Responses to interactive comments

Journal: Atmospheric Chemistry and Physics

Manuscript ID: acp-2020-903

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

Dear Referee #3,

We appreciate your comments to help improve the manuscript. We tried our best to address your comments and detailed responses and related changes are shown below. Our response is in blue and the modifications in the manuscript are in red.

Comments: The paper by Zhang et al. entitled "Impact of reduced anthropogenic emissions during COVID-19 on air quality in India" is on a very relevant and interesting topic which is to use the covid lockdown emission reductions for assessing impacts on air quality over India. Unfortunately, the analyses and interpretation are weak in several places (listed below). There are hardly any new trustworthy insights from this modelling study which have not been reported already by the authors in previous works on the same topic published recently (see Sharma, S., Zhang, M., Anshika, Gao, J., Zhang, H., and Kota, S. H.: Effect of restricted emissions during COVID-19 on air quality in India, Science of The Total Environment, 728, 138878, https://doi.org/10.1016/j.scitotenv.2020.138878, 2020).

Response:

We appreciate the comments. However, we don't agree that there are hardly any new trustworthy insights. We comprehensively evaluate the impact of the nationwide lockdown on air quality in India, which also provides reliable recommendations for the improvement of emission reduction policies.

First, we determined the response of air quality in India under the synergetic impacts from the meteorological conditions and anthropogenic emissions during the pre-lockdown and lockdown periods. For instance, we directly quantified the change in air quality during lockdown due to the reduced anthropogenic emissions through differences between Case 1 (without emission reductions) and Case 2 (with emission reductions) during the lockdown. This casts lights on the policy implementation in India, which may help to mitigate air pollution in the future.

Second, we are the first study that explored the impacts of COVID-19 lockdown on Indian air quality on a regional scale. It allows us to figure out the changes of primary and even secondary pollutants during two periods (pre-lockdown and lockdown) and illustrate their differences in urban and rural areas. This could be a great help to formulate the city-level control policy in India.

Third, in atmospheric chemistry, we developed a better understanding of the secondary pollutants formations by investigating their non-linear responses to the precursors' changes during the lockdown. In particular, the sensitivity of $PM_{2.5}$ secondary components (Fig. 6 in the revised manuscript) and the change of spatial distributions of O_3 production sensitivity (Fig. S5 in the revised supplement) due to emission changes during the lockdown give us a more in-depth discussion on secondary pollutants.

In the revised manuscript, we added such information to the Introduction to make it clear.

Changes in manuscript:

Introduction (Lines 64-68 in the revision): "However, the role of meteorological conditions 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 mainly in urban cities. Further, the number of monitoring stations in the country is way below the guidelines by the governing bodies and not uniformly distributed, which results in observation data limitations in India (Sahu et al., 2020)."

Introduction (Lines 69-74 in the revision): "In this study, the Community Multi-Scale Air Quality (CMAQ) model was used to investigate changes in 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 throughout Indian region. We explored the synergetic impacts from the meteorological conditions and anthropogenic emissions during the pre-lockdown and lockdown periods. Besides, we directly quantified the change in air quality during the lockdown due to the reduced anthropogenic emissions by comparing the differences between Case 1 (without emission reductions) and Case 2 (with emission reductions)."

Conclusion (Lines 309-310 in the revision): "However, more stringent mitigation measures are needed to achieve effective control of air pollution from secondary air pollutants and their components, particularly in rural areas."

Comments: Instead there are even discrepancies from the earlier work based on interpretation of what appears to be the same measured dataset. While in the previous work (Sharma et al., 2020) it was reported that there was a 17% increase in ozone during COVID, in the present work it has been reported that a significant decrease in surface ozone (MDA8 values) occurred, without even clarifying what changed between the two studies except for additional modelling analyses in this study.

Response:

We are sorry for being not clear enough. As described in the previous response, these two studies are different in many aspects and we are not aiming to show the same results.

First, the duration of lockdown considered in this study (from March 24 to April 24, 2020) is different from Sharma et al. (from March 15th to April 14, 2020). Second, the variations of MDA8 O_3 in this study included all India within the 36-km domain (117×117 grids) (Fig. 1 in the manuscript), while Sharma et al. only focused on urban measurements at 22 cities. Third, this study excluded the influence of meteorology by comparing Case 1 (without emission reductions) and Case 2 (with emission reductions) during the lockdown, while Sharma et al. concluded the increase in O_3 by comparing 2020 and the previous three years.

Moreover, the results of our study are consistent with Sharma et al. in the urban areas. In these areas that were under VOC-limited conditions, both studies concluded that O_3 increased during the lockdown. In our study, the increase in MDA8 O_3 was up to 21%, which was close to Shame et al.

In summary, different research methods and study periods result in different O_3 changing ratios, and it is not able to conclude that as discrepancies.

Changes in manuscript: Since the differences have been added in the previous comment, no special changes were made for this point.

Comments: There are several major issues with the present submission which need to be addressed/clarified for meriting further publication in ACP.

Response: We thank the reviewers for the detailed comments below and made necessary changes to the manuscript.

Comments: Validation of the model used in this work has not been done/described adequately: Authors use only 2 m level measurements of temperature and meteorological data and chemical data from 5 monitoring stations operated by the regulatory agency of India located within cities to compare their modelled output.

Response:

We collected all available observations to validate our WRF and CMAQ models. Since the observations are limited in India, it is important to conduct simulation studies like this one to improve our understanding and help design control strategies.

Changes in manuscript: No changes were made for this point.

Comments: Measured chemical data: The authors present only daily averaged data in the plots (Figures 2 and 3). This would be fine but I could find no details of the original high resolution primary data (presumably available at temporal resolution of few minutes from the analyzers in the monitoring stations) to build confidence in the reader about the trustworthiness of the primary data and its quality assurance. If they could provide such high resolution data for the five stations (even for few days in both periods) for ozone, NO, NO_2, $PM_{2.5}$ etc.. with gaps in measurements if any (after all there was a lockdown so maintenance could be difficult), and the calibration data of any of the analyzers, it would go a long way in instilling confidence in the highly averaged data. The reviewer looked up their previous study Sharma et al 2020 which has been cited for detailed description of the primary data and found that this reference did not contain these details and somewhat remarkably the Sharma et al. 2020 paper reported data until April 14th 2020 in that work, was submitted on April 16th, 2020 and accepted on April 19th, 2020. While this does not necessarily suggest that due diligence was not taken as given the nature of topic urgency to publish would have been a factor, the rapid turn-around time and lack of experimental details in the peer reviewed reference cited and which forms the basis of the daily averages does leave room for concern. So the authors should provide the original primary data as a time series for these 5 monitoring stations in the revised supplement along with details of calibration experiments and data quality control followed to allay such potential concerns about the primary measured dataset.

Response:

Although higher resolution from some sampling equipment is available, monitoring agencies worldwide only report hourly data. The data from the CPCB database can be downloaded at https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/data and we are not able to post the raw data here due to the CPCB's data access statement. Also, the temporal-resolution of the CMAQ model is hourly. Besides, daily average $PM_{2.5}$ is commonly used in previous studies due to its practical significance and data availability. For O_3, the maximum daily 8-hour concentration (MDA8 O_3) calculated as running the maximum continuous 8-hour data is used to represent pollution level (Lei et al., 2019). Thus, we used the daily average $PM_{2.5}$ and MDA8 O_3 specifically in Figures 2 and 3 in the manuscript.

We are sorry that the validity of the observation data is not clear. The observation data from CPCB has been through quality assurance or quality control programs by establishing strict procedures for sampling, analysis, and calibration before publication (Gurjar et al., 2016). Thus, studies usually use the data directly. In this study, we made additional checks to screen out outliers. For example, a cut-off value of 40 ppb was applied to hourly O_3 based on EPA's recommendations (EPA, 2005). For $PM_{2.5}$, abnormally high values of greater than 300 $\mu g\ m^{-3}$ were excluded. These explanations were added in different parts of the revised manuscript.

Changes in manuscript:

Methodology (Lines 82-83 in the revision): "The CPCB database provides data quality assurance (QA) or quality control (QC) programs by establishing strict procedures for sampling, analysis, and calibration (Gurjar et al., 2016)."

Results and discussion (Lines 150-153 in the revision): "For $PM_{2.5}$, the averaged mean fractional bias (MFB) (-0.48) and mean fractional error (MFE) (0.61) values met the criteria limits of ±0.6 and 0.75 claimed by the EPA (2007b) in all the five urban cites after excluding some abnormally high values of greater than 300 $\mu g\ m^{-3}$. For O_3, a cut-off value of 40 ppb is applied, which is based on EPA's recommendations (EPA, 2005)."

Comments: Also they should discuss whether data from 5 cities are adequate to make inferences about all of India with same degree of confidence which spans vast rural and countryside regions? It might be advisable to better focus on the 5 cities alone for which they have the data and even there they should acknowledge how data from one monitoring station may be limited for representing air quality of the entire city. In fact, a combination of monitoring station data and satellite data (agreed also can have issues but better than nothing) would be better.

Response:

Thanks for the comments. First of all, we agree that 5 cities were maybe inadequate to evaluate the model for the whole of India. However, it is an endless effort and impractical to monitor the whole country. Focusing only on areas with observations is a safe idea, but not a promising one. This actually is the advantage of modelling work and the merit of this study. Besides, when we validate our simulated results in urban areas, we have the confidence to investigate rural and countryside regions. The modelling work is not only to reproduce what has been observed but more importantly to investigate what is not been observed after enough validation.

If the monitoring station can represent the entire city is another good question for observation experts. In this study, the observation data we compare with the predicted values are an average of multiple sites in each city (Table 1), which can represent the $PM_{2.5}$ and O_3 levels of the entire city well. We also added more observations to make the results more representative in the revision (Fig. 1 also added as Fig. 2 in the revised manuscript & Fig. 2 added as Fig. 3 in the revised manuscript). The model performed well at simulating O_3, $PM_{2.5}$, CO, and NO_2 in these major cities in India (Table 2, also added as Table S7 in the revised supplement).

Thanks to the suggestion of using satellite, we compared model performance with satellite observations (TROPOMI) for HCHO and NO_2 (Fig. 3, also revised Fig. S1 in the supplement). The corresponding explanatory statements were added in the Results and discussion section.

Table 1: Cities and monitoring sites selected for observation data from the CPCB database.

City	Monitoring sites
Hyderabad	Bollaram Industrial Area, Hyderabad - TSPCB
	ICRISAT Patancheru, Hyderabad - TSPCB
	IDA Pashamylaram, Hyderabad - TSPCB
Chennai	Velachery Res. Area, Chennai - CPCB
	Alandur Bus Depot, Chennai - CPCB
	Manali Village, Chennai - TNPCB
Mumbai	Chhatrapati Shivaji Intl. Airport (T2), Mumbai - MPCB
	Bandra, Mumbai - MPCB
	Borivali East, Mumbai - MPCB
	Powai, Mumbai - MPCB
	Sion, Mumbai - MPCB
Bengaluru	BTM Layout, Bengaluru - CPCB
	BWSSB Kadabesanahalli, Bengaluru - CPCB
	Silk Board, Bengaluru - KSPCB
Delhi	Aya Nagar, Delhi - IMD
	Ashok Vihar, Delhi - DPCC
	Bawana, Delhi - DPCC
	Alipur, Delhi - DPCC
	Anand Vihar, Delhi - DPCC
	CRRI Mathura Road, Delhi - IMD
	DTU, Delhi - CPCB
	Dr. Karni Singh Shooting Range, Delhi - DPCC
	Dwarka-Sector 8, Delhi - DPCC
	ITO, Delhi - CPCB

	Jahangirpuri, Delhi - DPCC
	Jawaharlal Nehru Stadium, Delhi - DPCC
	Lodhi Road, Delhi - IMD
	Major Dhyan Chand National Stadium, Delhi - DPCC
	Mandir Marg, Delhi - DPCC
	NSIT Dwarka, Delhi - CPCB
	Najafgarh, Delhi - DPCC
	Nehru Nagar, Delhi - DPCC
	North Campus, DU, Delhi - IMD
	Okhla Phase-2, Delhi - DPCC
	Patparganj, Delhi - DPCC
	R K Puram, Delhi - DPCC
	Shadipur, Delhi - CPCB
	Sri Aurobindo Marg, Delhi - DPCC
	Vivek Vihar, Delhi - DPCC

Table 2: model performance of O_3 (ppb), $PM_{2.5}$ ($\mu g\ m^{-3}$), CO (ppb), and NO_2 (ppb) at Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru (OBS is mean observation; PRE is mean prediction; MFB is mean fractional bias; MFE is mean fractional error; MNB is mean normalized bias; MNE is mean normalized error).

Variable	Statistics	Delhi	Mumbai	Chennai	Hyderabad	Bengaluru	ALL	Benchmark
O_3	OBS	61.37	56.64	49.94	44.03	47.66	51.93	
	PRE	56.86	47.06	39.32	52.56	43.58	47.88	
	MNB	-0.04	-0.13	**-0.20**	**0.20**	-0.07	-0.05	≤±0.15
	MNE	0.20	0.28	0.29	0.22	0.26	0.25	≤0.30
	MFB	-0.08	-0.20	-0.27	0.17	-0.17	-0.11	
	MFE	0.21	0.32	0.36	0.18	0.33	0.28	
$PM_{2.5}$	OBS	58.08	33.10	23.09	33.09	33.79	36.23	
	PRE	38.10	21.46	15.57	18.28	16.29	21.94	
	MNB	-0.16	-0.28	-0.12	-0.41	-0.41	-0.28	
	MNE	0.54	0.44	0.46	0.47	0.51	0.48	
	MFB	-0.40	-0.46	-0.30	-0.59	**-0.66**	-0.48	≤±0.6
	MFE	0.62	0.59	0.51	0.63	0.73	0.61	≤0.75
NO_2	OBS	13.87	11.17	3.74	10.60	10.68	10.01	
	PRE	7.00	9.68	4.34	3.04	8.64	6.54	
	MNB	-0.51	0.42	0.46	-0.74	-0.30	-0.14	
	MNE	0.74	1.34	1.11	0.86	0.89	0.99	

	MFB	-1.00	-0.47	-0.20	-1.44	-0.90	-0.80
	MFE	1.13	1.18	0.82	1.51	1.19	1.17
CO	OBS	0.69	0.65	0.45	0.38	0.74	0.58
	PRE	0.26	0.16	0.12	0.13	0.14	0.16
	MNB	-0.59	-0.72	-0.71	-0.61	-0.78	-0.68
	MNE	0.59	0.72	0.71	0.61	0.78	0.68
	MFB	-0.88	-1.15	-1.13	-0.92	-1.32	-1.08
	MFE	0.89	1.15	1.13	0.92	1.32	1.08

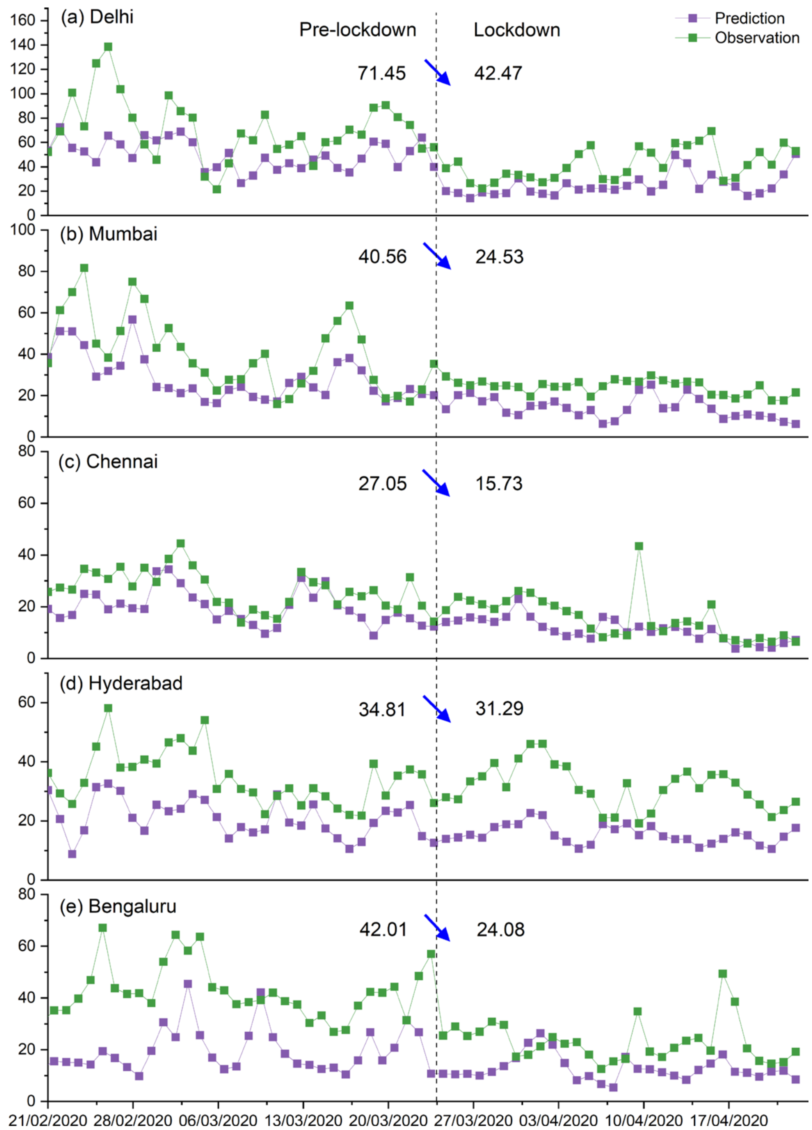

Figure 1: Comparison of predicted and observed $PM_{2.5}$ from February 21 to April 24, 2020 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The unit is μg m^{-3}.

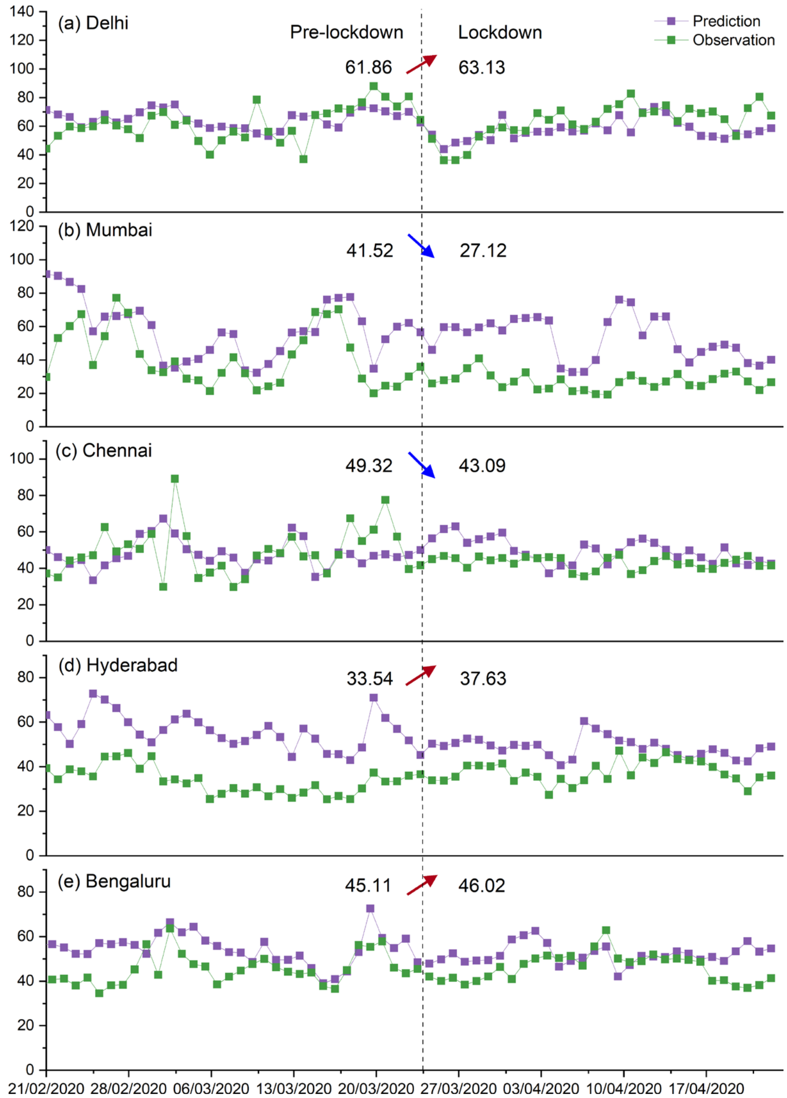

Figure 2: Comparison of predicted and observed MDA8 O_3 from February 21 to April 24, 2020 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The unit is ppb.

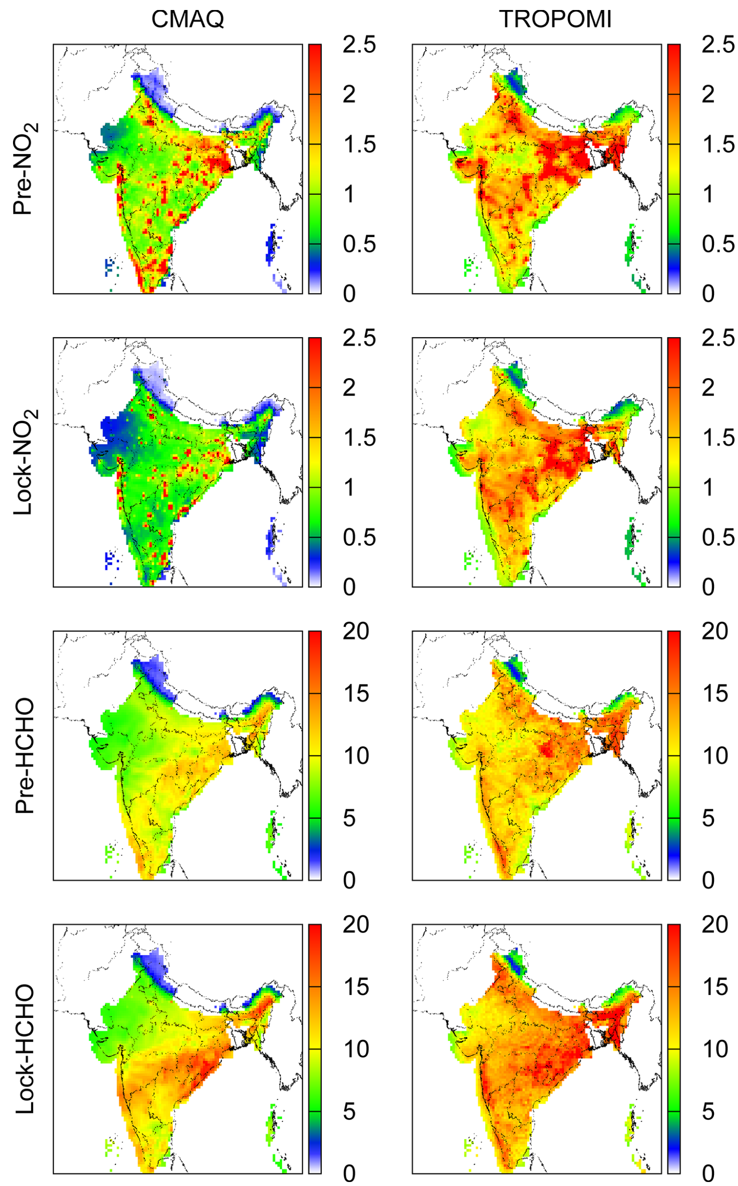

Figure 3: Comparison of the simulated and satellite-observed NO_2 and HCHO column number density before lockdown and during the lockdown in India. The unit is 10^{15} molec cm^{-2}.

Changes in manuscript:

Results and discussion (Lines 150-153 in the revision): "For $PM_{2.5}$, the averaged mean fractional bias (MFB) (-0.48) and mean fractional error (MFE) (0.61) values met the criteria limits of ±0.6 and 0.75 claimed by the EPA (2007b) in all the five urban cites after excluding some abnormally high values of greater than 300 μg m^{-3}. For O_3, a cut-off value of 40 ppb is applied, which is based on EPA's recommendations (EPA, 2005)."

Results and discussion(Lines 154-156 in the revision): "And averaged MFB (-0.05) and MFE (0.25) 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."

Results and discussion (Lines 168-172 in the revision): "Overall, sharp decreases are found in the observed $PM_{2.5}$ in all these cities, and the averaged $PM_{2.5}$ level drops from 43.18 μg m^{-3} to 27.62 μg m^{-3}. The mean observed $PM_{2.5}$ concentrations during lockdown are 42.47 μg m^{-3} (Delhi), 24.53 μg m^{-3} (Mumbai), 15.73 μg m^{-3} (Chennai), 31.29 μg m^{-3} (Hyderabad), 24.08 μg m^{-3} (Bengaluru), which are reduced by 41%, 40%, 42%, 10%, and 43% 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 57%) during the lockdown period."

Results and discussion (Lines 177-178 in the revision): "The observed average MDA8 O_3 during lockdown is higher than that of pre-lockdown in Delhi (2%), Hyderabad (12%), and Bengaluru (2%)."

Results and discussion (Lines 182-183 in the revision): "In contrast, the observed average MDA8 O_3 during lockdown is reduced compared with the pre-lockdown period in both Mumbai (-35%) and Chennai (-13%)."

Methodology (Lines 85-89 in the revision): "The satellite-observed NO_2 and formaldehyde (HCHO) column number density datasets are from the Sentinel-5 Precursor TROPOspheric Monitoring Instrument (S-5P TROPOMI) (https://scihub.copernicus.eu). Besides, we filter the satellite data under the recommended criteria of QA values greater than 75% for tropospheric NO_2 column number density datasets and 50% for HCHO (Apituley, 2018)."

Results and discussion (Lines 158-163 in the revision): "To further validate modeled HCHO and NO_2, we compared our simulated results with satellite-observed data during pre-lockdown and lockdown periods (Fig. S1). The tropospheric column densities of NO_2 and HCHO were calculated by summing their concentrations of 17 vertical layers in the CMAQ model (H. J. Eskes, 2020). The predicted regional distribution of tropospheric column NO_2 and HCHO is similar to satellite-observations. Overall, HCHO and NO_2 are higher in eastern and northern India than in other regions. And their variation trends from CMAQ and TROPOMI are consistent that NO_2 decreases while HCHO increases during the lockdown."

Comments: Validation of model using 2 m level measured meteorological data: For the kind of modelling investigation the authors are making namely, effect of emission changes on concentrations of pollutants use of only the 2 m level observations without comparison with satellite data, sonde data, mixing layer height data (see ERA5 products) seems to be a major shortcoming. Note that the changes in ventilation coefficient before and during lockdown and the changing season (Spring to Summer) can alone have big impacts on the concentrations.

Response:

Thanks for the referee's comments. The weather research and forecasting (WRF) ARW regional model is a high-resolution meteorology model that is widely used in Indian studies (Gevorgyan, 2018; Srinivas et al., 2013; Chawla et al., 2018; Ashrit and Mohandas, 2010). The capability of the WRF model has been validated for providing reliable meteorological inputs to air quality models even under extreme weather events (Zhang et al., 2020; Stella and Agnihotri, 2015; Pattanaik and Rama Rao, 2009; Rajeevan et al., 2010). In the manuscript, we validated the model performance of WRF using observation from the National Climate

Data Center (NCDC). Although only the near-surface meteorological factors are considered, our prediction also shows good performance with the comparison of satellite data (NO_2 and HCHO) (Fig. 3, also added as Fig. S1 in the revised supplement). As suggested by the referee, the reanalysis-based ERA5 product produces global atmospheric quantities at 31-km horizontal resolution combining model simulations and observations. However, there are uncertainties such as its high root-mean-square error values in the tropical and subtropical climate zone so that the reliability and applicability of the ERA5 dataset still need to be explored in India with a tropical monsoon climate (Jiang et al., 2020; Kolluru et al., 2020). In follow-up studies, we will consider its application to further validate the WRF model.

As for the impacts of ventilation coefficient and changing season, we added more analysis of the difference in meteorological conditions between pre-lockdown and lockdown periods including temperature (T), relative humidity (RH), planetary boundary layer (PBL) height, the average daily precipitation, and wind fields in Fig. 4 (also added as Fig. S3 in the revised supplement). The explanations about the impacts of these meteorological conditions on Indian air quality are also added in the Results and discussion section.

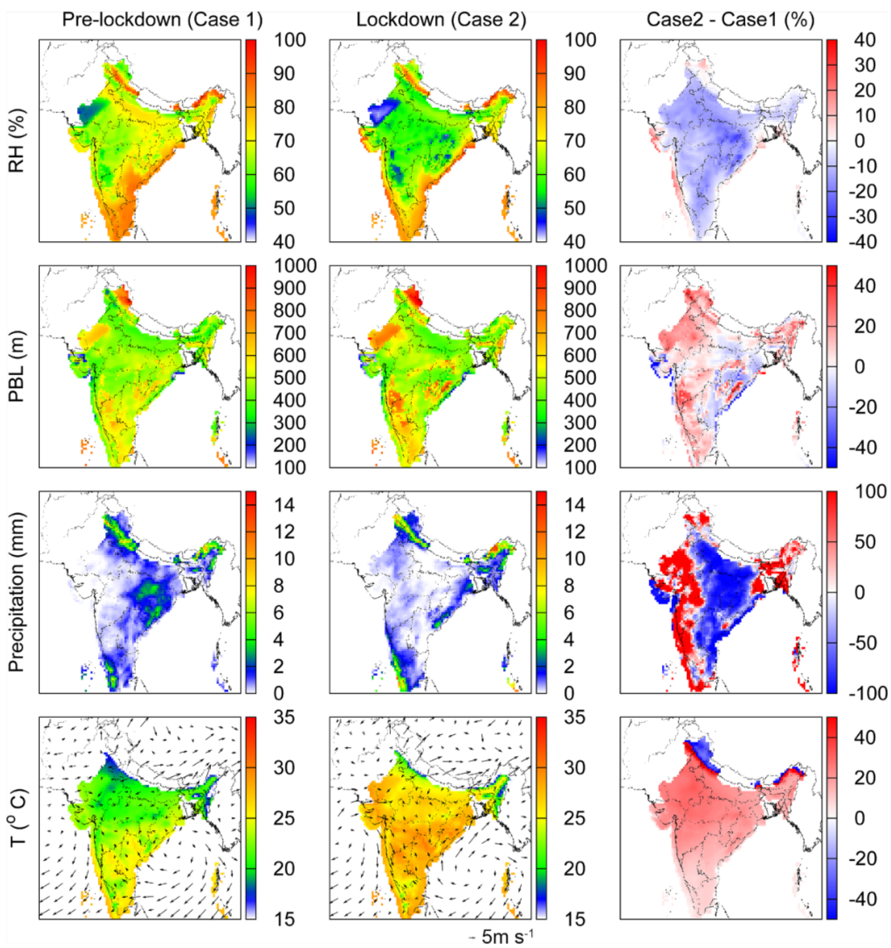

Figure 4: Distribution of simulated temperature (T), relative humidity (RH), planetary boundary layer (PBL) height, the average daily precipitation, and wind fields in India before and during the lockdown period. "Case2 - Case1" indicates (Case 2 – Case 1)/Case 1, reported as %.

Changes in manuscript:

Results and discussion (Lines 199-203 in the revision): "Variations in near-surface meteorological factors during lockdown also play an important role in $PM_{2.5}$ changes. As is shown in Fig. S3, lower $PM_{2.5}$ in urban areas during lockdown (Fig. 4) may attribute to the decrease of RH and increase of planetary boundary layer (PBL) height, while the decrease of precipitation and WS allows $PM_{2.5}$ to accumulate in some rural areas (Schnell et al., 2018; Le et al., 2020)."

Results and discussion (Lines 207-211 in the revision): "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 and decrease of RH (Fig. S3). Higher temperature speeds up photochemical processes that produce O_3, while higher RH reduces them (Chen et al., 2019; Zhao et al., 2017; Ali et al., 2012)."

Conclusion (Lines 302-303 in the revision): "It can be concluded that meteorological conditions play an important role in those increases according to the comparison between pre-lockdown (Case 1), and lockdown (Case 2)."

Comments: Changes in atmospheric chemistry of primary pollutant removal and formation of secondary pollutants:

Currently the study tends to attribute all the observed concentration changes in pollutants primarily to the emission reductions. However it has been documented elegantly in the following paper: Kroll, J.H., Heald, C.L., Cappa, C.D. et al. The complex chemical effects of COVID-19 shutdowns on air quality. Nat. Chem. 12, 777–779 (2020). https://doi.org/10.1038/s41557-020-0535-z, that several other processes play a big role. The purpose of using a model should be that these effects can be teased out through sensitivity experiments but unfortunately this has not been addressed in current version of the manuscript. For example the authors note that the temperature increased during the lockdown period. A key question is what effect the temperature change and the reduced emission of VOCs (no VOC measurements have been provided at all), NOx and CO would have on the removal rates of primary pollutants and formation of secondary pollutants.

Further have authors identified days when it rained in both pre covid lockdown and during lockdown periods which would cause strong biases for the comparisons.

Response:

We showed not only the impact of emission reductions but also that the meteorology conditions and explained that the specific chemical reactions can affect the change of pollutant concentrations. We also added more comprehensive discussions about the effects of chemical processes in the manuscript (see the Results and discussion section).

We utilized the model to explore the comprehensive effects of meteorology including the increased temperature on the change of primary and secondary pollutants by comparing pre-lockdown (Case 1) and lockdown (Case 2) rather than focusing on a single variable. We also investigated the nonlinear relationship between emissions and atmospheric composition. For example, Fig. 8 in the manuscript also showed the relative changes in concentrations of HCHO and NO_x and emissions of HCHO and NO_x in eight major cities of India in Case 2 to Case 1 and proved that the concentration of NO_x is appreciably highly sensitive to a primary NO_x emission reduction compared with HCHO.

In specific, though we can't obtain a reliable VOC observation dataset, the high correlation between HCHO and the total VOCs is shown in Fig. 5 below, and HCHO has been further validated by satellite-observations (Fig. 3, also added as Fig. S1 in the revised supplement).

We also added discussion on the effects of variations in precipitation during the lockdown on $PM_{2.5}$ and MDA8 O_3 changes in the Results and discussion results. In addition to the regional change of precipitation shown in Fig. 4 (also added as Fig. S3 in the revised supplement), we also added the daily average precipitation figures in these 5 major cities from February 21 to April 24, 2020 (Fig. 6 in the response). On average, the precipitation in India is relatively low from the pre-lockdown to lockdown periods (lower than 1 mm for each city). Although the few rainy days (such as March 5, 2020) may promote $PM_{2.5}$ removal, generally, it has little impact on the comparison of overall air quality before and during the lockdown.

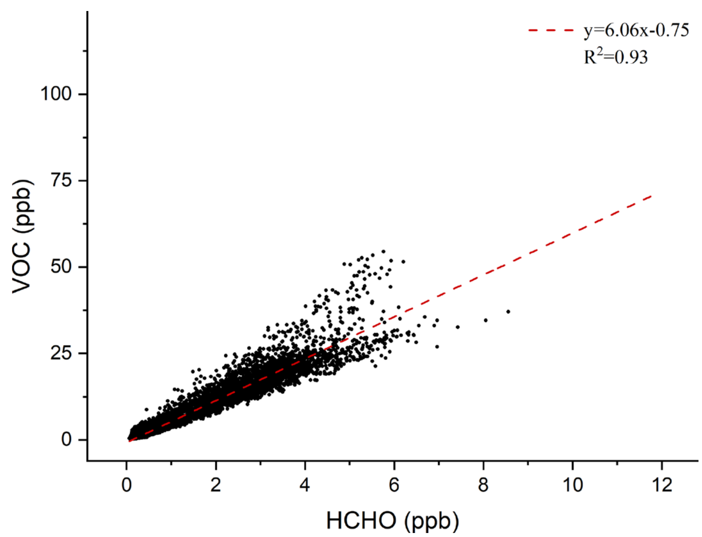

Figure 5: Scatter plots comparing the simulated average daily HCHO and the total VOCs at all 117×117 grids from February 21 to April 24, 2020.

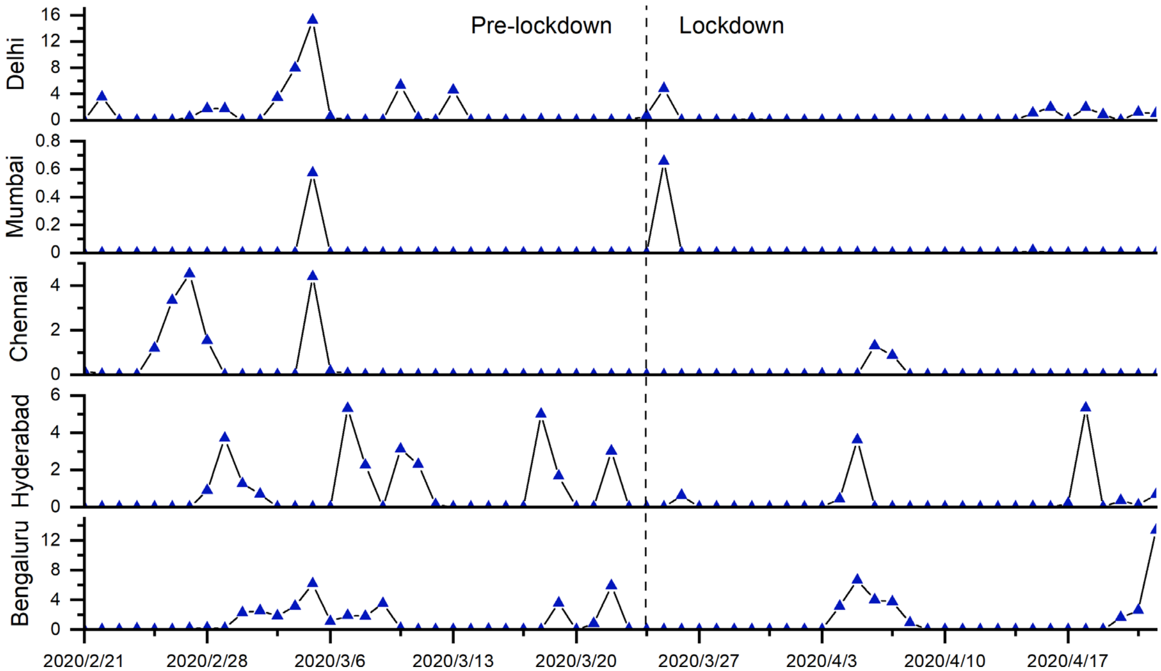

Figure 6: The predicted average daily precipitation from February 21 to April 24, 2020 in Delhi, Mumbai, Chennai, Hyderabad, and Bengaluru. The unit is mm.

Changes in manuscript:

Results and discussion (Lines 199-203 in the revision): "Variations in near-surface meteorological factors during lockdown also play an important role in $PM_{2.5}$ changes. As is shown in Fig. S3, lower $PM_{2.5}$ in urban areas during lockdown (Fig. 4) may attribute to the decrease of RH and increase of planetary boundary layer (PBL) height, while the decrease of precipitation and WS allows $PM_{2.5}$ to accumulate in some rural areas (Schnell et al., 2018; Le et al., 2020)."

Results and discussion (Lines 241-242 in the revision): "Besides, the reduction of NO_x may lead to an increase of SOA offsetting some of the influence caused by the reduction in VOC emissions (Kroll et al., 2020)."

Comments: Existing inadequacies in VOC emission inventories and modelled ozone simulations over India: While the authors are using pre lockdown and lockdown periods for comparison, it is a fact that of all emission inventories, VOC emissions are the most poorly constrained due to the absence of in-situ VOC data over many regions in India. A generic problem also seen is the tendency for overestimation of ozone by models over the Indian region. This suggests that the basic reactant mixture and chemistry are still inadequate for modelling ozone and secondary pollutant formation accurately over India. So how can one be sure that the changed chemical mixture between pre-lockdown and during lockdown are not skewed by these gaps in our basic undertanding? While it would be unfair to hold the authors to solve all these issues, one does expect that the limitations and existing issues are duly acknowledged in the work instead of making highly speculative and prescriptive measures for air quality mitigation based on such modelling results.

Response:

Thanks for pointing out this. We also acknowledge that there are still many limitations and deficiencies in the VOC emission inventories, and the simulation of O_3 is usually over-estimated (Kota et al., 2018; Hu et al., 2015). Much more needs to be done regarding emissions and chemical reactions to better simulate O_3 in India. But there is no denying that the simulation results of this model are acceptable compared with the standards recommended by EPA (EPA, 2005, 2007a). In the future, we will continuously apply new information to improve our modelling results once they are available.

Changes in manuscript:

Results and discussion (Lines 163-164 in the revision): "We also acknowledge that the uncertainty of emission inventory and chemical mechanism in the modelling may affect the simulated results (Dominutti et al., 2020; Kitayama et al., 2019)."

Comments: Use of formaldehye for constraining VOC emissions where a large number of more reactive primary VOC emissions occur should also be discussed and clarified. Trusting the formaldehye from the model in absence of in-situ formaldehye measurements to compare with or even satellite or columnar measurements which have been reported from India is recommended.

Response:

As one of the most abundant oxygenated VOCs, HCHO is one of the major contributors to total VOCs reactivity (Zhang et al., 2012; Steiner et al., 2008). Therefore, it is used to show the model performance on VOCs due to the lack of VOCs observations. Figure 5 (added as Fig.S4 in the revised supplement) shows scatter plots comparing the simulated average daily HCHO and the total VOCs at all 117×117 grids during the study period. It can be seen from the results that HCHO has a high correlation with VOCs, and R^2 reaches 0.93. We also compared the simulated HCHO and NO_2 with satellite observations (TROPOMI) to further verify the model (Fig. 3, also Fig. S1 in the revised supplement).

Changes in manuscript:

Methodology (Lines 85-89 in the revision): "The satellite-observed NO_2 and formaldehyde (HCHO) column number density datasets are from the Sentinel-5 Precursor TROPOspheric Monitoring Instrument (S-5P TROPOMI) (https://scihub.copernicus.eu). Besides, we filter the satellite data under the recommended criteria of QA values greater than 75% for tropospheric NO_2 column number density datasets and 50% for HCHO (Apituley, 2018)."

Results and discussion (Lines 158-163 in the revision): "To further validate modeled HCHO and NO_2, we compared our simulated results with satellite-observed data during pre-lockdown and lockdown periods (Fig. S1). The tropospheric column densities of NO_2 and HCHO were calculated by summing their concentrations of 17 vertical layers in the CMAQ model (H. J. Eskes, 2020). The predicted regional distribution of tropospheric column NO_2 and HCHO is similar to satellite-observations. Overall, HCHO and NO_2 are higher in eastern and northern India than in other regions. And their variation trends from CMAQ and TROPOMI are consistent that NO_2 decreases while HCHO increases during the lockdown."

Comments: Are benzene and toluene data available from the monitoring stations which could be included in the analyses? If so these should also be included in view of their health and SOA formation potential.

Response:

Thanks for the referee's comments. The available observational data for benzene and toluene from the CPCB dataset is extremely limited. For example, Chennai does not have a single monitoring site to provide its hourly observations. In the model simulation, the EDGAR emission inventory does not provide a separate benzene emission and toluene is lumped into ARO1 species in the SAPRC-11 photochemical mechanism (Carter, 2011; Hu et al., 2016). So it is a pity that the observation of benzene and toluene cannot be compared with the model simulation. Besides, our study is not focused on health risks or their specific impact on SOA formation, but on the impact of anthropogenic emission reductions on major air pollutants during the lockdown.

Changes in manuscript: No changes were made for this point.

Comments: Choice of scaling factors for emission reductions: The authors make several assumptions and justification for the use of scaling factors for emissions which are valid (see Equations 1 and 2).

For example:

Ammonia agricultural emissions: Several satellite studies have indicated high ammonia emissions from agriculture and a recent by G.K. Singh, P. Rajeev, D. Paul, et al., Chemical characterization and stable nitrogen isotope composition of nitrogenous component of ambient aerosols, Science of the Total Environment, https://doi.org/10.1016/j.scitotenv.2020.143032 showed that agriculture activities and waste generation are major sources of ammonia. The assumption by the authors that the agricultural emissions do not change between pre-lockdown and during lockdown is not valid for large parts of the India in particular the Indo-Gangetic Plain because during the pre-lockdown dates farmers were still applying fertilizers to the wheat crops, whereas by last week of March this completely stops. So infact the ammonia and hence ammonium ion source from agriculture is likely stronger in pre-lockdown period and so cannot be treated as constant between both periods. As ammonia is such an important emission for $PM_{2.5}$ too, this has large implications for the inferences currently drawn by the authors.

Response:

Thanks for the helpful suggestion from the referee. However, due to the data limitation, we cannot calculate a specific emission reduction ratio for agriculture due to the lockdown on a regional scale. As long as we can get more information, we will further refine the proportion of emission reduction in the lockdown in the follow-up study.

Changes in manuscript: No changes were made for this point.

Comments: Ozone production sensitivity indicator: The use of $HCHO/NO_2$ as based on Silman et al 1995 which the authors cite cannot be applied blindly because as noted by the original authors (Silman and He in their JGR paper in 2002) is suitable only for ambient ozone mixing ratios in the range of 80-200 ppb and then again for columns retrieved using satellite data. For ground based data, more robust proxies would be H_2O_2/ HNO_3 or even O_3/NO_y.

Response:

Thanks for the referee's comments. As shown in Fig. 7 (revised Fig. S5 in the supplement), we change the indicator of O_3 sensitivity to NO_x and VOCs into O_3/NO_y and Sillman (1995) suggested the transition value that separate NO_x-sensitive and VOC-sensitive locations (O_3/NO_y= 6-8). According to the value, we can find the most Indian region is NO_x-sensitive and the VOC-limited and transition regimes expand during the lockdown because of the reduction of anthropogenic emissions.

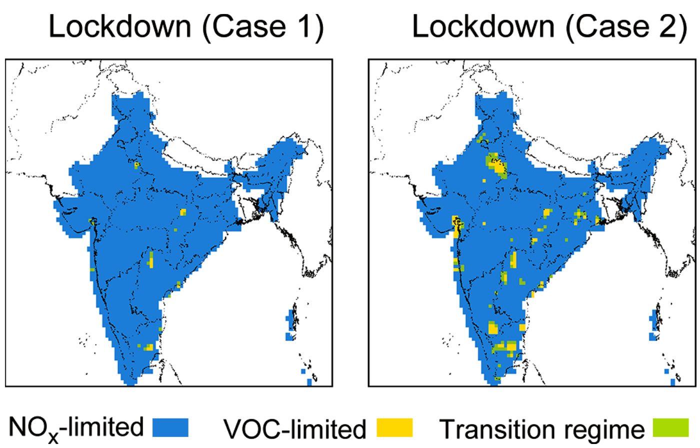

Figure 7: Spatial distributions of O_3 production sensitivity in India from March 24 to April 24, 2020.

Changes in manuscript:

Results and discussion (Lines 275-280 in the revision): "Figure S5 shows the O_3 production sensitivity (O_3/NO_y) in India during the lockdown, which is considered as an indicator of O_3 sensitivity to NO_x and VOCs (Sillman, 1995; Sillman and He, 2002). In India, NO_x-limited regimes ($O_3/NO_y > 8$) 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 ($O_3/NO_y < 6$) expands mainly in the northwest and south of India from Case 2 during the lockdown. The transition regimes ($6< O_3/NO_y < 8$) that O_3 formation is controlled by both NO_x and VOC emissions in the vicinity of the VOC-limited regions."

Comments: In the absence of measured VOC data presented by the authors to validate their model VOC data (note there are no measurements of HCHO presented), the authors should remove this discussion completely or present for each city site the high resolution O_3 Vs NO_x data from daytime for pre and during lockdown periods.

Response:

As shown in responses to previous comments, we compared the simulated HCHO with satellite observations (TROPOMI) to further verify the model (Fig. 3, also revised Fig. S1 in the supplement). We believe that the discussion is useful to readers, and we acknowledged that more studies are needed to better illustrate the relationship between O_3, VOCs, and NO_x.

Changes in manuscript:

Methodology (Lines 85-89 in the revision): "The satellite-observed NO_2 and formaldehyde (HCHO) column number density datasets are from the Sentinel-5 Precursor TROPOspheric Monitoring Instrument (S-5P TROPOMI) (https://scihub.copernicus.eu). Besides, we filter the satellite data under the recommended criteria of QA values greater than 75% for tropospheric NO_2 column number density datasets and 50% for HCHO (Apituley, 2018)."

Results and discussion (Lines 158-163 in the revision): "To further validate modeled HCHO and NO_2, we compared our simulated results with satellite-observed data during pre-lockdown and lockdown periods (Fig. S1). The tropospheric column densities of NO_2 and HCHO were calculated by summing their concentrations of 17 vertical layers in the CMAQ model (H. J. Eskes, 2020). The predicted regional distribution of tropospheric column NO_2 and HCHO is similar to satellite-observations. Overall, HCHO and NO_2 are higher in eastern and northern India than in other regions. And their variation trends from CMAQ and TROPOMI are consistent that NO_2 decreases while HCHO increases during the lockdown."

Comments: In several instances, the grammar and language also need to be corrected. I recommend the authors to consider the above major concerns to revise and improve the manuscript.

Response: As suggested, we made corresponding changes and improved the grammar and language in the revised manuscript.

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