Response to Referee #2 (acp-2022-729)

We Thank Reviewer for his/her constructive comments

Responses to the Specific comments

General comments: This manuscript evaluates the changes in air quality during the COVID-19 lockdown by dividing them into meteorological and emissions parts. The authors specifically assessed the impact of emission reduction during the lockdown using a multi-air pollutant inversion system and observational data, which is a unique approach compared to previous studies. The findings from this study will be valuable in evaluating the effectiveness of emission reduction policies by policymakers in polluted regions, including China. However, to be published in ACP, the authors must address the following issues:

Reply: The authors appreciate the reviewer for his/her constructive suggestions. In the revised manuscript we have considered each comment for improvement, revision, and correction. Please refer to our responses for more details given below.

Comment 1: Line 39: During the COVID-19 period, a haze event also occurred. However, the use of the term "COVID-19 haze" may convey the notion that the pandemic was the cause of the haze phenomenon. Thus, the authors should choose terms more carefully.

Reply: Thanks for pointing out this issue. The "COVID-19 haze" has been replaced by "unexpected $PM_{2.5}$ pollution during the COVID-19 lockdown" in the revised manuscript.

Comment 2: Lines 128-129: Despite removing unrealistic observations through the Wu et al. (2018) method, some extreme values persist in the time-series plots (e.g., Figures 3(b), S1(b), and S2(b)). Hence, the authors should thoroughly verify that the raw data has been properly filtered.

Reply: Thanks for this important comment. We have checked the quality of observation data we used in the assimilation. According to Fig. 3(b), S1(b) and S2(b), the possible extreme values for $PM_{2.5}$, PM_{10} and SO_2 mainly occurred over the NE region around 25th January. Figure R1(a) shows the spatial distributions of daily averaged $PM_{2.5}$ concentrations over the NE region. Obvious high $PM_{2.5}$ concentrations could be found at multiple monitoring sites over Liaoning province, where the $PM_{2.5}$ concentrations increased sharply (over 400 $\mu g/m^3$) at the night of 24th January and early morning of 25th January (Fig. R1(b)). Same phenomena also occurred in the city Baotou at Inner Mongolia (Fig.R1(c)). Considering that the 24th January was the 2020 Chinese New Year Eve and there are traditions of setting fireworks at the night of that day, the peak $PM_{2.5}$ concentrations at the night of 24th January may be related to the firework emissions. Meanwhile, there were multiple sites that showed same signals of high $PM_{2.5}$ concentrations. Thus, we think the high $PM_{2.5}$ concentrations during 24th January is reasonable, and did not treat them as outliers. Similar conclusions can be drawn for PM_{10} and SO_2, as seen in Fig. R2 and R3.

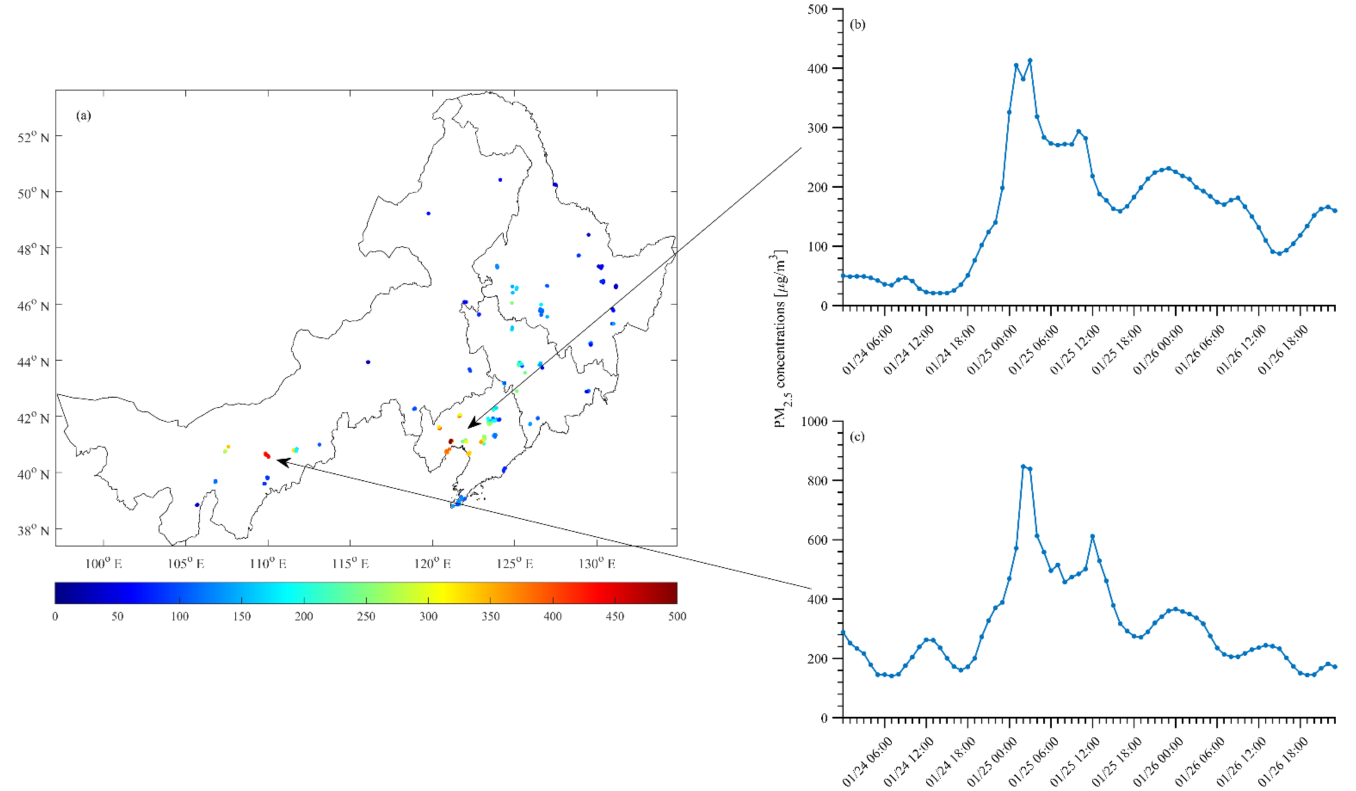

Figure R1: (a) spatial distribution of daily averaged $PM_{2.5}$ concentrations over the NE region at 25th January 2020, and the time series of averaged PM2.5 concentrations at (b) Liaoning province and (c) the city of Baotou at Inner Mongolia from 24th to 26th January 2020.

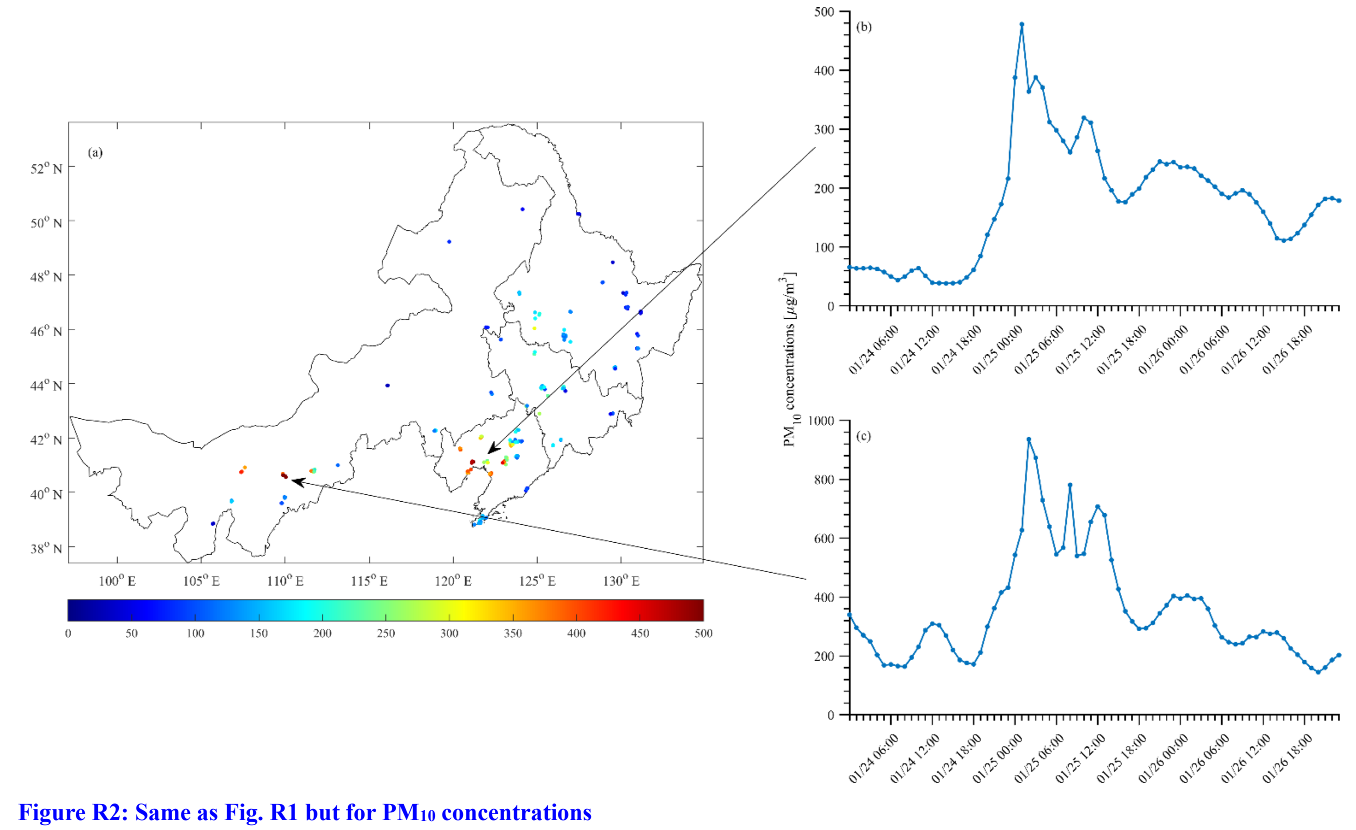

Figure R2: Same as Fig. R1 but for PM_{10} concentrations

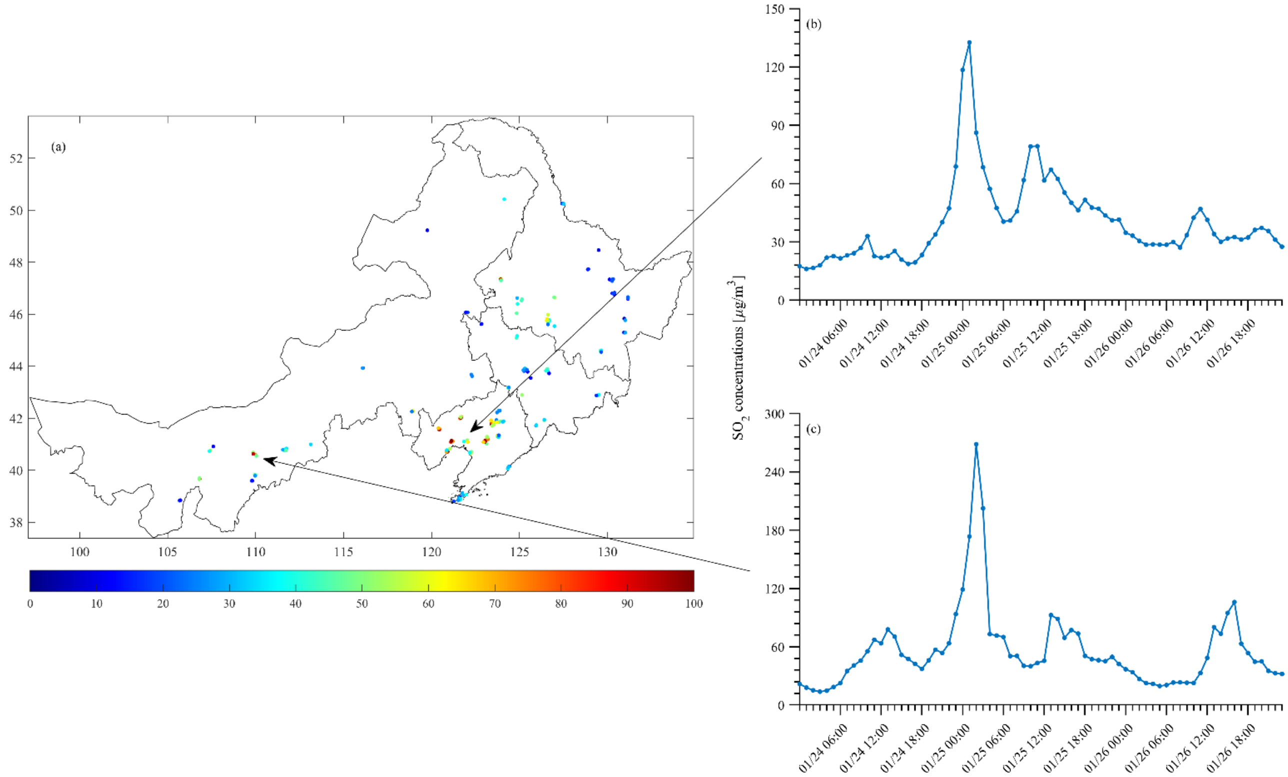

Figure R3: Same as Fig. R1 but for SO_2 concentrations

Comment 3: Lines 208-242: (Section 2.4) The authors assess both the MI and EI approaches for decreasing nonlinear effects. If the extent of nonlinearity (or sensitivity) demonstrated by the two methods is documented in the paper, it can provide a helpful reference for future research.

Reply: Thanks for the suggestion. We analyzed the differences between the MI and EI method in the revised manuscript (please see lines 393–398) and supplement (please see Figure S13 and S14). According to Figure R4 and R5, the calculated MI and EI changes of $PM_{2.5}$ and O_3 concentrations were consistent with each other over the Beijing and the NCP region, and indicates similar conclusions. The differences of calculated MI and EI were within 2 $\mu g/m^3$ for $PM_{2.5}$ concentrations, which were small in this application. In terms of O_3 concentrations, the differences were larger, which were around 5 $\mu g/m^3$ over the Beijing and NCP region. In addition, the sign of calculated MI using EMIS change scenario and MET change scenario were opposite although both suggested weak contributions of meteorological variation to the changes of O_3 concentrations, suggesting that the calculated MI and EI changes of O_3 concentrations could be more sensitive to the used scenarios. This may be due to the stronger nonlinearity of O_3 concentrations to the meteorology and emissions.

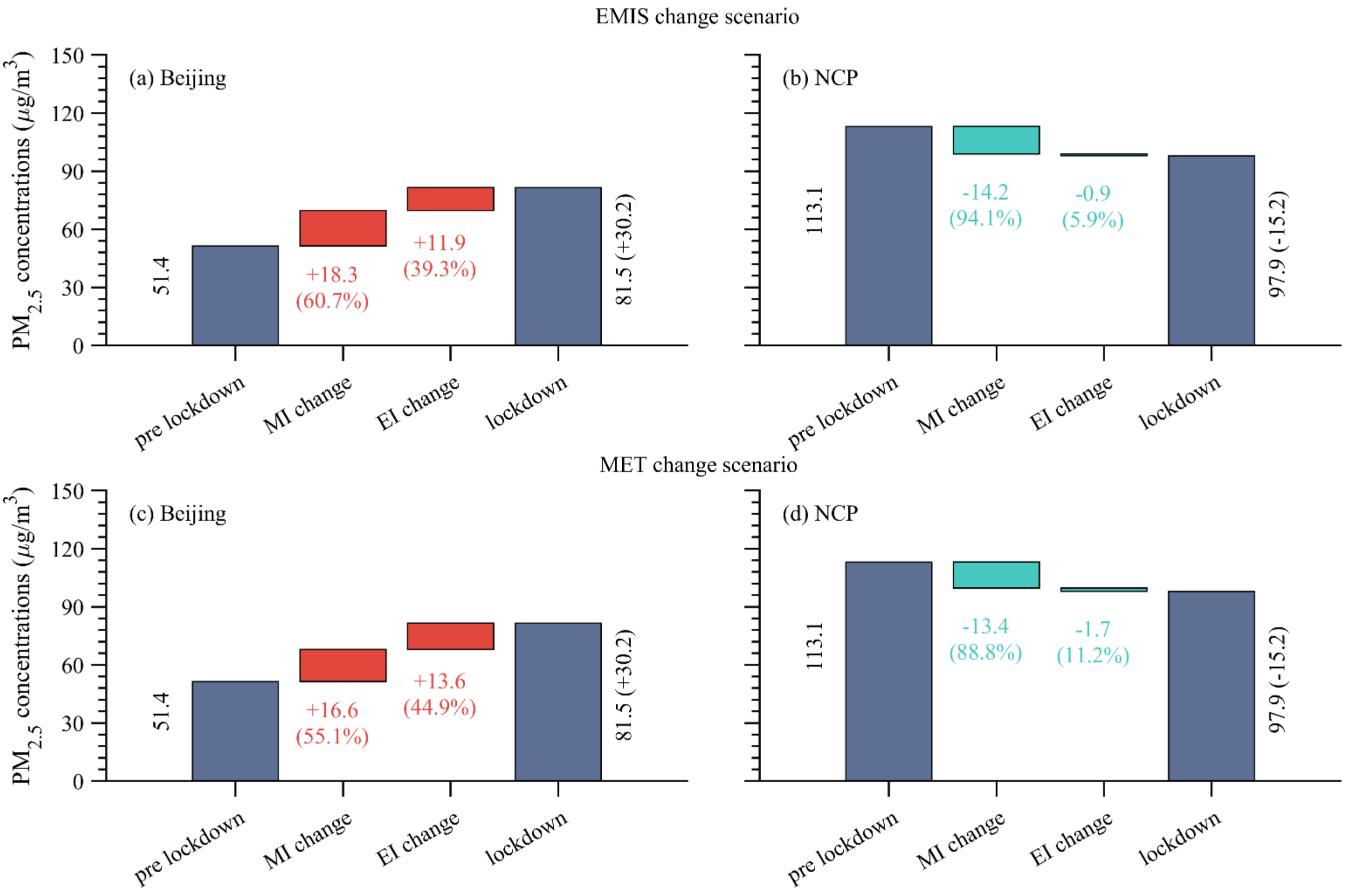

Figure R4: The calculated MI and EI changes of $PM_{2.5}$ concentrations over the (a, c) Beijing and (b, d) the NCP region using the EMIS change scenario (upper panel) and MET change scenario (lower panel).

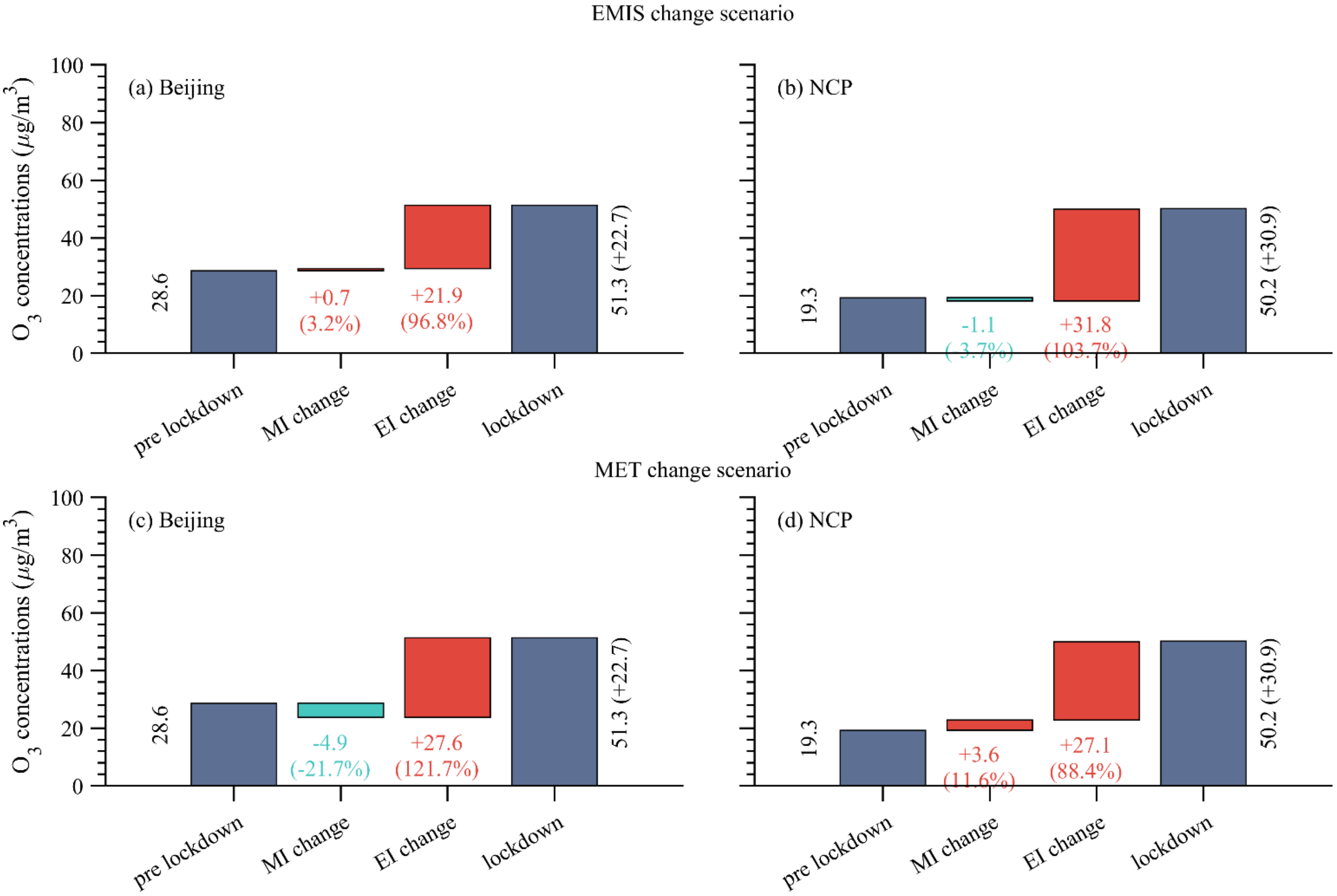

Figure R5: Same as Fig. R4 but for O_3 concentrations.

Comment 4: Lines 302-304: The authors posit that the rise in PM_{10} emissions in the NW and central regions during P3 is due to sandstorms but do not provide clear evidence. Furthermore, the simulation using a priori in the central region does not show a significant deviation from observation. Thus, the authors must provide further evidence for the sandstorm hypothesis.

Reply: Thanks for raising this issue. As we illustrated in Sect. 2.3 (lines 215–217 and table 1), the PM_{10} emissions were calculated by the sum of the emissions of $PM_{2.5}$ and PMC (coarse mode unspeciated aerosol) which were respectively constrained by the concentrations of $PM_{2.5}$ and $PM_{10-2.5}$. As we can see from Fig. R6, there were significantly larger deviations in the simulated PM_{10} – $PM_{2.5}$ concentrations from the observations over the Central region during the P3 period, which led to the rise in PM_{10} emissions over there. However, as the reviewer mentioned, the a priori PM_{10} simulation does not show a significant deviation from observations (Figure S2(f)), this may due to that the underestimations of $PM_{10-2.5}$ were partly compensated by the overestimated $PM_{2.5}$ concentrations over the Central region (Fig. S1(f)). Meanwhile, we used the $PM_{2.5}/PM_{10}$ ratio to investigate the potential causes of the increases in PM_{10} emission over the NW and Central regions in the revised manuscript, which is an indicator of the potential sources of particular matter. A lower $PM_{2.5}/PM_{10}$ ratio usually indicates significant contributions from natural sources such as dust (Wang et al., 2015; Fan et al., 2021). As we can see from Fig.R7, the $PM_{2.5}/PM_{10}$ ratio was stable during the P1 and P2 period, but it decreased substantially during the P3 period, from 0.81 to 0.48 over the NW region and from 0.77 to 0.53 over the Central region, which suggests larger contributions of dust emissions to the PM10 concentrations during the P3 period. Moreover, the NW and Central region are typical source areas of dust in China, therefore the increasing of PM_{10} emissions over NW and Central regions may be mainly related to the enhanced dust emissions. following the suggestion of reviewer, we added more explanations to the increased PM_{10} emissions over the NW and Central region in the revised manuscript (please see lines 342–348).

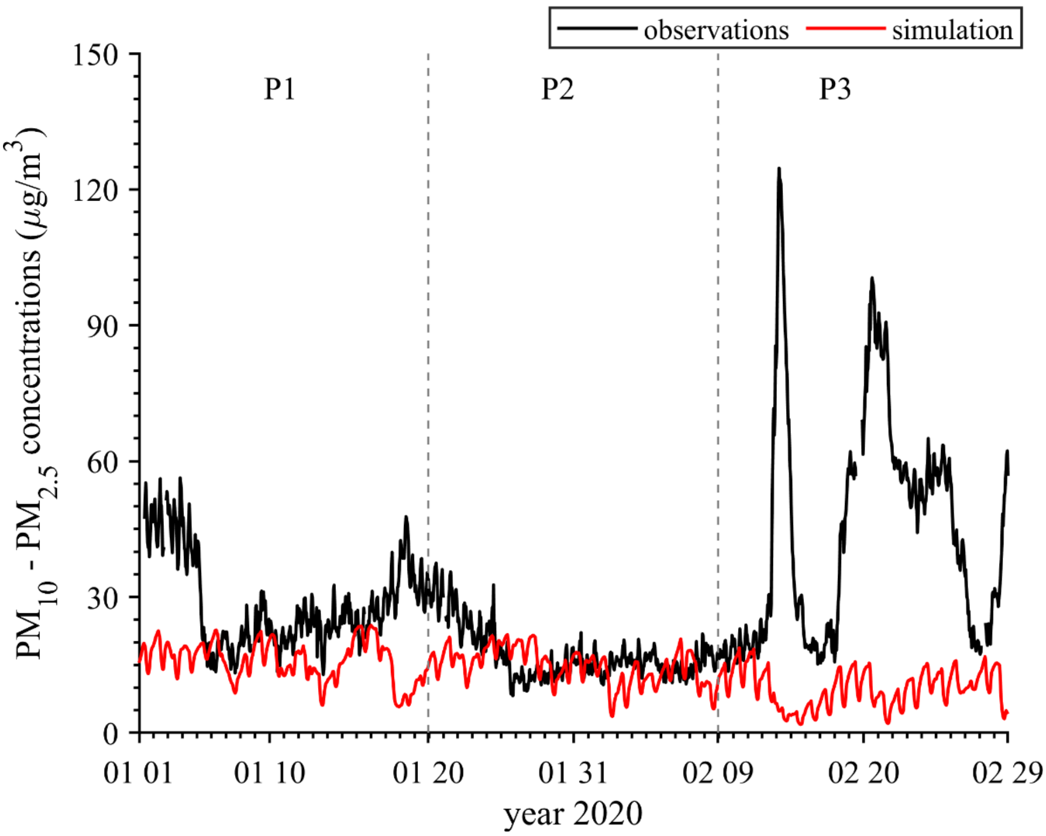

Figure R6: Timeseries of observed and simulated PM_{10} – $PM_{2.5}$ concentrations over Central region during COVID-19 pandemic.

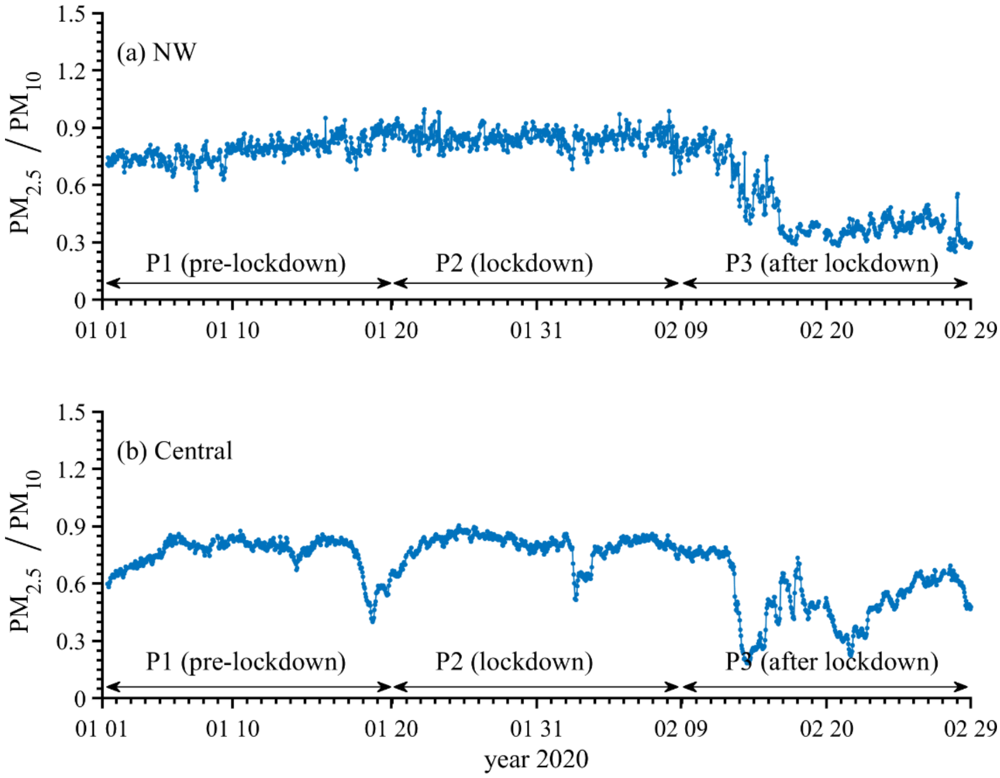

Figure R7: Timeseries of $PM_{2.5}/PM_{10}$ ratio during COVID-19 pandemic over (a) NW and (b) Central region

Comment 5: Line 306: east China -> southeast China

Reply: Done

Comment 6: Lines 311-312: There is a change in the values of SO_2 and PM_{10}.

Reply: Thanks for pointing out this mistake. The captions of Fig. 4d and Fig. 4f were wrongly labeled. We have corrected this in the revised manuscript.

Comment 7: Lines 313-315: The authors suggest that CO emissions decline significantly, as CO's transportation share (18%) is higher than SO_2(5%) and $PM_{2.5}$ (6%) (as shown in Figure 4). However, the percentage decrease in emissions is insignificant (-10.6% vs. -9.7% and -7.9%, as shown in Table 2). Furthermore, while the transportation share of PM_{10} emissions is only 2%, the emission decrease is -12.1%, which is greater than that of CO. Hence, other factors beyond transportation may have influenced the reduction in anthropogenic emissions during P2. Therefore, the authors should clarify their results.

Reply: Thanks for raising this important issue. We agree with the review that the percentage decrease in emissions of CO, SO_2 and $PM_{2.5}$ is not significant compared with the differences in their transportation share. This may be on the one hand due to the uncertainty in the estimated relative contributions of different sectors to the total emissions of CO, SO_2 and $PM_{2.5}$, on the other hand were possibly due to the uncertainty in the emission inversions, especially considering that the decreasing trend of CO, SO_2 and $PM_{2.5}$ were not significant. Also, other factors beyond transportation may have influenced the reductions of anthropogenic emissions during P2 period. For example, the larger reductions of PM_{10} emissions may be related in part to the reduced dust emissions due to shutting down of construction sites during the lockdown period (Li et al., 2020). Following the suggestion of reviewer, we have clarified it in the revised manuscript (please see lines 359–366).

Comment 8: Lines 317-318: The values given are incorrect (e.g., SO_2 is 77.6%, not 86%).

Reply: Thanks very much for your careful check. The given value is correct. It was that the caption of Fig. 4d and Fig. 4f was labeled wrongly, and we have corrected this mistake in the revised manuscript.

Comment 9: Lines 329-389: (section 3.3) The results presented by the authors, such as the significant contribution of meteorological fields to $PM_{2.5}$ during the pandemic and the titration effect on O_3, have been reported in previous studies. Hence, the authors should distinguish the difference between their results and previous studies using numerical values.

Reply: Thanks for this suggestion. However, it is difficult to directly compare our results with previous studies due the altered definition of meteorological contribution, different reference period that used to quantify the meteorological contributions and different targeted region. For example, in Song et al. (2021), the reference period used to determine the meteorological contribution is the corresponding period of COVID-19 pandemic in 2019. Le et al. (2020) used the multiyear climatology as the reference period. in Wang et al. (2020) and Sulaymon et al. (2021), the MI changes of $PM_{2.5}$ concentrations were defined as the difference between the modeled concentrations in high-pollution days and those in low-pollution days under hypothetical emission reduction scenario. Zhao et al. (2020) used a similar reference period to ours to determine the MI changes but they used the outdated emission inventory.

Table R1 summarized the studies that differentiated the contributions of meteorology and emission to the $PM_{2.5}$ concentrations over Beijing and the Beijing-Tianjin-Hebei (BTH) region. Note that some studies only provided the relative changes in the modeled $PM_{2.5}$ concentrations. It shows that due to the unknown emission changes during COVID-19 pandemic, the EI changes estimated by Zhao et al. (2020) were possibly largely overestimated compared to our studies (55% versus 24.7%). Both Sulaymon et al. (2021) and Wang et al. (2020) suggested negative EI changes during COVID-19 period in Beijing. This because they presumed that the emissions were largely reduced during COVID-19 lockdown which may deviate from the real changes of emissions according to our inversion results. Meanwhile, although they used same method and reference period, their results differed largely (-2.7 versus -13.4 $\mu g/m^3$) due to the different emission reduction scenario they assumed to represent the emissions during COVID-19 pandemic. Le et al. (2020) only considered the emission reductions of NO_x in their sensitivity simulations without considerations of other species, therefore their calculated EI changes may be underestimated compared to our results (almost 0% versus 24.7%). However, the calculated MI changes were consistent between our study and Le et al. (2020). In terms of O_3, the calculated EI changes by our study were also higher than that calculated by Zhao et al. (2020) in Beijing (85.7% versus 70%). These results suggested that the EI and MI changes calculated by our study could be more reasonable, as the emissions of different species were well constrained which could better represent the temporal variation and spatial heterogeneity of emission changes during COVID-19. Following the suggestions of reviewer, we have added the comparison of our results with previous studies in the revised manuscript (please see lines 442–464)

Table R1. calculated MI and EI changes in $PM_{2.5}$ concentrations during COVID-19 pandemic by previous studies

	MI changes	EI changes	Region	Reference period	Method	Reference
1	26.79 μg/m³	-21.84 μg/m³	Beijing	January 23-March 10, 2019 versus January 23-March 10, 2020	observation-based wind-decomposition method	Song et al. (2021)
2	Around 20 μg/m³	-2.7 μg/m³	Beijing	January 01 to February 29, 2020	CTM with hypothetical emission reduction scenario	Sulaymon et al. (2021)
3	Around 45 μg/m³	-13.4 μg/m³	Beijing	January 01 to February 29, 2020	CTM with hypothetical emission reduction scenario	Wang et al. (2020)
4	31.3%	Around 0%	Beijing-Tianjin-Hebei	January 01 to February 13, 2020	CTM sensitivity simulations using different emission rates and multiyear climatology	Le et al. (2020)
5	Around 5%	Around 55%	Beijing	January 16-22, 2020 versus January 26 to February 1, 2020	CTM with fixed emission inventory for 2017	Zhao et al. (2020)
6	17.5 μg/m³ (34.0%)	12.7 μg/m³ (24.7%)	Beijing	January 1-20, 2020 versus January 21 to February 9, 2020	CTM with inversion emission inventory	This study

Comment 9: Lines 340-341: The relative overestimation of ozone is not clear. Please provide a specific value. Also, Figure 7 is not related to this.

Reply: Thanks for this comment. The simulated increases in O_3 concentrations from pre-lockdown to lockdown period were 30.9 μg/m³ over the NCP region, which is slightly higher than the observed increases in O_3 concentrations (28.3 μg/m³). Following the suggestions of reviewer, we have clarified in the revised manuscript (please see lines 389–390), and we feel sorry for the wrong quotation of "Fig. 7" there which has been corrected in the revised manuscript (please see lines 390).

Comment 10: Lines 359-361: The author's assertion that the rise in $PM_{2.5}$ levels in the Beijing region is mainly due to fireworks during the Spring Festival is not supported by sufficient evidence, as there is no evidence that the increase in fireworks emissions is unique to Beijing.

Reply: Thanks for this important comment. Following the suggestions of reviewer, we have added more explanation of the possible causes of the $PM_{2.5}$ emission increases in the Beijing during the lockdown period through literature review and analysis of the $PM_{2.5}$ compositions. Zuo et al. (2022) analyzed the variations in the $PM_{2.5}$ sources based on the measurement of stable Cu and Si isotopic signature and metal concentrations of $PM_{2.5}$ in Beijing, which indicated that the primary $PM_{2.5}$ emissions did not decrease in Beijing during COVID-19 lockdown, and that the PM-associated industrial emissions may increase in Beijing and its upwind region during the lockdown period. Meanwhile, substantial high levels of potassium (K) and barium (Mg) were observed over Beijing during Spring Festival as seen from Fig. R8, which is an important fingerprint of the firework emissions. This suggest that the emissions from fireworks during Spring Festival were also a potential contributor to the increased of $PM_{2.5}$ emissions in Beijing, which is consistent with the measurement by Ma et al. (2022) and Dai et al. (2020). Therefore, the increased $PM_{2.5}$ emissions during lockdown period in Beijing may be attributed to the increased industrial $PM_{2.5}$ emissions and the firework emissions, which compensated the emission reductions from the traffic emissions. Following the suggestions of reviewer, we have clarified this in the revised manuscript (please see lines 416–418)

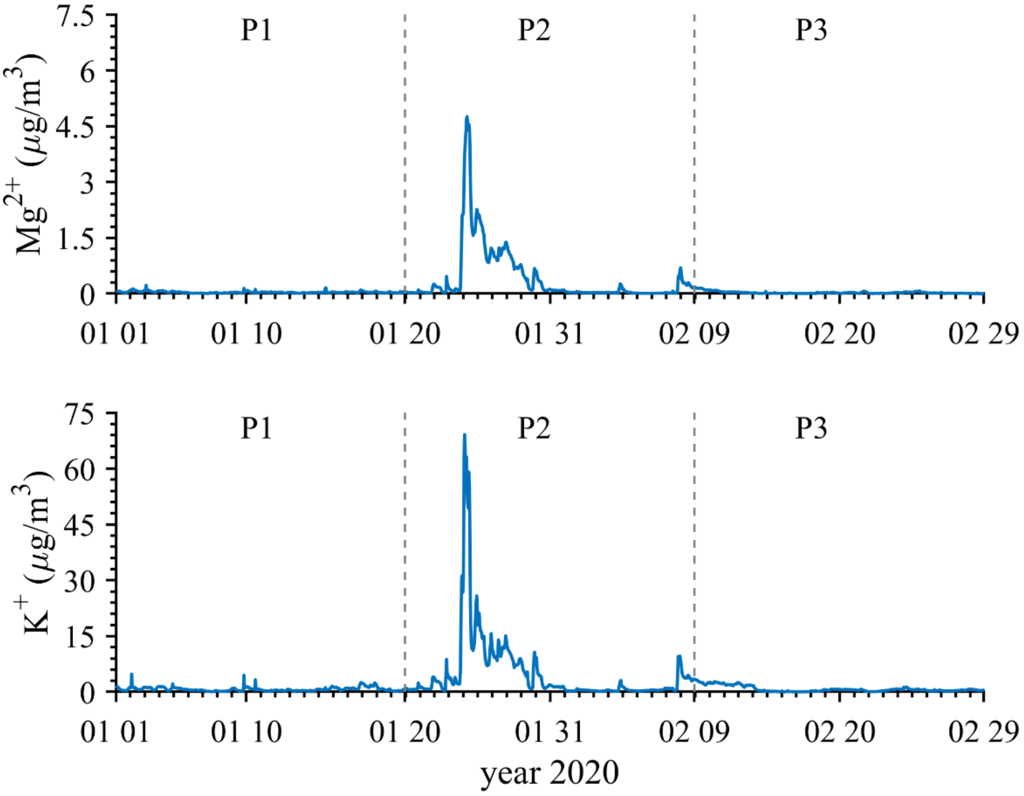

Figure R8: Timeseries of averaged concentrations of potassium and magnesium ion during COVID-19 pandemic over the Beijing.

Comment 11: Line 441: Some subscripts are misspelled.

Reply: We have corrected it in the revised manuscript.

Comment 12: Lines 460-461: Is the unit for ozone also μg m^{-3}?

Reply: Yes, the unit for ozone is also μg/m^3.

Comment 13: Line S36: Figure S4 -> Figure S7

Reply: We have corrected it in the revised manuscript.

References

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