Urban, 2020). Most Indian studies claimed the greatest reduction of particulate matter with an aerodynamic diameter of less than 2.5 μm ($PM_{2.5}$), up to 50% (Kumar et al., 2020; Mahato et al., 2020; Sharma et al., 2020). However, an increase of ozone (O_3) concentrations was observed (Collivignarelli et al., 2020; Sicard et al., 2020) and severe air pollution events still occurred after large emissions reduction due to unfavorable meteorological conditions (Wang et al., 2020). Moreover, another analysis showed that the effects of lockdown during the COVID-19 pandemic on $PM_{2.5}$ and O_3 pollution levels were less than the expected response to the enacted stay-at-home order (Bujin et al., 2020). Hence, the significance and impacts of lockdown measures are still not well understood.

Therefore, it is significant to understand the mechanisms involving in air pollution formation before and after dramatic emission changes comprehensively, which are limited in India. Many studies pointed out that the air quality was improved during the lockdown period and depends on the duration of the lockdown (Kumar et al., 2020). For instance, Mahato et al. (2020) concluded that air quality in India from March 24 to April 14 was improved sharply according to the change of the National Air Quality Index, especially for Delhi. also stated that the concentration of key pollutants such as $PM_{2.5}$ in both Delhi and Mumbai shows a decreasing trend. Compared with the same period in previous years, Gautam (2020) claimed that aerosol concentration levels are at its lowest in the last 20 years during lockdown based on satellite data. Srivastava et al. (2020) reported the concentrations of primary air pollutants are drastically lowed as a result of emission reduction. However, the role of meteorology and chemical reactions involving changes in air quality is not clear from these observation-based studies, which only showed the phenomenon of concentration reduction and switch of major primary pollutants. Further, the number of monitoring stations in the country is way below the guidelines by the governing bodies and not uniformly distributed.

In this study, the Community Multi-Scale Air Quality (CMAQ) model was used to investigate changes of air pollutants during the pre-lockdown (from February 21, 2020 to March 23, 2020) and lockdown (from March 24, 2020 to April 24, 2020) periods. Through two simulation scenarios, the air quality without emission reduction (Case 1) and with emissions reduction due to

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COVID-19 nationwide lockdown (Case 2) were compared to evaluate the impact of the reduction in anthropogenic emissions brought by nationwide lockdown on air quality and explore the specific impacts of meteorology and chemistry. The model performance was evaluated by comparing the simulation results with the observation data, which is collected by the Central Pollution Control Board (CPCB). This study has important implications for developing control strategies to improve air quality in India.

2 Methodology

2.1 Data collection

We used observed hourly $PM_{2.5}$, O_3, carbon monoxide (CO), and nitrogen dioxide (NO_2) data from February 21, 2020 to April 24, 2020 from the CPCB online database (https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/caaqm-landing, last access: July 07, 2020), which is widely applied in previous studies (Kumar, 2020;Sharma et al., 2020;Srivastava et al., 2020;Shehzad et al., 2020). The CPCB database provides data quality assurance or quality control programs by establishing strict procedures for sampling, analysis and calibration. Besides, the observed daily averages of $PM_{2.5}$ and maximum daily 8-h average ozone (MDA8 O_3) have been further calculated to analyse the change in air quality during the pre-lockdown (from February 21, 2020 to March 23, 2020) and lockdown (from March 24, 2020 to April 24, 2020).

2.2 Model description

This study applied CMAQ (Byun and Schere, 2006) version 5.0.2 with updated SAPRC-11 photochemical mechanism (Carter, 2011;Hu et al., 2016) and aerosol module (AERO6) (Binkowski and Roselle, 2003) to simulate air pollution across India with a horizontal resolution of 36 km × 36 km (117 × 117 grid cells). Figure 1 shows the simulation domain with positions of main Indian cities. The simulation was conducted from February 21 to March 23 as a pre-lockdown and March 24 to April 24 as a lockdown period.

The Weather Research & Forecasting model (WRF) version 3.6.1 was utilized to generate meteorology fields driven by the latest FNL (Final) Operational Global Analysis data. Anthropogenic emissions were from the monthly data from the Emissions Database for Global Atmospheric Research (EDGAR) version 4.3 (http://edgar.jrc.ec.europa.eu/overview.php?v=431). The monthly emissions from different source sectors were divided into six major groups of residential, industrial, agriculture, on-road, off-road, and energy before being adjusted from the base year of 2010 to 2019 based on population and economic growths similar to Guo et al. (2017) and the adjustment factors are shown in Table S1-S3. Weekly and diurnal profiles were used to convert monthly emissions to hourly inputs and the US EPA's SPECIATE 4.3 source profiles were used to speciate total particulate matters (PM) and volatile organic compounds (VOCs) to model species (Wang et al., 2014).

The biogenic emissions were derived from The Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012) and the emissions from biomass burning for 2018 were based on the Fire Inventory from the National Center for Atmospheric Research (FINN) (Wiedinmyer et al., 2011).

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2.3 Emission reduction during COVID-19

Due to the COVID-19 lockdown, human activities were limited and related anthropogenic emissions were reduced. Different sources were used to obtain changes in anthropogenic emissions from different sectors in comparison to 2019. For the sector of on-road and off-road, the vehicle emissions changes were based on the number of registered vehicles verified from the article (Bureau, 2020). The changes in energy demand were obtained from official data released by Power System Operation Corporation (POSOCO) (Abdi, 2020). Residential and agricultural emissions remain unchanged due to a lack of sufficient information.

For the industrial sector, we classify the Indian industries into 3 different classes based on the degree of air pollution caused (https://www.indianmirror.com/indian-industries/environment.html) (Table S4): 1) the red being the most polluting, 2) the orange, and 3) the green. The Pollution Index (PI) of any industry is a number ranging from 0 to 100 and the increasing value of PI denotes the increasing degree of pollution load from the industry. CPCB, State Pollution Control Boards (SPCBs) and the Ministry of Environment, Forest and Climate Change (MoEFCC) have finalized the following criteria on "Range of Pollution Index" for the purpose of categorization of the industrial sector (https://pib.gov.in/newsite/printrelease.aspx?relid=137373) which is shown in Table 1.

Based on the above definition of the red, orange, and green industry, the scores of 1, 0.6, and 0.4 have been assigned to each category. The emissions before lockdown can be expressed as:

$$E_1=25x+7y+31z \ , \tag{1}$$

where the ratio of x, y, and z is 1: 0.6: 0.4 as the scores and the numbers of red, orange, and green industries identified are 25, 7, and 31 before lockdown. Similarly, the emissions during the lockdown are as follows:

$$E_2=5x+y+5z \ , \tag{2}$$

Therefore, the percent reduction of industrial emissions can be calculated as:

$$\%\text{reduction}=\frac{E_1-E_2}{E_1}\times 100 \ , \tag{3}$$

In this study, two sensitivity simulations were conducted during the lockdown periods. Case 1 assumes business as usual with the same emissions as in 2019, while Case 2 adjusts anthropogenic emissions using factors obtained above for different sectors (Table 2). The differences between Case 2 and Case 1 can be assumed as the effects of COVID-19 lockdowns.

3 Results and discussion

3.1 WRF-CMAQ model validation

Meteorology plays an important role in emissions, transport, deposition, and formation of air pollutants (Zhang et al., 2015). Hence, the performance of WRF is validated to assure accurate air pollution simulation against available observation from the

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National Climate Data Center (NCDC). There are more than 1300 stations within the simulation domain with hourly observations. The considered variables contain temperature at 2 m above the surface (T2), wind speed (WS), wind direction (WD), and relative humidity (RH). Table S5 shows the statistics of mean observation and mean prediction of meteorological parameters, along with mean bias (MB), gross error (GE), and root mean squared error (RMSE), which are compared to benchmarks suggested by Emery et al. (2001b). All the statistical indexes are listed in Table S6.

In general, the WRF model performance is acceptable and similar to previous studies in India (Kota et al., 2018). For the pre-lockdown and lockdown period, predicted T2 was under-estimated with MB values of -1.5K and -1.2 K, respectively. The GE values for WS were 1.7% (pre-lockdown) and 1.8% (lockdown), satisfying the suggested criteria of 2.0%, and RMSE was slightly over the criteria. The MB values for WD were 3.2° and 2.6° during the two periods, which are within the criteria of ±10°. The GE and RMSE for WD were slightly out of the benchmarks. The under-predicted RH was also observed in this study, which was reported in other Asian studies (Hu et al., 2015). Those statistic values that did fall in the benchmark were mainly due to the resolution (36 km) applied in this study compared to the finer resolution (4–12 km) suggested in Emery et al. (2001a) (Sahu et al., 2020).

Table S7 shows the model performance of MDA8 O_3, $PM_{2.5}$, CO, and NO_2 in five major cities in India including Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. For $PM_{2.5}$, the averaged MFB (-0.37) and MFE (0.62) values met the criteria limits of ±0.6 and 0.75 claimed by the EPA (2007) in all the five urban cites. For O_3, a cut off value of 40 ppb is applied, which is based on EPA's recommendations. Besides, the model was able to reproduce the variation trends of observed hourly O_3 in all these major cities, although slightly over-estimations have occurred. And averaged MFB (-0.02) and MFE (0.29) values of O_3 also satisfy the benchmarks of ±0.15 and 0.30 set by the EPA (2005) in most of these cities with Chennai and Hyderabad exceeding the limits slightly. The performance of $PM_{2.5}$, NO_2, O_3, and CO in these urban areas were also similar to Kota et al. (2018), which could provide robust results for the following air quality study.

3.2 Changes in air quality from pre-lockdown to lockdown periods

Figure 2 shows predicted and observed $PM_{2.5}$ from February 21 to April 24 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The model succeeds in estimating the observed peak and valley values with slight under-estimation in all these cities. Overall, sharp decreases are found in the observed $PM_{2.5}$ in all these cities, and the averaged $PM_{2.5}$ level drops from 38.41 μg m^{-3} to 27.30 μg m^{-3}. The mean observed $PM_{2.5}$ concentrations during lockdown are 37.33 μg m^{-3} (Delhi), 26.93 μg m^{-3} (Mumbai), 15.49 μg m^{-3} (Chennai), 30.50 μg m^{-3} (Hyderabad), 26.23 μg m^{-3} (Bengaluru), which are reduced by 34%, 31%, 46%, 22%, and 10% respectively compared with that of the pre-lockdown period. Besides, the observed peak values of $PM_{2.5}$ in each city also decrease appreciably (up to 71%) during the lockdown period except Chennai. On March 24 that the first day of lockdown, a significant drop in $PM_{2.5}$ concentration due to the emission reduction of primary pollutants is observed (Fig. S1). However, most of the $PM_{2.5}$ concentrations are still above the WHO annual guideline values of 10 μg m^{-3} WHO (WHO, 2016) during the lockdown period, with peak values over 60 μg m^{-3} occasionally.

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Figure 3 shows the temporal variation of MDA8 O_3 in these five cities. The predicted MDA8 O_3 is consistent in trend with observation values in most days, while simulated concentrations are overall higher, particularly in Hyderabad. The observed average MDA8 O_3 during lockdown is higher than that of pre-lockdown in Delhi (11%), Hyderabad (3%), and Bengaluru (26%). This is likely due to the fact that O_3 formation in these cities is under VOC control (Sharma et al., 2020), and nitrogen oxide (NO_x) reduction leads to O_3 increase by enhanced hydrogen oxide radicals (HO_x) concentrations (Zhao et al., 2017). The increase of monthly average T2 from pre-lockdown (281.0 K) to lockdown (285.1 K) could also lead to an increase of O_3 (Chen et al., 2019). In contrast, the observed average MDA8 O_3 during lockdown is reduced compared with the pre-lockdown period in both Mumbai (-22%) and Chennai (-37%). This could be caused by the much larger reduction in emissions as Mumbai and Chennai are the most affected areas. In specific, Mumbai accounted for more than a fifth of infections in India (Mukherjee, 2020).

Figure 4 shows the comparison of predicted air pollutants before and during the lockdown throughout India. Generally, decreasing trends of key pollutants including particulate matter with an aerodynamic diameter of less than 10 μm (PM_{10}) (-16%), $PM_{2.5}$ (-26%), MDA8 O_3 (-11%), NO_2 (-50%) and sulfur dioxide (SO_2) (-14%) are observed across Indian. Changes of these pollutants present distinct regional variations. In the northern and western India, the lower levels of these pollutants are observed during the lockdown, with the reductions of $PM_{2.5}$ and PM_{10} up to 79%. In particular, the most significant decreases are found in the populated, industrialized, and polluted Indo-Gangetic Plain (IGP) during the lockdown. The averaged $PM_{2.5}$ even drops from approximately 35–70 μg m^{-3} (pre-lockdown) to 15–40 μg m^{-3} (lockdown) in these regions because local emissions are generally the largest contributor (38–78%) to $PM_{2.5}$ in India (David et al., 2019). However, rising trends of these key pollutants are found mainly in the northeastern, eastern, and parts of southern India.

Besides, changes in $PM_{2.5}$ also show prominent differences in the rural and urban areas. In India, rural areas have different emission sources from that of urban areas and are less influenced by lock measures (Garaga et al., 2020). In megacities such as Delhi, the predicted concentrations of $PM_{2.5}$ decline during the lockdown, which is consistent with previous results (Kumari and Toshniwal, 2020;Chauhan and Singh, 2020). For instance, over a 60% reduction of $PM_{2.5}$ is estimated in Delhi and Ahmedabad. However, rising trends of $PM_{2.5}$ (~20%) are observed in the far-flung northeastern part of India.

As gaseous precursors of major components to $PM_{2.5}$ (Jain et al., 2020), concentrations of NO_2 and SO_2 also decrease significantly in most regions by up to 90% and 87%, respectively. However, their levels increase in parts of the east and south India and thus leading to higher levels of $PM_{2.5}$ and PM_{10} in the same regions. MDA8 O_3 is also rising in eastern India by the highest increasing rate of 29%, while 30% reduction is observed in northern and western India. Although significant reductions are found in O_3 precursor emissions throughout India during the lockdown, the MDA8 O_3 has not shown comparable decreasing trends, which is affected by the meteorological conditions such as an increase of temperature (Fig. S2) and the influence of chemical processes (Chen et al., 2019;Zhao et al., 2017).

In summary, the decrease of $PM_{2.5}$, PM_{10}, NO_2, SO_2, and the increase of MDA8 O_3 during lockdown is consistent with previous results (Srivastava et al., 2020;Mahato et al., 2020). In case of Delhi, compared with the previous studies, the $PM_{2.5}$ reduction (34%) is comparable with 35% reported by Chauhan and Singh (2020), while less than 53% stated by Mahato et al. (2020) and

49% calculated by Kumari and Toshniwal (2020) during the first phase of lockdown (from March 24, 2020 to April 15, 2020). These differences highly depends on the duration of lockdown because there is an increase in traffic flow and some relaxation in the later lockdown period (after April 15, 2020) (Kumar, 2020). Moreover, the different characteristics of these air pollutants in rural and urban areas have not been investigated comprehensively in previous studies. Kumari and Toshniwal (2020) also concluded that concentrations of PM_{10}, $PM_{2.5,}$ and SO_2 tended to rise in Singrauli (rural area, located in central India) during the lockdown, contrary to the results of Delhi and Mumbai. Therefore, our results have important implications for the study of air quality changes and their regional distribution across India and indicate more strident emission reduction policies should be implemented across India, especially in the later phases of lockdown and in rural areas.

3.3 Effects of emission reductions on $PM_{2.5}$ during the lockdown

There are significant changes of $PM_{2.5}$ between the lockdown and pre-lockdown periods, but it remains unclear regards the direct impacts of emission reductions during the lockdown. Figure 5 shows the differences in major $PM_{2.5}$ components during the lockdown period with (Case 2) and without (Case 1) control measures.

Major components of $PM_{2.5}$ including nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium (NH_4^+), elemental carbon (EC), primary organic aerosol (POA), and secondary organic aerosol (SOA), decreased significantly in Case 2 compared to Case 1, indicating the positive effects of emission reduction. Primary components of $PM_{2.5}$ (EC and POA) are dropped by averaged 37% and 14%, respectively. EC is usually emitted from combustion sources and a drastic decrease of up to 74% directly reflected the impact of emission reductions from industry and transportation. Secondary inorganic aerosol (SIA) including NO_3^-, SO_4^{2-}, and NH_4^+ and SOA accounted for most of the $PM_{2.5}$ bulk mass (39%) and showed greater decreases than primary components. Moreover, the spatial distribution of SIA is apparently affected similar to $PM_{2.5}$ that the reduction is more significant in the north of India where the decrease of NO_3^-, SO_4^{2-}, and NH_4^+ are up to 92%, 57%, and 79% respectively. The largest reduction of NO_3^- by averaged 62% resulted from transportation reduction and SO_4^{2-} reduction (averaged 31%) is likely due to the falling release of industry (Gawhane et al., 2017;Wang et al., 2020). On average, NH_4^+ and SOA are decreased by 41% and 14%, respectively. The significant decrease in NH_4^+ cannot be attributed to the absence of reduced agricultural emissions in the simulation but may be due to the relatively reduced $(NH_4)_2SO_4$ and NH_4NO_3 in CMAQ chemistry-transport model (Fountoukis and Nenes, 2007). By contrast, compared with VOCs, an important precursor of SOA, the smaller reduction of SOA may be related to the weakening of the atmospheric oxidizing capacity (AOC), which plays an important role in the formation of SOA (Feng et al., 2019).

Figure 6 shows the predicted response of changes in concentration of primary $PM_{2.5}$ (PPM) and secondary components to the reduced emissions of related precursors in Delhi, Mumbai, Kolkata, Bengaluru, Hyderabad, Chennai, Ahmedabad, and Lucknow. Generally, all species decreased with the reduced emissions and the great sensitivity of $PM_{2.5}$ component concentrations to emissions showed the important role of meteorology and the effectiveness of stringent measures to reduce emissions.

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On average, NO_3^- shares the largest reduction of 77% mainly driven by the decrease of its gaseous precursor NO_x (71%). At least 27% decrease of SO_4^{2-} is found in each city caused by the largest reduction of SO_2 (averaged 59%). Over 70% average reduction of NO_x and NO_3^- may still relate to the reduction of vehicles. And SOA is dropped by averaged 18% because of the lack of precursors due to the emission reduction of VOCs (29%). Due to the reduction of emitting precursors, the concentration reduction of $PM_{2.5}$ secondary components is less than that of primary components. The ratios of PPM reduction in emission (averaged 39%) are larger than the reduction in concentration (averaged 43%) in five selected cities. Especially, a 7% reduction in emission of PPM caused a 43% decline in its concentration in Hyderabad. Emissions of EC and organic carbon (OC) have also been reduced by a certain proportion resulting in a similar or greater reduction in concentrations.

The response of concentration to emissions in all cities presented a nonlinear change that has been confirmed previously by Zhao et al. (2017), which is related to various meteorological conditions (Wang et al., 2020). For example, in Lucknow, PPM, EC, OC, SO_2, NO_x, and VOCs decreased by 14%, 25%, 8%, 39%, 55%, and 11% respectively, while the concentration of PPM, EC, OC, SO_4^{2-}, NO_3^-, and SOA dropped by 21%, 32%, 12%, 43%, 78%, and 18%. Besides, the concentration response to emission reduction is likely to be more prominent in highly polluted and industrialized areas. The highest reductions in PPM and these secondary components of $PM_{2.5}$ happened in Ahmedabad (an industrial city located in western India) with high vehicular populations. While Bengaluru, a major southern Indian city, is considered as one of the cleaner Indian major cities because of its low $PM_{2.5}$ concentrations with no heavy industries (Guttikunda et al., 2019). Consequently, the reduction in $PM_{2.5}$ and its major components (especially for secondary components) in Bengaluru is not as significant as Ahmedabad although a similar reduction in emissions is observed.

3.4 Effects of emission reductions on O_3 during the lockdown

We investigated the changes of MDA8 O_3 and its major precursors NO_x and formaldehyde (HCHO) that is an important component of total VOCs reactivity (Steiner et al., 2008) during the lockdown period. Figure 7 shows that MDA8 O_3, NO_x, and HCHO decreased all over India. The average reduction rates of MDA8 O_3, NO_x, and HCHO are approximately 15%, 50%, and 15%, respectively. For both Case 1 and Case 2, the higher levels of MDA8 O_3 are in eastern India (over 60 ppb, Case 1) in which the higher NO_x is also observed (over 12 ppb, Case 1) during the lockdown. Compared to $PM_{2.5}$, no significant north-south differences are found in the change of O_3. NO_x concentration has the greatest reduction that is mostly driven by the large cutting of energy emission by 26%, which is consistent with the decline of India's electricity consumption (9.2%) (Reuters, 2020).

Figure S2 shows the spatial distributions of O_3 production sensitivity in India during the lockdown, according to the ratio of $HCHO/NO_2$, which is considered as an indicator of O_3 sensitivity to NO_x and VOCs (Sillman, 1995). The criteria used to identify O_3 precursor sensitivity from the ratio of $HCHO/NO_2$ is suggested by Duncan et al. (2010). In India, NO_x-limited regimes ($HCHO/NO_2 > 2$) are found in vast areas from both Case 1 and Case 2, which was also reported in previous studies (Mahajan et al., 2015). Compared to Case 1, the VOC-limited area ($HCHO/NO_2 < 1$) expands mainly in the northwest and south of India from Case 2 during the lockdown. The transition regimes ($1 < HCHO/NO_2 < 2$) that O_3 formation is controlled

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by both NO_x and VOC emissions in the vicinity of the VOC-limited regions. Simultaneously, the rise of MDA8 O_3 (averaged 5% and up to 21%) is found sporadically in these VOC-limited areas in which more significant decreases of NO_x (compared with VOCs) reduce the O_3 consumption ($NO + O_3 = NO_2 + O_2$) and enhance HO_x concentrations resulting in an increase in O_3 levels.

Figure 8 compares the concentrations of MDA8 O_3, HCHO, and NO_x with emissions of VOCs, HCHO, and NO_x in eight major cities of India, Delhi, Mumbai, Kolkata, Bengaluru, Hyderabad, Chennai, Ahmedabad, and Lucknow. Generally, the decline in O_3 concentration in Delhi (14%), Mumbai (23%), Kolkata (24%), Bengaluru (20%), Hyderabad (17%), Chennai (20%), Ahmedabad (21%), and Lucknow (15%) showed that effectiveness of emission reductions that play an important role in the control of O_3 pollution, even in these VOC-limited areas.

The changes in emissions and concentrations of MDA8 O_3, HCHO, and NO_x showed a non-linear response. In Delhi, a 76% reduction in NO_x emissions resulted in a 77% reduction in its concentration, while a 29% reduction in HCHO resulted in only an 11% reduction. In a megacity like Delhi, about 7 million vehicles and many fossil fuel-based plants lead to high NO_x emissions, and local restricted transportation and industrial activities during lockdown could lead to a significant reduction of primary NO_x emissions (Sharma et al., 2016). The concentration of NO_x is appreciably highly sensitive to a primary NO_x emission reduction. However, the VOCs emission reduction resulting from the lockdown is relatively less than NO_x in each city. And most of the reduction of HCHO concentration is less than that of emission reduction, which is different from NO_x, which indicated that the change of HCHO concentrations is not dominated by primary HCHO emission reduction.

4 Conclusion

Compared with pre-lockdown, observed $PM_{2.5}$ during the lockdown in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru shows an overall decreasing trend. In contrast, MDA8 O_3 increases in three of these cities. The comparison of predicted air pollutants across India before and during the lockdown shows distinct regional characteristics. The most significant reductions of $PM_{2.5}$ and PM_{10} (up to 79%) are observed in most of northern and western India including all these megacities. However, increases of MDA8 O_3 (up to 29%) and other key pollutants are reported in northeastern, eastern, and parts of southern India covering most of the rural areas.

The drastic decline in $PM_{2.5}$ and its major components during the lockdown period in Case 2 compared with Case 1 shows the positive impacts of emission control measures, especially for SIA that the decrease of NO_3^-, SO_4^{2-}, and NH_4^+ are up to 92%, 57%, and 79%, respectively. During the lockdown, the decrease of MDA8 O_3 (averaged 15%) occurs in most regions in India, which is attributed to the lower emissions of NO_x (48%) and VOCs (6%) that are precursors of O_3. Our results demonstrate that the strident emissions controls due to the lockdown have mitigated air pollution in India. However, we also find the scattered increases in MDA8 O_3 (up to 21%) in some urban locations in the VOC-limited areas due to the emissions reduction. This indicates that a more localized control policy with the consideration of the O_3 sensitivity regime should be implemented in India to improve the air quality especially for secondary pollutants such as O_3.

Data availability. The datasets used in the study can be accessed from websites listed in the references or by contacting the corresponding authors (peng.ce.wang@polyu.edu.hk; zhanghl@fudan.edu.cn).

Author contribution. MZ conducted the modelling and led the writing of the manuscript. AK carried out the data collection and initial analysis. SZ, JS, and JM assisted with the data analysis. MX, SK assisted with the interpretation of the results and the writing of the paper. HZ and PW designed the study, discussed the results, and edited the paper.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1: The criteria on the "Range of Pollution Index" for the purpose of categorization of industrial sectors.

Categories	Pollution Index score
Red category	≥60
Orange category	41–59
Green category	21–40

Table 2: Percent reduction in anthropogenic emissions in India during COVID-19 lockdown.

Sector	%Reduction
Residential	0
Industrial	82
Agriculture	0
On-road	85
Off-road	85
Energy	26

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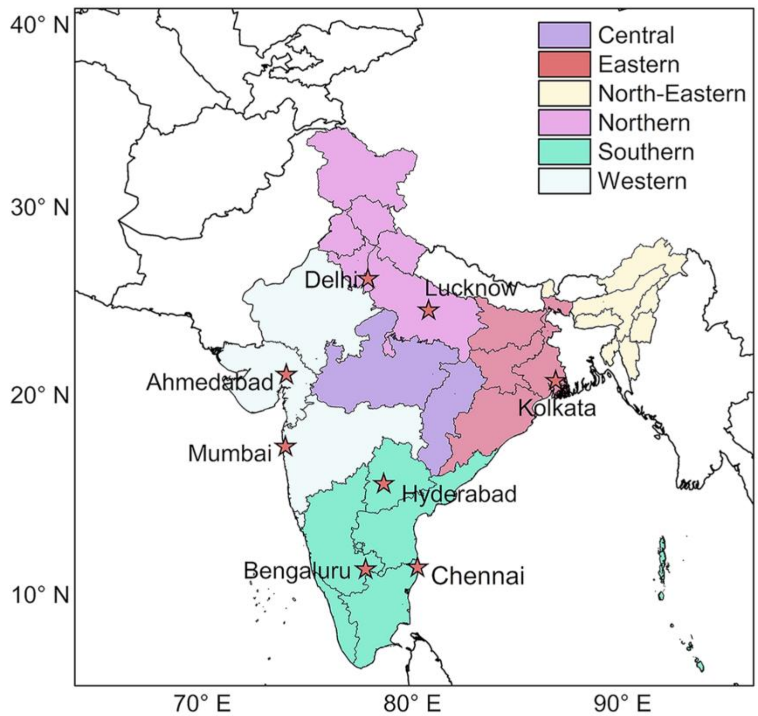

Figure 1: The simulation domain with the location of major Indian cities selected for analysis.

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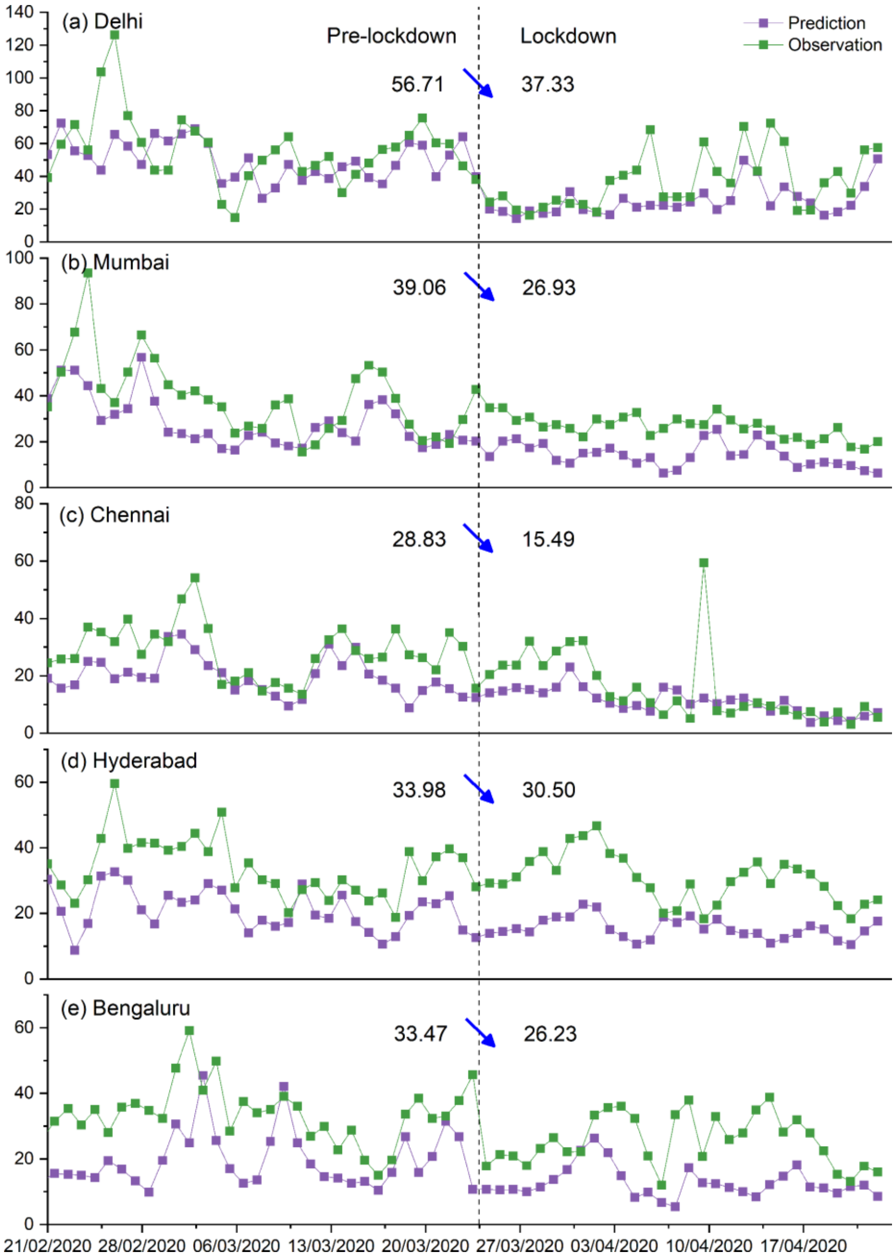

470 **Figure 2: Comparison of predicted and observed $PM_{2.5}$ from February 21 to April 24 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The unit is μg m^{-3}.**

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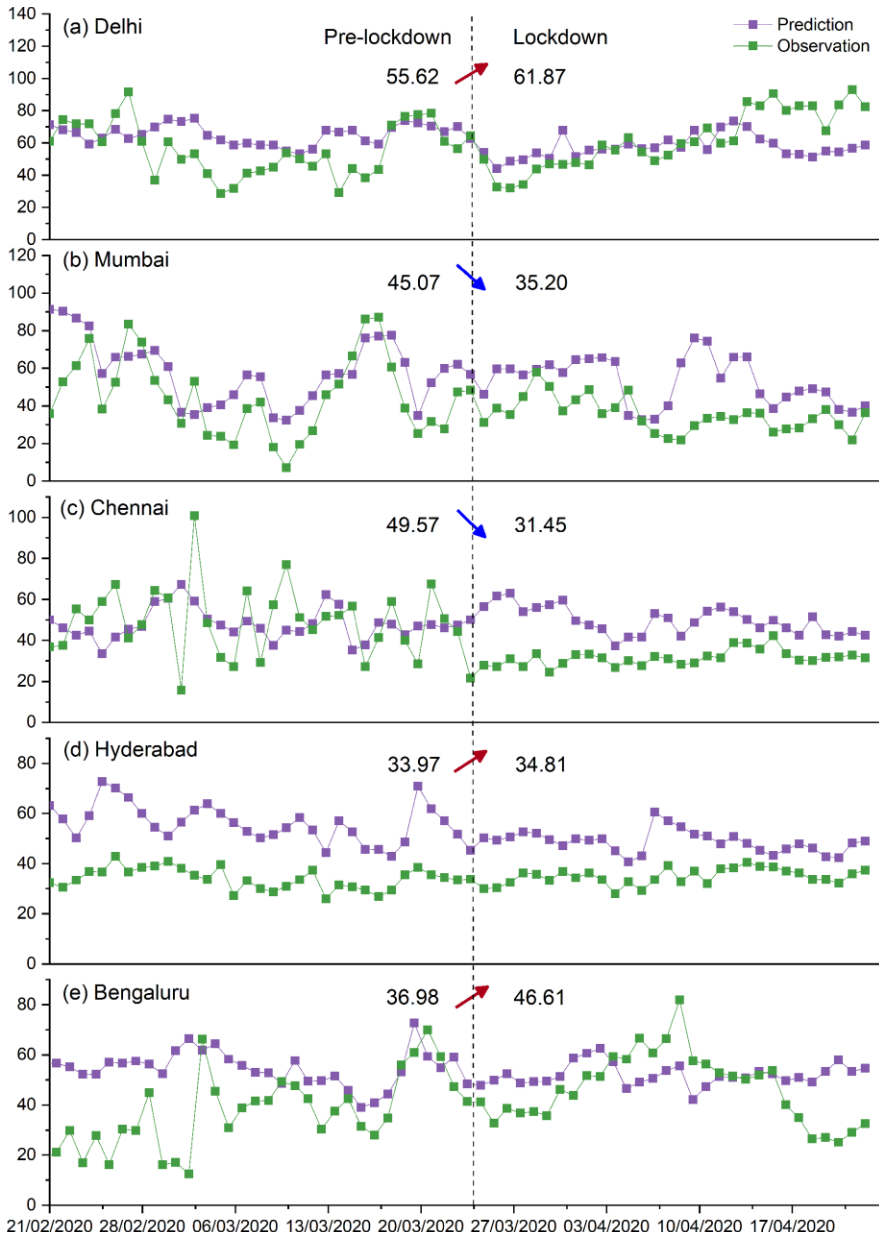

Figure 3: Comparison of predicted and observed MDA8 O_3 from February 21 to April 24 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The unit is ppb.

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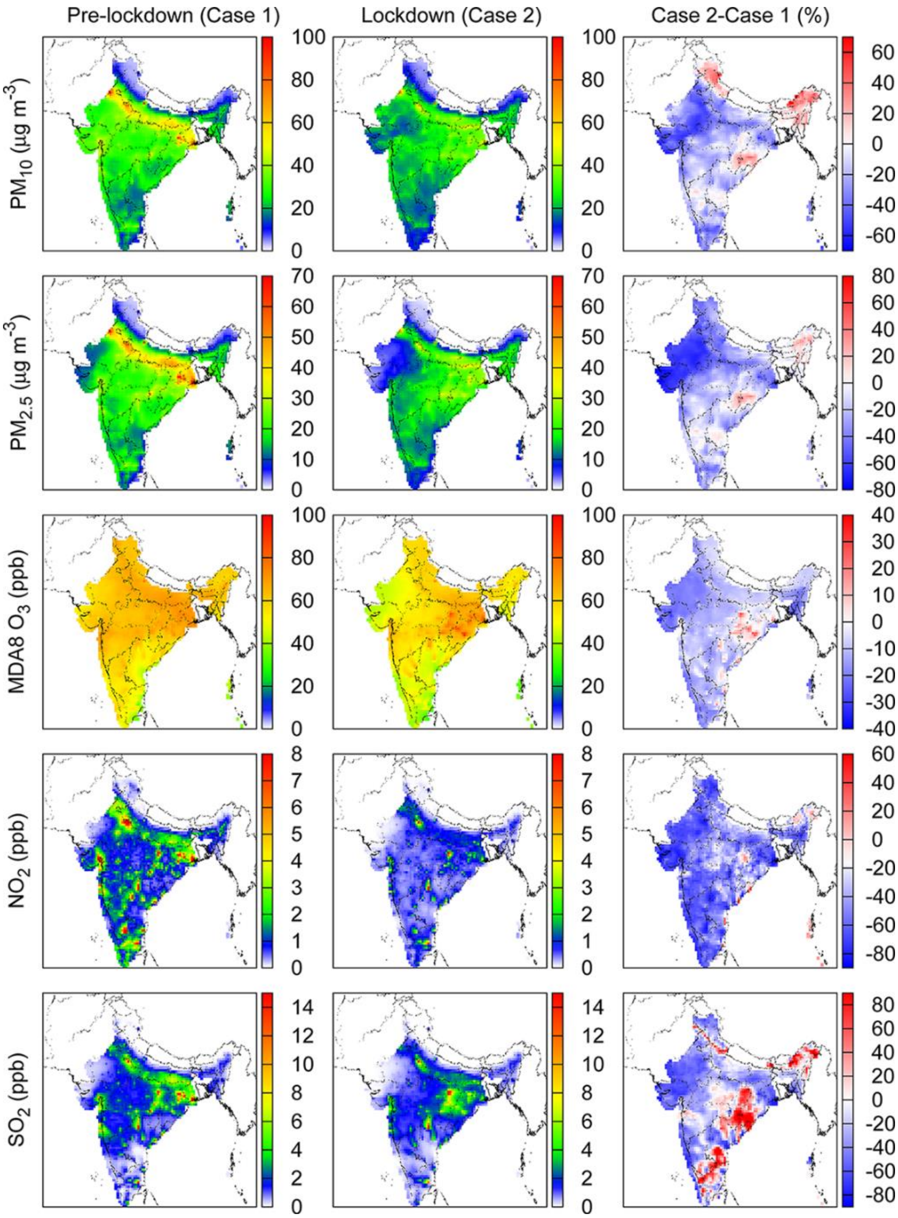

Figure 4: Predicted PM_{10} (µg m^{-3}), $PM_{2.5}$ (µg m^{-3}), MDA8 O_3 (ppb), NO_2 (ppb), and SO_2 (ppb) before lockdown, during the lockdown and the changes between them in India.

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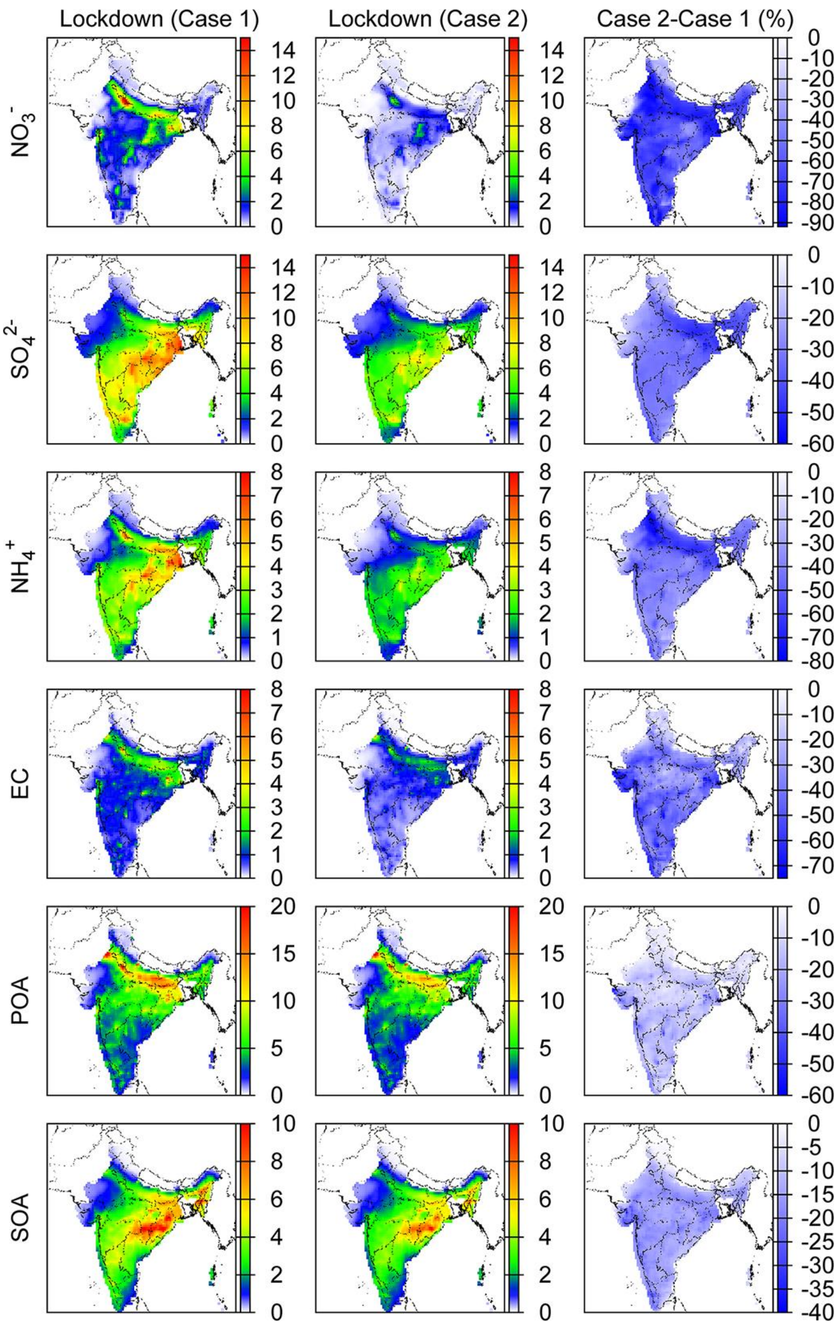

480 **Figure 5: Predicted $PM_{2.5}$ components and the changes caused by lockdown measures from March 24 to April 24, 2020 in India. The unit is μg m^{-3}.**

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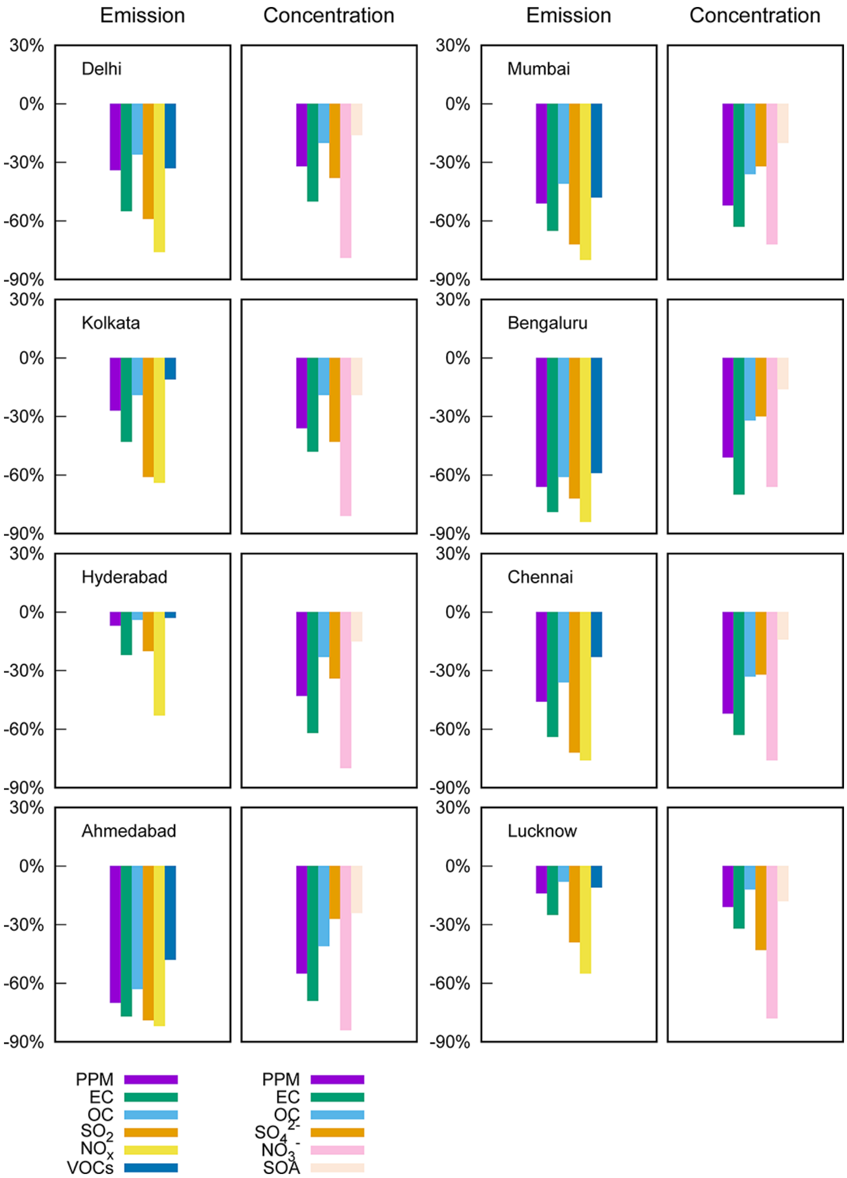

Figure 6: Predicted relative changes in concentrations of primary and secondary components, and emissions of their precursors in eight cities of India in Case 2 to Case 1.

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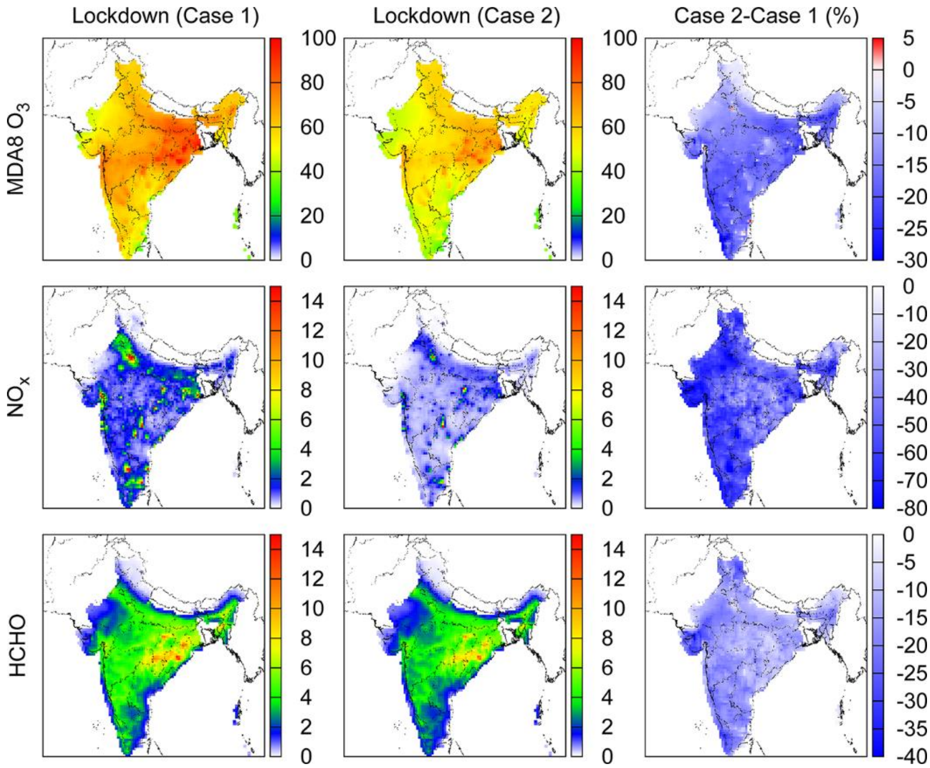

Figure 7: Predicted O_3, NO_x, HCHO, and the changes caused by nationwide lockdown measures from March 24 to April 24, 2020 in India. The unit is ppb.

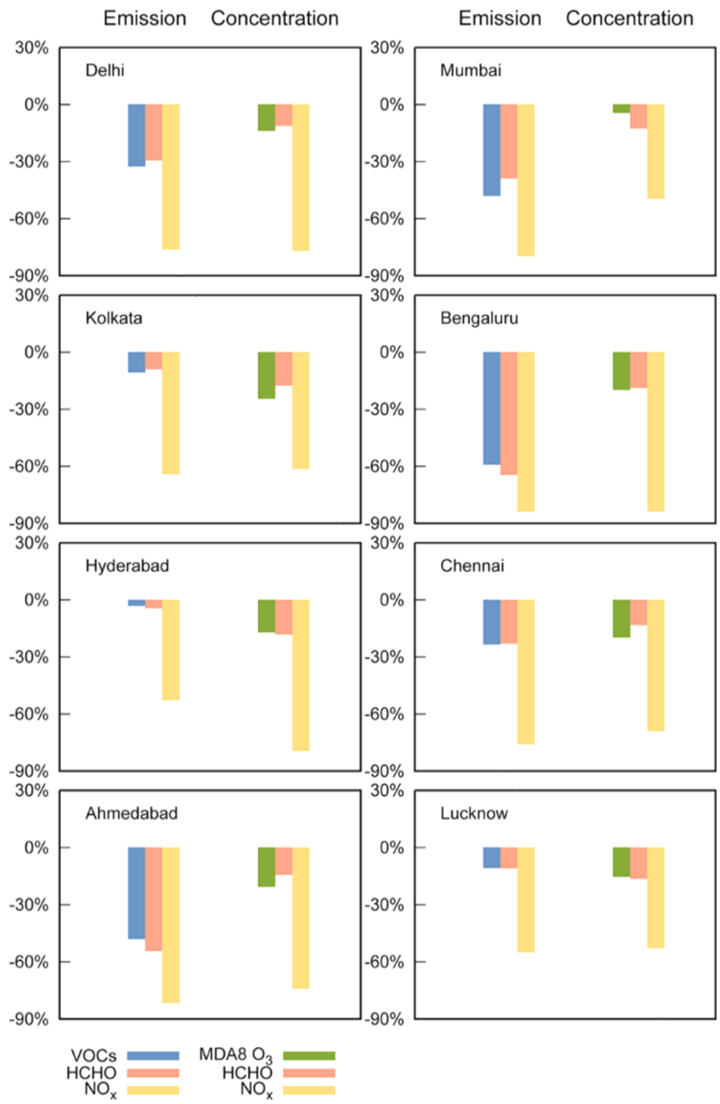

Figure 8: Predicted relative changes in concentrations of O_3, HCHO, and NO_x and emissions of VOCs, HCHO, and NO_x in eight
490 **major cities of India in Case 2 to Case 1.**