

Response to Reviewer #2 (ESSD-2023-477)

We Thank Reviewer for his/her constructive comments

Responses to the Specific comments

General comments: The manuscript presents a comprehensive study aimed at estimating air pollutant emissions in China through the assimilation of surface observations. The authors found that the emission reduction efforts during the 2018-2020 Action Plan generally exceeded those of the 2013-2017 Action Plan. They also conducted comparisons with various bottom-up emission inventories, and provided detailed explanations for differences and uncertainties. These findings are relevant and potentially important. However, while reading the manuscript, I encountered several unanswered questions, mainly related to the settings and parameters of the estimation technique, as well as potential uncertainties and biases in the inferred emission estimates. In particular, I have doubts about the credibility of the NMVOC emission inversion. I believe that further analysis and discussion addressing the major and specific issues outlined below are necessary to substantiate the authors' claims and make the manuscript suitable for publication in ESSD:

Reply: The authors appreciate the reviewer for his/her constructive and insightful comments. We have carefully considered the comments and revised the manuscript accordingly. Please refer to our responses for more details given below.

Major comments:

Comment 1: The authors compare the posterior results with other sources and frequently employ terms like "underestimate" and "overestimate" without explicitly specifying what is considered an under- or overestimate relative to a reference. For instance, in line 47, the use of these terms lacks clarity. More critically, the terms "underestimate" and "overestimate" imply that the posterior is inherently closer to the truth than the other sources, assuming that the other sources are less accurate. This assumption is not self-evident. In inversion, adjusting emissions to match observations does not conclusively prove that the posterior emissions are improved, nor does it inherently indicate biases in other bottom-up inventories. To claim that HTAP and other sources are less accurate and to justify the terms "overestimate" and "underestimate," the authors need to provide a more convincing argument. Simply relying on posterior simulations is not sufficient to demonstrate the improvement in posterior emissions and the existence of biases in other bottom-up inventories. If a more convincing argument cannot be made, the authors should consider using more neutral terms to avoid implying a hierarchy of accuracy among different emission sources. An example of inconsistency can be found in Section 4.3, where the authors, in comparing their emission inventory with others, occasionally use alternative inventories as a basis to highlight the agreement and reduced uncertainty of their inventory compared to

bottom-up inventories. At other times, however, they claim that these alternative inventories exhibit significant uncertainties. This contradiction raises concerns about the clarity and consistency of the manuscript. It is essential that the authors provide a more coherent explanation or rationale for the varying assessments of uncertainty in other inventories.

Reply: Thanks for this important comment. We apology for the ambiguous employment of the terms like "underestimate" and "overestimate" when we compare the posteriori results with other sources. As the reviewer suggested, we have made revisions to these expressions throughout the manuscripts to ensure greater clarity in our intended meaning. For example, the unclear expression in line 47 has been revised to “the CAQIEI suggested higher NMVOC emissions than the other emission inventories by about 30.4–81.4% over the NCP region but suggested lower NMVOC emissions by about 27.6–0.0% over the SE region.” Similar revisions have also been made throughout the manuscript.

We agree with the reviewer that adjusting emissions to match observations does not conclusively prove that the posterior emissions are more accurate than the other emission inventories. The purpose of comparisons of CAQIEI with other emission inventories in our manuscript is also not to prove the superiority of the inversion inventory, nor to rank the accuracy among different inventories. In fact, it is difficult to validate the estimated emission inventory because of the unavailability of the truth value or observations of air pollutant emissions, which is the main challenge faced in the research of emission inventory. Thus, one of the most important values of the top-down analysis is to provide valuable clues for verifying the emission inventory (Zhang et al., 2009; Streets et al., 2006) through the assimilations of observations of air pollutant concentrations, as demonstrated in many inversion studies, such as Miyazaki et al. (2017); Zheng et al. (2019); Goldberg et al. (2019) and so on. Therefore, similar to pervious top-down studies, the primary objectives of our manuscript are to utilize the inversion method to investigate the changes of air pollutant emissions in China, and to provide information about potential uncertainty in current understandings of the Chinese air pollutant emissions. The inversion emissions are also useful for improving the emission inventories. We feel sorry that the inappropriate use of the terms like "underestimate" or "overestimate" in the manuscript gives the implications about the hierarchy of the accuracy among different emission sources. This is not the intendency of our work. Following the suggestions of reviewer, we have made a throughout revision to our manuscript, especially for Sect 4.3 by using more neutral terms to make our intendency clearer. In the revised Sect 4.3, we only highlight the similarity and differences among the different inventories, and delete the terms like underestimate and overestimate. Meanwhile, the consistency in Sect 4.3 has also improved in the revised manuscript by giving a more coherent explanation. (please see lines 650–794 in the revised manuscript)

Comment 2: In particular, the authors' comparison of natural and anthropogenic species emissions (such as PM10 and NMVOC) reveals a significant issue. Natural sources inherently exhibit considerable uncertainty, and in many regions, natural sources contribute significantly more than anthropogenic sources. Therefore, using the uncertainty in natural sources as a basis does not necessarily indicate large uncertainties in anthropogenic sources within the bottom-up inventories. An inconsistency arises in Line 790, where the authors' explanation appears contradictory. They simultaneously assume minimal variations in natural sources and cite literature indicating an increasing trend in natural sources. Additionally, the manuscript attributes emission changes to anthropogenic sources while acknowledging substantial uncertainty in natural sources. If it is acknowledged that natural sources indeed carry significant uncertainty (which is indeed the case), the manuscript should avoid using terms such as "not captured," "overestimated," or "underestimated" concerning the bottom-up inventories. These terms imply a clear attribution of error that may not be justified given the uncertainties associated with natural sources. Clear and consistent handling of uncertainties in both natural and anthropogenic sources is crucial for maintaining the credibility of the manuscript.

Reply: Thanks for this suggestion. We acknowledge the significant uncertainty existed in the natural sources and its implications for the assessment of anthropogenic emissions. As we illustrated in the responses to Comment 1, we are not intended to conclusively prove the biases in existing emission inventories, but to provide clues for their possible deficiencies through the comparison of our inversion results with the other emission inventories. However, since our inversion inventory does not differentiate the natural and anthropogenic sources, the natural sources have to be considered to make different emission inventories comparable. We apology the inappropriate use of the terms “not captured”, “overestimated” and “underestimated” in the comparisons of different emission inventories, which did not accurately convey our true intent and caused some inconsistencies and contradictions in the manuscripts as the reviewer mentioned. Following the suggestions of reviewer, we have made following revisions to provide a clear and consistent handling of uncertainties in the natural and anthropogenic sources in our manuscript:

- 1) Add the discussions about the uncertainty in the natural sources and its implications on the comparisons of inversion results with other anthropogenic emission inventories. We also highlighted in the revised manuscript that the comparisons conducted in this study is on the basis of the natural sources estimated by CAMS and GFAS inventories, which would be sensitive to the used natural emission inventories, and does not necessarily indicate large uncertainties in anthropogenic sources within the bottom-up inventories. (please see lines 657–665, 786–788, 994 – 1001 in the revised manuscript).
- 2) Rewrote the expressions including the terms such as "not captured," "overestimated," or "underestimated" concerning the bottom-up inventories throughout the manuscript. For example, the lines 766–767 in the original manuscript has been revised to “In particular, CAQIEI suggests increases of NO_x

emissions over the Central region, which is opposite to the previous emission inventories.” The lines 781–782 in the original manuscript has also been deleted in the revised manuscript. More revision is available in the Sect 4.3 in the revised manuscript.

- 3) The inconsistent or contradictory expressions in the manuscript have also been revised to maintain the consistency of the manuscript. For example, we do not simultaneously attribute the increase in NMVOC emissions to biogenic sources while also stating that the variation in biogenic sources is not significant. Instead, in the revised manuscript, the emission trends of biogenic NMVOC (also for other species) are only estimated based on the CAMS emission inventory, and further analysis is also made based on this assumption (lines 593–597). The uncertainty of this assumption and its potential impacts on the comparisons of our inversion results with previous emission inventories were then discussed in the manuscript (please see lines 657–665, 786–788, 994 – 1001 in the revised manuscript). This would help increase the consistency of the manuscript. Regarding to the inconsistency that we attribute the emission changes to anthropogenic sources while acknowledging substantial uncertainty in natural sources. Since our inversion result do not differentiate the anthropogenic and natural sources, it is difficult to directly compare our inversion results with previous emission inventories. To deal with this issue, the natural sources estimated by CAMS and GFAS were used in this study to account for the influences of natural sources. Therefore, our primary intention is to make an attempt to compare our inversion results with previous emission inventories on the basis of state-of-art estimations of natural source. However, as we are aware of that despite the use of state-of-art estimations of natural sources, there is still significant uncertainty in the estimated emission trends of natural sources, which would influence the comparison results of our inversion inventory with previous emission inventories. That’s why we acknowledge the large uncertainty in the estimated natural sources. This could help the potential reader better understand the comparison results of CAQIEI with previous emission inventories. We feel sorry that the inappropriate expression in our original manuscript did not correctly convey our intention, and lead to inconsistency. In the revised manuscript, we explicitly pointed out that the comparison conducted in our study is on the basis of natural emissions estimated by CAMS and GFAS at the beginning of Sect. 4.3 (Lines 657 – 665), which would be sensitivity to the used natural sources. This would help potential readers better understand our comparison results and improve the consistency with the discussion of the uncertainty in natural sources in our manuscript. More revision is available in the Sect 4.3 in the revised manuscript.

Comment 3: In Line 272, it is mentioned that VOC emissions are optimized through assimilating ground-level O₃ observations. However, several factors need consideration. On one hand, VOC-O₃ interactions involve strong nonlinear chemical reactions, and emission adjustments exhibit bidirectionality (Tang et al., 2016). Despite the convergence of simulations and observations, VOC inversion results may deteriorate due to these complexities. On the other hand, the majority of the national monitoring stations are situated in urban areas, whereas VOC primarily originates from suburban or rural regions. I am skeptical about the feasibility of assimilating O₃ to constrain VOC emissions. As evident from Figure 3, the posterior simulations do not show a significant improvement in O₃. As the authors noted, O₃ cannot effectively constrain precursor NO_x (L278). Therefore, I recommend deleting the VOC emission inversion.

Reply: Thanks for this comment. We agree with the reviewer that the NO_x-VOC-O₃ nonlinear interaction would influence the inversion of NMVOC emission based on the O₃ concentration if were not well addressed. On the one hand, the O₃ concentrations are dependent not only on the NMVOC emissions but also on the NO_x emissions. The errors in the a priori emissions of NO_x would also contribute to the simulation errors of O₃, and deteriorate the inversion of NMVOC. This concern has been considered in our inversion method through two approaches. Firstly, the emissions of NO_x and NMVOC were perturbed independently in our study, thus their contributions to the simulation errors of O₃ concentrations could be isolated through the use of ensemble simulations. Secondly, the use of iteration inversion method can further reduce the influence of the errors in NO_x emissions on the inversion of NMVOC emission, since the errors in NO_x emission would be constrained by its own observations during the iterations as we illustrated in lines 320–324 in the revised manuscript. This is in fact similar to the approach used by Xing et al. (2020) who firstly constrained the NO_x emissions based on observations of NO₂, and then constrained the NMVOC emissions based on O₃ concentrations. Also, in Feng et al. (2024), the NO₂ concentrations were also assimilated to constrain the NO_x emissions to account for the influences of errors in NO_x emissions on the NMVOC emissions. These studies indicates that the iteratively nonlinear joint inversion of NO_x and NMVOCs using multi-species observations adopted in our study is an effective and commonly used way to address the intricate relationship among VOC-NO_x-O₃ (Feng et al., 2024). On the other hand, the emission adjustments exhibit bidirectionality dependent on the VOC-limited or NO_x-limited regimes. According to the Fig 3 in the revised manuscript, the NMVOC emissions were adjusted in alignment with the direction of the O₃ errors, suggesting a VOC-limited regime over urban areas in China, given that the O₃ observation sites are predominantly situated in the urban areas. This agrees with Ren et al. (2022) who diagnosed the NO_x-VOC-O₃ sensitivity based on the satellite retrievals and found that the VOC-limited regimes are mainly located in the urban areas in China. This suggests that the relationship between the O₃ concentrations and VOC emissions could be reasonably reflected by inverse modeling. Moreover, considering there are transport of VOCs from suburban or rural areas (Liu et al., 2022), the O₃

concentrations in the urban areas could also provide information on the NMVOC emission over the suburban or rural areas. Therefore, although the majority of monitoring stations are located in urban areas, the NMVOC emissions over remote regions lacking observations could still be constrained to some extent through the utilization of covariance relationships estimated by the ensemble simulations. However, we agree with the reviewer that the lack of observation sites over the remote areas significantly hinders the fully constrains of the NMVOC emission over there and may lead to larger uncertainty. More observations over the suburban and rural areas are required to better constrain the NMVOC emissions in the future.

To date, we think there remains feasibility in utilizing the O₃ observations to constrain the VOC emissions. Besides our study, the assimilation of surface O₃ observations to constrain the VOC emissions has also been performed in other inversion studies, such as Xing et al. (2020) and Ma et al. (2019). Both of these studies have demonstrated the effectiveness of assimilating surface O₃ concentrations on the inversion of VOC emissions. For example, Ma et al. (2019) found that the assimilation of O₃ concentration could adjust the NMVOC emissions in the direction resembling the bottom-up inventories, and the forecast skill of O₃ concentrations were also improved, indicating that the constrained NMVOC emissions are improved relative to their priori. Our inversion results suggest similar effectiveness of the assimilation of O₃ concentrations on the NMVOC emissions as reflected by the improvement of O₃ simulations (Table 2 in the revised manuscript) and the overall consistency with the bottom-up inventories and top-down emission inventories using the satellite observation data (Souri et al., 2020). Meanwhile, there are limited ways to constrain the NMVOC emissions due to the lack of NMVOC observations. Previous inversion studies are mainly relied on the satellite observations of formaldehyde and glyoxal. However, these inversion studies are also hindered by the NO_x-VOC-O₃ chemistry and the inherent uncertainty in the satellite observations of formaldehyde and glyoxal (Cao et al., 2018; Stavrou et al., 2015), leading to uncertainty in their estimates. Given that, we think it is still worth a try to advance our understanding of the NMVOC emissions in China by assimilating the surface O₃ concentrations. Therefore, we lean towards retaining the inversion results of NMVOCs. This on the one hand could provide the users of interest with some potential valuable information on the NMVOC emissions in China, and on the other hand can serve as a comparable reference for future VOC inversion studies based on other methods or observation data, which could help the development of the inversion method of NMVOC. However, we acknowledge the complexity of the inversion of NMVOC emission due to the nonlinear NO_x-VOC-O₃ interactions and the limited observation sites which were not fully addressed in our study. Therefore, more descriptions on the rationale and uncertainty in the inversion of NMVOC emissions based on O₃ concentrations have been added in the revised manuscript to assist the potential readers in properly utilizing the inversion results of NMVOC. In addition, more robust analysis of the effects of nonlinear NO_x-VOC-O₃ interactions and the number of observation sites should be performed in future to better illustrate the feasibility

of assimilating O₃ to constrain NMVOC emissions. Detailed revisions to the manuscript are available in lines 308–337 and 1002–1011 in the revised manuscripts.

Comment 4: The changes in observation coverage each year can significantly impact emission estimates. If the authors intend to include the years 2013-2014 in this study, they should compare the impact of site differences on emissions for a more robust analysis. If the authors aim to investigate trends, it is advisable to delete emissions in the 2013-2014 period, as this might otherwise potentially mislead readers, given that the changes during this period do not contribute meaningfully to the study's overall trend analysis. There also appears to be some discrepancies in the manuscript where emission changes are often stated as occurring from 2015-2017, while the text descriptions indicate the period as 2013-2017, as seen in lines 452, 485, and 561, among others. Furthermore, it is important to note that the changes in emissions observed from 2015-2017 not necessarily reflect the overall reduction rate of the action plan for the entire period of 2013-2017. Additionally, the data from 2015-2017 alone may not be sufficient to conclude that the emission reduction rate during the 2013-2017 period is lower than that during the 2018-2020 action plan.

Reply: Thanks for this comment. Following the suggestions of the reviewer, we added more analysis on the influences of the site differences on the emission inversions in the revised manuscript. Figure R1 shows the spatial distributions of the observation sites used in inversion during 2013–2015 when the number of observation sites changed rapidly. It can be seen that the observation sites were mainly concentrated in the megacity clusters (e.g., North China Plain, Yangtze River Delta and Pearl River Delta) and the capital cities of each province in 2013. The number of observation sites continued to increase across the China in 2014 and 2015. In particular, many areas that were previously unobserved in 2013 have added monitoring stations, which significantly increased the observation coverage in China especially over the NW, NE, SW and Central regions. Figure R2 shows the calculated emission increments at the observation sites (a posteriori minus a priori) for different species in China from 2013 to 2015 under the scenario of fixed observation sites (blue lines) and varying observation sites (orange). In the fixed-site scenario, it is assumed that the number of observation sites remains constant at the 2013 level while in the varying-site scenario, the number of observation sites increases over time. The differences in emission increments between these two scenarios are used to analyze the impact of changes in the observation coverage on the emission inversions. Please note that, to simplify calculations, we only computed the emission increments at the locations of the observation sites. Therefore, they may not be equal to the emission increments calculated for the entire grid as reported in the paper. However, they are still useful indicators for the effects of emission inversion. In addition, since we did not consider the temporal variation in the a priori emissions, the changes of emission increments at the observation sites can be used to approximate the temporal variations of the a posterior emissions. It can be

clearly seen that there are obvious differences in the emission increments between the two scenarios. The emission increment is larger in the varying-site scenario than that in the fixed-site scenario for all species due to the increases of observation sites. Moreover, as indicated in Fig. R2, the changes of observation sites were shown to significantly affect the estimation of the emission trend in 2013 and 2014. Most of species showed decreasing trends in their inversed emission under the fixed-site scenario. However, under the varying-site scenario, the decreasing trends were smaller for PM_{2.5}, NO_x and NMVOC, and the emissions of PM₁₀ and CO even showed increasing trends. This is due to that the emission increments were positive over most of observation sites for these species as demonstrated in Fig.3 in the revised manuscript. Thus, the increases of observation site would lead to increases of positive emission increments and higher a posteriori emissions, which may counteract the decreasing trends or even lead to an opposite trend. These results provide the evidences that the increasing trends in the total emissions of PM₁₀ and CO from 2013 to 2015 seen in Fig. 6 and Fig. 7 are highly likely to be a spurious trend caused by the changes of observation coverage. The weak emission changes in PM_{2.5} and NO_x (Fig. 6 and Fig. 7) may also be related to the changes in the number of observation sites. The SO₂ emission is an except that its calculated trend is larger under the varying-site scenario than that under the fixed-site scenario. This is because that the emission increment for the SO₂ is generally negative over the most of sites, thus the increased observation sites would lead to larger decreasing trend in the inversed emissions of SO₂. These results highlighted the significant influences of the site differences on the estimated emissions and their trends. Therefore, as the reviewer suggested, we recommend not to use the emission in 2013 and 2014 when analyze the trends of the emissions, which has been written in the user notes of our data products. we also only investigated the emission changes from 2015 to 2020 in our manuscript to avoid misleading the potential users. Following the suggestions, the analysis on the influences of the site differences on the emission inversion has been added in the revised manuscript to remind potential users to be aware of this issue. Please see lines 199–207 in the revised manuscript, lines 3–28 and Fig. S1–S2 in the revised supplement.

We feel sorry for the discrepancies in the manuscript. The 2013-2017 is merely used as the names for the clean air action plans during 2013–2017, rather than referring to the years calculating emission changes. This confusion has been revised by using more accurate expression. For example, the lines 36 – 37 in the original manuscript have been revised to “It is also estimated that the emission reductions were larger during 2018–2020 (from -26.6% to -4.5%) than during 2015–2017 (from -23.8% to 27.6%) for most species.” in the revised manuscript (lines 36–37). Also, we agree with the reviewer that the changes in emissions observed from 2015-2017 not necessarily reflect the overall reduction rate of the action plan for the entire period of 2013-2017, and that they were not sufficient to conclude that the emission reduction rate during the 2013-2017 period is lower than that during the 2018-2020 action plan. Thanks for the reviewer’s reminder. We have softened the

statement of this conclusion and added relevant discussions in the revised manuscript to enhance the rigor of our paper (please see lines 423–427 and 984–987 in the revised manuscript).

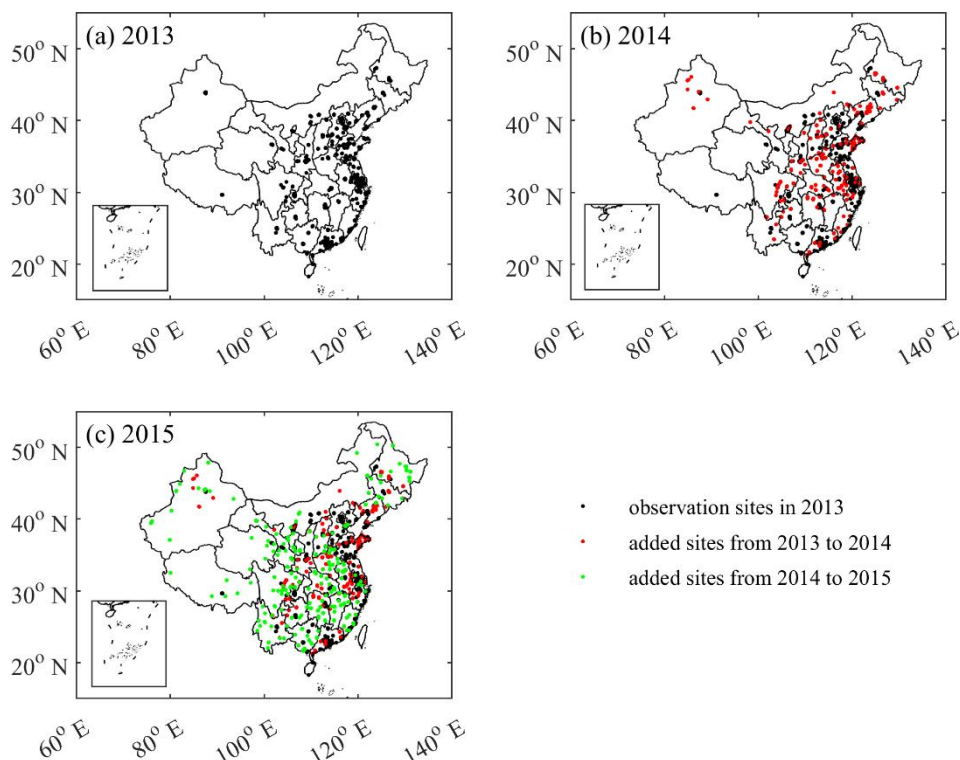


Figure R1 Spatial distributions of observation sites in (a) 2013, (b) 2014 and (c) 2015. The observation sites in 2013 were marked as black dots, while the added observation sites from 2013 to 2014 and those from 2014 to 2015 were marked as red and green dots respectively.

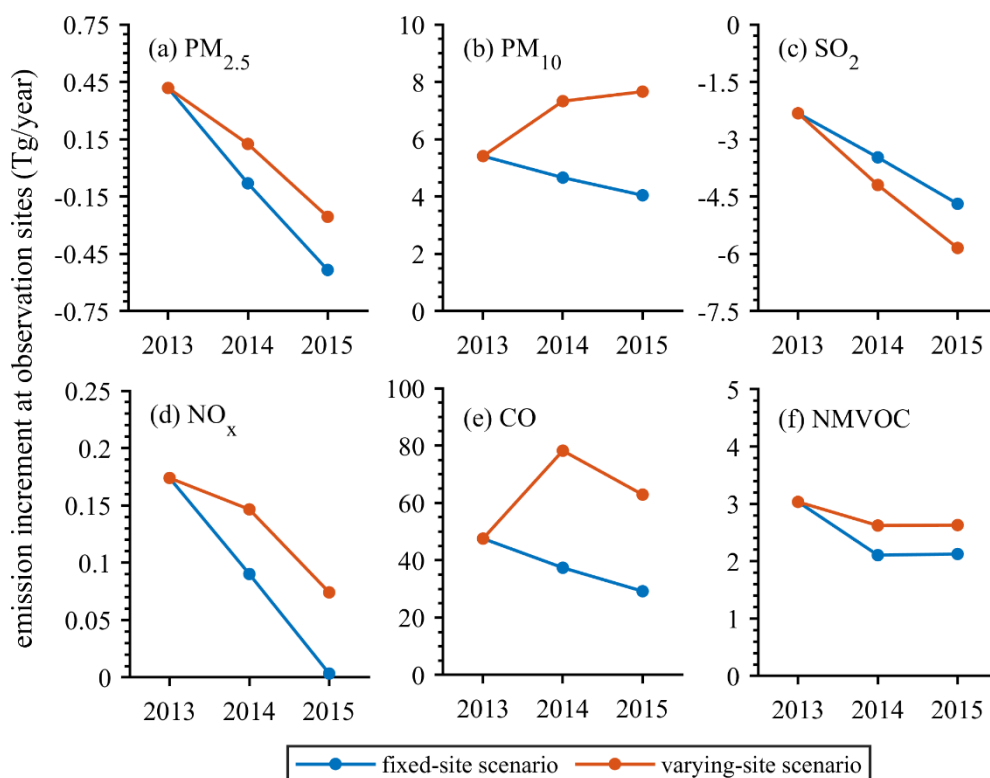


Figure R2: the calculated total emission increments at the observation sites for different species under the fixed-site scenario and varying-site scenario.

Comment 5: The authors use PM_{2.5} observations to simultaneously constrain BC, OC, and primary PM_{2.5}. If they do not consider inter-species correlations or use random perturbations, and, for instance, if BC and OC increase while PM_{2.5} decreases in one ensemble member. How do they constrain emissions when the simulated PM_{2.5} and observations are the same.

Reply: Thanks for this comment. Since we aim to estimate the emissions separately for BC, OC and primary PM_{2.5}, it is necessary to perturb the a priori emissions of BC, OC and primary PM_{2.5} randomly during the inversion to avoid the spurious correlations between the non- or weakly related variables. This enables us to statistically differentiate the contributions of their emission errors to the simulation errors of PM_{2.5} concentration through the use of ensemble simulation, making the emissions of BC, OC, primary adjusted by different scaling factors (i.e., β_{BC} , β_{OC} and β_{PMF}). Also, it is feasible that using same perturbation coefficient to perturb their emissions. As I understand it, this treatment is closer to what the reviewer mentioned regarding considering the inter-species correlations. This is equivalent to perturbing only the total PM_{2.5} emissions and allows the estimations of total PM_{2.5} emissions by using a same scaling factor. Therefore, applying independent perturbations or using the same perturbation coefficient are both commonly employed methods in the inversion studies. We agree with the reviewer that it is possible for BC and OC to increase while PM_{2.5} decreases in one ensemble member under the conditions of random perturbation. However, as we used the deterministic form of EnKF (DEnKF), the ensemble member is only used to calculate the background perturbation \mathbf{X}_i^b and the subsequent background covariance matrix \mathbf{B}_e^b . The behavior of single ensemble would not significantly influence of the statistical properties of the ensemble, unless there is spurious correlation among the emissions of different PM_{2.5} components. For example, if there is a spurious negative correlation between the perturbed emissions of BC and PMF, there would lead to a false negative correlation between the PM_{2.5} concentrations and the emissions of BC. That's why the emissions of different PM_{2.5} component should be perturbed randomly during the assimilation. Also, in the DEnKF, the observation innovation (observation minus simulation) is only determined by the observation and ensemble mean of the simulated PM_{2.5}. Therefore, whether to adjust the a priori emission is only determined by the deviations between the ensemble mean and observations, rather than the simulation results in one ensemble member. Meanwhile, since the emissions were perturbed unbiasedly in our study, the ensemble mean of perturbed emissions is equal to the a priori emission. Thus, the ensemble mean of the model simulation is mainly determined by the a priori emission. If the ensemble mean of PM_{2.5} simulations equals the observed values, it suggests that the a prior emission may have no error, and thus, we won't make adjustments to the prior emission. However, we acknowledge that in such cases, there may still be errors in the emissions of BC, OC, and primary PM_{2.5}, such as the underestimation of BC and OC while the overestimation of primary PM_{2.5}. This is primarily due to that we only assimilate the observations of total PM_{2.5} mass without the assimilation of speciated PM_{2.5}

observations. In the absence of detailed speciated PM_{2.5} observations, assimilating only total PM_{2.5} concentration observations cannot adjust the proportions of emissions for different PM_{2.5} components when the observations and simulations are equal, which is a specific manifestation of the uncertainty resulting from adjusting PM_{2.5} emissions solely based on total PM_{2.5} concentration. This limitation has been explicitly pointed out in our manuscript and thus only the total PM_{2.5} emissions were provided to prevent the potential misuse of PM_{2.5} component emissions without sufficient validation. Following the suggestions of reviewer, we give more discussions about the limitations of only assimilation total PM_{2.5} mass in the revised manuscripts (please see lines 299–303 in the manuscript).

Comment 6: The authors simultaneously constrain concentrations and emissions, emphasizing that concentration errors arise from emission uncertainties, implying a shared source of uncertainty (L222). In this context, the question arises whether optimizing concentrations would diminish emission uncertainties, thereby affecting emission estimates.

Reply: Thanks for this comment. We feel sorry for this confusion. Since we used the modified EnKF method to constrain the emissions (Wu et al., 2020), the concentrations were not optimized simultaneously with the emissions. As we written in the manuscript, the modified EnKF is an offline application of the EnKF method that decouples the analysis step from the ensemble simulation. In this method, the ensemble simulation was performed firstly with the perturbed emissions, thus the concentration errors estimated by the ensemble simulation mainly stem from the emission uncertainty as we written in line 240–241 in the revised manuscript. After that, the observations were assimilated to constrain the emissions. During this step, the concentration was not required to be optimized but was used to estimate the covariance between the emission and concentration. Therefore, although the concentration was included in the state variable as illustrated in Eq. (1), it was not optimized during the inversion step and thus would not diminish emission uncertainties. The feasibility of this method in the emission inversion has been discussed and tested in Wu et al. (2020) through the observation system simulation experiments, which shows good performances of this method in reducing the errors in the a priori emission inventory. To avoid this confusion, we have added relevant explanations regarding to the optimization of the state of concentrations in the revised manuscript. Please see lines 215–219 and lines 231–233 in the revised manuscript.

Comment 7: The NO_x emission changes optimized by the authors appear to contradict existing research findings and are inconsistent with recent emission reduction policies. Despite citing the study by Zheng et al., (2018), the actual NO_x emissions reported by Zheng show a significant decrease. Could this discrepancy be attributed to the bottom-up inventory lacking sufficient statistics on mobile vehicle emissions? Moreover,

according to Zheng's study, industrial and power plant emissions collectively contribute to over 50% of total emissions. Hence, the second reason provided by the authors may not be suitable if the industrial and power plant emissions are substantial contributors.

Reply: Thanks for this comment. The NO_x emission changes are determined by the combined effects of pollution control and growth of activity. If the effects of air pollution control exceed the additional emissions caused by the growth of activity, the NO_x emission would decrease and vice versa. According to Zheng et al. (2021), the increases of activity levels has offset the mitigation effects of the emission controls for the traffic and industrial sectors. For example, the vehicle growth yielded increases of 1.4 Tg NO_x emission compared with its 2010 level, which exceeded the emission reductions of NO_x (1.3 Tg) achieved by the pollution control on the traffic section (Zheng et al., 2018). This indicates that the increase in the activity levels and the insufficient effectiveness of emission control in industrial and traffic sectors do exist, and has been considered in some bottom-up emission inventories, such as MEIC. The discrepancy in the estimated NO_x emission changes between inversion results and other emission inventories thus reflect the uncertainty in the quantification of the combined effects of NO_x emission control and activity growth. Our inversion results suggest that the offset effects of activity growth may be larger than the mitigation effects of the pollution control during 2015–2017, while previous emission inventories suggest larger mitigation effects of air pollution control than the offset effects of activity growth. As the reviewer mentioned, this discrepancy could be attributed to the bottom-up inventory lacking sufficient statistics on the mobile vehicle or other sectors. For example, previous inversion study by Kong et al. (2022) found there are numerous small-to-medium local sources of NO_x emission related to the minor roads or small human settlements in China that are unclear or missing in the MEIC, EDGAR and CEDS emission inventory. The emission trends of these unaccounted local sources are thus not able to be considered by these emission inventory, which could be an important factor for the differences between our inversion results and previous inventories. Following the suggestions of the reviewer, more discussions have been added in the revised manuscript to better explain the discrepancy between our inversion results and previous inventories (please see lines 826–831 in the revised manuscript). We agree with the reviewer that industrial and power plant emissions are substantial contributors to the NO_x emission. Our second reason is mainly related to the control of traffic sector. Following the suggestions of the reviewer, we have deleted this reason in the revised manuscript. Please see lines 566 – 568 in the revised manuscript.

Comment 8: "Figure 12 shows significant discrepancies between the results of the author's inversion and other bottom-up inventories. According to other literature, it is known that China experienced two peaks in VOC emissions in May and July 2015. The variation in VOC emissions closely follows the changes in O₃

levels, suggesting a strong dependence of VOC on O₃ variations. This raises the question of whether non-linear changes are being overlooked.

Reply: Thanks for this comment. Firstly, we feel sorry that there is an error in the description of Fig. 12. It actually presents the monthly profiles of the averaged air pollutant emissions from 2015 to 2018 rather than just for 2015. Figure R3 shows the comparisons of the standardized monthly profile of the averaged a posteriori NMVOC emission and MDA8h O₃ concentrations in China from 2015 to 2018. The standardized monthly profiles were calculated by dividing them by their mean values. It shows that the monthly variation of the a posteriori NMVOC emissions have significant similarity to the monthly variation of the observed MDA8h O₃ concentrations, as the reviewer mentioned. However, there are still obvious differences in their month variations. For example, the peak values of the observed MDA8h O₃ concentrations occur in May, while the peak values of the a posteriori NMVOC emissions occur in July. This suggest that the monthly profile of a posteriori NMVOC emissions is not solely dependent on the variations of the MDA8h O₃ concentrations, thus some non-linear changes, such as unfavorable meteorological conditions that lead to high O₃ concentrations even with relatively low NMVOC emissions, could be represented in our method to some extent. This is also the advantage of the EnKF method which provide an effective way to consider the flow dependent non-linear relationships between the concentrations and emissions. For example, the sensitivity of O₃ concentrations to the NMVOC emissions under different meteorological conditions could be represented in the EnKF through the use of ensemble simulation. Nevertheless, we acknowledge that there could still be some unknown nonlinear-changes in the model or the EnKF method that were not well considered during inversion, which leads to uncertainty in the a posteriori NMVOC emissions.

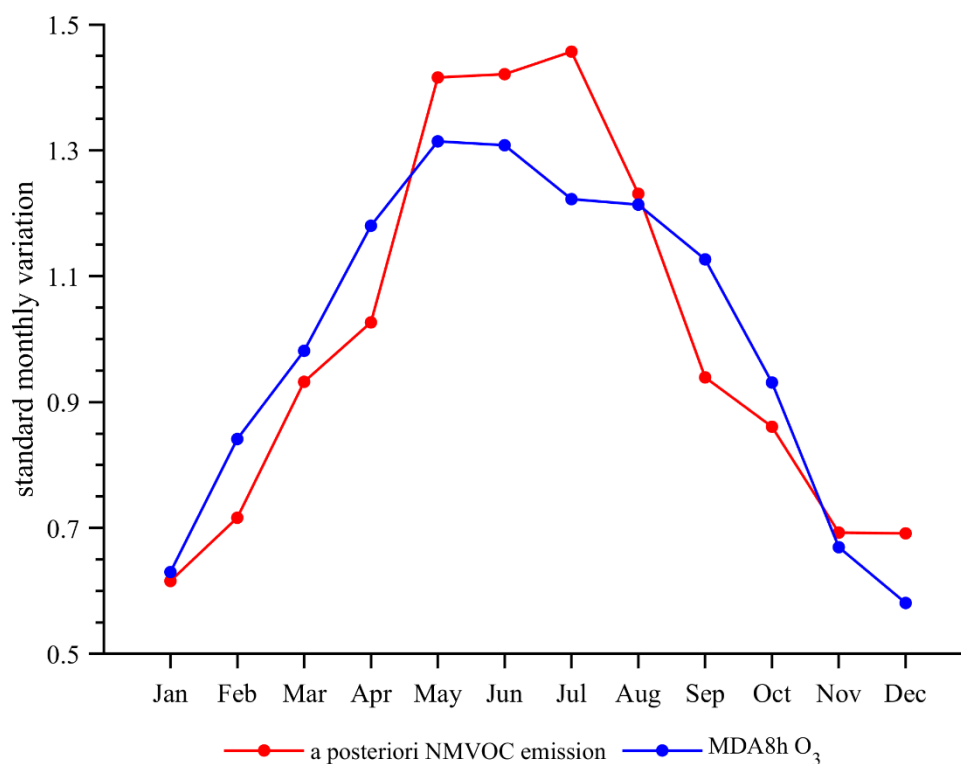


Figure R3: the standard monthly variation of the averaged a posteriori NMVOC emission and MDA8h O₃ in China during 2015–2018.

Comment 9: The author has provided a high-resolution, multispecies emission inventory. To facilitate users' understanding of the data's accuracy, could you please provide information on the uncertainties associated with different species, allowing users to assess the error range in the data?

Reply: Thanks for this suggestion. Within the framework of the EnKF assimilation, the information on the uncertainty of the a posteriori emission for different species could be provided by the analysis ensemble spread estimated from the standard deviation across the analysis ensemble (Miyazaki et al., 2020). According to Sakov and Oke (2008), the analysis ensemble can be calculated as follows:

$$\mathbf{X}^a = \mathbf{X}^b - \frac{1}{2} \mathbf{K} \mathbf{H} \mathbf{X}^b \quad (\text{R1})$$

Based on the analysis ensemble, the uncertainty of the a posteriori emission was estimated as follows: 101.4% (PM_{2.5}), 102.5% (PM₁₀), 26.7% (SO₂), 46.8% (CO), 31.8% (NO_x) and 65.5% (NMVOC). However, it should be noted that such uncertainty was only calculated under the framework of the EnKF constructed in this study, which is dependent on the assigned value of the a priori emission uncertainty, observation errors and the number of assimilated observations. In addition, we only considered the a priori emission uncertainty and the observation errors during the inversion. The influences of the other error sources, such as uncertainty in the chemistry transport model, meteorology simulations and the inversion method were not considered. Therefore, the current estimated uncertainty should be considered as a lower bound for the real uncertainty. More systematic analysis that thoroughly consider the uncertainty sources regarding the emission inversion should be conducted in future to give a more accurate estimation of the uncertainty in our products. Following the suggestions, we have added the descriptions on the uncertainty on the a posteriori emission in the revised manuscript (please see lines 885–899 in the revised manuscript).

Specific comments:

Comment 1: Change "Fengwei Plain" to "Fenwei Plain".

Reply: Done.

Comment 2: In L212, the VOC adjustment factor was omitted.

Reply: Done.

Comment 3: Since MOZART data products are no longer updated, are the boundary conditions in this study

based on simulations conducted by the author's team? Additionally, maritime shipping emissions have a significant impact on the generation of NO₂ and O₃ in coastal provinces. Has the model taken into account inputs from maritime emissions? Why was a localized scale of 180 km chosen?

Reply: We feel sorry for the lack of clear explanation regarding the use of MOZART data products. As mentioned by the reviewer, the MOZART data products have not been updated since 2018. Therefore, the multi-year average results from the MOZART were used for the simulations after 2018. Because most of the model boundaries were set in the clean areas and are located at distance from China, we assumed that the differences in boundary conditions would not significantly affect the modeling results in China. following the suggestions of reviewer, we have clarified the use of MOZART data in the revised manuscript (please see lines in 160–165). The ship emissions have been considered in our inversion study as a part of the HTAP emission inventory as we illustrated in lines 145 – 147 in the revised manuscript. The localized scale of 180km was chosen according our previous inversion study (Kong et al., 2023), and is similar to the localization scales used in Feng et al. (2020) and Ma et al. (2019) which were determined based on the wind speed and the lifespan of the species (please see lines 282 – 284 in the revised manuscript).

Comment 4: In L291, is it necessary to reassemble simulations for each iteration, or is it multiple inversions on the original ensemble? If multiple iterations are performed, does it imply that the posterior approaches the observations more closely with each iteration? Why was the choice made to iterate twice?

Reply: Thanks for raising this important issue. In this method, we conduct a new simulation by using the a posteriori emission from the previous iteration to update the ensemble mean of the original ensemble. This enables the observational information and the adjusted emissions to be promptly incorporated into the model, thereby providing feedback for the adjustments of emission in the next iteration. However, we did not reassemble the ensemble simulation for each iteration due to the expensive computational cost of the ensemble simulation. Therefore, in each iteration calculation, the ensemble perturbation that were used to calculate the background error covariance matrix remains the same with only the ensemble mean being updated based on the inversion results of the previous iteration.

As mentioned by the reviewer, it is implied that the posteriori should approach the observations more closely with each iteration, which has been demonstrated in our previous inversion studies (Kong et al., 2023). As seen in the Fig.3 of Kong et al. (2023) (fig. R4), four times of iterations were conducted to adjust the SO₂ emissions. We can clearly see that due to the large positive biases in the a priori SO₂ emissions, the model still has large positive biases in simulated SO₂ concentration over all regions of China even after assimilation (first iteration). With the increases in the iteration times, the biases and errors continued to decrease which is consistent with the implication in the iteration inversion. However, the degree of improvement will gradually

diminish with an increasing number of iterations until convergence is achieved. Our previous study shows that the improvement become no longer significant after two iterations. Thus, we choice the two times of the iteration in this study maintain a balance between the filter performance and the computational cost. Following the suggestions of reviewer, we have added more description on the implementation of iteration inversion scheme and the determination of the times of iteration in the revised manuscript. Please see lines 345–354 and 358–360.

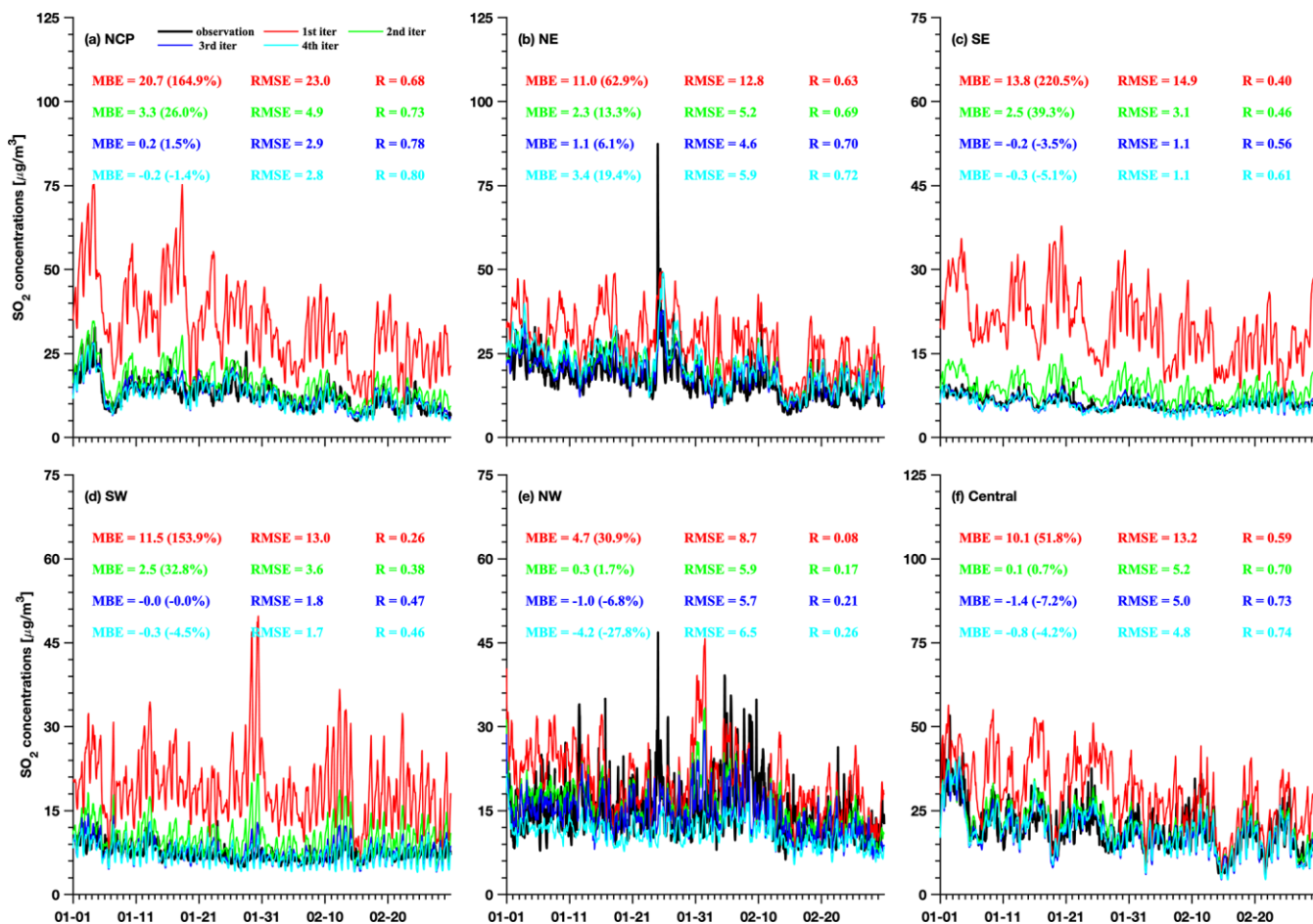


Figure R4: Comparisons of the observed and simulated mean SO₂ concentrations using emissions of different iteration time over (a) the NCP region, (b) NE region, (c) SE region, (d) SW region, (e) NW region and (f) central region (taken from Fig 3 in Kong et al. (2023)).

Comment 5: With such a high grid resolution of 15 km, how does the computational cost for the inversion of multiple years in the ensemble calculations? Additionally, what is the size of the assimilation window?

Reply: Thanks for this comment. We have added more details related to this issue in the revised manuscript. The computational cost is still expensive for the inversion of multiple years in the ensemble calculations with a high grid resolution of 15km. According to our estimation, we used about 12000 CPUs in the ensemble simulation, and the computational time for one-year ensemble simulation reaches approximately 2 million core-hours. Thanks to the “Earth System Science Numerical Simulator Facility” (EarthLab) which provide us

with ample computational resources to complete this research. Since we constrained the daily emissions, the size of the assimilation window is 24h in our study. Please see lines 1255–1258 in the revised manuscript.

Comment 6: The inflation factor 'r' varies for each window. Is 'r' a matrix or a scalar? If it is a scalar, could the author provide the specific range of 'r'?

Reply: The inflation factor is a scalar but its value varies in the space and time, which is calculated by using the method of Wang and Bishop (2003):

$$\lambda = \frac{(\mathbf{R}^{-1/2}\mathbf{d})^T \mathbf{R}^{-1/2}\mathbf{d} - \mathbf{p}}{\text{trace}\{\mathbf{R}^{-1/2}\mathbf{H}\mathbf{B}_e^b(\mathbf{R}^{-1/2}\mathbf{H})^T\}} \quad (\text{R2})$$

$$\mathbf{d} = \mathbf{y}^o - \mathbf{H}\overline{\mathbf{x}^b} \quad (\text{R3})$$

where λ is the inflation factor, \mathbf{d} is the observation innovation, \mathbf{R} is the observation error covariance matrix, and \mathbf{p} is the number of observations. following the suggestions of the reviewer, we analyze the calculated value of inflation factor in the revised manuscript. Table R1 shows the calculated average value (standard deviation) of the used inflation factor for the different species over different regions of China. It shows that the inflation factor over the east China (including NCP and SE region) was generally round 1.0, suggesting that the original ensemble can well represent the simulation errors of the different air pollutants over these regions. The inflation factor is larger over the western China (including SW, NW and Central regions), especially for PM₁₀ and SO₂, suggesting that the original ensemble may underestimate the simulation errors of the air pollutants. This is associated with the large biases in the simulated air pollutant concentrations over there and reflect that the emission uncertainties assumed in our studies may be underestimated over these regions. it also highlighted the importance of the use of inflation method during the inversion, otherwise it would lead to filter divergency caused by the underestimations of the background error covariance. Following the suggestions of the reviewer, we have added the discussions of the inflation factors in the revised manuscript. Please see lines 269 – 277 in the revised manuscript and Table S1 in the revised supplement.

Table R1 The average mean (standard deviation) of the calculated factor for the inflation of the ensemble member over different regions of China for different species

	NCP	NE	SE	SW	NW	Central
PM _{2.5}	1.0 (0.2)	1.7 (1.6)	1.0 (0.0)	6.8 (8.5)	3.1 (3.8)	3.9 (3.9)
PM ₁₀	1.4 (0.7)	7.2 (8.0)	2.4 (0.8)	78.1 (108.2)	26.3 (36.5)	36.0 (49.0)
SO ₂	1.4 (0.7)	4.1 (3.2)	2.3 (0.8)	176.1 (254.6)	7.8 (6.5)	58.6 (72.5)
NO _x	1.0 (0.1)	1.7 (0.7)	1.2 (0.3)	8.1 (5.3)	2.8 (1.3)	5.4 (4.1)

CO	1.0 (0.1)	2.8 (2.3)	1.4 (0.4)	18.8 (16.8)	6.8 (6.9)	8.6 (10.0)
NMVOOC	1.4 (0.6)	4.5 (4.4)	1.6 (0.5)	8.1 (8.6)	6.5 (5.8)	8.1 (10.1)

Comment 7: Table 3 lacks information regarding the year.

Reply: Thanks for this comment. we have added the year information in the revised Table 3.

Comment 8: How were diurnal variations of the emissions specified?

Reply: Since the a priori emission inventory did not provide the information on the diurnal variations of the emissions, and it is difficult to estimate the diurnal variations of the emissions for different sectors over the whole China, we used the constant diurnal variation during the assimilation. We acknowledge that the uncertainty in the diurnal variations of the emission would lead to uncertainty in our inversion results. However, the diurnal variations of the emission may not significantly influence the simulation results of the daily mean concentrations of air pollutants according to the sensitivity experiments conducted by Wang et al. (2010) in China and Mues et al. (2014) in Europe. As shown in Wang et al. (2010), the differences in the simulated concentrations of SO₂, NO₂ and O₃ with or without considerations of diurnal variation were estimated to be within 1 ppbv in China. Therefore, the diurnal variation may not significantly influence our inversion results. Following the suggestion of reviewer, we have added the description of the settings of the diurnal variation of the emissions in the revised manuscript. Please see lines 155–159.

Comment 9: How is the optimization of VOC components conducted when VOC consists of multiple components?

Reply: Since we did not have the observations of the VOC components, we only optimize the gross emissions of the VOC during our assimilation which has been pointed out in the revised manuscript (lines 336–337).

Comment 10: Region name in Figure 1 refers to specific areas. Consider a different expression to avoid potential ambiguity.

Reply: Thanks for this suggestion. Since the region names in Fig. 1 were also used in our other papers, we are intended to keep their name to guarantee the consistency among our works.

Comment 11: Please consider adopting a clearer representation for Figure 11.

Reply: Thanks for this suggestion. We have redrawn the Figure 11 in revised manuscript for a clearer representation as shown in Fig. R5:

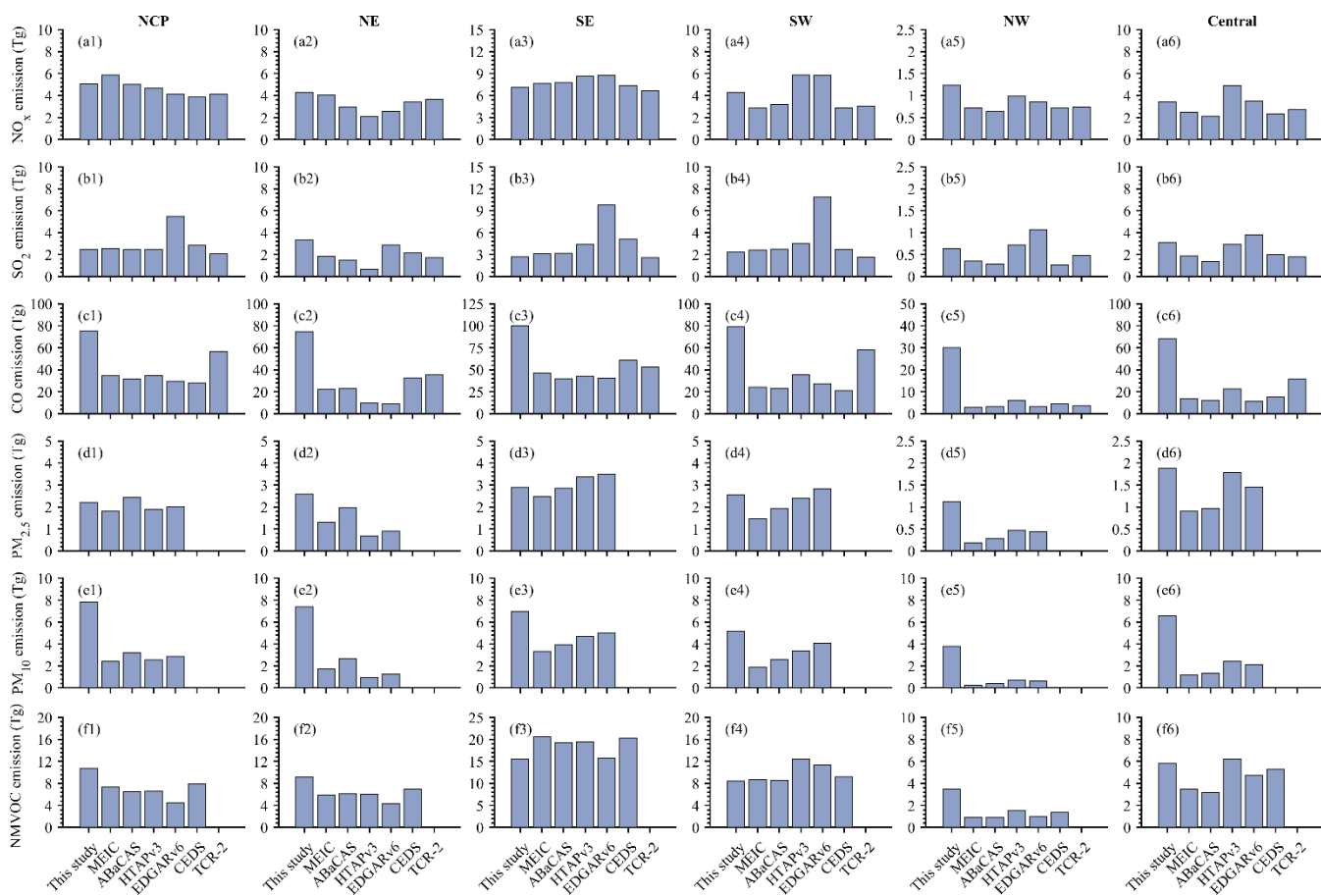


Figure R5: Comparisons of the averaged emissions of (a) NO_x , (b) SO_2 , (c) CO , (d) $\text{PM}_{2.5}$, (e) PM_{10} , and (f) NMVOCs over different regions in China from 2015 to 2018 between CAQIEI and previous inventories added with natural sources.

Comment 12: L483 Change "Fig.3" to "Fig. 4"

Reply: Done

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