Response to Reviewer #1 (ESSD-2023-477)

We Thank Reviewer for his/her constructive comments

Responses to the Specific comments

General comments: This is a valuable high-resolution emission dataset for China, and the paper is well-structured. However, I have some questions and suggestions:

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

Comment 1: In Figure 4, compared to other pollutants, NOx and VOC emissions still show distributions in the Tibet region of China. Could you provide more information about the sources of NOx and VOC in this region?

Reply: Thanks for this suggestion. Besides the anthropogenic emissions of NO_x and VOC which are mainly located over the urban areas of the Tibet region of China like the other pollutants, there are also distributions of natural sources distributed over there, for example, the soil NO_x emissions and the biogenic NMVOC emissions. Thus, the differences in the distributions of the emissions of NO_x, and VOC compared to other pollutants over the Tibet region of China could be mainly attributed to the natural sources of these two species, considering that the contributions of natural sources to the other pollutants are much smaller. Following the suggestions of the reviewer, we have added more information about the sources of NO_x and VOC in this region in the revised manuscript (please see lines 438–440)

Changes in the manuscript: lines 438–440.

Comment 2: Could you display the temporal trends in emissions from 2013 to 2020 for China and its sub-regions, comparing different inventories (multiple lines) to better illustrate the changes in emission patterns over time?

Reply: Thanks for this suggestion. we have added the temporal trends in the emissions of different air pollutants in China (Fig. R1) and its sub-regions (Fig. R2–R7) obtained from our inversion results and other emission inventories in the revised manuscript to better illustrate the changes in emission patterns over time. Please see Fig. 13 in the revised manuscript and Fig. S10–S15 in the revised supplement.

Changes in the manuscript: Fig.13

Changes in the supplement: Fig. S10–S15

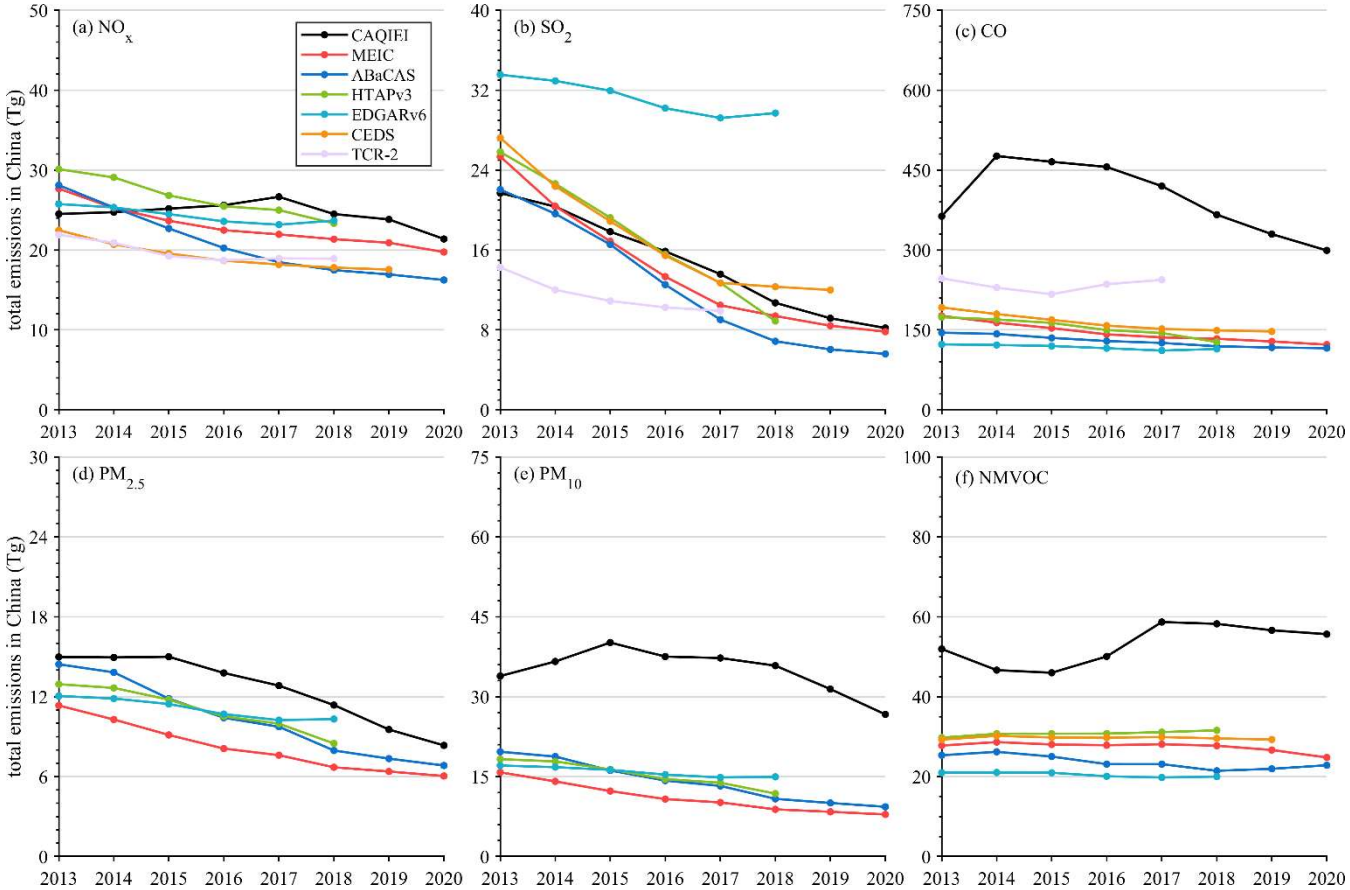

Figure R1: Time series of annual emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10} and (f) NMVOC over China from 2013 to 2020 obtained from CAQIEI and previous inventories. Note that the natural sources were not included in the previous inventories in this figure.

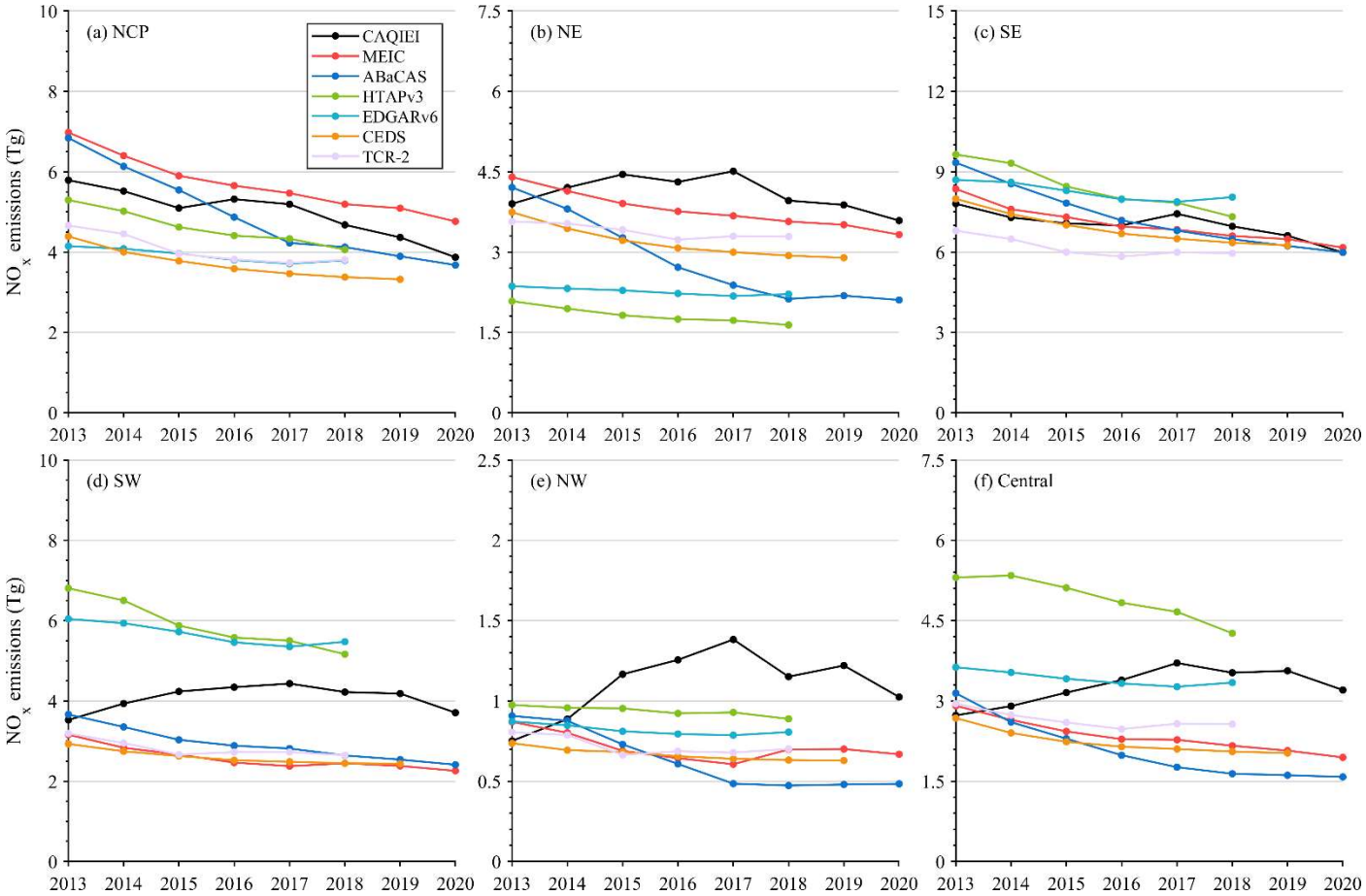

Figure R2: Time series of annual NOx emissions over of different regions of China: (a) NCP, (b) NE, (c) SE, (d) SW, (e) NW and (f) Central from 2013 to 2020 obtained from CAQIEI and previous inventories. Note that the natural sources were not included in the previous inventories in this figure.

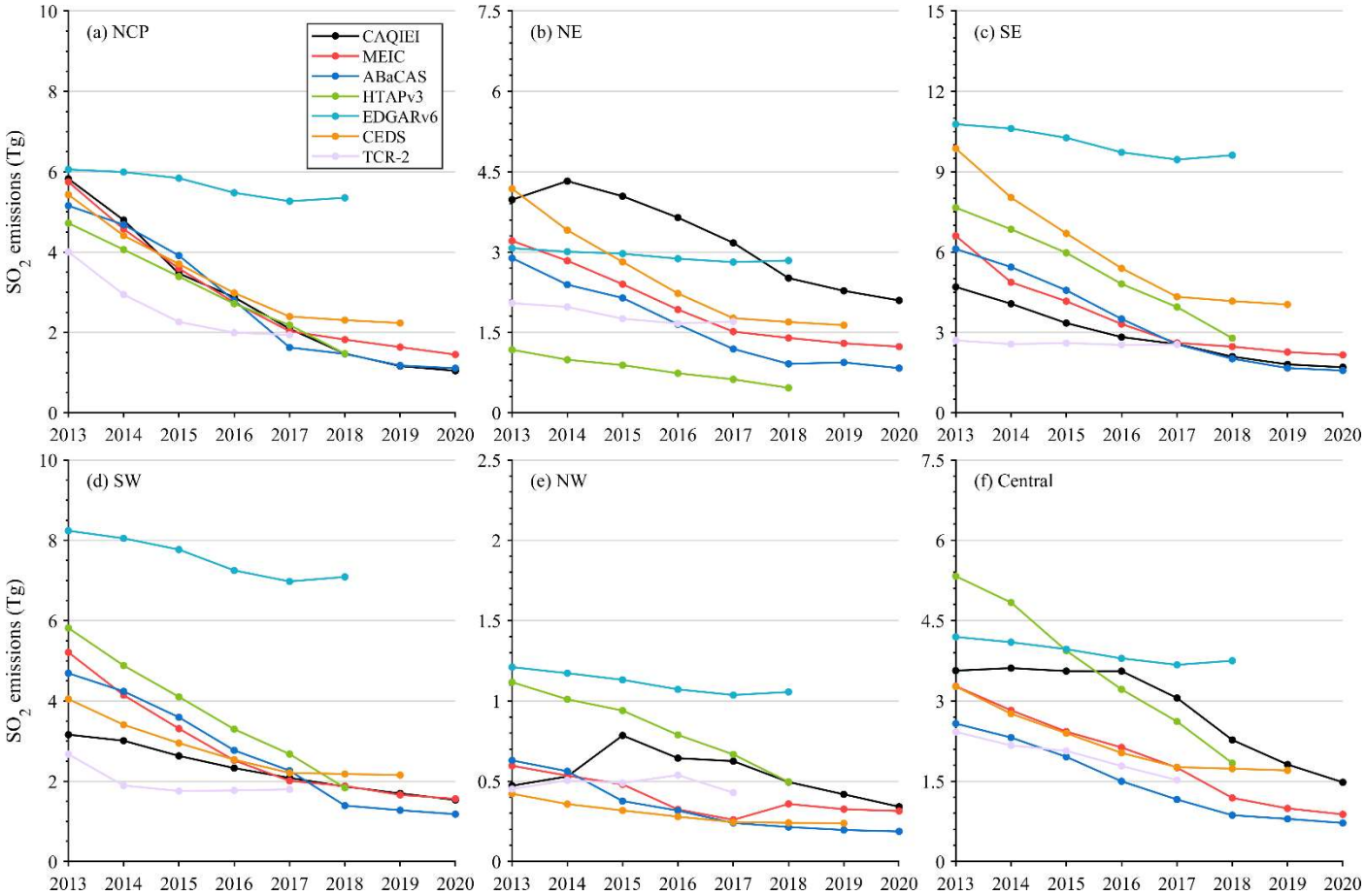

Figure R3: Same as Fig. R2 but for SO_2.

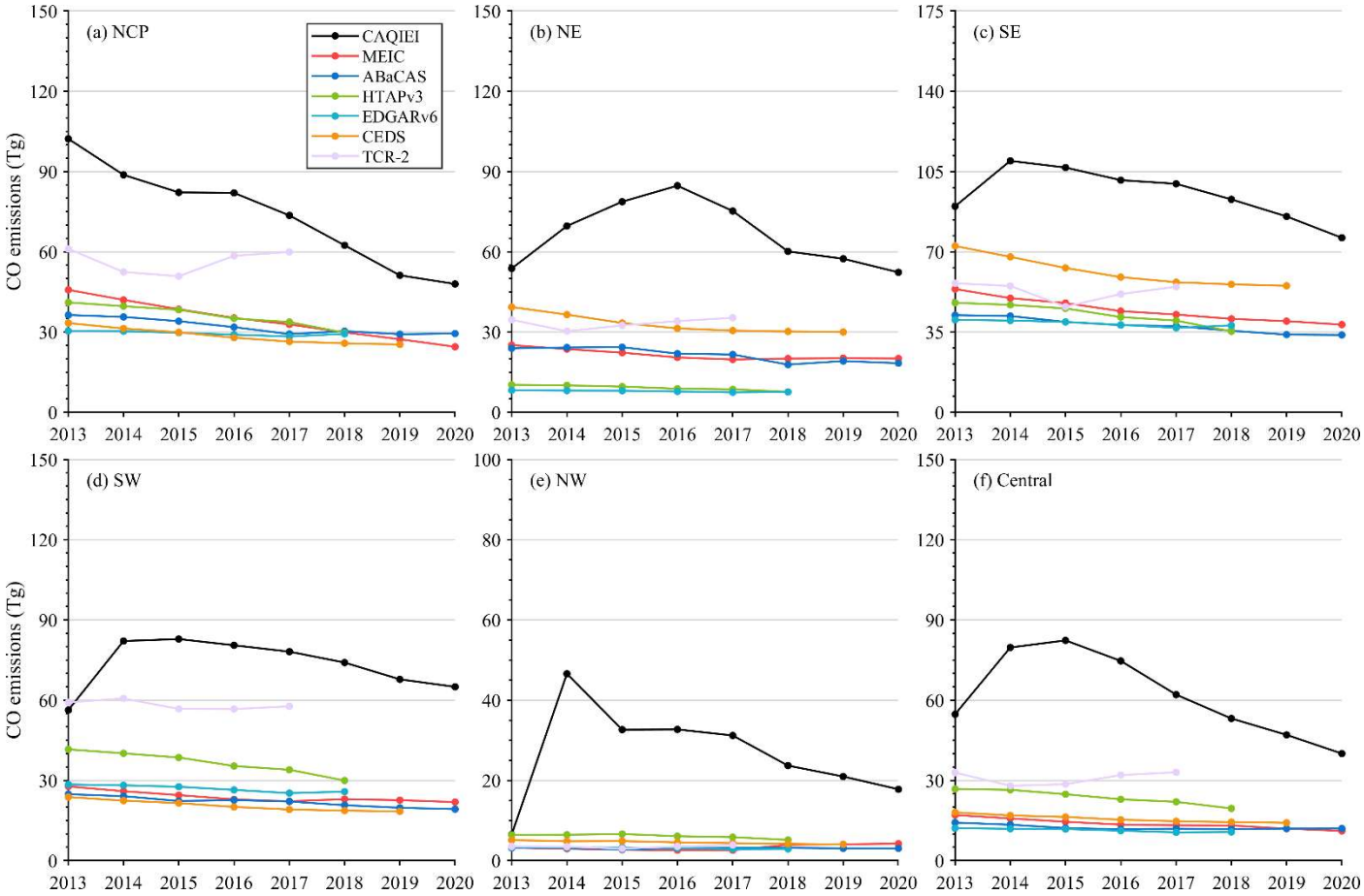

Figure R4: Same as Fig. R2 but for CO.

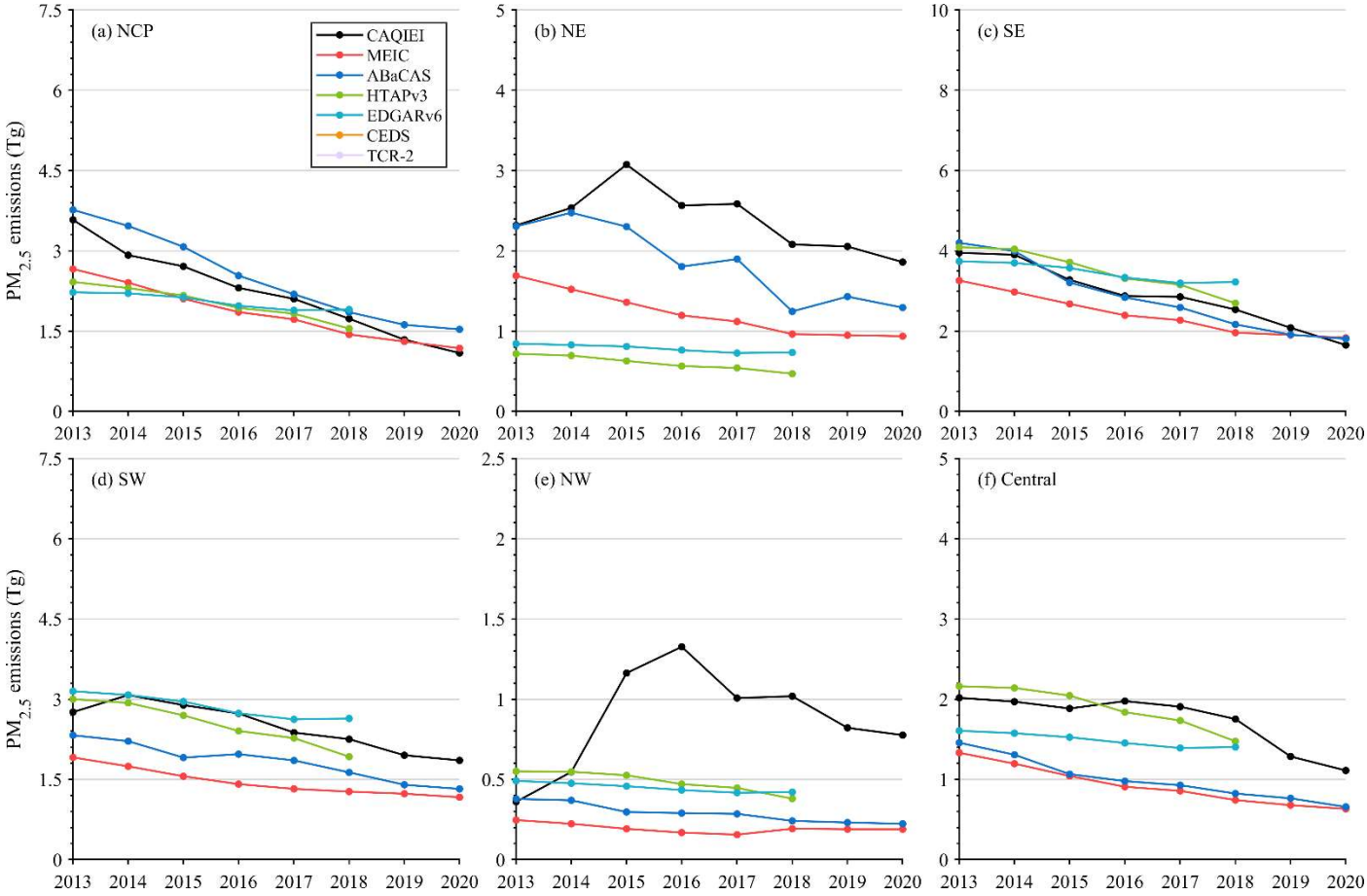

Figure R5: Same as Fig. R4 but for $PM_{2.5}$.

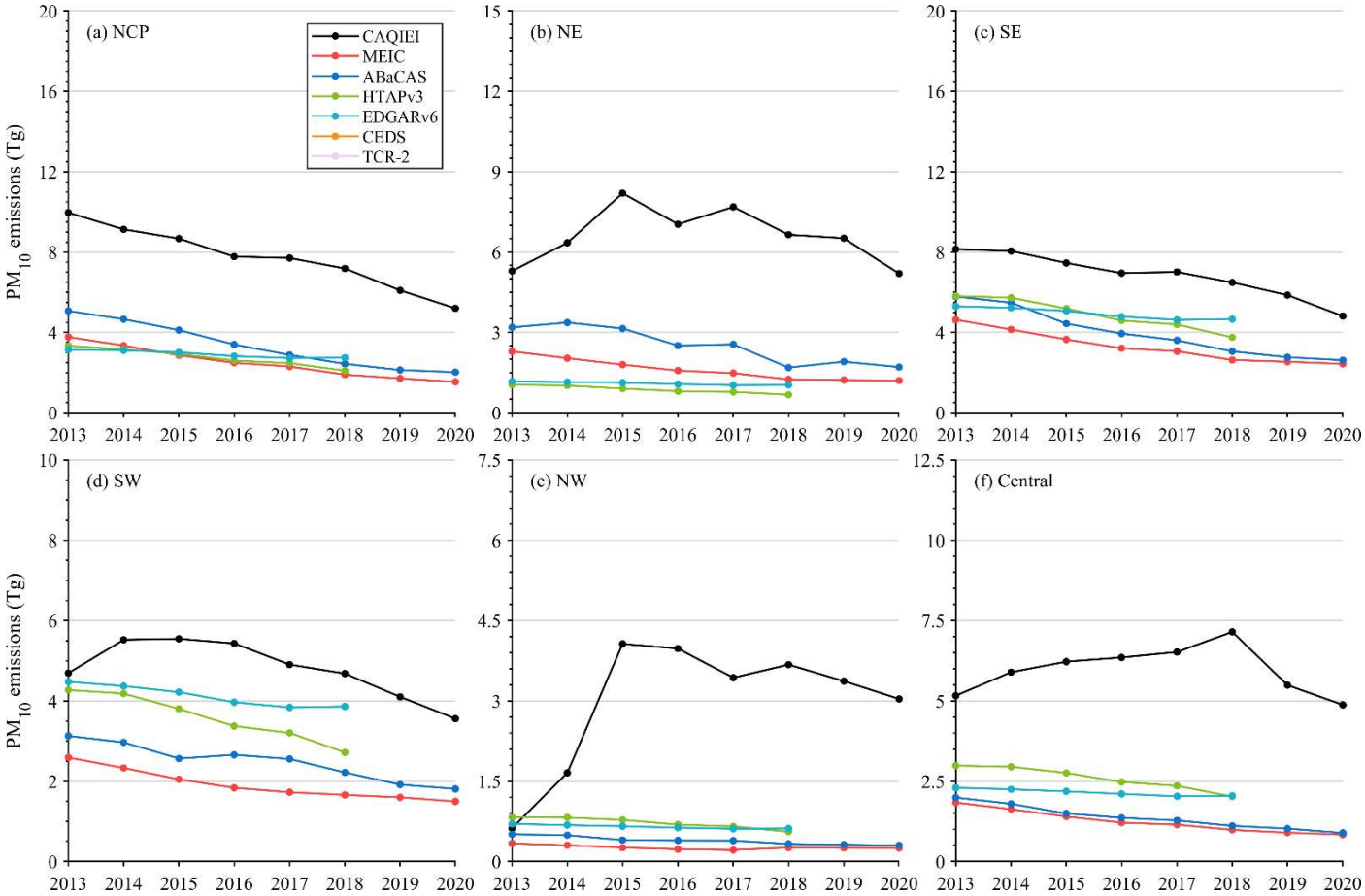

Figure R6: Same as Fig. R5 but for PM_{10}

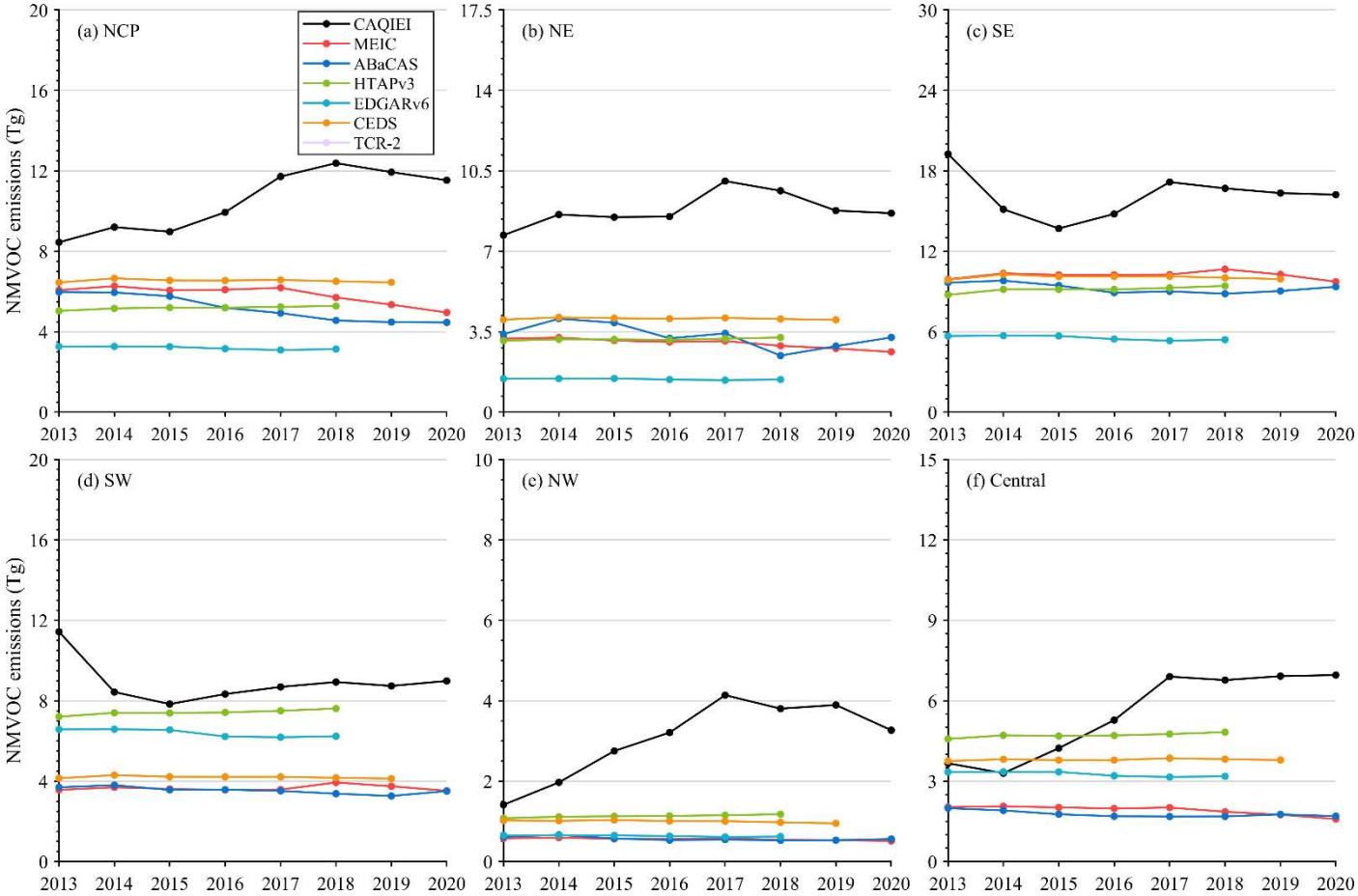

Figure R7: Same as Fig. R2 but for NMVOC

Comment 3: As a high-resolution grid product, is it possible to include a comparison at the grid scale with other inventories?

Reply: Thanks for this comment. We have added the comparisons of the inversion results with the other emission inventory at the grid scale in the revised manuscript by drawing the spatial distributions of the emissions of different pollutants obtained from different emission inventory (Fig. R8). Please see Fig. S9 in the revised supplement.

Changes in the supplement: Fig. S9

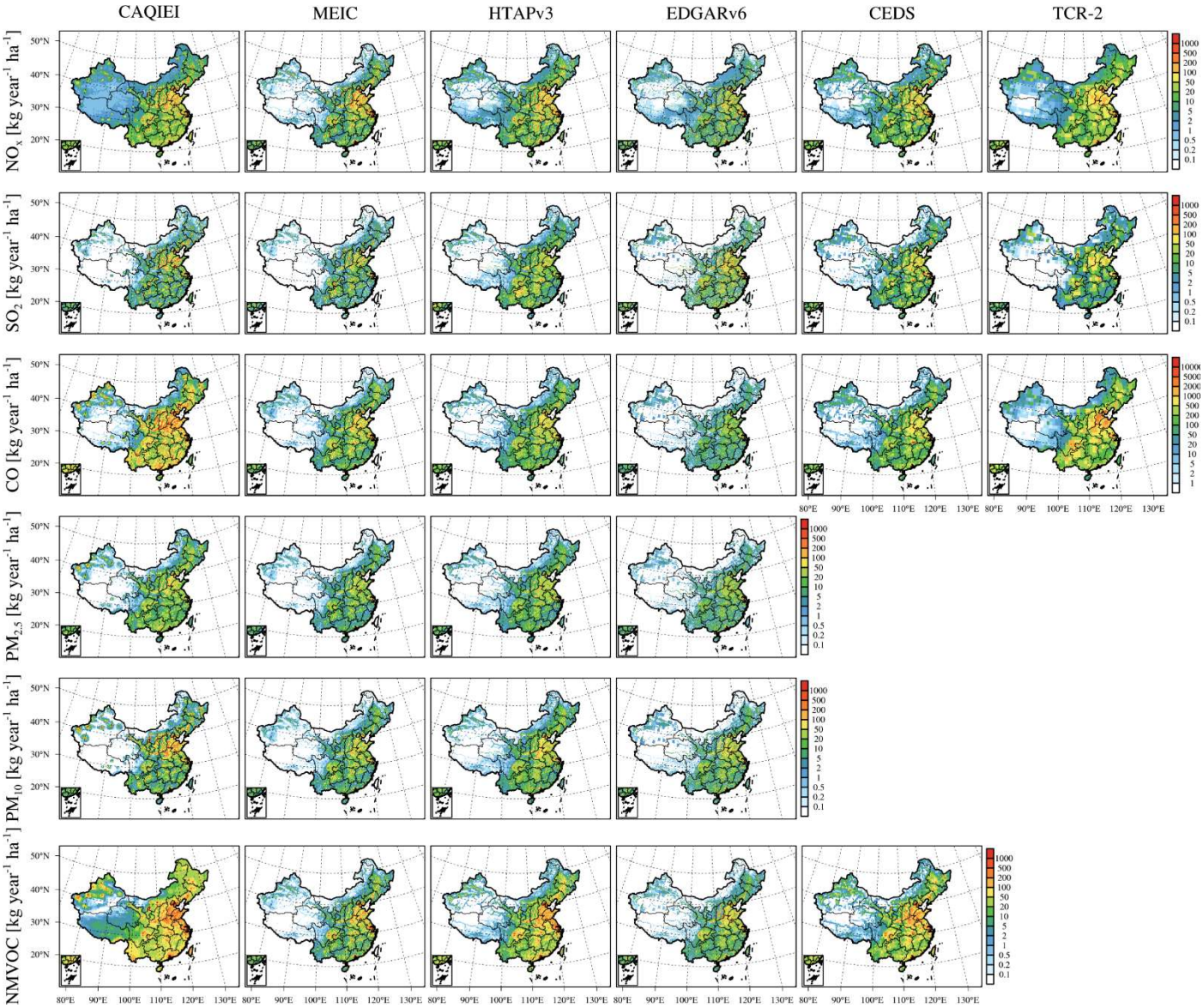

Figure R8: Spatial distributions of the averaged emissions of different air pollutants in China during 2015–2018 obtained from CAQIEI, MEIC, HTAPv3, EDGARv6, CEDS and TCR-2. Note the due to absence of gridded products of the ABaCAS inventory, we did not provide its spatial distributions. Also, the natural sources were not added to the previous emission inventories in this figure because of the different spatial resolutions among these inventories.

Comment 4: In Figure 12, the profiles presented in this study differ from previous research, especially for SO2 and PM10. Are the monthly simulation results for SO_2 and PM_{10} superior to the simulation results using other inventories?

Reply: Thanks for this comment. Since the monthly profiles of the a priori SO_2 and PM_{10} emissions are very similar to those of the other inventories (Fig. R9), the comparisons of the a priori and a posterior simulation could be used to investigate whether the monthly simulation results for SO_2 and PM_{10} are superior to the simulation results using other inventories. Figure R10 and Figure R11 then show the comparison of the a priori and a posterior monthly simulation of SO_2 and PM_{10} over different regions of China. It can be clearly seen that the performance of monthly simulations of SO_2 and PM_{10} are improved significantly by using the a posteriori simulation, suggesting that the monthly simulation results of SO_2 and PM_{10} would be superior to the simulation results using other inventories.

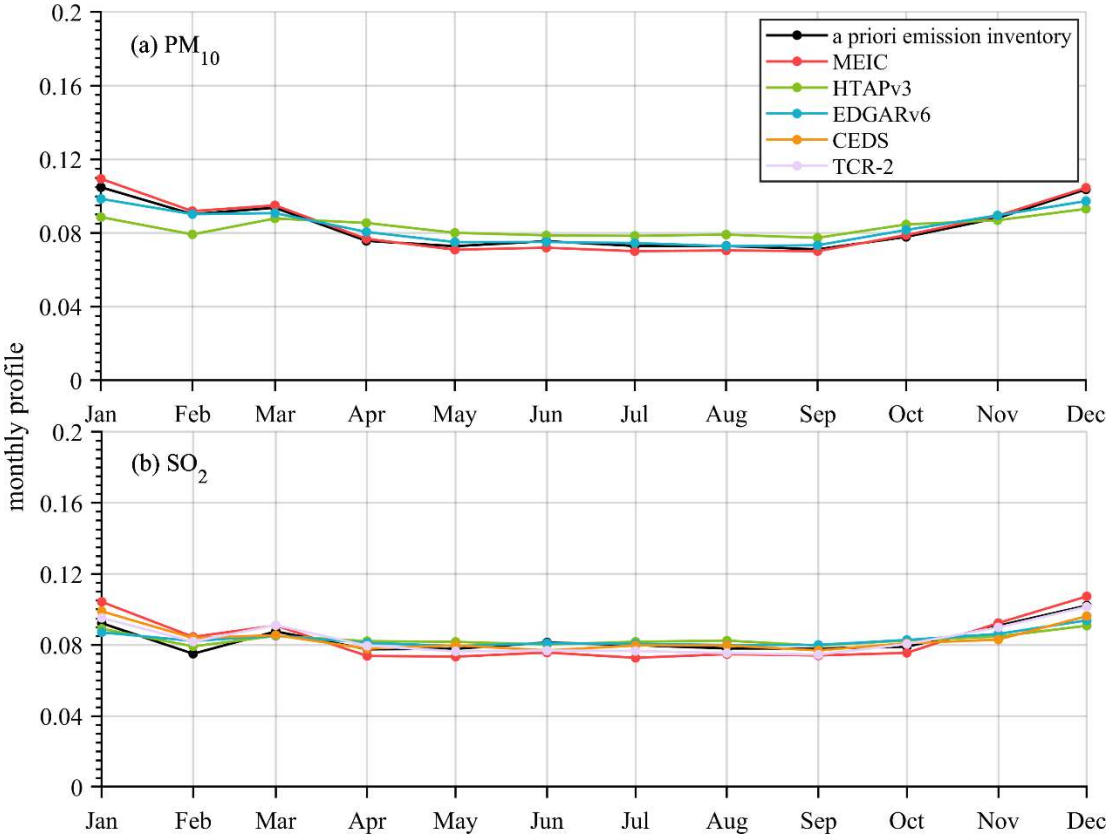

Figure R9: Comparisons of the monthly profiles of the a priori SO_2 and PM_{10} emissions with previous emission inventories.

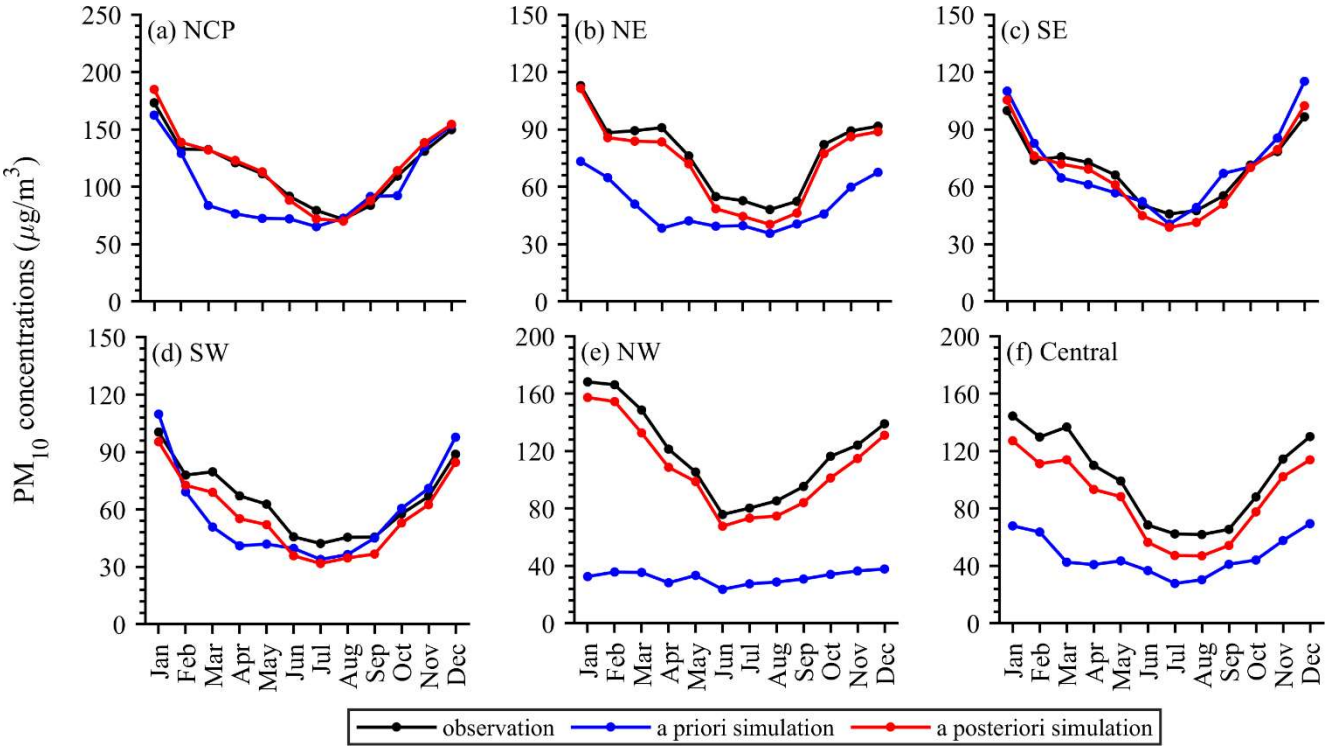

Figure R10: comparison of the a priori and a posterior monthly simulation of PM_{10} concentration over different regions of China.

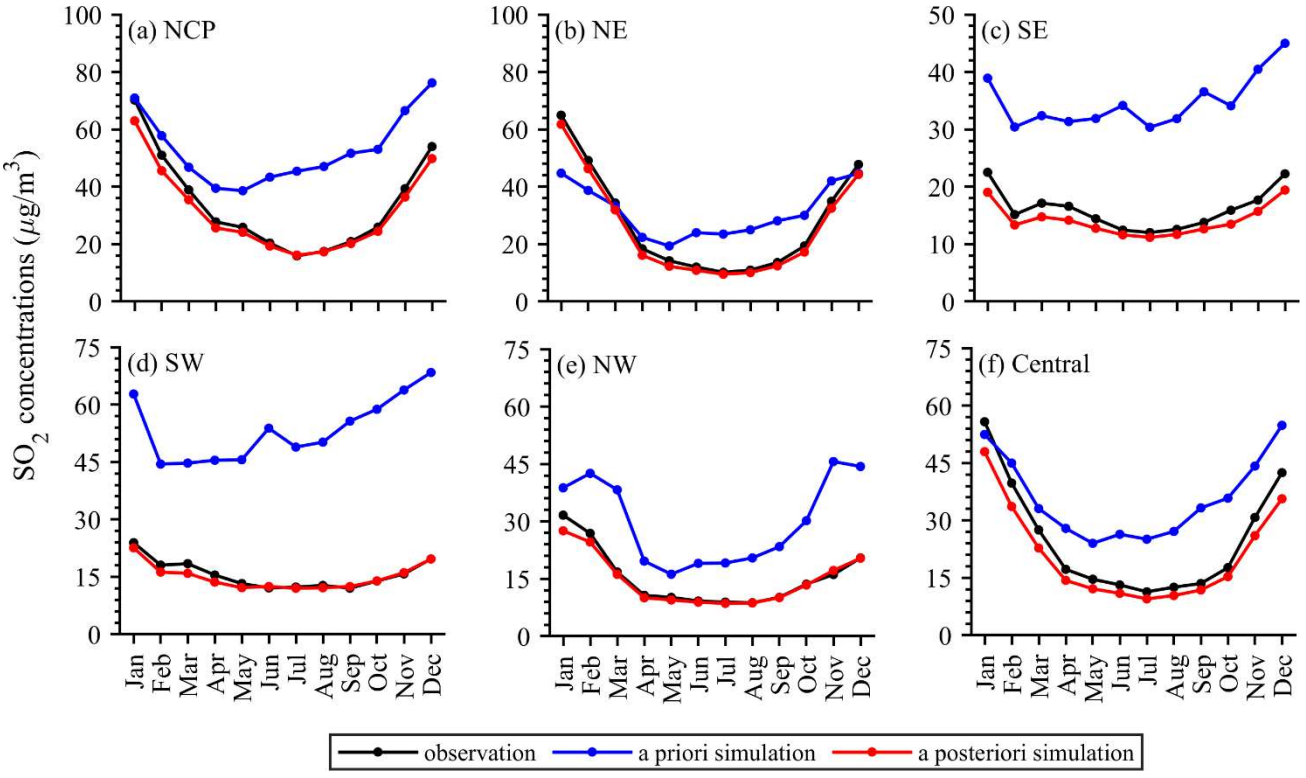

Figure R11: comparison of the a priori and a posterior monthly simulation of SO_2 concentration over different regions of China.

Comment 5: Additionally, it would be interesting to see further validation results from more models (e.g., WRF-CMAQ) using this dataset as input in the future.

Reply: Thanks for this suggestion. Using other models to validate the inversion inventory is a worthwhile endeavor because the results can better validate our inversion inventory and also provide us with interesting information about the impacts of model uncertainty on the emission inversions. As suggested by the reviewer, further validation results by using the other models would be analyzed and provided in the future which has been mentioned in the revised manuscript (please see lines 1015–1016).

Changes in the manuscript: lines 1015–1016.

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 up-to-point 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)

Changes in the manuscript: throughout the Sect 4.3

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 natura sources. Therefore, our primary intendency 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.

Changes in the manuscript: lines 657–665, 786–788, 994–1001

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-O_3 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 O_3 to constrain VOC emissions. As evident from Figure 3, the posterior simulations do not show a significant improvement in O_3. As the authors noted, O_3 cannot effectively constrain precursor NOx (L278). Therefore, I recommend deleting the VOC emission inversion.

Reply: Thanks for this comment. We agree with the reviewer that the NO_x-VOC-O_3 nonlinear interaction would influence the inversion of NMVOC emission based on the O_3 concentration if were not well addressed. On the one hand, the O_3 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 O_3, 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 O_3 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 NO_2, and then constrained the NMVOC emissions based on O_3 concentrations. Also, in Feng et al. (2024), the NO_2 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*-O_3 (Feng et al., 2024). On the other hand, the emission adjustments exhibit bidirectionality dependent on the VOC-limited or NOx-limited regimes. According to the Fig 3 in the revised manuscript, the NMVOC emissions were adjusted in alignment with the direction of the O_3 errors, suggesting a VOC-limited regime over urban areas in China, given that the O_3 observation sites are predominantly situated in the urban areas. This agrees with Ren et al. (2022) who diagnosed the NO_x-VOC-O_3 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 O_3 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 O_3

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 O_3 observations to constrain the VOC emissions. Besides our study, the assimilation of surface O_3 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 O_3 concentrations on the inversion of VOC emissions. For example, Ma et al. (2019) found that the assimilation of O_3 concentration could adjust the NMVOC emissions in the direction resembling the bottom-up inventories, and the forecast skill of O_3 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 O_3 concentrations on the NMVOC emissions as reflected by the improvement of O_3 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-O_3 chemistry and the inherent uncertainty in the satellite observations of formaldehyde and glyoxal (Cao et al., 2018; Stavrakou 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 O_3 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-O_3 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 O_3 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-O_3 interactions and the number of observation sites should be performed in future to better illustrate the feasibility

of assimilating O_3 to constrain NMVOC emissions. Detailed revisions to the manuscript are available in lines 308–337 and 1002–1011 in the revised manuscripts.

Changes in the manuscript: lines 308–337, 1002–1011

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 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 PM_{10} 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 PM_{10} 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 SO_2 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 SO_2 is generally negative over the most of sites, thus the increased observation sites would lead to larger decreasing trend in the inversed emissions of SO_2. 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).

Changes in the manuscript: lines 199–207, 423–427, 984–986

Changes in the supplement: lines 3–28 and Fig. S1–S2

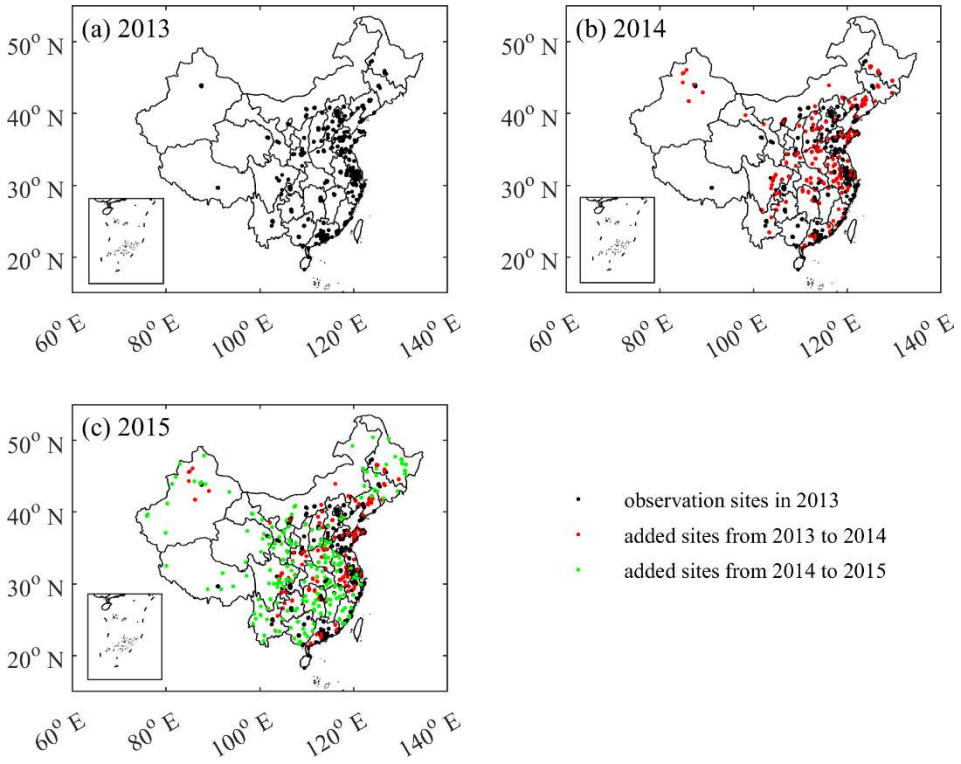

Figure R1 Spatial distributions of observation sites in (a) 2013, (b) 2014 and (c) 2015. The observation sites in 2013 were marked as black dots, while the added observation sites from 2013 to 2014 and those from 2014 to 2015 were marked as red and green dots respectively.

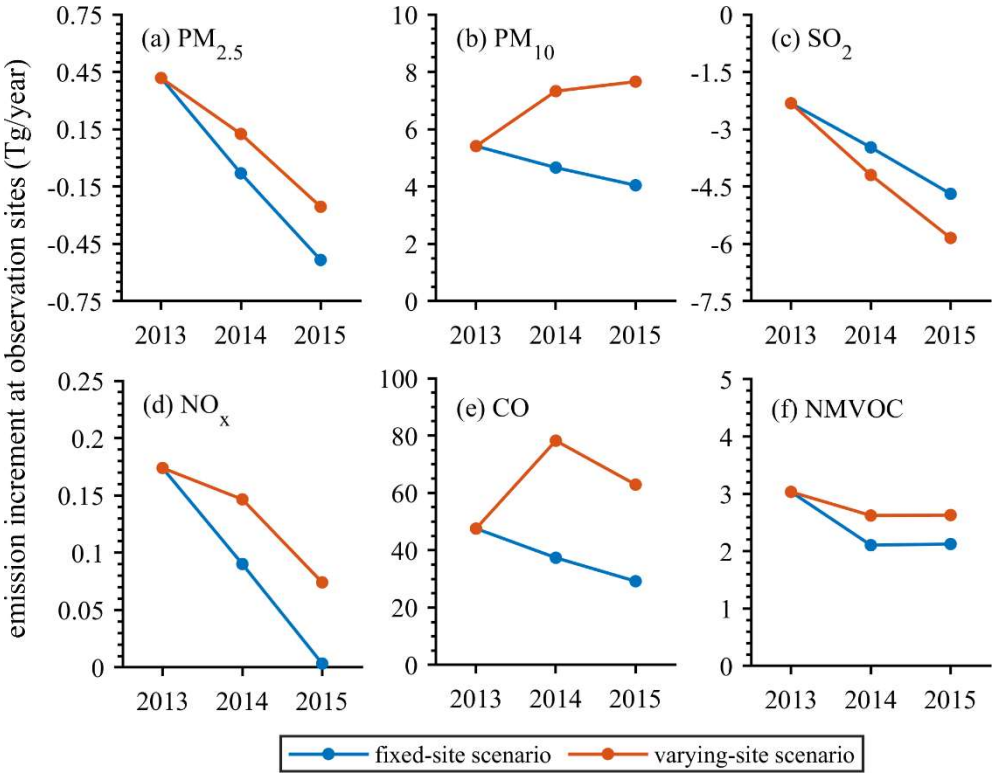

Figure R2: the calculated total emission increments at the observation sites for different species under the fixed-site scenario and varying-site scenario.

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 PM2.5 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 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).

Changes in the manuscript: lines 299–303

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.

Changes in the manuscript: lines 215–219, 231–233

Comment 7: The NOx 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 NOx 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.

Changes in the manuscript: lines 826–831, 566–568

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 O_3 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 O_3 concentrations, as the reviewer mentioned. However, there are still obvious differences in their month variations. For example, the peak values of the observed MDA8h O_3 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 O_3 concentrations, thus some non-linear changes, such as unfavorable meteorological conditions that lead to high O_3 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 O_3 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.

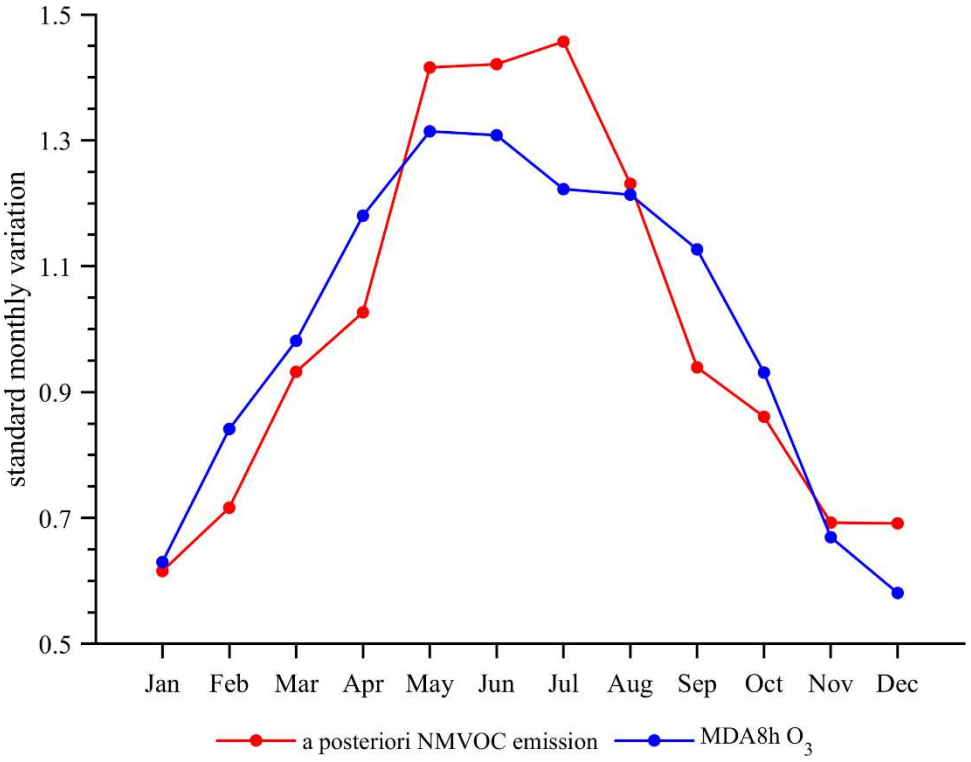

Figure R3: the standard monthly variation of the averaged a posteriori NMVOC emission and MDA8h O_3 in China during 2015–2018.

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 form 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:

$$\mathbf{X^a = X^b - \frac{1}{2}KHX^b} \quad \textbf{(R1)}$$

Based on the analysis ensemble, the uncertainty of the a posteriori emission was estimated as follows: 101.4% ($PM_{2.5}$), 102.5% (PM_{10}), 26.7% (SO_2), 46.8% (CO), 31.8% (NOx) 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).

Changes in the manuscript: lines 885–899

Specific comments:

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

Reply: Done.

Changes in the manuscript: lines 38, 490, 633, 635 and 958

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

Reply: Done.

Changes in the manuscript: lines 229

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 NO_2 and O_3 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).

Changes in the manuscript: lines 282–284

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 SO_2 emissions. We can clearly see that due to the large positive biases in the a priori SO_2 emissions, the model still has large positive biases in simulated SO_2 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.

Changes in the manuscript: lines 345–354 and 358–360

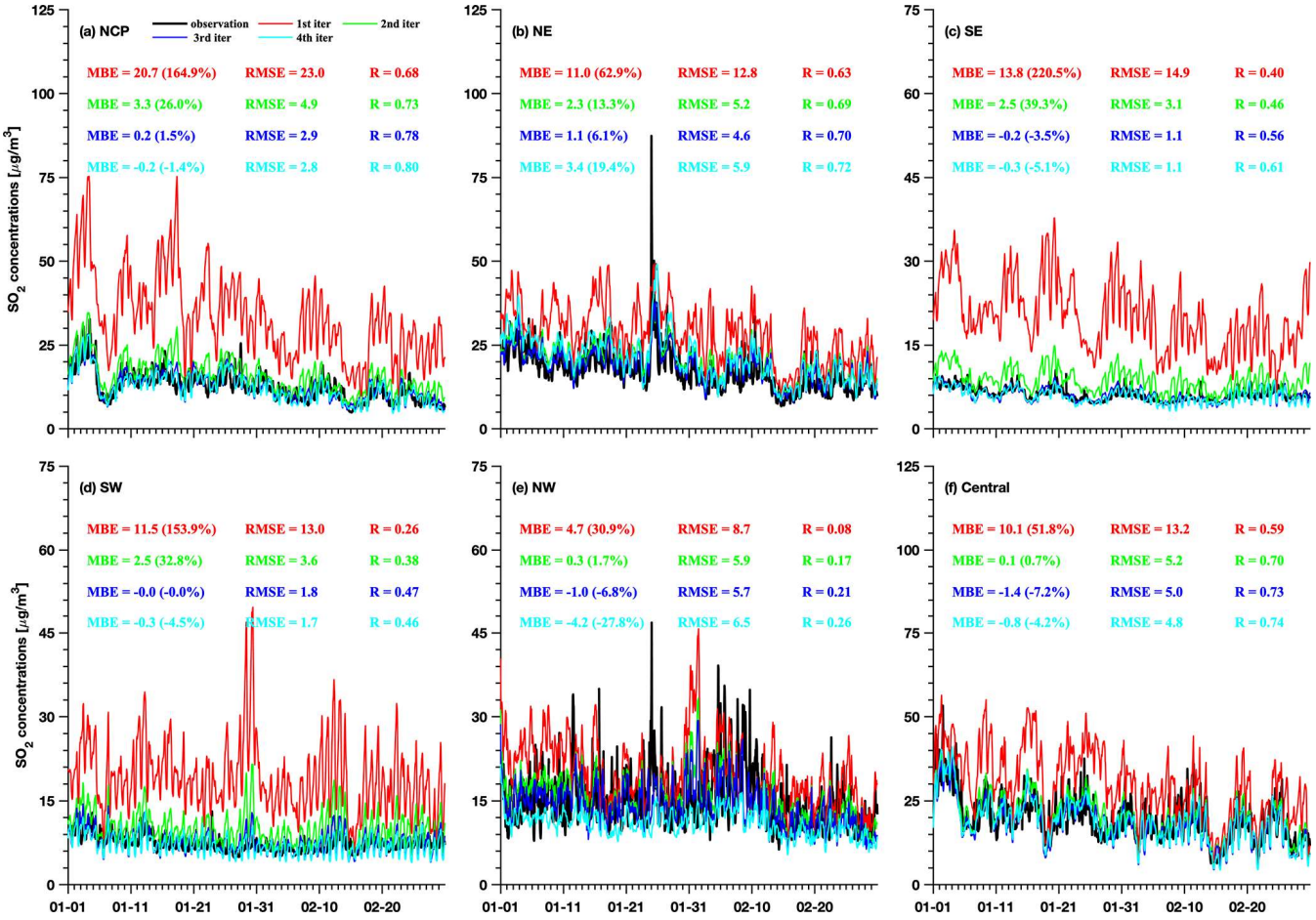

Figure R4: Comparisons of the observed and simulated mean SO2 concentrations using emissions of different iteration time over (a) the NCP region, (b) NE region, (c) SE region, (d) SW region, (e) NW region and (f) central region (taken from Fig 3 in Kong et al. (2023)).

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.

Changes in the manuscript: lines 1255–1258

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):

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

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

where λ is the inflation factor, $\boldsymbol{d}$ is the observation innovation, $\mathbf{R}$ is the observation error covariance matrix, and $\boldsymbol{p}$ 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 PM_{10} and SO_2, 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.

Changes in the manuscript: lines 269 – 277

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)
NMVOC	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.

Changes in the manuscript: lines 1081

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 SO_2, NO_2 and O_3 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.

Changes in the manuscript: 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).

Changes in the 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:

Changes in the manuscript: Figure 11

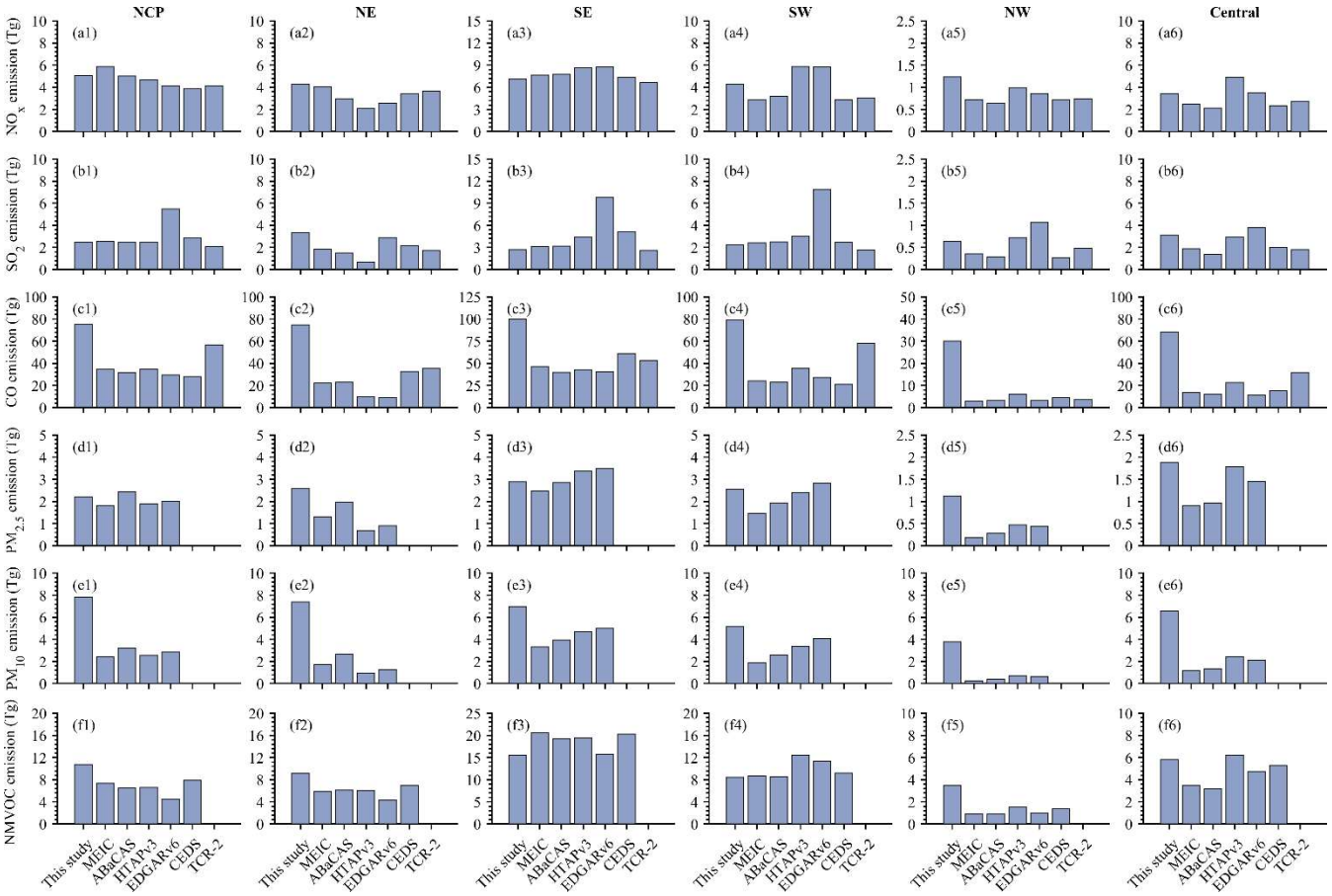

Figure R5: Comparisons of the averaged emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over different regions in China from 2015 to 2018 between CAQIEI and previous inventories added with natural sources.

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

Reply: Done

Changes in the manuscript: lines 548

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Changes of air pollutant emissions in China during two clean air action periods derived from the newly developed Inversed Emission Inventory for Chinese Air Quality (CAQIEI)

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Abstract

A new long-term emission inventory called the Inversed Emission Inventory for Chinese Air Quality (CAQIEI) was developed in this study by assimilating surface observations from the China National Environmental Monitoring Centre (CNEMC) using the ensemble Kalman filter ~~(EnKF)~~ and the Nested Air Quality Prediction Modeling System ~~(NAQPMS)~~. This inventory contains the constrained monthly emissions of NO_x, SO_2, CO, primary $PM_{2.5}$, primary PM_{10}, and NMVOCs in China from 2013 to 2020, with a horizontal resolution of 15 km × 15 km. This paper documents detailed descriptions of the assimilation system and the evaluation results for the emission inventory. The results suggest that CAQIEI can effectively reduce the biases in the *a priori* emission inventory, with the normalized mean biases ranging from −9.1% to 9.5% in the *a posteriori* simulation, which are significantly reduced from the biases in the *a priori* simulations (−45.6% to 93.8%). The calculated RMSEs (0.3 mg/m^3 for CO and 9.4–21.1 μg/m^3 for other species, on the monthly scale) and correlation coefficients (0.76–0.94) were also improved from the *a priori* simulations, demonstrating the good performance of the data assimilation system~~suggesting that CAQIEI can reasonably reproduce the magnitude and variation of emissions of different air pollutants in China~~. Based on CAQIEI, we estimated China's total emissions (including both natural and anthropogenic emissions) of the 6 species in 2015 to be as follows: 25.2 Tg of NO_x, 17.8 Tg of SO_2, 465.4 Tg of CO, 15.0 Tg of $PM_{2.5}$, 40.1 Tg of PM_{10}, and 46.0 Tg of NMVOCs. From 2015 to 2020, the total emissions reduced by 54.1% for SO_2, 44.4% for $PM_{2.5}$, 33.6% for PM_{10}, 35.7% for CO, and 15.1% for NO_x, but increased by 21.0% for NMVOCs. It is also estimated that the ~~Larger~~ emission reductions were ~~achieved~~larger during ~~the~~ 2018–2020 (from -26.6% to -4.5%) ~~action plan~~ than during ~~the~~ 2015~~3~~–2017 (from -23.8% to 27.6%) ~~action plan~~ for most species. ~~In~~ Pparticularly, the total Chinese NO_x and NMVOC emissions were shown to increase during ~~the~~ 2015~~3~~–2017, ~~action pla~~ especially over the ~~in, and there were obvious emission increases in the~~ Fengwei Plain area (FW) where the emissions of particulate matter (PM) also increased ~~over the Central China region~~. The situation changed during 2018–2020 when the upward trends were contained and reversed to a downward trends for both the total emissions of NO_x and NMVOC, and the PM emissions over FW. ~~However, NO_x and NMVOC emissions declined during the 2018–2020 action plan, and the emissions over the Fengwei Plain area also decreased.~~ This suggests that the emission control policies may be~~were~~ improved

in the 2018–2020 action plan. We also compared the CAQIEI with ~~previous~~ other air pollutant emission inventories in China, which verified our inversion results in terms of total emissions of NO_x, SO_2 and NMVOCs, and more importantly identified the potential uncertainties in ~~our~~ current ~~understanding of China's air pollutant~~ emission inventoriess. Firstly, the CAQIEI suggested higher CO emissions in China ~~may be substantially underestimated by current inventories~~, with ~~the~~ CO emissions estimated by CAQIEI (426.8 Tg) being more than twice the amount in previous inventories (120.7–237.7 Tg). Significantly higher emissions were also suggested ~~Significant underestimations for other air pollutant emissions may also exist~~ over the western and northeastern China for other air pollutants. Secondly~~In addition~~, the CAQIEI suggested higher ~~the~~ NMVOC emissions than previous emission inventories by about 30.4–81.4% over the NCP region ~~were shown to be substantially underestimated over northern China~~ but suggested lower NMVOC emissions by about 27.6–0.0%~~but overestimated in~~ over the SE region~~southern China~~. Thirdly~~Secondly~~, the CAQIEI suggested smaller ~~the~~ emission reduction rates during 2015–2018 ~~during 2015–2018 estimated by CAQIEI are generally smaller~~ than ~~those estimated by previous~~previous emission inventories for most species except of CO~~, especially for NO_x, PM_{10} and NMVOCs, suggesting that the mitigation effects of the air pollution control may be overestimated currently~~. ~~In P~~particularly, China's NMVOC emissions were shown to have increased by 26.6% from 2015 to 2018, especially over the NCP~~North China Plain~~ (by 38.0%), n~~N~~ortheast China (by 38.3%), and c~~C~~entral China (60.0%). ~~In contrast, the emissions reduction rate of CO may be underestimated.~~ These results provide us with ~~Overall, our emissions inventory sheds~~ new ~~light~~ insight into~~on~~ the complex variations of air pollutant emissions in China during its two recent clean air action ~~period~~s, which has the potential to~~could significantly~~ improve our understanding of air pollutant emissions in China and ~~related changes~~their impacts on ~~in~~ air quality ~~in China~~. The whole datasets are available at https://doi.org/10.57760/sciencedb.13151 (Kong et al., 2023).

1 Introduction

Air pollution is a serious environmental issue owing to its substantial impacts on human health, ecosystems, and climate change (Von Schneidemesser et al., 2015; Cohen et al., 2017; Bobbink et al., 1998). According to the World Health Organization, air pollution–induced strokes, lung cancer, and heart disease are causing millions of premature deaths worldwide every year (WHO, 2016). The fine particulate matter ($PM_{2.5}$) in the atmosphere not only degrades visibility but also affects the radiative forcing of the climate, both directly and indirectly (Martin et al., 2004). After removal from the atmosphere through dry and wet deposition, air pollutants such as sulfur, nitrate, and ammonium contribute significantly to soil acidification, eutrophication, and even biodiversity reduction (Krupa, 2003; Hernández et al., 2016).

China has experienced severe $PM_{2.5}$ pollution in recent decades, due to its large emissions of air pollutants associated with rapid urbanization and high consumption of fossil fuels (Kan et al., 2012; Song et al., 2017). The annual concentrations of $PM_{2.5}$ in 2013 reached 106, 67 and 47 $\mu g/m^3$ over the Beijing–Tianjin–Heibei, Yangtze River Delta, and Peral River Delta region, respectively, which were all higher than China's national standard (35 $\mu g/m^3$), and 5–10 times higher than that of the World Health Organization (10 $\mu g/m^3$). To tackle this problem, strict emission control policies (so-called "clean air action plans") have been proposed by China's government, including the "Action Plan on the Prevention and Control of Air Pollution" from 2013 to 2017 (hereinafter called the "2013–2017 a~~A~~ction p~~P~~lan"), and the "Three-year Action Plan for Winning the Bule Sky War" from 2018–2020 (hereinafter called the "2018–2020 a~~A~~ction p~~P~~lan"). With the successful implementation of these two action plans, the air quality was substantially improved in China, as evidenced in both observational and reanalysis datasets (Li et al., 2020b; Zheng et al., 2017; Krotkov et al., 2016; Zhong et al., 2021; Li et al., 2017a; Kong et al., 2021). However, with the deepening of air pollution control, unexpected changes have occurred in China, bringing about new challenges for the mitigation of air pollution in the future. On the one hand, despite a significant decline in $PM_{2.5}$ concentrations in China, severe haze still occasionally occurs during the wintertime (Zhou et al., 2022b; Li et al., 2017c). In addition, field measurements in cities over different regions of China consistently show different responses of aerosol chemical compositions to the emission

control policies (Tang et al., 2021; Zhou et al., 2019; Wang et al., 2022; Zhang et al., 2020; Li et al., 2019a; Xu et al., 2019b; Lei et al., 2021; Zhou et al., 2022a). Compared with other aerosol species that showed substantial decreases during the clean air action plans, nitrate has shown a weaker response to the control measures, remaining at high levels and in some cases having even increased slightly. As a result, nitrate is playing an increasingly important role in heavy haze episodes in winter, and dominates the chemical composition of $PM_{2.5}$ (Fu et al., 2020; Xu et al., 2019a), leading to a rapid transition from sulphate- to nitrate-driven aerosol pollution (Li et al., 2019a; Wang et al., 2019b). On the other hand, photochemical pollution has deteriorated in China, with ozone (O_3) concentrations having increased substantially in eastern China during 2013–2017 (Li et al., 2019b; Lu et al., 2018; Lu et al., 2020; Wang et al., 2020b).

These unexpected changes have raised considerable concern among the scientific community and policymakers regarding the overall effects of the clean air action plans, and how to coordinate the control of $PM_{2.5}$ and O_3 pollution. Addressing this problem requires a comprehensive understanding of the effects of the clean air action plans on the emissions of different chemical species. In this respect, previous studies have compiled several long-term air pollutant emission inventories in China using the bottom-up approach – for example, the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University for 2010–2020 (Zheng et al., 2018); the Air Benefit and Cost and Attainment Assessment System-Emission Inventory version 2.0 (ABaCAS-EI v2.0) developed by Tsinghua University for 2005–2021 (Li et al., 2023); the Regional Emission Inventory in Asia (REAS) for 1950–2015 developed Kurokawa and Ohara (2020); the Emissions Database for Global Atmospheric Research (EDGAR) for 1970–2018 developed by Jalkanen et al. (2012); the Hemispheric Transport of Air Pollution (HTAP) Inventory for 2000–2018 developed by Crippa et al. (2023); and the Community Emissions Data System (CEDS) Inventory for 1970–2019 developed by Mcduffie et al. (2020). These emission inventories have provided the community with important insights into the long-term changes in the emissions of different air pollutants in China, thus playing an indispensable role in our understanding of the effects of the country's clean air action plans on emissions and air quality. However, due to the lack of accurate activity data and emission factors, bottom-up emission inventories are ~~still~~ subject to large uncertainties, particularly during the clean air action periods when the activity data and emission factors changed considerably and were difficult to track. Consequently, the estimated emission rates from different bottom-up emission inventories could differ by more than a factor of 2 (Elguindi et al., 2020). For example, the estimated emissions for the year 2010 from different bottom-up inventories were 104.9–194.5 Tg for carbon monoxide (CO), 15.6–25.4 Tg for nitrogen oxides (NO_x), 22.9–27.0 Tg for non-methane volatile organic compounds (NMVOCs), 15.7–35.5 Tg for sulfur dioxide (SO_2), 1.28–2.34 Tg for black carbon (BC), and 2.78–4.66 Tg for organic carbon (OC), reflecting the large uncertainty in current bottom-up estimates of air pollutant emissions in China, which hinders the proper assessment of the effects of the clean air action plans.

Inverse modeling of multiple air pollutant emissions (i.e., a top-down approach) provides an attractive way to constrain bottom-up emissions by reducing the discrepancy between the model and observation through the use of data assimilation. Numerous studies have confirmed the effectiveness of such a top-down method in verifying bottom-up emission estimates and reducing their uncertainties (e.g., Elbern et al., 2007; Henze et al., 2009; Miyazaki and Eskes, 2013; Tang et al., 2013; Koohkan et al., 2013; Koukouli et al., 2018; Jiang et al., 2017; Muller et al., 2018; Paulot et al., 2014; Qu et al., 2017. Based on long-term satellite observations, the top-down method has also been used to track the long-term variations of emissions. For example, Zheng et al. (2019) estimated the global emissions of CO for the period 2000–2017 based on a multi-species atmospheric Bayesian inversion approach; Qu et al. (2019) constrained global SO_2 emissions for the period 2005–2017 by assimilating satellite retrievals of SO_2 columns using a hybrid 4DVar/mass balance emission inversion method; by assimilating satellite observations of multiple species, Miyazaki et al. (2020b) simultaneously estimated global emissions of CO, NO_x, and SO_2 for the period 2005–2018; and, most recently, a regional top-down estimation of $PM_{2.5}$ emissions in China during 2016–2020 was carried out by Peng et al. (2023) by assimilating surface observations. These studies provide us with valuable clues for evaluating bottom-up emissions and improving our knowledge on the changes in emissions of different species in China during

the clean air action plans. However, most of these studies focused on emission trends at the global scale, which involved the use of coarse model resolutions (>1°) that may be insufficient to capture the spatial variability of emission variations at the regional scale. Meanwhile, current long-term, top-down estimates mainly focus on single species and do not fully cover the two clean air action periods in China. Indeed, to date, there are still no long-term, top-down estimates of major air pollutant emissions in China that fully cover the two clean air action periods.

In a previous study performed by our group, we developed a high-resolution air quality reanalysis dataset over China (CAQRA) for the period 2013–2020 to track the air quality trends in China during the clean air action periods (Kong et al., 2021). In the present study, as a follow up to this work, we constrained the long-term emission trends of major air pollutants in China for 2013–2020 (which will be extended in the future on a yearly basis) by assimilating surface observations of air pollutants from the China National Environmental Monitoring Centre (CNEMC) using an ensemble Kalman filter and the Nested Air Quality Prediction and Forecasting System (NAQPMS). In the following sections, we present detailed descriptions of the chemical data assimilation, the evaluation results of the inversed emissions inventory, and the estimated emission trends of different air pollutants in China during the clean air action periods.

2 The chemical data assimilation system

We used the chemical data assimilation system (ChemDAS) developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences, to constrain the long-term emission trends of different air pollutants in China, which was used in the development of CAQRA in our previous work (Kong et al., 2021). Since the chemical transport model (CTM) and the observations used in the top-down estimation were the same as those used in CAQRA, we only briefly describe these two components in the following two subsections, instead concentrating on providing a fuller description (in the third subsection) of the inversion scheme in ChemDAS.

2.1 Chemical transport model

The NAQPMS model was used as the forecast model to represent the atmospheric chemistry in this study, and the Weather Research and Forecasting (WRF) model was used as the meteorological model to provide the meteorological input data. NAQPMS contains comprehensive modules for the emission, diffusion, transportation, deposition, and chemistry processes in the atmosphere, and has been used in previous inversion studies (Tang et al., 2013; Kong et al., 2019; Wu et al., 2020a; Kong et al., 2023). Detailed configurations of the different modules used in NAQPMS are available in these publications.

Figure 1 shows the domain of the inverse model, which is the same as that used in CAQRA, with a fine-scale horizontal resolution of 15 km. The *a priori* emissions inventory includes the anthropogenic emissions obtained from the HTAP v2.2 emissions inventory that provides the emissions from energy, industry, transport, residential, agriculture, air and ship sectors, with a base year of 2010 (Janssens-Maenhout et al., 2015); biogenic emissions obtained from the Monitoring Atmospheric Composition and Climate (MACC) project (Sindelarova et al., 2014); biomass burning emissions obtained from the Global Fire Emissions Database (GFED), version 4 (Van Der Werf et al., 2010; Randerson et al., 2017); soil and lightning NO_x emissions obtained from Yan et al. (2003) and Price et al. (1997); and marine volatile organic compound emissions obtained from the POET database (Granier et al., 2005). The dust emissions were calculated online in NAQPMS as a function of the relative humidity, frictional velocity, mineral particle size distribution, and the surface roughness (Li et al., 2012), while the sea salt emissions were calculated using the scheme of Athanasopoulou et al. (2008). Note that we did not consider the temporal variation in the *a priori* emission inventory, so that the top-down estimated emission trends were only derived from the surface observations. In addition, we used the constant diurnal variation of the emissions in this study due to the lack of information on the diurnal variation of the emissions from different sectors, which is a potential limitation in our current work. However, since the emission inversion was performed on the daily basis (Sect. 2.3.3), the diurnal variations of the emission may not

significantly influence the simulation results of the daily mean concentrations of air pollutants (less than 1 ppbv for SO_2, NO_2 and O_3) according to the sensitivity experiments conducted by Wang et al. (2010). The initial condition was treated as clean air in NAQPMS, with a 2-week spin-up time. Top and boundary conditions were provided by the Model for Ozone and Related Chemical Tracers (MOZART) (Brasseur et al., 1998; Hauglustaine et al., 1998) data products provided by National Center for Atmospheric Research (NCAR). Note that since the MOZART data products were not available for years after 2018, the multi-year average results from 2013 to 2017 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 over the China. To improve the performance of meteorological simulation, a 36-h free run of the WRF model was conducted for each day by using the NCAR/NCEP 1°×1° reanalysis data. The simulation results of the first 12 h were treated as the spin-up run, and the remaining 24 h were used to provide the meteorological inputs for the NAQPMS model.

2.2 Assimilated observations

The assimilated observational dataset in this study was the same as that used in CAQRA, which includes surface concentrations of $PM_{2.5}$, PM_{10} (coarse particulate matter), SO_2, NO_2 (nitrogen dioxide), CO, and O_3, from 2013 to 2020, obtained from CNEMC (Fig. 1). Before the assimilation, outliers of the observations were filtered out by using an automatic quality control method developed by Wu et al. (2018). Four types of outliers characterized by temporal and spatial inconsistencies, instrument-induced low variances, periodic calibration exceptions, and lower PM_{10} concentrations than those of $PM_{2.5}$, were filtered out to prevent adverse impacts on the inversion process. As estimated in Kong et al. (2021), about 1.5% of observational data were filtered out after quality control, but further assessment showed that it had few effects on the average concentrations of different species, which were estimated to be less than 1 $\mu g/m^3$ for the gaseous air pollutants and less than 5 $\mu g/m^3$ for the particulate matter. Estimation of observation error is also important to the inversion of emissions since the observational error and background errors determine the degree of adjustment to the emissions. The observational error comprises the measurement error and the representativeness error induced by the different spatial scales that the model and observations represent. The estimations of these two components of observational error were the same as those used in CAQRA, detailed descriptions of which are available in Kong et al. (2021).

It should be noted that the number of observation sites were not constant throughout the whole inversion period, being approximately 510 in 2013 and then increasing to 1436 in 2015. According to Fig. S1, the observation sites were mainly concentrated in the megacity clusters in China (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 and~~, which~~ could lead to spurious trends in the top-down estimated emissions. To investigate the potential impacts of this on the top-down estimations, the changes in the coverage of observations over different regions of China from 2013 to 2020 were~~-~~ firstly calculated by the ratio of areas that were influenced by observations to the total area of each region (Fig. 2). It can be clearly seen that the observational coverage increased from 2013 to 2015 with the expansion of the air quality monitoring network in China, and became stable after 2015. However, the influence of the variation in the number of observation sites varied among different regions. Over the North China Plain (NCP) region, the observational coverage was approximately 90% in 2013, and reached 100% in 2014, suggesting that the variation in the observation sites may have little influence on the estimated changes in emissions there. A similar conclusion can be drawn for the Southeast China (SE) region, where the observational coverage was about 75% in 2013 and reached 100% in 2015. Elsewhere, in the other four regions, the influence of the variation in observation sites is expected to be larger because of the low observational coverage in both 2013 and 2014. For example, the observational coverage over the Northwest China (NW) region was less than 10% in 2013, but increased to about 60% in 2015.~~-~~ To better illustrate the impact of changes in observation coverage on

the inversions, the sensitivity analysis of the emission increments with the fixed observation sites or varying observation sites is performed in this study ~~Such large changes in observational coverage are believed to significantly influence the estimated changes in emissions over these regions~~(Text S1 and Fig. S2). It shows that the additional emission increments caused by the increases of observation sites would weaken the decreasing trends estimated in the fixed-site scenario for the emissions of $PM_{2.5}$, NO_x and NMVOC and even lead to increasing trends for the emissions of PM_{10} and CO. In contrast, the increases of observation sites would enhance the decreasing trends of SO_2 estimated in the fixed-site scenario. Such different behaviors are mainly related to the different sign of the emission increment of different species as we illustrated in Text S1. These results highlighted the significant influences of the site differences on the estimated emissions and their trends, which should be noted by the potential users. Therefore~~us~~, in order to reduce this influence on the estimated emission trends, in our following analysis we mainly present the emission trends after 2015, when the observational coverage had stabilized in all regions.

2.3 Data assimilation algorithm

We used the modified EnKF coupled with state augmentation method to constrain the long-term emissions of different air pollutants. EnKF is an advanced data assimilation method originally proposed by Evensen (1994) that features representing the background error covariance matrix with a stochastic ensemble of model realizations. Through the use of ensemble simulations, it has the ability to consider the indirect relationship between the emissions and chemical concentrations caused by the complex physical and chemical processes in the atmosphere. It also allows for the estimation of flow-dependent emission–concentration relationships that vary in time and space depending on the atmospheric conditions. The modified EnKF is an offline application of the EnKF method that works by decoupling the analysis step from the ensemble simulation, which has benefits in the reuse of costly ensemble simulations and makes high-resolution long-term inversion affordable (Wu et al., 2020a). In this method, the ensemble simulation was performed firstly with the perturbed emissions, and then the observations were assimilated to constrain the emissions (Wu et al., 2020a). The state augmentation method is a commonly used parameter estimation method (Tandeo et al., 2020) in which the air pollutant emissions are taken as the state variable and are updated according to the error covariance between the emissions and the concentrations of related species.

2.3.1 State variable and ensemble generations

The state variable used in this study was chosen following our previous multi-species inversion study (Kong et al., 2023), which included the scaling factors for the emissions of fine-mode unspeciated aerosol (PMF), coarse-mode unspeciated aerosol (PMC), BC, OC, NO_x, SO_2, CO, and NMVOC~~s~~, as well as the chemical concentrations of $PM_{2.5}$, $PM_{10\text{-}2.5}$ (PM_{10} minus $PM_{2.5}$), NO_2, SO_2, CO, and daily maximum 8-h O_3 (MDA8h O_3), which are formulated as follows:

$$\boldsymbol{x} = [\boldsymbol{c},\ \boldsymbol{\beta}]^{\boldsymbol{T}}, \quad (1)$$

$$\boldsymbol{c} = [\mathbf{PM_{2.5}}, \mathbf{PM_{10-2.5}},\ \mathbf{NO_2},\ \mathbf{SO_2}, \mathbf{CO},\ \mathbf{MDA8h\ O_3}], \quad (2)$$

$$\boldsymbol{\beta} = [\boldsymbol{\beta}_{\mathbf{PMF}},\ \boldsymbol{\beta}_{\mathbf{PMC}},\ \boldsymbol{\beta}_{\mathbf{BC}},\ \boldsymbol{\beta}_{\mathbf{OC}}, \boldsymbol{\beta}_{\mathbf{NO_x}}, \boldsymbol{\beta}_{\mathbf{SO_2}}, \boldsymbol{\beta}_{\mathbf{CO}}, \boldsymbol{\beta}_{\mathbf{NMVOC}}], \quad (3)$$

where $\boldsymbol{x}$ denotes the vector of the state variable, $\boldsymbol{c}$ denotes the vector of the chemical concentrations of different species, and $\boldsymbol{\beta}$ denotes the vector of the scaling factors for the emissions of different species. Note that although the chemical concentration variables are included in the state variable, they are not optimized simultaneously with the emission in the analysis step and are only used to estimate the covariance between the emission and concentrations. Detailed descriptions of the state variables are available in Table 1.

The ensemble of the scaling factors was generated using the same method of Kong et al. (2021), which has a medium size of 50 and considers the uncertainties of major air pollutant emissions in China, including SO_2, NO_x, CO, NMVOCs, ammonia, PM_{10}, $PM_{2.5}$, BC, and OC. The uncertainties of these species were considered to be 12%, 31%, 70%, 68%, 53%, 132%, 130%, 208% and 258%, respectively according to the estimates of Li et al. (2017b) and Streets et al. (2003). The ensemble of the

chemical concentrations was generated through an ensemble simulation based on NAQPMS and the perturbed emissions calculated by multiplying the *a priori* emissions by the ensemble of the scaling factors. This treatment implicitly assumes that the uncertainty in the chemical concentration is mainly caused by the emission uncertainty. This makes sense on a monthly or yearly basis, considering that substantial changes in emissions are expected to have taken place during the clean air action plans, which are subject to large uncertainty. However, the lack of consideration of other error sources, such as those of the meteorological simulation and the model itself, may lead to underestimation of the background error covariance and overcorrection of the emissions, which is a potential limitation of this study. In addition, the dust and sea salt emissions were not perturbed and constrained in this study, and thus the errors in the simulated fine and coarse dust emissions would influence the inversion of $PM_{2.5}$ and PM_{10} emissions. As a result, the top-down estimated $PM_{2.5}$ and PM_{10} emissions will contain errors in the simulated dust and sea salt emissions. Particularly, we did not consider the emission of coarse dust during the inversion process since we found large errors in the simulated coarse dust concentration that could have significantly influenced the inversion of PM_{10} emissions. Consequently, the top-down estimated PM_{10} emissions in this study comprise all coarse dust emissions. A detailed description of the ensemble generation is available in Kong et al. (2021).

2.3.2 Inversion algorithm

We used a deterministic form of EnKF (DEnKF) proposed by Sakov and Oke (2008) to update the scaling factors of the emissions of different species, which is formulated as follows:

$$\overline{\boldsymbol{x}^{\boldsymbol{a}}} = \overline{\boldsymbol{x}^{\boldsymbol{b}}} + \mathbf{K}\lambda\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}\mathbf{H}^{\mathrm{T}}\left(\mathbf{H}\lambda\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R}\right)^{-1}\left(\boldsymbol{y}^{\boldsymbol{o}} - \mathbf{H}\overline{\boldsymbol{x}^{\boldsymbol{b}}}\right), \quad (42)$$

$$\mathbf{X}^{\mathbf{a}} = \mathbf{X}^{\mathbf{b}} - \frac{\mathbf{1}}{\mathbf{2}}\mathbf{K}\mathbf{H}\mathbf{X}^{\mathbf{b}} \quad (5)$$

$$\mathbf{K} = \lambda\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}\mathbf{H}^{\mathrm{T}}\left(\mathbf{H}\lambda\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}\mathbf{H}^{\mathbf{T}} + \mathbf{R}\right)^{-1}, \quad (6)$$

$$\overline{\boldsymbol{x}^{\boldsymbol{b}}} = \frac{1}{N}\sum_{i=1}^{N}\boldsymbol{x}_{\boldsymbol{i}}^{\boldsymbol{b}}\,;\boldsymbol{X}_{\boldsymbol{i}}^{\boldsymbol{b}} = \boldsymbol{x}_{\boldsymbol{i}}^{\boldsymbol{b}} - \overline{\boldsymbol{x}^{\boldsymbol{b}}}, \quad (3)$$

$$\mathbf{B}_{\mathbf{e}}^{\mathbf{b}} = \frac{1}{N-1}\sum_{i=1}^{N}\boldsymbol{X}_{\boldsymbol{i}}^{\boldsymbol{b}}\left(\boldsymbol{X}_{\boldsymbol{i}}^{\boldsymbol{b}}\right)^{\mathrm{T}}, \quad (74)$$

$$\overline{\boldsymbol{x}^{\boldsymbol{b}}} = \frac{1}{N}\sum_{i=1}^{N}\boldsymbol{x}_{\boldsymbol{i}}^{\boldsymbol{b}}\,;\boldsymbol{X}_{\boldsymbol{i}}^{\boldsymbol{b}} = \boldsymbol{x}_{\boldsymbol{i}}^{\boldsymbol{b}} - \overline{\boldsymbol{x}^{\boldsymbol{b}}}, \quad (8)$$

where $\overline{\boldsymbol{x}}$ denotes the ensemble mean of the state variable; the superscript $\mathbf{b}$ and $\boldsymbol{a}$ respectively denote the *a priori* and *a posteriori* estimate; $\mathbf{X}^{\mathbf{a}}$ is the analysed anomalies that can be used to calculate the uncertainty of the a posteriori emissions. $\mathbf{K}$ is the Kalman gain matrix; $\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}$ is the background error covariance matrix calculated by the background perturbation $\boldsymbol{X}^{\boldsymbol{b}}$; $\boldsymbol{y}^{\boldsymbol{o}}$ is the vector of the observation and $\mathbf{R}$ is the observation error covariance matrix; $\mathbf{H}$ is the linear observation operator, which maps the model space to the observation space; $\boldsymbol{\lambda}$ is the inflation factor used to compensate for the underestimation of the background error caused by the limited ensemble size and unaccounted error sources, which is calculated using the method of Wang and Bishop (2003),

$$\lambda = \frac{\left(\mathbf{R}^{-1/2}\boldsymbol{d}\right)^{\mathrm{T}}\mathbf{R}^{-1/2}\boldsymbol{d} - \boldsymbol{p}}{\boldsymbol{trace}\left\{\mathbf{R}^{-1/2}\mathbf{H}\mathbf{B}_{\mathbf{e}}^{\mathbf{b}}\left(\mathbf{R}^{-1/2}\mathbf{H}\right)^{\mathrm{T}}\right\}} \quad (95)$$

$$\boldsymbol{d} = \boldsymbol{y}^{\boldsymbol{o}} - \mathbf{H}\overline{\boldsymbol{x}^{\boldsymbol{b}}} \quad (106)$$

where $\boldsymbol{d}$ is the observation innovation and $\boldsymbol{p}$ is the number of observations. Table S1 summarized the calculated average value (standard deviation) of the used inflation factor for different species. 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 PM_{10} (36.0–78.1) and SO_2 (7.8–176.1), 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. This 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.

In order to reduce the influence of the spurious correlations on the performance of data assimilation, the EnKF was performed locally in this study in that the analysis was calculated grid by grid with the assumption that only measurements located within a certain distance (cutoff radius) from a grid point would influence the analysis results of this grid. The use of this local analysis method also allowed the inflation factor to be calculated locally and to vary in time and space, which can help characterize the spatiotemporal variations of errors as we illustrated above. Similar to in Kong et al. (2021) and Kong et al. (2023), the cutoff radius was chosen as 180 km for each species based on the wind speed and the lifespan of the species (Feng et al., 2020).~~, and~~ T~~t~~he same local scheme with a buffer area was also employed during the inversion to alleviate the discontinuities in the updated state caused by the cut-off radius.~~,~~ A detailed description of the local analysis scheme is available in Kong et al. (2021).

Table 1 ~~then~~ summarizes the corresponding relationships between the emissions and chemical concentrations. Similar to in Ma et al. (2019) and Miyazaki et al. (2012), we did not consider the inter-species correlation during the assimilation, to prevent the spurious correlations between non- or weakly related variables. In most cases, observations of one particular species were only allowed to adjust the emissions of the same species. The assimilation of $PM_{2.5}$ mass observation was more complicated ~~than that of other species~~ as there are multiple error sources in the simulated mass concentrations of $PM_{2.5}$, not only from primary emission, but also from secondary production. In this study, the $PM_{2.5}$ mass observation was used to constrain the emissions of PMF, BC and OC but not used to constrain the emissions of its precursors to avoid the spurious correlations and nonlinear chemistry effects, which is similar to the scheme used in Ma et al. (2019). This is feasible since the emissions of primary $PM_{2.5}$ (i.e., PMF, BC and OC) and the emissions of $PM_{2.5}$ precursors (e.g., SO_2, NO_2) were perturbed independently in our method, thus the contributions of primary $PM_{2.5}$ emission and the secondary $PM_{2.5}$ productions to the $PM_{2.5}$ mass could be isolated through the use of ensemble simulations. Meanwhile, the use of iteration inversion method (which will be introduced later) can further reduce the influence of the errors in the precursors' emissions on the inversion of primary $PM_{2.5}$ emission, since the precursors' emission would be constrained by their own observations during the iterations. However, the lack of assimilation of speciated $PM_{2.5}$ observations may lead to uncertainties in the estimated emissions of PMF, BC and OC, which is a potential limitation in current work. For example, if the a priori simulated $PM_{2.5}$ equals the observations, the emissions of PMF, BC and OC would not be adjusted by using the current method. However, in such cases, there may still be discrepancies in the proportions of the emissions of different $PM_{2.5}$ components. To adjust the emissions of PMC, we used the observations of $PM_{10-2.5}$ to avoid the potential cross-correlations between $PM_{2.5}$ and PM_{10} (Peng et al., 2018; Ma et al., 2019). For the NO_x emissions~~Meanwhile~~, although the O_3 concentration are chemically related to the NO_x emissions, we did not use the O_3 concentrations to constrain the NO_x emission in this study since there is nonlinear relationship between the O_3 concentration and NO_x emission which would lead to wrong adjustment of NO_x emissions (Tang et al., 2016).

The inversion of NMVOC emission is more difficult than other species d~~D~~ue to the lack of long-term nationwide NMVOC observations and the strong chemical activity. Previous studies usually assimilated the satellite observations of formaldehyde and glyoxal to constrain the NMVOC emissions, such as Cao et al. (2018) and Stavrakou et al. (2015). However, these inversion studies are hindered by the NO_x-VOC-O_3 chemistry and the inherent uncertainty in the satellite observations of formaldehyde and glyoxal. Considering the strong chemical relationship between the O_3 and NMVOC, some pioneer studies have also explored the method of assimilating ground-level O_3 concentrations to constrain the NMVOC emissions (Ma et al., 2019; Xing et al., 2020), and demonstrated the effectiveness of this approach. For example, Ma et al. (2019) found that the assimilation of O_3 concentration could adjust the NMVOC emissions in the direction resembling the bottom-up inventories, and the forecast skill of O_3 concentrations were also improved, indicating that the constrained NMVOC emissions are improved relative to their a priori. Inspired by these studies, we have made an attempt to constrain the NMVOC emissions based on ~~,~~the

MDA8h O_3 ~~was used to constrain the NMVOC emissions considering its strong chemical relationship with the NMVOC emissions. Meanwhile,~~ Tthe use of MDA8h O_3 rather than the daily mean O_3 concentration is to~~could~~ avoid the effects of the nighttime O_3 chemistry. An important issue that should be noted when using the MDA8h O_3 to constrain the NMVOC emission is the nonlinear interactions among NO_x, NMVOC and O_3. On the one hand, the O_3 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 O_3, and deteriorate the inversion of NMVOC. ~~Another important issue that should be noted when using the MDA8h O_3 to constrain the NMVOC emission is that the errors in the simulation results of MDA8h O_3 are also caused by the errors in NO_x emissions.~~ The iteration inversion scheme could help deal with this issue as the errors in the NO_x emissions will be constrained by the NO_2 observations in the next iteration, which can reduce the influences of errors in the NO_x emission on the inversion of NMVOC emission based on ~~the~~ MDA8h O_3 concentrations. 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 NO_2, and then constrained the NMVOC emissions based on O_3 concentrations. Also, in Feng et al. (2024), the NO_2 observations were simultaneously assimilated to constrain the NO_x emissions to account for the influences of errors in NO_x emissions on the NMVOC emissions, suggesting that the iteratively nonlinear joint inversion of NO*x* and NMVOCs is an effective way to address the intricate relationship among VOC-NO*x*-O_3 (Feng et al., 2024). On the other hand, the emission adjustments of NMVOC may exhibit bidirectionality dependent on the VOC-limited or NO_x-limited regimes. According to Fig. 3, the NMVOC emissions were adjusted in alignment with the direction of the O_3 errors, suggesting a VOC-limited regime over urban areas in China, given that the O_3 observation sites are predominantly situated in the urban areas. This agrees with Ren et al. (2022) who diagnosed the NO_x-VOC-O_3 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 O_3 concentrations and VOC emissions could be reasonably reflected by our inversion system, providing the feasibility in utilizing the O_3 observations to constrain the VOC emissions. Note that due to the lack observations of the VOC components, we only optimize the gross emissions of the VOC during the assimilation. ~~Meanwhile, although the O_3 concentration are chemically related to the NO_x emissions, we did not use the O_3 concentrations to constrain the NO_x emission in this study since there is nonlinear relationship between the O_3 concentration and NO_x emission which would lead to wrong adjustment of NO_x emissions (Tang et al., 2016).~~

As we illustrated before, there exists nonlinear effects in the atmospheric chemistry which could influence the inversion results of different species. In addition, since we did not consider the temporal variations in the *a priori* emissions, it was expected that there would be significant biases in the *a priori* emissions for the years after 2013, as substantial changes in emissions were expected owing to the implementation of strict emission control measures. Such bias in the *a priori* emissions does not conform to the assumption of the EnKF that the *a priori* estimate is unbiased, which could thus lead to incomplete adjustments of the *a priori* emissions and degrade the performance of the data assimilation (Dee and Da Silva, 1998). To address these issues, an iteration inversion scheme was employed in this study, which has been used previously in Kong et al. (2023). The main idea of the iteration inversion scheme is to preserve the background perturbation $\mathbf{X^b}$ but to update the ensemble mean of the state variable $\overline{\boldsymbol{x^b}}$ based on the inversion results of the kth iteration and corresponding model simulation. According to this, a new single model simulation is required to be conducted by using the a posteriori emission from the previous iteration as the input 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. The state variable used in the $(k+1)$th inversions is then formulated as follows:

$$\boldsymbol{x_i^{b,k+1}} = \left[\boldsymbol{c^k} + \boldsymbol{c_i^e} - \overline{\boldsymbol{c^e}}, \boldsymbol{\beta^k} + \boldsymbol{\beta_i^e} - \overline{\boldsymbol{\beta^e}}\right]^{\boldsymbol{T}}, \quad (11\text{~~7~~})$$

where $\boldsymbol{c^k}$ represents the model simulations using the inversed emissions of the kth iteration, $\boldsymbol{c_i^e}$ represents the ith member of ensemble simulations with an ensemble mean of $\overline{\boldsymbol{c^e}}$, $\boldsymbol{\beta^k}$ represents the updated scaling factors at the kth iteration, and $\boldsymbol{\beta_i^e}$ represents the ith member of the ensemble of scaling factors with a mean value of $\overline{\boldsymbol{\beta^e}}$. Two rounds of iteration were conducted in this study based on our previous inversion study to maintain a balance between the inversion performance and the computational cost of the long-term inversions (Kong et al., 2023).~~, which is enough for reducing the biases in the *a priori* emissions.~~

2.3.3 Setup of inversion estimation

Based on this inversion scheme, we firstly constrained the daily emissions of PMF, PMC, BC, OC, NO_x, SO_2, CO, and NMVOCs, from 2013 to 2020, based on the daily averaged observations of $PM_{2.5}$, $PM_{10-2.5}$, NO_2, CO, and MDA8h O_3. However, similar to in Kong et al. (2023), we only provide the emissions of $PM_{2.5}$ (PMF+BC+OC) and PM_{10} ($PM_{2.5}$+PMF) for the aerosol species since the lack of speciated $PM_{2.5}$ observations would lead to uncertainties in the estimated emissions of PMF, BC, and OC as we mentioned before. Meanwhile, as mentioned in subsection 2.3.1, the meteorological and model uncertainty were not considered in the ensemble simulation. Thus, the errors in the meteorological simulation would cause fluctuations in the daily emissions that could contaminate the inversion results and are~~, which would be~~ difficult to isolate from the inherent variations of emissions (Tang et al., 2013). Considering this, the daily emissions were averaged to monthly values to reduce the influences of random model errors after the assimilation.

3 Performance of the chemical data assimilation system

3.1 Analysis of OmF and emission increment

The observation-minus-forecast (OmF) and emission increment (*a posteriori* emission minus *a priori* emission) were firstly analyzed to demonstrate the performance of the data assimilation. As shown in Fig. 3, the *a priori* simulation generally underestimated the $PM_{2.5}$ concentrations over the NCP, SE and SW regions (positive OmF values) during 2013–2014, but overestimated the $PM_{2.5}$ concentrations from 2016, reflecting the effects of the emission control measures during these years. In the NE, NW and central China (hereafter, "Central") regions, obvious underestimation of the $PM_{2.5}$ concentration was found (positive OmF values) throughout almost the entire assimilation period. Similarly, the OmF values of PM_{10} were positive throughout the whole assimilation period over all regions of China. In contrast, the OmF values for SO_2 were negative for most regions, and the negative OmF values over the NCP region became larger as the years progressed, which reflects the effects of the emission control measures. The OmF for NO_2 reveals a seasonal variation over the NCP and SE regions, with negative values during summer and positive values during winter, while there were obvious positive OmF values over the NE, SW, NW and Central regions. In terms of CO, large positive OmF values were found over all regions of China, and there were decreasing trends in the OmF values of CO over different regions of China associated with the emission control policies during these years. The OmF values for O_3 were positive over most regions of China, except the NW region. These results provide us with valuable information on the potential deficiencies in the ~~suggest that the~~ *a priori* emissions~~ may underestimate the emissions of $PM_{2.5}$, PM_{10}, CO, NO_2 and NMVOCs in China, but overestimate the SO_2 emissions~~. However, since our inversion method did not differentiate between anthropogenic and natural emissions, the biases in the model simulation may also be attributable to the errors in natural emissions such as dust, especially over the major dust-source areas of China (e.g., the NW and Central regions). In addition, the effects of emission control were not considered in the *a priori* emissions, which is another important contributor to the errors in the model simulation for the later years. Thus, the emission increments calculated by the assimilation should reflect the combined effects of errors in the anthropogenic and natural emissions, as well as the emission control.

The calculated emission increments were consistent with the OmF values for all species, which indicates that the data assimilation method can probably constrain the emissions based on the observations. According to Fig. 3, the emission

increments were positive for $PM_{2.5}$ over the NE, NW and Central regions, for NO_2 over the NE, SW, NW and Central regions, and for PM_{10}, CO and NMVOC over almost all regions throughout the assimilation period. In contrast, the emission increments were negative for the SO_2 emissions for most cases. Consistent with the OmF values, the emission increments were positive for $PM_{2.5}$ over the NCP, SE and SW regions during 2013–2014, but became negative from 2016 owing to the implementation of strict emission control measures. The emission increments for NO_x also showed significant seasonal variation over the NCP and SE regions, being positive during winter and negative during summer. The *a posteriori* biases for the model simulations of different species were also plotted to assess the performance of the data assimilation. It can be clearly seen that the biases were substantially reduced for all species, and the calculated root-mean-square error (RMSE) reduced by 23.2–52.8% for $PM_{2.5}$, 19.9–37.8% for PM_{10}, 36.4–77.3% for SO_2, 18.3–25.2% for NO_2, 29.9–40.5% for CO, and 4.4–26.1% for O_3 over the different regions of China, suggesting a good performance of the data assimilation system.

3.2 Evaluation of the inversion results

Table 2 shows the calculated evaluation statistics for the inversion at different temporal scales. It can be clearly seen that the model simulation with the *a posteriori* emission inventory reproduced well the magnitude and temporal variations of the different air pollutants in China, with calculated correlation coefficients of approximately 0.77, 0.72, 0.64, 0.67, 0.69 and 0.71, and normalized mean biases of approximately 4.5%, −4.6%, −9.0%, −3.9%, −8.8% and 9.5%, for the hourly concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and O_3, respectively. Moreover, the *a posteriori* model simulation achieved comparable accuracy with the air quality reanalysis data we developed in Kong et al. (2021) in terms of the RMSE, which was 32.4 $\mu g \cdot m^{-3}$, 53.1 $\mu g \cdot m^{-3}$, 24.9 $\mu g \cdot m^{-3}$, 19.9 $\mu g \cdot m^{-3}$, 0.56 $mg \cdot m^{-3}$ and 34.9 $\mu g \cdot m^{-3}$, respectively, for these species at the hourly scale. At the daily, monthly and yearly scales, the constrained model simulation performed better, with RMSEs of about 9.1–20.0 $\mu g \cdot m^{-3}$ ($PM_{2.5}$), 18.5–31.6$\mu g \cdot m^{-3}$ (PM_{10}), 11.5–16.0$\mu g \cdot m^{-3}$ (SO_2), 8.1–12.8$\mu g \cdot m^{-3}$ (NO_2), 0.28–0.39$mg \cdot m^{-3}$ (CO), and 14.2–26.1 $\mu g \cdot m^{-3}$ (O_3), which were respectively reduced by 56.7–67.3%, 49.2–52.1%, 68.8–72.8%, 36.3–39.8%, 47.0–58.0%, and 22.9–30.5% compared to the RMSEs of the *a priori* simulations. These validation results confirm the good performance of the data assimilation method and suggest~~indicate~~ that the inversed emissions inventory has the capability to~~can~~ reasonably represent the magnitude and long-term trends of the air pollutant emissions in China during 2013–2020.

4 Results

Based on the top-down estimation, the gridded emissions for $PM_{2.5}$, PM_{10}, SO_2, CO, NO_x and NMVOCs over China from 2013 to 2020 were developed into what we have called the Inversed Emissions Inventory for Chinese Air Quality (CAQIEI). In the following sections, we first analyze the magnitude and seasonality of the air pollutant emissions in China by taking 2015 as a reference year~~ for~~ when the number of observation sites became stable. After that, the changes in emissions of different air pollutants from 2015 to 2020 are analyzed and compared between the two clean air action plans in China. Note that due to the impacts of the changes in observation coverage, it is difficult to estimate the overall emission reduction rates during the 2013–2017 action plan by using our inversion results. The emission change rates during 2015–2017 were then sampled in this study to assess the mitigation effects during the 2013–2017 action plan and to be compared with the emission change rates during 2018–2020. Finally, CAQIEI is compared to the previous bottom-up and top-down emission inventories to validate our top-down estimation and identify the potential uncertainties in the current understanding of China's air pollutant emissions.

4.1 Top-down estimated Chinese air pollutant emissions in 2015

The top-down estimated emissions of different species in 2015 are as follows: 25.2 Tg of NO_x, 17.8 Tg of SO_2, 465.4 Tg of CO, 15.0 Tg of $PM_{2.5}$, 40.1 Tg of PM_{10}, and 46.0 Tg of NMVOCs. Note that these values not only contain anthropogenic

emissions but also natural (e.g., dust and biogenic NMVOC) emissions. Thus, the top-down estimated emissions of PM and NMVOCs were higher than those estimated by previous studies, as we mention in the following sections. Emission maps of all species in 2015 are shown in Fig. 4, and the calculated emissions of different species over different regions are presented in Table 3. According to Fig. 4, higher air pollutant emissions are widely distributed in the megacity clusters (e.g., NCP, Yangtze River Delta and Pearl River Delta) and the developed cities in China, reflecting the influences of human activities. NCP was ~~was~~ the region with the largest emission intensity of air pollutants in China, contributing 5.1 Tg of NO_x, 3.5 Tg of SO_2, 82.2 Tg of CO, 2.7 Tg of $PM_{2.5}$, 8.7 Tg of PM_{10} and 9.0 Tg of NMVOCs to the total emissions in China. ~~There were also obvious emission hotspots distributed over the large cities of other regions, reflecting the influences of human activities.~~ The inversion results also demonstrate the contribution of natural sources to the air pollutant emissions, such as the soil NO_x emissions and the biogenic NMVOC emission distributed in the Tibet Plateau region. In general, the majority of air pollutant emissions were located in eastern China (including the NCP, NE and SE regions), where the economy is relatively well developed, which in total accounted for 66.0% of NO_x, 60.9% of SO_2, 57.5% of CO, 60.4% of $PM_{2.5}$, 60.5% of PM_{10}, and 67.8% of NMVOC emissions in China. However, although the GDP of western China (including the SW, NW and Central regions) is less than one third that of eastern China, the top-down estimation indicates that the air pollutant emissions in western China could have accounted for about 32.2–42.5% of the total emissions, which reflects the low emission control levels over these regions.

Figure 5 shows the monthly variations of air pollutant emissions in China for year 2015. The monthly profile of NO_x emissions was relatively flat among the six species. SO_2 and CO showed higher emissions during wintertime because of the enhanced residential emissions associated with higher coal consumption for heating during that time of year. Meanwhile, the emission factor for CO from vehicles in winter was also higher than in other seasons, due to additional emissions from the cold-start process (Kurokawa et al., 2013; Li et al., 2017b). $PM_{2.5}$ and PM_{10} had higher emissions during winter and spring, which, on the one hand was due to the enhanced emissions from the residential and industrial sectors during wintertime (Li et al., 2017b), whilst on the other hand was due to the enhanced dust emissions during the spring season (Fan et al., 2021). Emissions of NMVOCs exhibited strong monthly variations, with higher emissions mainly in summer because of the enhanced NMVOC emissions from biogenic sources.

4.2 Top-down estimated emission changes of different air pollutants

4.2.1 Emission changes of particular matter

Figure 6 shows the top-down estimated emission changes of $PM_{2.5}$ and PM_{10} over China during two clean air action periods. Both $PM_{2.5}$ and PM_{10} emissions decreased substantially, by 44.3% and 21.2% respectively, from 2013 to 2020. On the contrary, the top-down estimates showed increases of $PM_{2.5}$ and PM_{10} emissions in 2014 and 2015, but this would be ~~likely to be~~ a spurious trend caused by the changes of observation sites as we ~~seen by the good correlation between the inversed $PM_{2.5}$ emissions and the observational coverage over the NW region (Fig. 2 and Fig. S1)~~discussed in Text S1. Therefore, the emissions in 2013 and 2014 were discarded to prevent the spurious trends. According to Fig. 6, the $PM_{2.5}$ emissions decreased by 14.5% from~~during~~ ~~the~~ 2015~~3~~ (15.0 Tg) to –2017 (12.8 Tg) ~~clean air action period, from 15.0 Tg in 2015 to 12.8 Tg in 2017~~, and the reduction in emissions was roughly uniform throughout the period, which was about 8% compared to previous years. The PM_{10} emissions showed a smaller reduction rate (−7.2%) than that of $PM_{2.5}$, decreasing from 40.1 Tg in 2015 to 37.2 Tg in 2017. Compared with the emission reduction rate during ~~the~~ 2015–2017~~3–2017 action plan~~, both $PM_{2.5}$ and PM_{10} showed larger emission reduction rates during ~~the~~ 2018–2020 ~~action period~~, estimated to be 27.2% and 25.5%, respectively~~, from 2018 to 2020~~. The emission reductions in each year were also larger, especially for PM_{10}. For example, $PM_{2.5}$ and PM_{10} emissions reduced by about 19.3% and 14.0% in 2019 compared to 2018. This may have been due to that in addition to thee strict controls imposed on the industrial and power sectors during the 2013–2017 action period, ~~along with the strengthened controls~~ theon

residential emissions have been strengthened during the 2018–2020 action period. In particular, "coal-to-electricity" and "coal-to-gas" strategies were vigorously implemented in northern China during~~–~~ the 2018–2020 action~~wintertime~~ to reduce coal consumption and related air pollutant emissions (Liu et al., 2016; Wang et al., 2020a). Thus, our inversion results confirm the effectiveness of the controls on residential emissions in terms of reducing the emissions of $PM_{2.5}$ and PM_{10}. In addition, the control of non-point sources, such as blowing-dust emissions, was also strengthened during the 2018–2020 action period, which is consistent with the faster reduction of PM_{10} emissions during 2018–2020. The annual trends of $PM_{2.5}$ and PM_{10} emissions were also calculated in China using the Mann–Kendall trend test and the Theil–Sen trend estimation method, the results of which are summarized in Table 4. The calculation of emission trends can help extend the existing emission datasets forward in time to produce up-to-date products. The top-down estimated trends of $PM_{2.5}$ and PM_{10} emissions were −1.4 and −2.6 Tg/year during 2015–2020, attributable to the strict emission control measures imposed during the two clean air action plans. As mentioned, the decreasing trends were larger during ~~the~~ 2018–2020 ~~action plan~~ (−1.5 and −4.6 Tg/year) than during ~~the~~ 20153–2017 ~~action plan~~ (−1.1 and −1.5 Tg/year).~~.~~

On the regional scale (Fig. S31), it can be clearly seen that the $PM_{2.5}$ emissions decreased consistently over all regions, by 59.8% in NCP, 49.6% in SE, 39.5% in NE, 35.8% in SW, 33.2% in NW, and 41.0% in Central, from 2015 to 2020. The NCP region showed the largest reduction in emissions among the six regions, with its emission reduction rate being almost larger than 10% in each year. This is consistent with the strictest emission control policies having been imposed over the NCP region. The SE region showed a similar reduction in emissions to the NCP region, with its emission reduction rate being larger than 10% in most years. Obvious increases of $PM_{2.5}$ emissions could be found over the NW region from 2013 to 2015 ~~possibly~~ owing to the increase in the number of observation sites in those years. After 2015, $PM_{2.5}$ emissions generally decreased over the NW region, while there was a slight rebound in $PM_{2.5}$ emissions in 2016 and 2018, possibly due to the influences of the errors in fine dust emission. The Central region showed different characteristics of emission changes to the other regions insofar as it showed little change in $PM_{2.5}$ emissions during 2015–2018 but large reductions in 2019. This may be consistent with the control of emissions over the Feng~~g~~wei Plain area (the part of the Central region where the emission intensity is largest) being weak during the 2013–2017 action plan but strengthened during the 2018–2020 action plan. In terms of the $PM_{2.5}$ emission trends over the different regions, the calculated $PM_{2.5}$ emission trends were about −0.32 Tg/year in NCP, −0.32 Tg/year in SE, −0.24 Tg/year in NE, −0.21 Tg/year in SW, −0.09 Tg/year in NW, and −0.15 Tg/year in Central, from 2015 to 2020.

The changes of PM_{10} emissions were generally similar to those of $PM_{2.5}$, i.e., with decreases in all regions from 2015 to 2020 (Fig. S42). The top-down estimated PM_{10} emission reductions from 2015 to 2020 were about 3.5 Tg (40.0%) in NCP, 2.6 Tg (35.5%) in SE, 3.0 Tg (36.6%) in NE, 2.0 Tg (35.9%) in SW, 1.0 Tg (25.3%) in NW, and 1.3 Tg (21.6%) in Central; and the calculated trends were about −0.64 Tg/yr, −0.52 Tg/yr, −0.51 Tg/yr, −0.40 Tg/yr, −0.20 Tg/yr, and −0.27 Tg/yr, respectively. However, due to the influences of the changes in the number of observation sites, the PM_{10} emissions over the NE, SW and NW regions increased substantially from 2013 to 2015, while they decreased in almost all years after 2015. Different from the other regions, the Central region showed increases in PM_{10} emissions from 2015 to 2018, by about 0.92 Tg (14.9%), but substantial decreases in 2019 and 2020. The result~~s~~ also shows that most PM_{10} emission reductions were achieved during the 2018–2020 action plan. According to CAQIEI, the PM_{10} emissions decreased by 0.64–2.3 Tg (17.4–31.8%) from 2018 to 2020, which accounted for 48.4–169.0% of the total reduction in emissions from 2015 to 2020. This again emphasizes the effectiveness of the control of blowing-dust emissions during the 2018–2020 action plan.

4.2.2 Emission changes of gaseous air pollutants

4.2.2.1 SO_2 and CO

Figure 7 shows the emission changes of different gaseous air pollutants in China from 2013 to 2020. Similar to the PM emissions, SO_2 and CO emissions decreased continuously during the two action plan periods, with top-down estimated

emission reductions of about 9.6 Tg (54.1%) and 166.3 Tg (35.7%) for SO_2 and CO from 2015 to 2020, respectively. Meanwhile, both SO_2 and CO showed a significant decreasing trend from 2015 to 2020, with estimated trends of approximately −2.1 Tg/yr and −36.0 Tg/yr, respectively (Table 5). The reductions in SO_2 and CO emissions are closely consistent with the strict emission control measures imposed during the action plan periods, such as the phasing out of outdated industrial capacity and high-emitting factories, the strengthening of emission standards for industry and the power sector, the elimination of small coal-fired industrial boilers, and the replacement of coal with cleaner energies, which reflects the effectiveness of the emission control measures during the two action plan periods. Reductions of SO_2 emission were generally steady during the two action plan periods, which were approximately 4.2 Tg (23.8%) from 2015 to 2017 and 2.5 Tg (23.5%) from 2018 to 2020. However, CO showed a different emission reduction rate during the two action plan periods, with its emission reductions (67.1 Tg, 18.3%) during ~~the~~ 2018–2020 ~~action plan~~ being larger than those (45.6 Tg, 9.8%) during 2015–2017~~the 2013–2017 action plan~~. This contrast may reflect~~s~~ the different emission control policies during the two clean air action periods, as well as the different emission distributions among the sectors between SO_2 and CO. According to the estimates of Zheng et al. (2018), the share of emissions from the industrial and power sector for SO_2 (77%) is nearly double that for CO (39%). Thus, the smaller reduction of CO emissions ~~during the 2013–2017 action plan~~ than that of SO_2 during 2015–2017 provides evidence that the 2013–2017 action plan mainly focused on controlling the emissions from the industrial and power sectors. During the 2018–2020 action plan, strict control measures targeted on the residential and transportation sectors were also implemented, which together account for 61% of CO emissions but only 23% of SO_2 emissions. As a result, CO showed a larger emission reduction rate during ~~the~~ 2018–2020 ~~action plan~~, while the emission reduction rate for SO_2 was similar to that during~~in the~~ 2015~~3~~–2017 ~~action plan~~. The calculated trends of SO_2 and CO emissions during the two action plans are presented in Table 4, which are −2.1 Tg/yr and −1.3 Tg/yr for SO_2, and −22.8 Tg/yr and −33.5 Tg/yr for CO, respectively.

The reduction of SO_2 and CO emissions was also evident on the regional scale (Fig. S5~~3~~ and S6~~4~~). According to the top-down estimation, the reduction of SO_2 emissions ranged from 0.44 to 2.42 Tg (41.7–69.9%) from 2015 to 2020, with the NCP region exhibiting the largest reductions. The calculated decreasing trend of SO_2 emissions was also significant over all regions, ranging from −0.08 Tg/yr over the NW region to −0.57 Tg/yr over the NCP region (Table 5). With regards to the emission reduction rate during the different action plans, the results suggest that the emission reduction rate of SO_2 was higher during ~~the~~ 2015 – 2017~~3–2017 action plan~~ (by 20.8–39.8%) than that during~~during the~~ 2018–2020 ~~action plan~~ (16.6–29.0%) over the NCP, SE, NE and SW regions. This may have been because, after the strict emission controls imposed upon industry and power plants during the 2013–2017 action plan, the room for further reductions in SO_2 emissions become smaller during the 2018–2020 action plan over these regions. Although residential and vehicle emissions were controlled more strictly during the 2018–2020 action plan, in total they account for ~20% of anthropogenic SO_2 emissions in China (Zheng et al., 2018). Thus, the enhanced reductions in SO_2 emissions from the residential and transportation sectors may not have been able to fully compensate for the weakened reductions from the industrial and power sectors, leading to a smaller SO_2 emission reduction rate over these regions. In contrast, the SO_2 emission reduction rate during~~the~~ 2018–2020 ~~action plan~~ (31.1–34.8%) was higher than that during ~~the~~ 2201~~3~~–2017 ~~action plan~~ (14.1–20.4%) over the NW and Central regions. This may have been due to the fact that the emission controls over the NW and Central regions were relatively weak during the 2013–2017 action plan (as also evidenced by the emission reduction rates of other species) owing to its less-developed economy. During the 2018–2020 action plan, the emission controls over these two regions were strengthened, which led to their higher emission reduction rates. Accordingly, the enhanced SO_2 emission reduction rates over the NW and Central regions compensated for the weakened reduction rates over the other regions, leading to a steady SO_2 emission reduction rate on the national scale.

The reductions of CO emissions from 2015 to 2020 were approximately 14.9–42.3 Tg (21.6–51.4%) over the different regions of China, with significant decreasing trends ranging from −3.0 to −8.7 Tg/yr (Fig. S6~~3~~ and Table 5). Consistent with the comparisons of national CO emission reduction rates between the two action plans, the emission reduction rates during ~~the~~ 2015~~3~~–2017 ~~action plan~~ (4.4–24.6%) were estimated to be smaller than those during ~~the~~ 2018–2020 ~~action plan~~ (12.2–24.6%)

over all the different regions except the Central region, where the CO emission reduction rate was similar during the two action plans (Fig. S64).

4.2.2.2 NO_x and NMVOCs

The top-down estimated NO_x and NMVOC emissions showed different changes to the other four species, by increasing during ~~the~~ 20153–2017 ~~action plan~~ but declining during ~~the~~ 2018–2020 ~~action plan~~. Specifically, NO_x emissions increased slightly by 5.9% from 2015 (25.2 Tg) to 2017 (26.6 Tg), with a non-significant increasing trend of 0.74 Tg/yr. Then, NO_x emissions began to decrease in 2018, with a top-down estimated emission reduction and calculated trend of approximately 3.1 Tg (12.7%) and −1.6 Tg/yr, respectively, from 2018 to 2020. NMVOCs showed stronger emission increases than did NO_x, with top-down estimated emission increases of approximately 12.7 Tg (27.6%) and a calculated emission trend of about 6.3 Tg/yr from 2015 to 2017. Similar to NO_x, NMVOC emissions began to decrease after 2018, with a top-down estimated reduction of approximately 2.6 Tg (−4.4%) from 2018 to 2020, and a calculated trend of about −1.3 Tg/yr.

The increases of NO_x and NMVOC emissions during 2015–2017 suggest~~indicate~~ that the 2013–2017 action plan may not have achieved desirable mitigation effects on these two species. For NO_x emissions, the upward trend may have been associated with the following ~~three~~ factors. ~~Firstly~~On the one hand, vehicle exhaust is one of the most important sources of NO_x in China, accounting for 31% of all NO_x emissions nationally (Zheng et al., 2018). From 2013 to 2017, the number of vehicles in China continued to increase and reached 310 million in 2017, approximately 33.5% higher than in 2013 (MEE, 2017), which led to increases of NO_x emissions from vehicles in China. ~~Secondly, as discussed, the control measures implemented during the 2013–2017 action plan mainly focused on power plants and industrial sources. Controls on vehicle sources were relatively weak. In particular, vehicles with high NO_x emissions, such as heavy-duty diesel trucks, were not controlled strictly during the 2013–2017 action plan.~~ On the other hand~~Thirdly~~, although the 2013–2017 action plan was effective in reducing the NO_x emissions from coal-fired power plants by promoting denitrification facilities and an ultra-low emission standard, the mitigation impacts on industrial NO_x emissions may have been relatively small. For example, Wang et al. (2019a) compiled a unit-based emissions inventory for China's iron and steel industry from 2010 to 2015, based on detailed survey results of approximately 4900 production facilities in mainland China. They found that there were almost no NO_x control measures in China's iron and steel industry during 2010–2015, resulting in a 12.4% increase in China's NO_x emissions from the iron and steel industry in 2015 compared to 2010. In addition, although the penetration rate of denitrification facilities in China's cement industry reached 92% in 2015, the actual operating rate of denitrification facilities in the cement industry was not desirable, due to the lack of online emission monitoring systems. According to the research results of the Ministry of Ecology and Environment, 800, 1300, and 1400 cement production kilns were equipped with selective non-catalytic denitrification facilities from 2013 to 2015, but the actual operating rates were only 51%, 54% and 73%, respectively (Liu et al., 2021). In addition, the new precalciner kilns used in the cement industry have a higher NO_x emission factor, such that the shift from traditional vertical kilns to precalciner kilns has to some extent increased the cement industry's emissions of NO_x (Liu et al., 2021). Thus, there is evidence that the mitigation effects of the industrial control measures on NO_x emissions may not be as significant as expected. Overall, the increased number of vehicles may have offset the emission mitigation effects brought about by the control of power plants, and ~~. Meanwhile,~~ the mitigation effects of controlling ~~vehicle and~~ industrial NO_x emissions were also undesirable. Consequently, NO_x emissions in China may not have decreased, and even increased slightly, during the 2013–2017 action plan. Figure S75 further shows the changes in NO_x emissions over different regions of China, revealing that NO_x emissions over the NCP, SE, NE and SW regions were roughly unchanged (by less than 5%) from 2015 to 2017, while they increased over NW (18.6%) and Central (17.5%). This is consistent with previous results and indicates that NO_x emissions may have increased over the NW and Central regions, possibly due to their increased human activities and weak emission controls.

In terms of NMVOC emissions, since the inversion results did not differentiate between anthropogenic and biogenic sources, the changes in NMVOC emissions may have been related to both anthropogenic and biogenic emissions. With respect to anthropogenic emissions, previous bottom-up studies have suggested that China's NMVOC emissions did not decline during the 2013–2017 action plan, due to the lack of effective control measures on the chemical industry and solvent use (Zheng et al., 2018; Li et al., 2019c). According to the estimates of Li et al. (2019c), China's NMVOC emissions from solvent use increased by 11.1% in 2017 compared to those in 2015. Meanwhile, the increase in the number of vehicles in China may also have led to an increase in NMVOC emissions from transportation. Thus, ~~Therefore,~~ the~~the~~ increases of NMVOC emission during 2015–2017 estimated by our inversion inventory may be related to the increases in anthropogenic NMVOC emissions from the chemical industry, solvent use, and vehicles~~, together with the increase in biogenic NMVOC emissions, may be the main reasons for the increased NMVOC emissions during the 2013–2017 action plan~~. For the trends of biogenic NMVOC emissions, the CAMS global emission inventory shows that there were only little changes in the biogenic NMVOC emissions in China from 2013 to 2018 (Sect. 4.3.3), suggesting little contributions of the biogenic sources to the increased NMVOC emission in China.~~According to the estimations of Li et al. (2020a), there was also an upward trend in China's biogenic NMVOC emissions from 2008 to 2018 because of the increased vegetation cover and air temperature. Compared to the emissions in 2008, China's biogenic NMVOC emissions increased by 20.18% in 2017, with an average annual rate of increase of 2.03%. Therefore, the increases in NMVOC emissions from the chemical industry, solvent use, and vehicles, together with the increase in biogenic NMVOC emissions, may be the main reasons for the increased NMVOC emissions during the 2013–2017 action plan.~~Figure S~~8~~6 further shows the changes in NMVOC emissions over different regions of China, which suggests consistent increases in NMVOC emissions from 2015 to 2017 over different regions. According to the top-down estimations, NMVOC emissions increased by 30.5%, 25.2%, 18.5%, 10.9%, 50.5% and 63.1% over the NCP, SE, NE, SW, NW and Central regions, respectively. Again, the NW and Central regions exhibited the largest emission increases among the six regions, which is consistent with their elevated levels of human activity and weak emission controls.

The decrease in NO_x and NMVOC emissions after 2018 suggests that the emission control strategy of the Chinese government had reached a point of optimization. The 2018–2020 action plan not only strengthened the controls over the industrial and power sectors, but also the transportation sector, especially for diesel vehicles with high NO_x emissions. For example, the Chinese government released the "Action Plan for the Control of Diesel Trucks", and vigorously promoted an adjustment of the transportation structure of China by gradually improving the availability of rail transport. As a result, there was a downward trend in NO_x emissions in China. The top-down estimated reductions of NO_x emissions were approximately 0.81 Tg (17.2%) over NCP, 0.98 Tg (14.0%) over SE, 0.37 Tg (9.4%) over NE, 0.51 Tg (12.2%) over SW, 0.13 Tg (11.0%) over NW, and 0.32 Tg (9.2%) over Central (Fig. S~~7~~5). The decrease in NMVOC emissions after 2018 may on the one hand have been related to the strengthening of vehicle controls during the 2018–2020 action plan, whilst on the other hand it may have been related to the promotion of clean heating plans in the northern region, which reduced the emissions of NMVOCs from residential sources. However, the decreases in NMVOC emissions were smaller than those in NO_x, which were estimated to be 0.84 Tg (6.9%) over NCP, 0.47 Tg (2.8%) over SE, 0.98 Tg (10.1%) over NE, and 0.53 Tg (14.1%) over NW (Fig. S6). Different from other regions, the NMVOC emissions over the SW and Central regions remained almost unchanged during the 2018–2020 action plan (Fig. S~~8~~6).

4.2.3 Changes in the distribution pattern of emissions in China

Due to the different emission control intensities over the different regions of China, the emission distribution patterns of the different species may also have been altered, which could have influenced the distributions of air pollution in China. Based on CAQIEI, we further investigated the emission distribution patterns, as well as their changes, during the two action plans. Maps of the emission changes of different species during 2015–2017~~the 2013–2017 action plan~~ and ~~the~~ 2018–2020~~ action plan~~ are presented in Fig. 8. The shares of emissions in 2015, 2017 and 2020 by each subregion of China are also presented (Fig.

9). It can be seen that the emission changes during the 2015–2017~~3–2017 action plan~~ were more heterogenous than those during ~~the~~ 2018–2020~~ action plan~~. The air pollutant emissions after the 2018–2020 action plan showed consistent reductions over most regions of China, while there were obvious emission increases detected from 2015 to 2017~~over the 2013–2017 action plan~~. This is consistent with the different emission control effectsi~~ciencies~~ during the two clean air action plans as mentioned in previous sections. Due to its strictest emission control policies, the NCP region showed consistent emission reductions of SO_2, NO_x, CO, $PM_{2.5}$ and PM_{10} during the two clean air action plans. Accordingly, the shares of emissions in the NCP region continued to decrease during the two action plan periods (Fig. 9). For example, the share of SO_2 emissions in the NCP region decreased from 19.4% to 15.4% ~~after the~~ during the period of 2015~~3~~–2017~~ action plan~~, and from 15.4% to 12.7% during~~after~~ the 2018–2020 action plan. In contrast, NMVOC emissions increased obviously over the NCP region from~~during the~~ 2015~~3~~ to –2017~~ action plan~~, and decreased during~~ the~~ 2018–2020~~ action plan~~. However, its share did not change significantly~~ during the two action plans~~, being roughly 20% throughout both periods. As for other regions, increases of SO_2, NO_x, $PM_{2.5}$, PM_{10} and NMVOC emissions during 2015–2017~~ the 2013–2017 action plan~~ could be found over the Central region. More specifically, the emission increases were mainly located in the Fenwei Plain area of the Central region, which was due to the fact that this area was not included as a key region of emission controls during the 2013–2017 action plan. However, the Fenwei Plain area was added as a key emission control region during the 2018–2020 action plan, which is consistent with the emission reductions for these species over the Central region (Fig. 8). As a result, the shares of SO_2 and $PM_{2.5}$ emissions in the Central region increased during 2015–2017~~in the 2013–2017 action plan~~ but decreased ~~in the~~during 2018–2020~~ action plan~~ (Fig. 9). However, the shares of NO_x, PM_{10} and NMVOC emissions continued to increase over the Central region during the two clean air action plans, which suggests larger roles of air pollutant emissions in that region. In contrast, the share of CO emissions in the Central region continued to decrease in the two action plans, from 17.7% in 2015 to 13.4% in 2020.

In terms of the shares of emissions in eastern and western China, the top-down estimation suggests an increased share of NO_x, $PM_{2.5}$, PM_{10} and NMVOC emissions in western China after the two clean air action plans (Fig. 9), which indicates slower emission reductions for these species in western China. However, the share of CO emissions in western China was reduced after the two clean air action plans. Although the share of SO_2 emissions in western China increased during~~ the~~ 2015~~3~~–2017~~ action plan~~, it turned to a decrease during~~in the~~ 2018–2020~~ action plan~~.

4.3 Comparisons with different ~~bottom-up~~ emission inventories

In this section, the CAQIEI is compared with the previous long-term bottom-up and top-down emission inventories in China to validate our inversion results and provide the clues for the potential uncertainty in the current air pollutant emission inventories. The bottom-up emission inventories used in the comparison include MEIC (Zheng et al., 2018), ABaCAS (Li et al., 2023), HTAPv3 (Crippa et al., 2023), EDGARv6 (Jalkanen et al., 2012) and CEDS (Mcduffie et al., 2020), while the top-down emission inventory is obtained from the updated Tropospheric Chemistry Reanalysis (TCR-2) (Miyazaki et al., 2020b). However, it is difficult to directly compare our inversion results with these emission inventories considering that the inversion emission includes both anthropogenic and natural emissions. To better compare our inversion results with previous inventories, the natural emission sources, including soil NO_x emissions and biogenic emissions obtained from the CAMS global emission inventory (https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-emission-inventories?tab=overview; last accessed 26 July 2023) and the biomass burning emissions obtained from the Global Fire Assimilation System (GFAS) (Kaiser et al., 2012) are taken as a reference to account for the influences of natural sources. The CAMS and GFAS emission inventory are used because they are state-of-art natural emission inventories and can provide us with long-term estimations of natural emissions. Since the latest year of most emission inventories is 2018, the comparisons were conducted between 2015 and 2018. Note that due to the complexity in the estimations of natural sources, significant uncertainty exists in the estimated natural

emissions. As a result, the comparison results would be sensitive to the used natural emission inventories, especially for the species with large amount of natural emission, such as the NMVOC and particulate matter. Therefore, it should be aware of that the comparison conducted here and the derived implications are on the basis of the natural emissions estimated by CAMS and GFAS. In addition, the natural dust emissions are not considered in the comparisons, which would influence the comparisons of the PM emissions.

4.3.1 Magnitude

~~In this subsection, we compare CAQIEI with previous long-term bottom-up and top-down emission inventories in China to validate our inversion results and identify the potential uncertainty in the current understanding of China's air pollutant emissions. The emission inventories adopted were MEIC (Zheng et al., 2018), ABaCAS (Li et al., 2023), HTAPv3 (Crippa et al., 2023), EDGARv6 (Jalkanen et al., 2012), CEDS (Meduffie et al., 2020), and the top-down emission estimates from the updated Tropospheric Chemistry Reanalysis (TCR-2) (Miyazaki et al., 2020b). (https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-emission-inventories?tab=overview; last accessed 26 July 2023)Since the latest year of most emission inventories is 2018, the comparisons were conducted between 2015 and 2018. In particular, the comparison with MEIC is highlighted considering its wide application in Chinese air pollution studies. Considering that the top-down estimation includes both anthropogenic and natural sources, the natural emission sources, including soil NO_x emissions and biogenic emissions obtained from the CAMS global emission inventory (https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-emission-inventories?tab=overview; last accessed 26 July 2023) and the biomass burning emissions obtained from the Global Fire Assimilation System (GFAS) (Kaiser et al., 2012), were also analyzed to help explain the discrepancies between our inversion results and previous inventories.~~

4.3.1.1 NO_x

Figure 10 shows the average emissions of different air pollutants in China during 2015–2018 obtained from CAQIEI and the previous emission inventories plus the natural sources we considered. ~~.~~Comparisons of the emission estimations on the regional scale and gridded scale are also presented (Fig. 11 and Fig. S9). The results show that the CAQIEI has slightly higher NO_x emissions in China than the other inventories. Considering that CAQIEI includes both anthropogenic and natural sources, this discrepancy could be explained by the natural NO_x sources. According to the estimations of CAMS and GFAS, the soil and biomass-burning NO_x emissions are approximately 1.9 and 0.08 Tg/yr, which explains well the higher NO_x emissions given by CAQIEI. After consideration of the natural sources, MEIC, HTAPv3 and EDGARv6 agree well with our inversion results on the national scale, with their differences within 1.0–7.4%.~~ This confirms well our inversion results and suggests that there is no significant bias in the estimations of total NO_x emissions in China for these inventories.~~ The NO_x emission estimated by ABaCAS, CEDS and TCR-2 ~~may have low bias in their estimated NO_x emissions considering their~~are slightly lower~~smaller values~~ than CAQIEI and other emission inventories. However, the differences between CAQIEI and these inventories were found to range from 15.9% to 21.3%, which is ~~consistent with~~within the previous estimated uncertainties of NO_x emissions in China (Kurokawa and Ohara, 2020; Li et al., 2017b; Li et al., 2023). These results suggest that the total NO_x emissions in CAQIEI are generally consistent with the current estimations of the anthropogenic and natural NO_x emissions in China. On the regional scale, the top-down estimated NO_x emissions show good agreement with the previous emission inventories over the NCP and SE regions, with their differences ranging from 1.0%–26.8%, suggesting good consistency~~lower uncertainties~~ in the estimations of NO_x emissions over these two regions. This makes sense because NCP and SE are the two most developed regions in China, and where surveys and research on emissions are most sufficient. The differences~~uncertainties~~ are larger over the other regions. In the NE region, CAQIEI has~~has~~ higher NO_x emissions than the other inventories~~,~~ by 5–70%, suggesting higher anthropogenic or biomass-burning emissions over there.~~.~~ The estimations made by MEIC, CEDS and TRC-2 are closer to our estimates, with their differences being approximately 5.4–23.3%, while the differences are larger for ABaCAS, HTAPv3 and EDGARv6 (36.7–70.0%).~~ This suggests that anthropogenic or biomass burning emissions may be underestimated over the NE region.~~ Over the SW and Central regions, there are large diversity in the previous emission inventories with estimations by HTAPv3 and EDGARv6 almost double those of MEIC, ABaCAS, CEDS and TCR-2. The CAQIEI suggests a midst estimation which is~~is~~ within the range of previous emission inventories~~higher than MEIC, ABaCAS,~~

~~CEDS and TCR-2 by 29.4–40.8% and 22.4–47.4%, respectively, but is lower than HTAPv3 and EDGARv6 by about 30%, suggesting higher uncertainties of estimated NO_x emissions over these two regions~~. In the NW region, CAQIEI is consistently higher than other~~the previous~~ inventories, by 22.7–64.2%, which ~~which~~ suggests a potential missing source of the NO_x emissions ~~significant low bias may exist in current estimations of NO_x emissions~~ over this region.

4.3.1.2 SO_2

For SO_2 emissions, since natural sources contribute little (only about 0.02 Tg/yr) to them in China, the discrepancies between CAQIEI and previous emission inventories are mainly attributable to the differences in anthropogenic emissions. As shown in Fig. 10, CAQIEI agrees well with HTAPv3 and CEDS on the national scale, with their differences being approximately ±2%, but is higher than MEIC, ABaCAS and TCR-2 by 17.4–32.9%~~, suggesting a negative bias in their estimated SO_2 emissions in China~~. In contrast, EDGARv6 may have a positive bias in its estimated SO_2 emissions, which are roughly double those of CAQIEI and other inventories. On the regional scale, our results agree well with MEIC, ABaCAS, HTAPv3, CEDS and TCR-2 over the NCP region, with their differences ranging from 1.0 to 18.1%~~, suggesting lower uncertainties in SO_2 emissions over this region~~. In the SE region, CAQIEI suggest~~is~~ lower SO_2 emissions than previous emission inventories, except TCR-2. The differences are relatively smaller ~~(by around −15%)~~ for the MEIC and ABaCAS inventories by around −15%, but larger for HTAPv3, EDGARv6 and CEDS (ranging from −47.3% to −113.2%). ~~This suggests that current global emission inventories may overestimate the SO_2 emissions in the SE region.~~ In contrast, CAQIEI ~~is~~ suggests higher SO_2 emissions than all previous emission inventories over the NE region by about 14.8–132.0%, indicating~~which suggests~~ a possible missing sources ~~consistent negative bias may exist in current estimations of SO_2 emissions~~ over ~~this region~~there. Similarly, the CAQIEI and HTAPv3 suggest~~is~~ higher SO_2 emissions than the MEIC, ABaCAS, CEDS and TCR-2~~,~~ by 27.0–75.6%~~,~~ in the NW region, and by 44.3–77.7% in the Central region~~, suggesting a negative bias in estimations of SO_2 emissions over these two regions~~. ~~The SO_2 emissions estimated by HTAPv3 are closer to our inversion results, with their differences being about 6.9–12.6%.~~

4.3.1.3 CO

For CO emissions, CAQIEI is substantially higher than the previous emission inventories, with the estimated CO emissions of CAQIEI being about three times higher than the bottom-up inventories and more than double those of the top-down estimates made by TCR-2. According to GFAS, the average rate of CO biomass-burning emissions in China from 2015 to 2018 was about 3.4 Tg/yr. Yin et al. (2019), based on MODIS fire radiative energy data, also estimated China's CO biomass-burning emissions to be about 5.0 (2.3–7.8) Tg/yr. The biogenic CO emissions obtained from the CAMS global emission inventory were approximately 2.3 Tg/yr. According to these estimates, natural CO emissions in China have a magnitude of about 10^1, which is rather small compared with anthropogenic sources, and cannot explain the large discrepancies between CAQIEI and other inventories. Thus, the CAQIEI suggest much higher anthropogenic CO emissions in China than the existing emission inventories~~there may be a large negative bias in current estimations of anthropogenic CO emissions in China~~. In fact, the potential underestimation of CO anthropogenic emissions has been investigated~~revealed~~ in previous studies and is regarded as the main reason for the negative bias in global or hemispheric CO simulations (Stein et al., 2014; Gaubert et al., 2020). Regionally, Kong et al. (2020) compared a suite of 13 modeling results from six different CTMs—namely, NAQPMS, CMAQ, WRF-Chem, NU-WRF, NHM-Chem and GEOS-Chem—with observations over the NCP and Pearl River Delta regions under the framework of the Model Inter-Comparison Study for Asia III (MICS-Asia III), and found consistent negative biases in the CO simulations of all models, pointing toward potential underestimations of CO emissions in China. Previous inversion studies have also reported ~~a significant underestimation of~~higher a posteriori CO emissions than~~in~~ their *a priori* emission inventories (Bergamaschi et al., 2000; Miyazaki et al., 2012; Petron et al., 2002; Petron et al., 2004; Tang et al., 2013; Gaubert et al., 2020). For example, the constrained CO emissions~~inversion results~~ reported by Gaubert et al. (2020) are 80% higher than ~~suggested~~

thecat CEDS ~~underestimates CO emissions by 80% over their~~ northern China. ~~Therefore, O~~our inversion results are consistent with ~~these~~previous inversion studies, suggesting higher ~~which supports the point on the underestimation of~~ anthropogenic CO emissions in China. However, direct evidence in support of such high CO emissions in China reported by our study is still limited currently. Thus, we compiled more inversion results within the period of 2013–2020 from previous studies to further validate our inversion results, which are summarized in Table 6. It can be clearly seen that there are large differences in the estimated CO emissions between the inversion results based on surface observations and those based on satellite data. Our inversion results are consistent with the results of Feng et al. (2020), with China's CO emissions in December 2017 estimated at approximately 1500.0 kt/day and 1388.1 kt/day, respectively. In addition, Feng et al. (2020) used the CMAQ model to constrain CO emissions, which is different from the model we used. This may indicate that the model uncertainty would not significantly influence the inversion results of CO emissions. However, the top-down estimated CO emissions based on satellite data (163.6–553.4 kt/day) are much lower than those based on surface observations, although they are all higher than their *a priori* emissions. The lower CO emission estimations based on satellite data assimilation may be attributable to the lower sensitivities of satellite data to surface concentrations, suggesting that the assimilation of satellite data alone may not be adequate to correct the negative biases in the *a priori* emissions. This deficiency has also been revealed by Miyazaki et al. (2020b), who found undercorrected surface CO emissions in the extratropic~~s~~ of the Northern Hemisphere in TCR-2. However, the assimilation of surface observations can be influenced by the uncertainties in the modeled vertical mixing, which could lead to the uncertainties in the inversed CO emissions based on surface observations. Therefore, the inversed CO emissions in CAQIEI could be partly supported by previous inversion studies based on surface observations, but more evidence is still needed to justify the magnitude of the inversed CO emissions. Besides anthropogenic sources, the chemical production of CO via oxidation of methane (CH_4) and NMVOCs, as well as the CO sinks via the hydroxyl radical (OH) reaction, also influence the simulation of CO (Stein et al., 2014; Gaubert et al., 2020; Müller et al., 2018). Due to the important role of OH in the chemical production and sinks of CO, the inversion of CO emissions is sensitive to the modeled OH abundance and the emissions of CH_4 and NMVOCs. According to the estimation of Müller et al. (2018), the magnitude of inversed CO emissions in China could differ by more than 40% when different levels of OH concentrations are used in the model. Thus, the much higher estimations of CO emissions in our inversion results may also be partly explained by the underestimation of CO chemical production or the overestimation of the CO sink.

4.3.1.4 $PM_{2.5}$

In terms of $PM_{2.5}$, the CAQIEI suggests~~is about 20%~~ higher emissions than ABaCAS, HTAPv3 and EDGARv6 by about 20%, and by 47.7% ~~higher~~ than MEIC on the national scale~~, suggesting that anthropogenic, biomass-burning, and/or fine-dust emissions may be underestimated~~. Larger discrepancies~~underestimations~~ mainly occur in the NE and NW regions, where CAQIEI is about 27.2–114.9% and 83.2–143.2% higher than the previous inventories. The differences in the estimated~~underestimated~~ $PM_{2.5}$ emissions may be related to the uncertainties~~underestimations~~ in the~~of~~ biomass-burning or anthropogenic sources in the NE region (Wu et al., 2020b), while in the NW region, ~~besides the possible underestimation of anthropogenic sources as seen from the other species,~~ the errors in the~~underestimated~~ fine-dust emissions may also contribute to the differences~~underestimation~~ in the estimated~~of~~ $PM_{2.5}$ emissions there. The differences in the estimated $PM_{2.5}$ emissions are relatively smaller in the NCP and SE regions, ranging from −18.9% to 20.4%, suggesting~~which shows~~ better agreement in the estimated $PM_{2.5}$ emissions over these two regions. ~~This confirms our inversion results and indicates lower uncertainty in the estimated $PM_{2.5}$ emissions over the NCP and SE regions.~~ In the SW region, CAQIEI is closer to HTAPv3 and EDGARv6, with their differences being about 6.3% and −9.5% respectively, and is higher than MEIC and ABaCAS by 54.2% and 28.6%, suggesting higher uncertainty in the estimated $PM_{2.5}$ emissions over there.~~.~~

4.3.1.5 PM_{10}

For PM_{10} emissions, it is difficult to directly compare CAQIEI with previous emission inventories since CAQIEI not only contains anthropogenic and biomass-burning emissions, but also coarse-dust emissions. As a result, the estimated emissions of PM_{10} by CAQIEI are substantially higher than those by previous inventories, especially over the NW, Central and NE regions (Fig. 11), which are the typical natural windblown dust-source regions in China (Zeng et al., 2020). Besides the naturally windblown dust of arid desert regions (Prospero et al., 2002), large amounts of coarse-dust emissions also stem from anthropogenic sources, including anthropogenic fugitive, combustion and industrial dust from urban sources (AFCID) (Philip et al., 2017), and anthropogenic windblown dust from human-disturbed soils due to changes in land-use practices, deforestation and agriculture (Tegen et al., 1996). Therefore, although the other regions are not typical natural windblown dust-source regions in China, there are still high levels of coarse-dust emissions from anthropogenic sources there (also called "urban dust"), which may be the main reason for ~~. t~~The large deviation in the estimated PM_{10} emissions~~differences~~ between CAQIEI and previous inventories~~ over these regions may reflect the underestimated and/or the unconsidered urban dust in previous emission inventories~~. On the one hand, a~~A~~lthough AFCID is included in MEIC, ABaCAS, HTAPv3 and EDGARv6, it is difficult for current bottom-up emission inventories to completely represent fugitive sources (Philip et al., 2017). On the other hand,~~In addition, the~~ anthropogenic windblown dust emissions ~~has not~~are beennot included in current bottom-up emission inventories, which is an important source of coarse dust in urban areas according to the estimations of Li et al. (2016) and another important contributor to ~~, which is an important source of coarse dust in urban areas according to the estimations of Li et al. (2016)~~the differences between CAQIEI and previous emission inventories.~~. Besides, similar to the situation with $PM_{2.5}$ emissions, anthropogenic and biomass-burning emissions may also be underestimated for PM_{10} emissions, which could partly explain the large differences between CAQIEI and previous inventories.~~

4.3.1.6 NMVOCs

For NMVOC emissions, since CAQIEI includes both anthropogenic and natural sources, its estimated NMVOC emissions are much higher than those estimated by previous emission inventories. After consideration of natural sources, the CAQIEI suggests close estimations of the NMVOC emissions~~agrees well~~ with the MEIC, HTAPv3 and CEDS inventories on the national scale, with their differences being about 1.5–12.5%~~, which validates our inversion results and the estimated NMNOV emissions for these inventories~~. The estimated NMVOC emission by ABaCAS and EDGARv6 ~~may have a negative bias in their estimated NMVOC emissions, which~~ are slightly lower than CAQIEI by 17.8% and 24.6%, respectively. On the regional scale, the ~~comparison of~~ CAQIEI ~~with previous inventories~~ suggests ~~that there may be~~ higher NMVOC emissions over the northern China (NCP, NE and NW),~~.~~ with ~~T~~the top-down estimated NMVOC emissions ~~are~~ about 30.4–81.4%, 27.3–72.1%, 79.3–116.8%, and 8.7–57.5% higher than those of the previous emission inventories~~ over these regions~~. ~~This suggests that NMVOC emissions may be underestimated in northern China, especially over the Central region.~~ In contrast, the CAQIEI suggests lower NMVOC emissions over the SE region~~ may be overestimated~~, with the estimated NMVOC emissions of CAQIEI being about 21.2–27.6% lower than those of MEIC, ABaCAS, HTAPv3 and CEDS. These results are consistent with the previous inversion results based on the satellite observations, which suggest higher NMVOC emissions over the NCP region and lower NMVOC emissions over the south China (Souri et al., 2020). Over the SW region, CAQIEI shows good agreement with MEIC, ABaCAS and CEDS, with CAQIEI being slightly lower than these inventories by 1.0–8.9%, but is lower than~~.~~ HTAPv3 and EDGARv6 by ~~may overestimate the NMVOC emissions over the SW region, with their results being~~ about 38.6% and 29.1%, respectively ~~higher than those of CAQIEI~~. Again, it should be noted that the comparisons of NMVOC emission are conducted on the basis of natural emissions estimated by CAMS and GFAS, and could be more sensitive to the used natural sources than other species considering the larger contributions of the natural source to the NMVOC emissions.

4.3.2 Seasonality

Figure 12 presents the monthly profiles of different air pollutants obtained from different emission inventories. Note that the natural sources have been added to the previous inventories to facilitate the comparisons. The results show that different emission inventories give similar monthly profiles of NO_x and CO emissions, with higher emissions during wintertime and lower emissions during summertime, which suggests relatively lower uncertainty in the estimated monthly profiles for these two species. For SO_2 emissions, CAQIEI yields stronger monthly variation than the other inventories, with a higher proportion from January to March and lower proportion during summertime. Due to the influences of dust emissions, the top-down estimated $PM_{2.5}$ and PM_{10} emissions show higher proportions than the other emission inventories during the spring season, especially for PM_{10}. However, the proportion of emissions during autumn and winter are lower than in the other inventories. The monthly profiles of NMVOC emissions are generally consistent, with higher emissions during summer due to the enhanced biogenic emissions. However, the profile of CAQIEI is flatter than the previous inventories, and suggests a higher proportion during springtime. In addition, the timings of peak values of NMVOC emissions are also different between CAQIEI and the previous inventories, with CAQIEI showing peak values during May–July but the other inventories suggesting peaks during June–August.

4.3.3 Emission changes during 2015–2018

The top-down estimated emission changes of different air pollutants during 2015–2018 were also compared with previous emission inventories~~, the results of which are shown in Fig. 13~~. Figure 13 shows the time series of the total emissions of different species from 2013 to 2020 obtained from the CAQIEI and other emission inventories. Comparisons of the emission changes over the regional scales are also presented in Fig. S10–S15. Before the comparison, we firstly analyze the trends of natural sources in China to investigate their influences on the emission changes of different species based on the CAMS emission inventory and GFAS. Note that we only consider the soil, biogenic and biomass-burning emissions for the natural sources; the trends of dust emissions in China are not analyzed, which may lead to uncertainty when comparing the emission changes of $PM_{2.5}$ and PM_{10}. As shown in Fig. S16~~7~~, the natural sources of NO_x and NMVOC emissions changed little during 2013–2018.~~, suggesting that the emission trends of these two species would be mainly driven by anthropogenic sources.~~ T~~T~~he other species had small decreasing trends from 2013 to 2018. However, considering the small contributions of natural sources to their emissions, these small trends would not significantly influence their emission trends. For the dust emissions, previous studies have indicated a declining trend in dust activity in China from 2001 to 2020 (Wu et al., 2022; Wang et al., 2021), due to weakened surface wind and increased vegetation cover and soil moisture. These results suggest that the emission trends in the CAQIEI would be mainly driven by the anthropogenic sources for the gaseous air pollutants based on the estimations of CAMS and GFAS, while ~~Thus,~~ its estimated emission trends of $PM_{2.5}$ and PM_{10}~~there~~ would be influenced by the declining trends in dust emissions in China, which should be noted when comparing the emission changes of $PM_{2.5}$ and PM_{10}.

As shown in Fig. 1~~4~~3, all the emission inventories agree that the NO_x, SO_2, CO, $PM_{2.5}$ and PM_{10} emissions in China were reduced from 2015 to 2018, except for the increases of CO emissions estimated by TCR-2, which confirms the effectiveness of the emission control policies implemented during the clean air action plans. Meanwhile, most emission inventories agree that SO_2 is the species with the largest emission reduction rates~~s~~, followed by $PM_{2.5}$, indicating better emission mitigation effects of these two species (Fig. 14). However, the CAQIEI suggested lower emission reduction rates than the other emission inventories for most species~~emission reduction rates estimated by CAQIEI are generally lower than those estimated by previous emission inventories~~, especially for NO_x, PM_{10} and NMVOCs (Fig. 14)~~, which suggests that the mitigation effects of the air quality control during 2015–2018 may be overestimated by previous inventories~~. T~~T~~he estimated emission reduction rate of NO_x obtained from CAQIEI is about −2.7%, which is lower than the values of MEIC (−9.7%), ABaCAS (−23.0%), HTAPv3 (−13.0%) and CEDS (−9.0%).~~.~~ As we discussed in Sect. 4.2.2.2, the small reductions of NO_x emission in CAQIEI would be related to the increased vehicle emissions and the undesirable mitigation effects of the industry control. In fact, these

factors have been considered in some bottom-up emission inventories, such as MEIC. The differences between our inversion results and previous inventories thus reflect uncertainty in the quantifications of the effects of these factors on the NO_x emissions due to the lack of sufficient statistics on mobile vehicle or other sectors. Our inversion results suggest larger adverse effects of these two factors on the reductions of NO_x emissions in China. According to Fig. S178, the differences between CAQIEI and these inventories mainly occur in the SE, SW, NW and Central regions, with the emission reduction rate estimated by CAQIEI being substantially lower than those estimated by previous inventories. In particular, CAQIEI suggests increases of NO_x emissions over the Central region, which is opposite to the previous emission inventories~~which was not captured by previous inventories~~. Better agreement is achieved over the NCP and NE regions, with the emission reduction rate estimated by CAQIEI being closer to those of MEIC, HTAPv3 and CEDS~~, suggesting lower uncertainty in the estimated NO_x emission reduction rate over these two regions~~. The NO_x emission reduction rates estimated by EDGARv6 (−3.3%) and TCR-2 (−1.7%) are closer to our results on the national scale, but they ~~underestimate the~~estimated lower NO_x emission reduction rate than our estimate over the NCP and NE regions.

Similarly, the emission reduction rate of PM_{10} obtained from CAQIEI (−10.8%) is ~~much~~ lower than those estimated by MEIC (−27.9%), ABaCAS (−33.0%) and HTAPv3 (−27.8%) on the national scale (Fig. 143). A lower PM_{10} emission reduction rate of CAQIEI than these inventories also exist~~s~~ in the different regions of China, except SW (Fig. S178). In particular, different from previous emission inventories, CAQIEI suggests that PM_{10} emissions may have actually increased over the Central region. Considering that dust emissions may have decreased from 2015 to 2018 owing to weakened dust events (Wang et al., 2021), the increase in PM_{10} emissions over the Central region may reflect the increases in anthropogenic sources. Meanwhile, we also found that CAQIEI estimated the emission reduction rate of PM_{10} to be smaller than that of $PM_{2.5}$. This is different from previous emission inventories, which show similar emission reduction rates for $PM_{2.5}$ and PM_{10}. Considering that PM_{10} emissions include $PM_{2.5}$ and PMC emissions, the lower emission reduction rate of PM_{10} than $PM_{2.5}$ in CAQIEI suggests that PMC emissions may have decreased slower than $PM_{2.5}$ emissions from 2015 to 2018~~, which was not captured by previous inventories~~.

In terms of NMVOCs, most previous inventories, including MEIC, EDGARv6 and CEDS, suggest a weak decrease in China, with the estimated rates of change in emissions ranging from −0.8% to −4.6%. The emission reduction rate estimated by ABaCAS is larger, reaching up to −14.2%. In contrast, the CAQIEI suggest~~indicate~~s an opposite emission change to these inventories, with estimated NMVOC emissions increasing by 26.6% from 2015 to 2018. HATPv3 also suggests an increase in NMVOC emissions, but with a much lower rate of increase (2.7%). Similar results could also be found on the regional scale (Fig. S178), especially over the NCP, NE and Central regions, where NMVOC emissions could have increased by 38.0%, 38.3% and 60.0%, respectively, according to the estimates of CAQIEI. As we discussed in Sect. 4.2.2.2, the increases of NMVOC emission estimated in CAQIEI may be related to the increased anthropogenic NMVOC emissions from the chemical industry, solvent use, and vehicles. Therefore, similar to the NO_x emissions, the differences between CAQIEI and previous inventories reflects the uncertainty in the quantifications of the impacts of these factors, and suggest larger adverse effects of these factors on the emission reductions of NMVOC emission than the previous inventories. ~~However, none of the previous emission inventories captured this increasing trend over these regions. Considering that biogenic NMVOC emissions changed little from 2015 to 2018 according to the estimates of the CAMS inventory, the increases of NMVOC emissions possibly arise from the increased anthropogenic sources. This is consistent with the estimate of Li et al. (2019c), who found persistent growth of anthropogenic NMVOC emissions in China from 1900 to 2017. However, the drivers of the increased NMVOC emissions still need to be investigated, considering the uncertainty in the estimated trends of biogenic NMVOC emissions. Different from the CAMS inventory, Li et al. (2020a) found an increasing trend in biogenic emissions in China from 2008 to 2018, especially over northern China, which can partly explain the increased NMVOC emissions in China. Therefore, more analysis is needed to better understand the potential drivers of the increased NMVOC emissions in China.~~

The differences in the estimated emission reduction rates between CAQIEI and previous inventories are relatively smaller for SO_2 and $PM_{2.5}$ emissions~~, suggesting lower uncertainty in the estimated emission reduction rates for these two species~~. The emission reduction rate of SO_2 estimated by CAQIEI is close to that estimated by MEIC and CEDS, ranging from −34.7% to −44.3%. ABaCAS and HTAPv3 estimate a larger emission reduction rate of about −58.5% and −53.7%, respectively. EDGARv6 and TCR-2 may ~~greatly~~ underestimate the reduction rate of SO_2, with estimates of only about −7.0% and −9.1%, respectively. This may be because EDGARv6 underestimates the FGD (flue-gas desulfurization devices) penetration or SO_2 removal efficiencies of FGD in China. On the regional scale (Fig. S1~~7~~8), the top-down estimated SO_2 emission reduction rate agrees reasonably with that of MEIC over the NCP, NE and SE regions, but these inventories estimate different SO_2 emission reduction rates over the SW, NW, and Central regions. The reduction rates estimated by MEIC over the SW and Central regions is higher than those given by CAQIEI, but lower over the NW region. The other emission inventories also give different emission reduction rates. ~~This~~ suggestings ~~there are~~ large~~high levels of~~ uncertainty in the estimated SO_2 emission reduction rates over these three regions.~~.~~ ~~For example, CEDS shows similar results to our estimate over the SW and Central regions, while the rate given by HTAPv3 is closer to MEIC. This suggests there are high levels of uncertainty in the estimated SO_2 emission reduction rates over these three regions.~~ In terms of $PM_{2.5}$, CAQIEI's estimated emission reduction rate agrees well with those of MEIC and HTAPv3 on the national scale, which is ~~suggesting that the emission reduction rate of $PM_{2.5}$ in China was~~ about 24–27% from 2015 to 2018. The emission reduction rate of $PM_{2.5}$ estimated by EDGARv6 are lower than our estimates and other inventories, which were ~~may underestimate the reduction rate of $PM_{2.5}$, at~~ about 9%. On the regional scale, our results show good consistency with ~~consistency with~~ MEIC and HTAPv3 over the NCP, NE, SE and SW regions, but they have large differences over the NW and SW regions~~, indicating higher uncertainty in the estimated reduction rate over western China~~.

Different from the other species, the CO emission reduction rate estimated by CAQIEI (−21.3%) is higher than in most of the previous inventories, including MEIC (−13.0%), ABaCAS (−11.6%), EDGARv6 (−4.7%), and CEDS (−11.7%), suggesting ~~that th~~larger~~e~~ mitigation effects on CO emissions ~~may be underestimated by these~~than other inventories. HTAPv3 agrees with our results, with an estimated emission reduction rate of about −22.0%. On the regional scale (Fig. S1~~7~~8), our result is consistent with MEIC over the NCP and SE regions, with estimated emission reduction rates for CO of around 24% and 15%, respectively, while in other regions the emission reduction rate estimated by CAQIEI is higher than that estimated by MEIC. The ~~The larger emission reduction rate for CO in our results is supported by HTAPv3 over the NE, SW, NW and Central regions, as well as by CEDS over the SW, NW and Central regions. This suggests that the emission reduction rates for CO may be underestimated by MEIC over these regions.~~ TCR-2 shows opposite changes in CO emissions compared with the other inventories insofar as it suggests increases of CO emissions over different regions of China. Since the emissions in TCR-2 are constrained by satellite observations, the differences between our results and those of TCR-2 highlight that the observations used to constrain the emissions may have a large influence on the estimated emission changes. In this case, the assimilation of surface observations (our study) is shown to be superior to the assimilation of satellite observations (TCR-2), as our results are more consistent with other bottom-up inventories.

4.4 Uncertainty estimation of CAQIEI

Finally, the uncertainty of the inversed emission inventory product is estimated in this section to facilitate users' understanding of the data's accuracy. Within the framework of EnKF, the analysis perturbation $\mathbf{X}^a$ estimated by using Eq. (3) could provide the information regarding the uncertainty of the inversed emission inventory. The Coefficient of variation (hereinafter, CV), defined as the standard deviation divided by the average, with a larger value denoting higher uncertainty, is calculated based on the analysis perturbation to measure the uncertainty of the inverse emission inventory. Based on this method, the uncertainty (CV) of the a posteriori emission was estimated as follows: 92.3% ($PM_{2.5}$), 88.8% (PM_{10}), 26.7%

(SO_2), 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.

5 Discussion and conclusion

A long-term, top-down emissions inventory of major air pollutants in China was developed and validated in this study by assimilating surface observations from CNEMC using the modified EnKF method and NAQPMS. It includes gridded emission maps of NO_x, SO_2, CO, primary $PM_{2.5}$, primary PM_{10}, and NMVOCs in China from 2013 to 2020, on a monthly basis, with a horizontal resolution of 15 km × 15 km. This new top-down emissions inventory, named CAQIEI, provides new insights into the air pollutant emissions and their changes in China during the country's two clean air action periods~~, which has not been reported by previous inventories~~. The estimated total emissions for the year 2015 in China are 25.2 Tg of NO_x, 17.8 Tg of SO_2, 465.4 Tg of CO, 15.0 Tg of $PM_{2.5}$, 40.1 Tg of PM_{10} and 46.0 Tg of NMVOCs. Comparisons of~~Comparisons of~~ CAQIEI with previous inventories, including MEIC, ABaCAS, HTAPv3, EDGARv6, CEDS and TCR-2, on the basis of the natural emissions obtained from CAMS and GFAS ~~, including MEIC, ABaCAS, HTAPv3, EDGARv6, CEDS and TCR-2,~~ showed reasonable agreement for the estimation of NO_x, SO_2 and NMVOC emissions in China~~, which confirms our inversion results and suggests lower uncertainty in the estimated total emissions in China for these species.~~ . The $PM_{2.5}$ emissions obtained from CAQIEI (13.2 Tg) are slightly higher than in the previous emission inventories (8.3–11.1 Tg)~~, suggesting possible underestimations of the anthropogenic, biomass-burning or fine-dust emissions in current estimations~~, while~~.~~ t~~T~~he CO emissions estimated by CAQIEI (426.8 Tg) are substantially higher than in previous inventories (120.7–237.7 Tg)~~.~~, However, the reasons for such a large gap are still not clear~~indicating that CO emissions may be greatly underestimated currently. Although previous model simulation and inversion studies generally support the underestimation of CO emissions in China, the reasons for such a large underestimation are still not clear~~, but might be attributable to both the underestimation of CO sources (e.g., anthropogenic, biomass-burning and chemical-production sources) according to previous model simulation and inversion studies (Bergamaschi et al., 2000; Miyazaki et al., 2012; Petron et al., 2002; Petron et al., 2004; Tang et al., 2013; Gaubert et al., 2020), ~~e.g., anthropogenic, biomass-burning and chemical-production sources,~~ and/or the overestimation of CO sinks in the model (Müller et al., 2018). In addition, comparisons with previous inversion studies suggest there are larger differences in the top-down estimated CO emissions based on surface and satellite observations. Our inversion results are consistent with previous inversions based on surface observations, but are much higher than those based on satellite observations, suggesting large uncertainty in inversion-estimated CO emissions in China. Therefore, more research is needed to better understand the reasons behind the negative biases in CO simulation, and to explain the differences between our results and those of previous inventories. Similar to situation with CO emissions, the PM_{10} emissions estimated by CAQIEI (37.7 Tg) are also substantially higher than in previous inventories (11.1–15.9 Tg). However, this will be mainly associated with the emissions of coarse dust, which were not included in previous inventories. The estimation of dust emissions in China is subject to high levels of uncertainty, with the estimated dust fluxes based on different dust emission schemes differing by several orders of magnitude (Zeng et al., 2020). Therefore, our inversion results could provide a reference for the magnitude of coarse-dust emissions in China, which could then help to reduce the large uncertainty in estimations of dust emissions in China.

Several potential important deficiencies in current emission estimations were also ~~revealed~~ indicated by CAQIEI on the regional scale. For example, the CAQIEI suggests substantially higher ~~there are significant negative biases in the estimated~~ air pollutant emissions than the previous emission inventories over the NW and Central regions~~, suggesting that the air pollutant emissions in western China may be greatly underestimated by current emission inventories~~. ~~As a result~~Thus, the~~significant~~ air pollutant issues may be more severe than we expected ~~neglected~~ over these two regions~~, which may have led to serious adverse impacts in terms of human health and ecosystems~~. Meanwhile, our inversion results suggest higher NMVOC emissions ~~are shown to be substantially underestimated~~ over the northern China but suggest lower NMVOC emissions ~~overestimated~~ in southern China, which is consistent with the previous inversion studies based on the satellite. China is now facing increasingly severe O_3 pollution and has an urgent need for a coordinated control of O_3 and $PM_{2.5}$. Our results ~~shed new light on the nature of~~may provide valuable information on the NMVOC emissions in China, which is important for a proper understanding of O_3 pollution and the development of effective control strategies nationally. Higher emissions~~Consistent negative biases~~ were also found~~identified~~ in the NE region ~~for the emissions of all species~~based on our inversion results. The NE region is a typical area for open-area biomass burning, with significant emissions from straw combustion (Wu et al., 2020b). The higher emissions estimated by our inversion result may indicate higher ~~underestimation of emissions there may reflect the underestimation of~~ biomass-burning emissions over there. This is consistent with recent estimations of biomass-burning emissions by Xu et al. (2023) and Wu et al. (2020b), who showed higher biomass-burning emissions in China than previous estimations, including those of GFEDv4.1s (https://www.globalfiredata.org/data.html), FINNv1.5 (https://www.acom.ucar.edu/Data/fire/), and GFASv1.2 (https://www.ecmwf.int/en/forecasts/dataset/global-fire-assimilation-system).

Based on CAQIEI, we further quantified the emission changes of different air pollutants in China during the two clean air action plans. The results confirmed the effectiveness of these campaigns on the mitigation of air pollutant emissions in China, with estimated emission reductions of 15.1% for NO_x, 54.5% for SO_2, 35.7% for CO, 44.4% for $PM_{2.5}$, and 33.6% for PM_{10} from 2015 to 2020. In contrast, NMVOC emissions increased by 21.0% from 2015 to 2020. Comparisons of the estimated emission reduction rates during the two clean air action plans suggested that emission reductions were larger during the 2018–2020 ~~action plan~~ than during ~~the~~ 2015~~3~~–2017 ~~action plan~~. The estimated rates of change in emissions were 5.9% for NO_x, −23.8% for SO_2, −9.8% for CO, −14.5% for $PM_{2.5}$, −7.2% for PM_{10}, and 27.6% for NMVOCs during ~~the~~ 2015~~3~~–2017 ~~action plan~~, which were smaller than the −12.1% for NO_x, −23.5% for SO_2, −18.3% for CO, −26.6% for $PM_{2.5}$, −25.5% for PM_{10}, and −4.5% for NMVOCs during ~~the~~ 2018–2020 ~~action plan~~. On the one hand, this is due to the fact that more sectors were controlled during the 2018–2020 action plan. Besides the industrial and power sectors, which were the main points of control in the 2013–2017 action plan, the residential sector, transportation sector, and non-point sources like blowing-dust emissions, were also strengthened in the 2018–2020 action plan. Consequently, the emission reduction rates of CO, $PM_{2.5}$ and PM_{10} during ~~the~~ 2018–2020 ~~action plan~~ were higher than those during the 2015~~8~~–2017 when the 2013–2017~~20~~ action plan was implemented. However, the reduction of SO_2 emissions was similar during the two action plan periods. This is because most SO_2 emissions stem from the industrial sector and power plants, which together contribute about 77% of all emissions (Zheng et al., 2018). Thus, the additional control of other sectors in the 2018–2020 action plan may not have ~~significant~~significantly impacted the mitigation of SO_2 emissions. On the other hand, strict emission controls were implemented or strengthened in more areas of China during the 2018–2020 action plans. For example, the inversion results indicated that there were obvious increases of SO_2, NO_x, $PM_{2.5}$, PM_{10} and NMVOC emissions during ~~the~~ 2015~~3~~–2017 ~~action plan~~ over the Central region, especially in the Fen~~g~~wei Plain area, where the emission controls were relatively weak during the 2013–2017 action plan. However, all species showed obvious emission reductions almost the whole China~~over the Fengwei Plain area, and almost the whole of China,~~ during the 2018–2020 action plan.

The estimated rates of change in emissions during 2015–2018 were also compared with those estimated by previous emission inventories. Although both CAQIEI and previous inventories showed declines of air pollutant emissions in China, the emission reduction rates estimated by CAQIEI were generally smaller than those estimated by previous inventories,

especially for NO_x, PM_{10} and NMVOCs, suggesting a smallerthat the mitigation effects of the air pollution control measures than the previous emission inventories suggestedmay be overestimated currently. In particular, China's NMVOC emissions were shown to have increased by 26.6% from 2015 to 2018, especially over NCP (38.0%), NE (38.3%) and Central (60.0%).~~, which was not captured by all previous inventories. The potential overestimation of the NO_x emission reduction rate was mainly a feature of the SE, SW, NW and Central regions; however, over the NCP and NE regions, our results agreed well with those of MEIC, HTAPv3 and CEDS, suggesting smaller uncertainty in the estimated reduction of NO_x emissions over these two regions. The potential overestimation of the PM_{10} emission reduction rate was a feature in most regions of China, possibly related to the smaller reduction rate of the country's PMC emissions.~~ CO was found to be an exception insofar as the emission reduction rate estimated by CAQIEI was larger than that of most previous emission inventories, ~~suggesting that the mitigation effects of CO emission control measures may be underestimated,~~ except in the NCP region. The estimated emission reduction rates of SO_2 and $PM_{2.5}$ were relatively closer to those of previous inventories, suggesting better consistency~~relatively lower uncertainty~~ in the estimated ~~pace of~~ emission reduction for these two species.

Overall, the inversion inventory developed in this study ~~sheds new light~~could provide us with value information on the complex variations of air pollutant emissions in China during its two recent clean air action periods, which could ~~significantly~~help improve our understanding of air pollutant emissions and related changes in air quality in China. For example, the increases of O_3 and nitrate concentrations may be associated with the undesirable emission reduction effects of the 2013–2017 action plans. The estimated lower~~possible overestimation of the~~ NO_x emission reduction rate by ~~previous inventories~~CAQIEI may also help explain the weak responses of nitrogen deposition fluxes to the clean air action plans. Meanwhile, this top-down emissions inventory can be used to supply the input data for CTMs or server as a comparable reference for future inversion studies based on other methods or observation data, which is expected to improve the performance of model simulations and air quality forecasts, and facilitate the development of inversion method.

6 Limitations

However, due to the complexity of the emission estimation, it is inevitable that there are some limitations in our inversion results. Here We summarise some issues that might affect the quality of the CAQIEI which were known at the time of publication to assist the potential users in properly using this data products. ~~However, there are some limitations to our inventory that potential users should be aware of.~~

(1) ~~Firstly,~~ Tthe changes in the number of observation sites ~~may have~~would induce~~d~~ spurious emission trends during 2013–2014, especially over western China, although the influence of the number of observation sites is smaller over the NCP and SE regions because of their higher density of observation sites. Therefore, it is recommended that not to use the emissions in 2013 and 2014 when analyzing the emission trends in China. This limitation makes it difficult to estimate the overall emission control effects of 2013 – 2017 action plan. Consequently, the emission change rate during the 2015–2017 were sampled in this study to represent the emission control effects of the 2013–2017 action plan, but it may not necessarily reflect the overall reduction rate of the action plan for the entire period. In addition, although the number of observation sites has become stable since 2015, the limited number of observation sites makes it difficult to fully constrain China's air pollutant emissions, especially for the natural sources considering that the majority of the observation sites are located in the urban areas.~~with respect to natural sources in remote areas~~ Therefore, the uncertainty in the estimated emissions over the remote areas are expected to be higher than those over the urban areas, especially for the species with large amount of natural emission, such as PM and NMVOC. ~~-~~For example, the coarse-dust emissions over western China are expected to be underestimated by CAQIEI because of the limited availability of observation sites.~~-~~ Therefore, adding observations there will help improve the accuracy of the inversion estimates.

(2) ~~Secondly,~~ The natural and anthropogenic emissions are not differentiated in our inversion method, leading to higher emissions of PM_{10} and NMVOCs than in other emission inventories. This also hinders the comparisons of our inversion results with the previous inventories. Therefore, potential readers should be aware of that the current comparisons of our inversion results and previous inventories are on the basis of the natural emissions estimated by CAMS and GFAS, which does not necessarily indicate large uncertainties in anthropogenic sources within the bottom-up inventories. The impacts are expected to be smaller for the NO_x, SO_2 and CO due to the small contributions of natural sources to their emission, but would be larger for NMVOC and PM which has large amount of natural emission.~~Consequently, the estimated changes in emissions of different air pollutants are also influenced by natural emissions, which should be considered in the comparisons of our inversion results with those of previous emission inventories.~~ Assimilation of isotope data, speciated $PM_{2.5}$ and NMVOC observations may help- differentiate the natural and anthropogenic emissions, and address this problem in future.

(3) The NMVOC emissions may have larger uncertainty than the other species. On the one hand, a significant amount of NMVOC emission would originate from suburban or rural regions. Therefore, although the O_3 observations at the urban sites could provide information on the NMVOC emissions over the suburban or rural areas according to covariance estimated by the ensemble simulation, the NMVOC emissions may not be fully constrained due to the lack of observation sites over the suburban or rural areas. On the other hand, due to the lack of long-term NMVOC observations, the NMVOC emissions were constrained by the O_3 concentrations in this study. Although the feasibility of this approach has been demonstrated by previous inversion studies, the nonlinear NO_x-VOC-O_3 interactions could inevitably introduces greater uncertainty into the inversion of NMVOC than other species. Therefore, more attention should be paid while using the inversion results of NMVOC, and more robust analysis of the effects of nonlinear NO_x-VOC-O_3 interactions and the number of observation sites should be performed in future to better illustrate the feasibility of assimilating O_3 to constrain the NMVOC emissions.

(4) ~~Thirdly,~~ ~~T~~the errors in the meteorological simulation and the CTMs were not considered in the emission inversions, which would lead~~ing~~ to uncertainty in our estimated emissions. However, it is difficult to consider the meteorological and model errors in the assimilation process. A multi-model inversion framework, for example that of Miyazaki et al. (2020a), may help alleviate the influences of model errors on emission inversions in future. ~~Meanwhile,~~ Using other models (e.g., WRF-Chem, CMAQ) to validate our inversion inventory could also help us assess the impacts of model uncertainty on the emission inversions. Meanwhile, because of the many uses that require a rapid update of emissions, it may be time to organize an intercomparison study focused on the emission inversions.

~~7~~6 data availability

The CAQIEI inventory can be freely download at https://doi.org/10.57760/sciencedb.13151 (Kong et al., 2023), which includes monthly grid maps of the air pollutant emissions from 2013 to 2020. The contained species include NO_x, SO_2, CO, primary $PM_{2.5}$, primary PM_{10} and NMVOC. The horizontal resolution is 15km. There are totally 8 Network Common Data Form files (NetCDF), which were named by the date and contains the monthly emissions of different air pollutants in China in each year. The description of the content of each NetCDF file and some important notes when using this dataset are also available in README.txt on the website.

Tables

Table 1. Corresponding relationships between the chemical observations and adjusted emissions

Species	Description	Observations used for inversions of this species
BC	Black carbon	$PM_{2.5}$
OC	Organic carbon	$PM_{2.5}$
PMF	Fine-mode unspeciated aerosol	$PM_{2.5}$
PMC	Coarse-mode unspeciated aerosol	$PM_{10} - PM_{2.5}$
NO_x	Nitrogen oxide	NO_2
SO_2	Sulfur dioxide	SO_2
CO	Carbon monoxide	CO
NMVOCs	Non-methane volatile organic compounds	MDA8h O_3

Table 2. Evaluation statistics of the *a posteriori* (*a priori*) model simulation for different species

	$PM_{2.5}$ ($\mu g/m^3$)				PM_{10} ($\mu g/m^3$)			
	R	MBE	NMB (%)	RMSE	R	MBE	NMB (%)	RMSE
Hourly	0.77 (0.53)	2.1 (13.3)	4.5 (28.6)	32.4 (55.6)	0.72 (0.44)	−3.7 (−11.5)	−4.6 (−14.3)	53.1 (74.4)
Daily	0.89 (0.61)	2.1 (13.3)	4.4 (28.4)	20.0 (46.3)	0.88 (0.51)	−3.7 (−11.2)	−4.6 (−14.1)	31.6 (62.2)
Monthly	0.94 (0.68)	2.1 (13.3)	4.5 (28.3)	11.7 (32.5)	0.90 (0.56)	−3.6 (−11.3)	−4.5 (−14.1)	21.2 (44.1)
Yearly	0.94 (0.62)	2.2 (11.9)	4.4 (24.3)	9.1 (27.7)	0.89 (0.52)	−3.8 (−13.4)	−4.6 (−16.1)	18.5 (38.7)
	SO_2 ($\mu g/m^3$)				NO_2 ($\mu g/m^3$)			
	R	MBE	NMB (%)	RMSE	R	MBE	NMB (%)	RMSE
Hourly	0.64 (0.16)	−1.8 (19.0)	−9.1 (93.8)	24.9 (58.7)	0.67 (0.45)	−1.2 (−0.9)	−3.9 (−2.7)	19.9 (25.5)
Daily	0.80 (0.20)	−1.8 (19.0)	−9.2 (94.5)	16.0 (51.4)	0.80 (0.51)	−1.2 (−0.8)	−3.7 (−2.6)	12.8 (20.1)
Monthly	0.85 (0.20)	−1.9 (18.9)	−9.3 (93.1)	12.4 (45.8)	0.84 (0.57)	−1.2 (−0.8)	−3.8 (−2.6)	9.4 (15.6)
Yearly	0.83 (0.18)	−2.4 (17.0)	−10.8 (75.9)	11.6 (42.4)	0.82 (0.63)	−1.3 (−1.6)	−3.9 (−5.0)	8.1 (13.0)
	CO (mg/m^3)				O_3 ($\mu g/m^3$)			
	R	MBE	NMB (%)	RMSE	R	MBE	NMB (%)	RMSE
Hourly	0.69 (0.38)	−0.1 (−0.4)	−8.8 (−45.6)	0.6 (0.8)	0.71 (0.51)	5.6 (−8.4)	9.5 (−14.0)	34.9 (41.6)
Daily	0.81 (0.42)	−0.1 (−0.4)	−8.6 (−45.5)	0.4 (0.7)	0.71 (0.40)	5.7 (−8.4)	9.5 (−14.1)	26.1 (33.8)
Monthly	0.83 (0.42)	−0.1 (−0.4)	−8.7 (−45.7)	0.3 (0.7)	0.76 (0.47)	5.6 (−8.4)	9.4 (−14.1)	19.6 (26.0)
Yearly	0.82 (0.27)	−0.1 (−0.5)	−9.0 (−47.6)	0.3 (0.7)	0.53 (0.11)	5.1 (−7.8)	8.7 (−13.4)	14.2 (20.5)

Table 3. Inversion-estimated emissions (Tg/yr) of different species in China as well as the six regions for year 2015

	China	NCP	SE	NE	SW	NW	Central
NO_x	25.2	5.1	7.1	4.5	4.2	1.2	3.2
SO_2	17.8	3.5	3.3	4.0	2.6	0.8	3.6
CO	465.4	82.2	106.7	78.7	82.8	32.6	82.3
$PM_{2.5}$	14.9	2.7	3.3	3.1	2.9	1.2	1.9
PM_{10}	40.1	8.7	7.5	8.2	5.5	4.1	6.2
NMVOC	46.0	9.0	13.7	8.5	7.8	2.7	4.2

Table 4. The calculated annual trends of $PM_{2.5}$ and PM_{10} emissions in China based on CAQIEI

	$PM_{2.5}$ (Tg/year)			PM_{10} (Tg/year)		
	2015–2020	2015–2017	2018–2020	2015–2020	2015–2017	2018–2020
China	−1.4*	−1.1	−1.5	−2.6*	−1.4	−4.6
NCP	−0.32*	−0.30	−0.32	−0.64*	−0.88	−0.99
SE	−0.32*	−0.21	−0.44	−0.52*	−0.48	−0.84
NE	−0.24*	−0.25	−0.11	−0.52*	−0.22	−0.73
SW	−0.21*	−0.26	−0.20	−0.40*	−0.26	−0.56
NW	−0.09	−0.08	−0.12	−0.20*	−0.32	−0.32
Central	−0.15	0.01	−0.32	−0.27	−0.32	−1.14

* Trend is significant at the 0.05 significance level

Table 5. The calculated annual trends of the four gaseous emissions in China based on CAQIEI

	SO_2 (Tg/year)			CO (Tg/year)		
	2015–2020	2015–2017	2018–2020	2015–2020	2015–2017	2018–2020
China	−2.1*	−2.1	−1.3	−36.0*	−22.8	−33.5
NCP	−0.57*	−0.69	−0.21	−8.4*	−4.30	−7.23
SE	−0.34*	−0.39	−0.20	−6.1*	−3.54	−8.37
NE	−0.44*	−0.44	−0.21	−6.2*	−1.74	−3.91
SW	−0.22*	−0.27	−0.17	−3.8*	−2.36	−4.54
NW	−0.08*	−0.08	−0.08	−3.0*	−0.73	−2.95
Central	−0.46*	−0.25	−0.40	−8.7*	−10.14	−6.55
	NO_x (Tg/year)			NMVOC (Tg/year)		
	2015–2020	2015–2017	2018–2020	2015–2020	2015–2017	2018–2020
China	−0.67	0.74	−1.6	1.9	6.3	−1.3
NCP	−0.32	0.05	−0.40	0.66	1.37	−0.42
SE	−0.22	0.18	−0.49	0.50	1.73	−0.24
NE	−0.17	0.03	−0.19	0.03	0.79	−0.49
SW	−0.06	0.10	−0.26	0.23*	0.43	0.03
NW	−0.03	0.11	−0.06	0.10	0.69	−0.27
Central	0.04	0.28	−0.16	0.55*	1.33	0.09

* Trend is significant at the 0.05 significance level

Table 6 The top-down estimated CO emissions in China from previous inventories

Reference	Region	Period	Method	Assimilated observation	*A priori* CO emission (kt/day)	*A posteriori* CO emission (kt/day)
Feng et al. (2020)	China Mainland	December 2013	EnKF with CMAQ model	Surface observation	586.4	1678.0
		December 2017			499.3	1388.1
	NCP	December 2013			143.9	394.3
		December 2017			120.5	340.7
Muller et al. (2018)	China	2013	4DVar with IMAGES model	IASI CO observation with different constraints on OH levels	454.8	367.1–553.4
Gaubert et al. (2020)	Central China	May 2016	DART/CAM-CHEM	MOPITT CO observation	193.6	220.3
	North China				93.5	163.6
Jiang et al. (2017)	East China	2013	4DVar with GEOS-Chem	MOPITT CO observation	564.5	439.5–484.4
		2014				430.4–481.1
		2015				397.5–439.7
Zheng et al. (2019)	China	2010–2017 average	Bayesian inversion	MOPITT CO, OMI HCHO, and GOSAT CH_4 observation	-	444.4

Figures

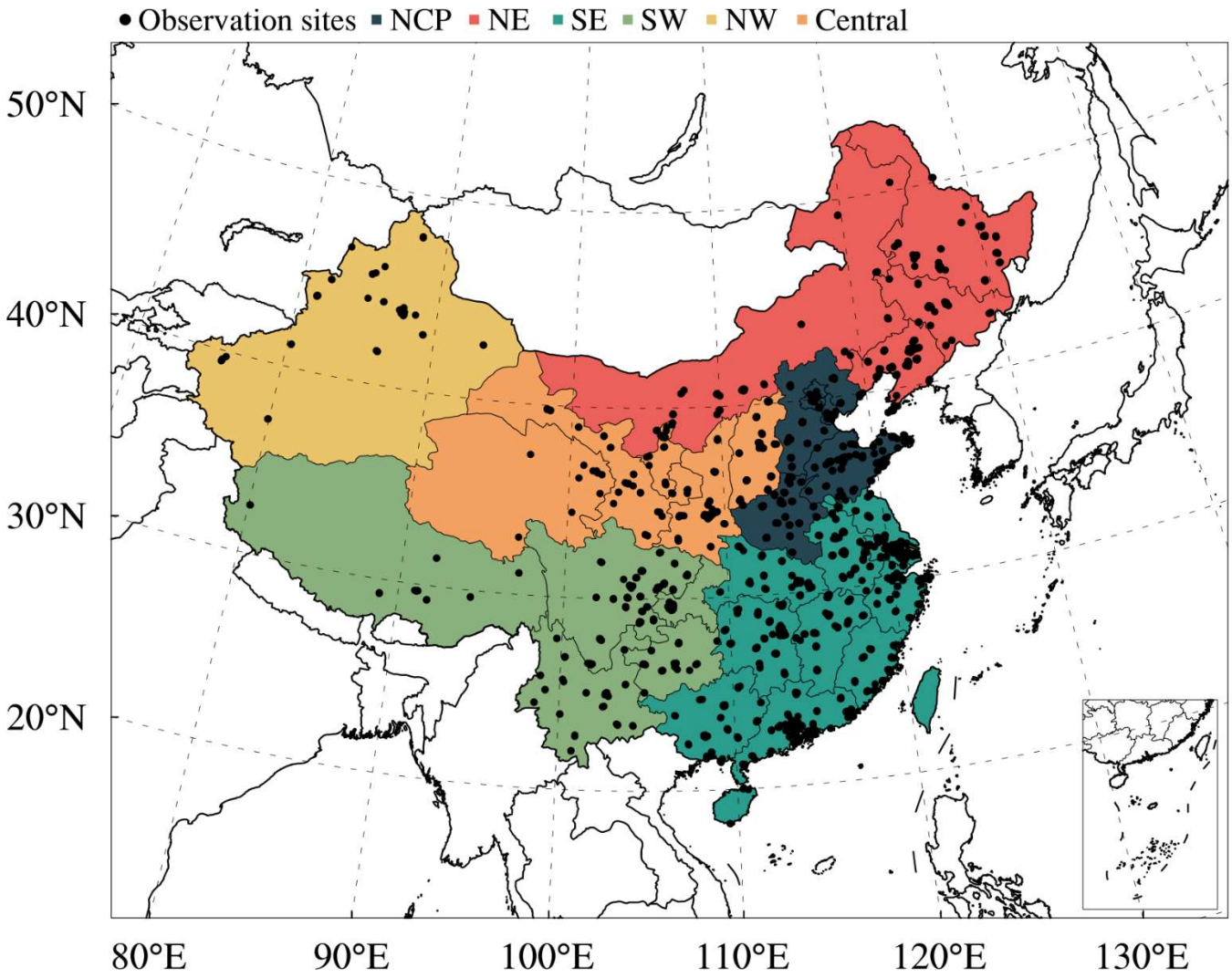

Figure 1: Modeling domain of the ensemble simulation overlaid with the distributions of observation sites from CNEMC. Different colors denote the different regions in mainland China—namely, the North China Plain (NCP), Northeast China (NE), Southwest China (SW), Southeast China (SE), Northwest China (NW) and Central China (Central).

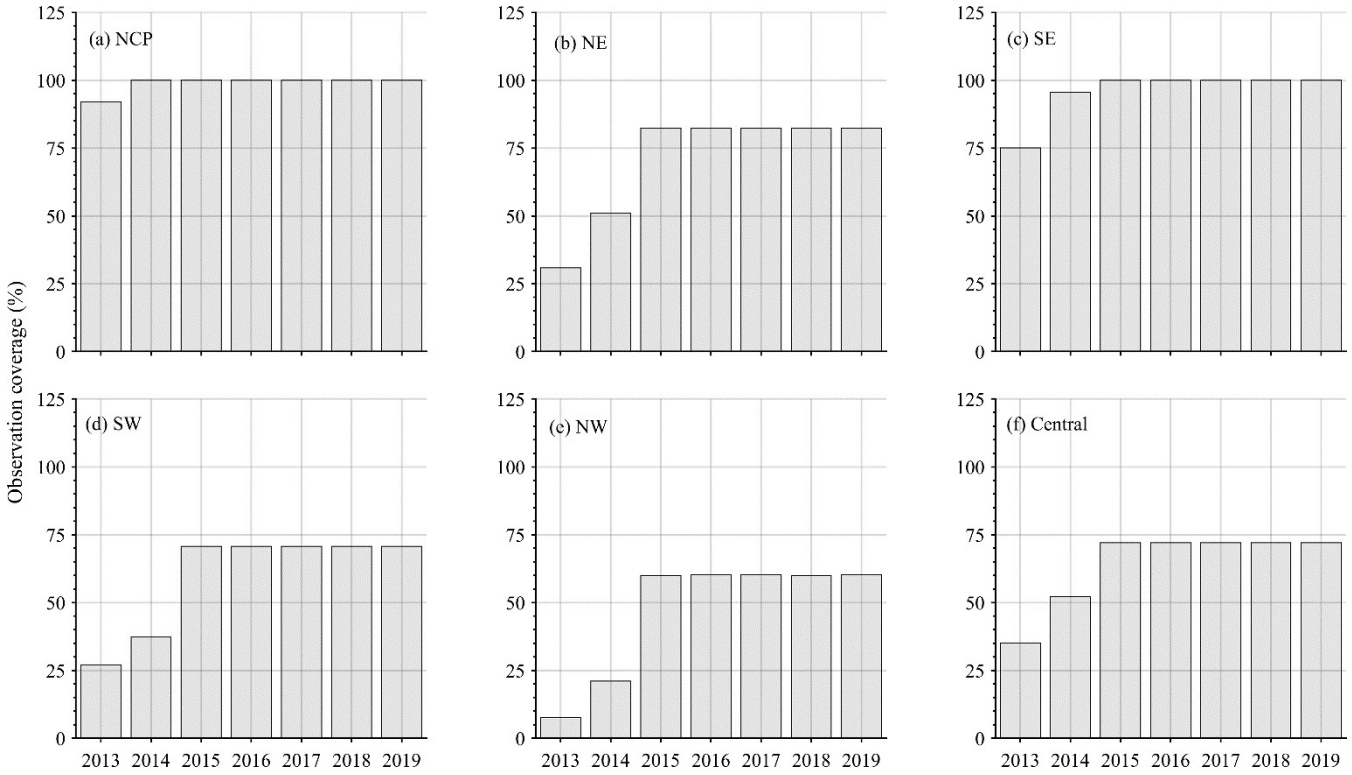

Figure 2: Time series of the observational coverage from 2013 to 2020 over different regions of China.

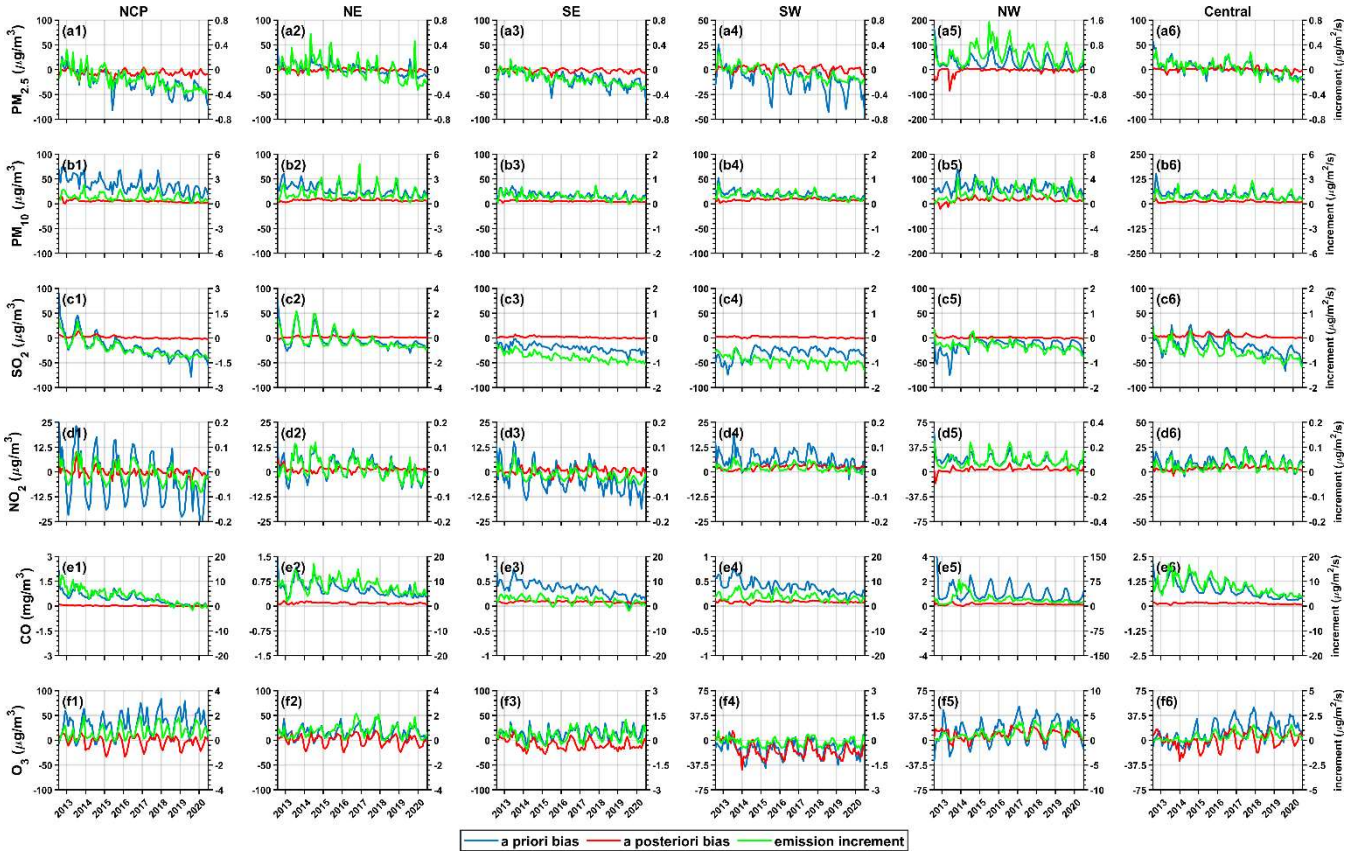

Figure 3: Time series of the *a priori* bias (blue lines), the *a posteriori* bias (red lines), and the emission increment (green lines) from 2013 to 2020 for different species over the six regions of China.

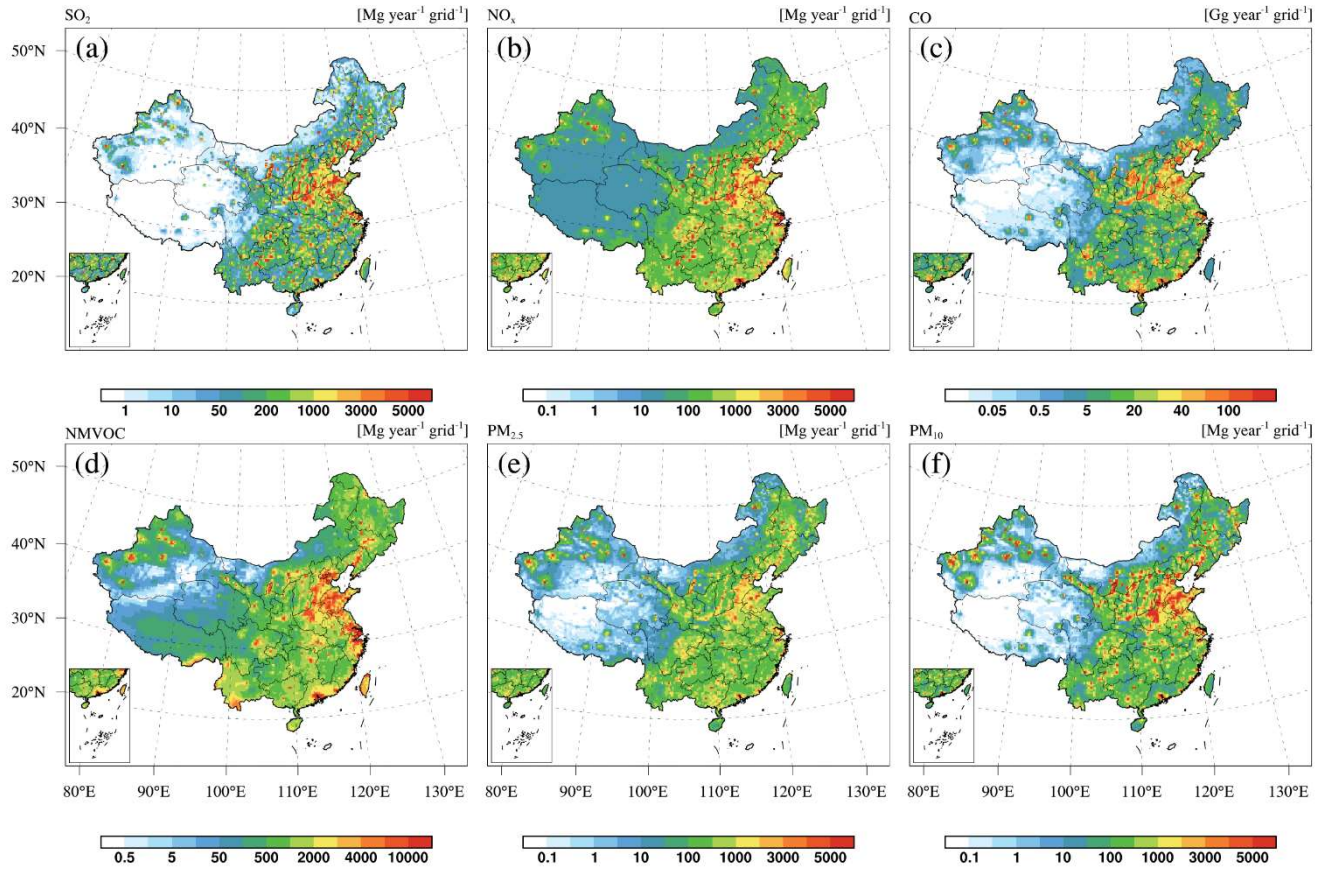

Figure 4: Spatial distributions of the emissions of (a) SO_2, (b) NO_x, (c) CO, (d) NMVOCs, (e) $PM_{2.5}$, and (f) PM_{10} in 2015 obtained from CAQIEI.

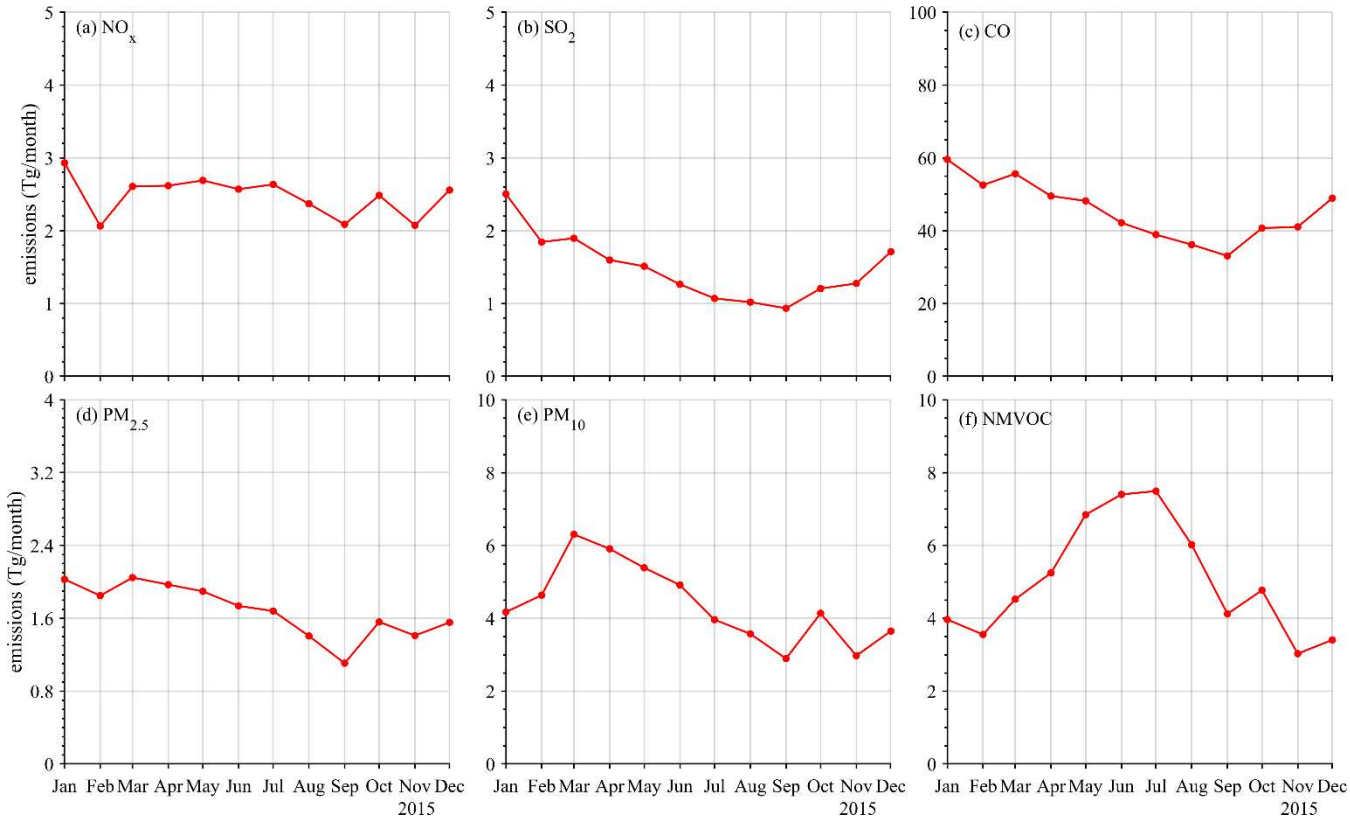

Figure 5: Monthly series of emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs in 2015 obtained from CAQIEI.

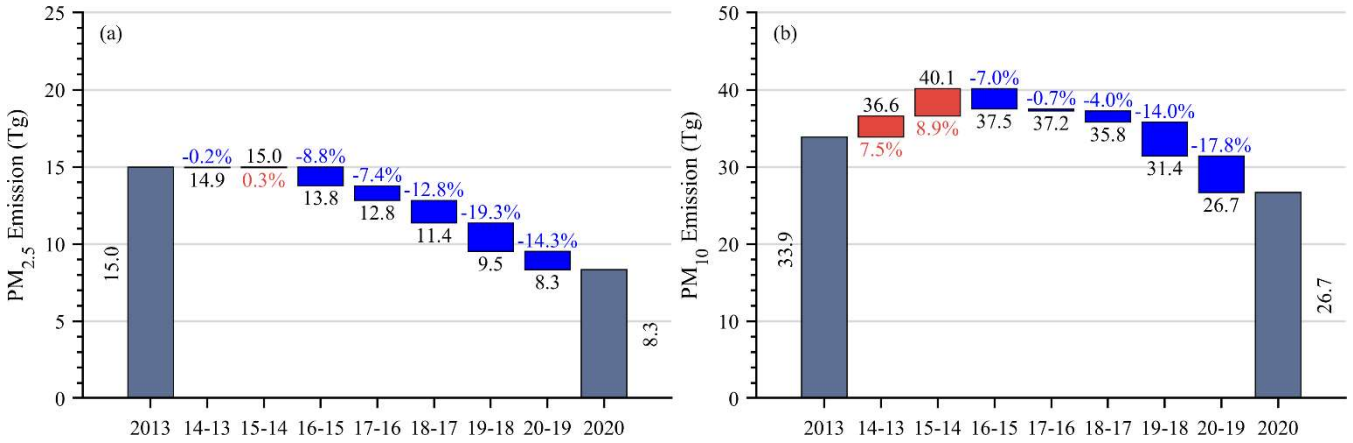

Figure 6: Emission changes in (a) $PM_{2.5}$ and (b) PM_{10} obtained from CAQIEI from 2013 to 2020.

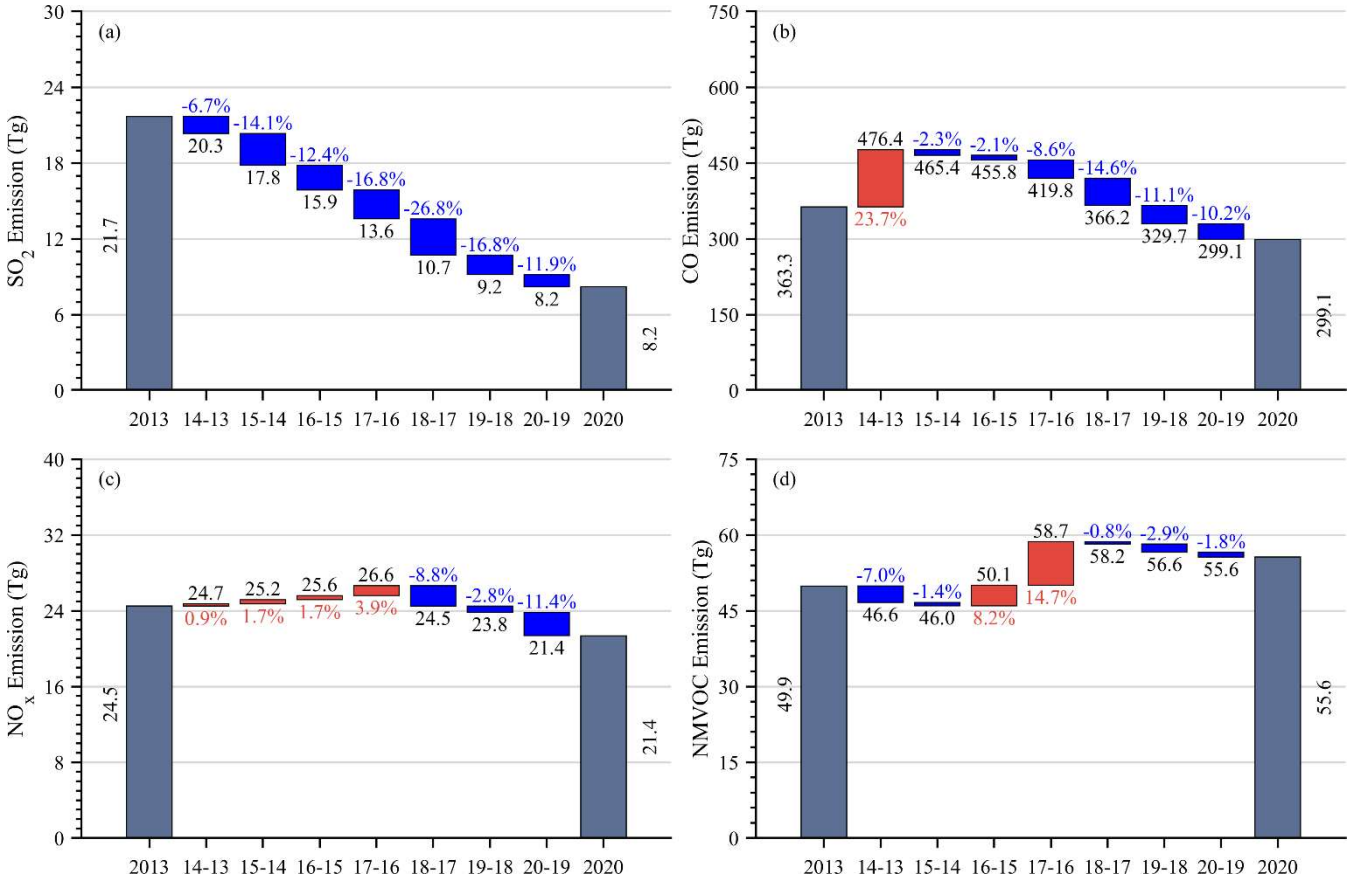

Figure 7: Emission changes in (a) SO_2, (b) CO, (c) NO_x, and (d) NMVOCs obtained from CAQIEI from 2013 to 2020.

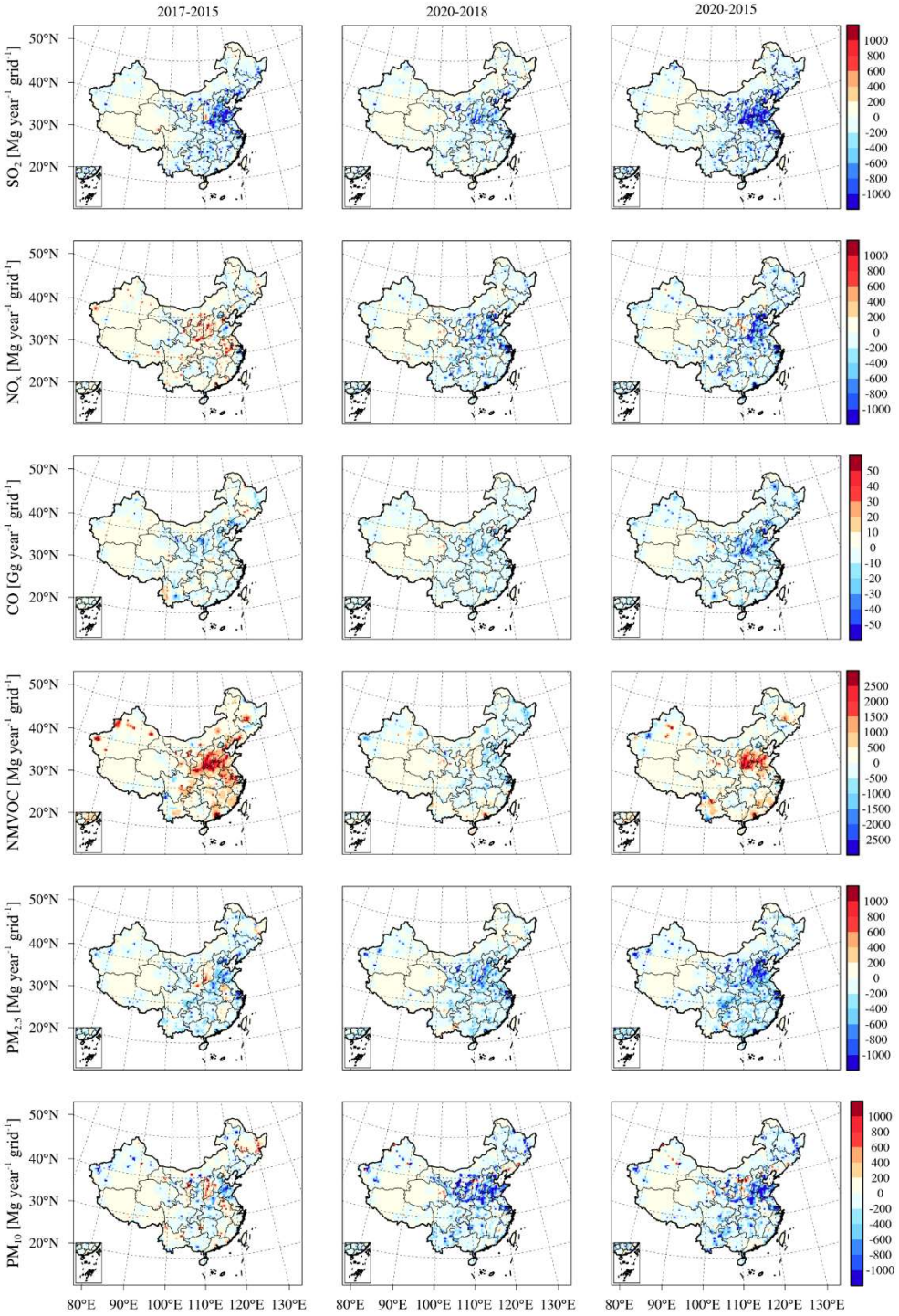

Figure 8: Spatial distributions of the emission changes of different species during 2015–2017 (left panels), 2018–2020 (middle panels), and 2015–2020 (right panels) obtained from CAQIEI from 2013 to 2020.

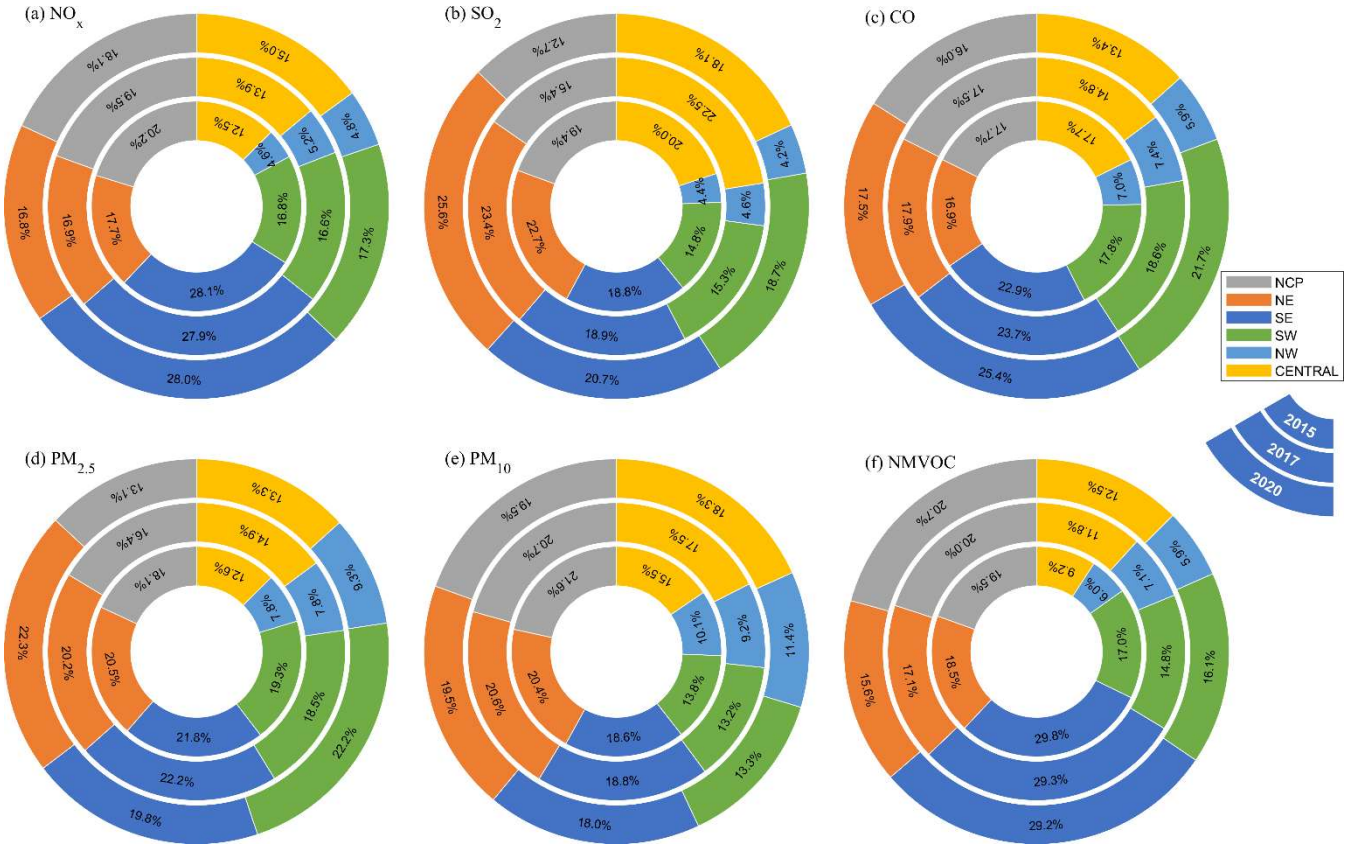

Figure 9: Emission distributions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, € PM_{10}, and (f) NMVOCs among different regions in China obtained from CAQIEI in 2015, 2017 and 2020.

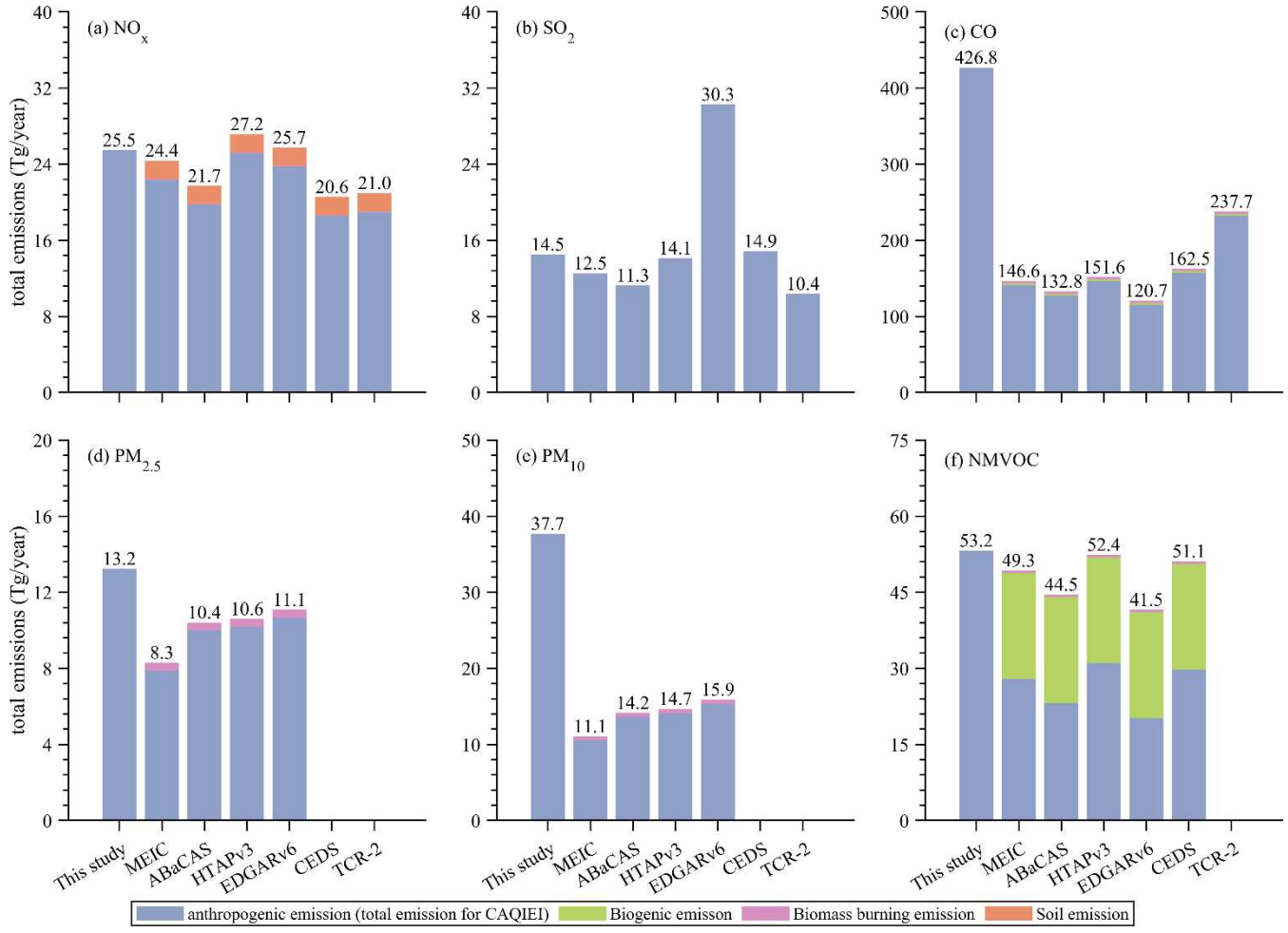

Figure 10: Comparisons of the averaged emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over China from 2015 to 2018 between CAQIEI and previous inventories added with natural sources.

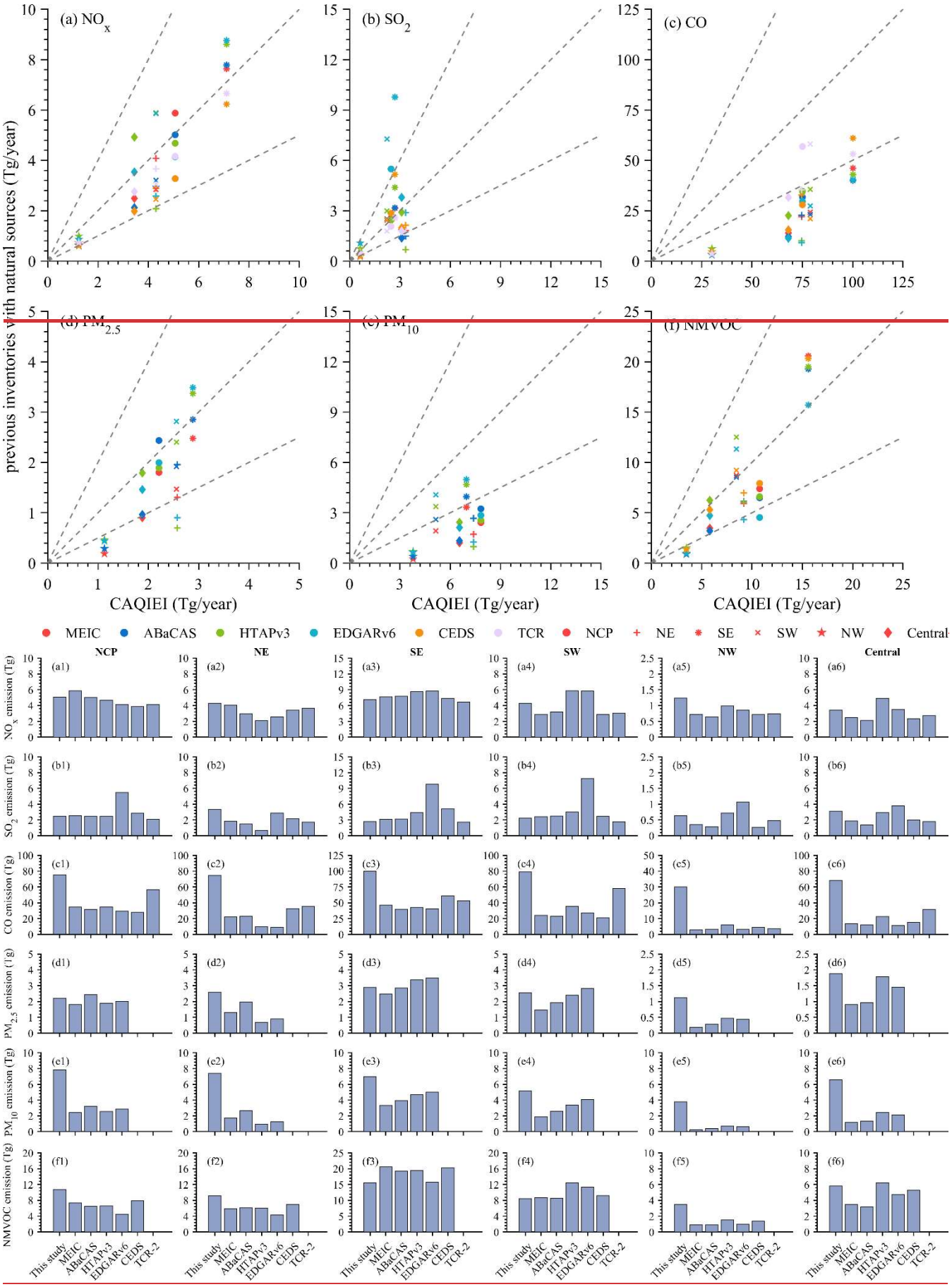

Figure 11: Comparisons of the averaged emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over different regions in China from 2015 to 2018 between CAQIEI and previous inventories added with natural sources.

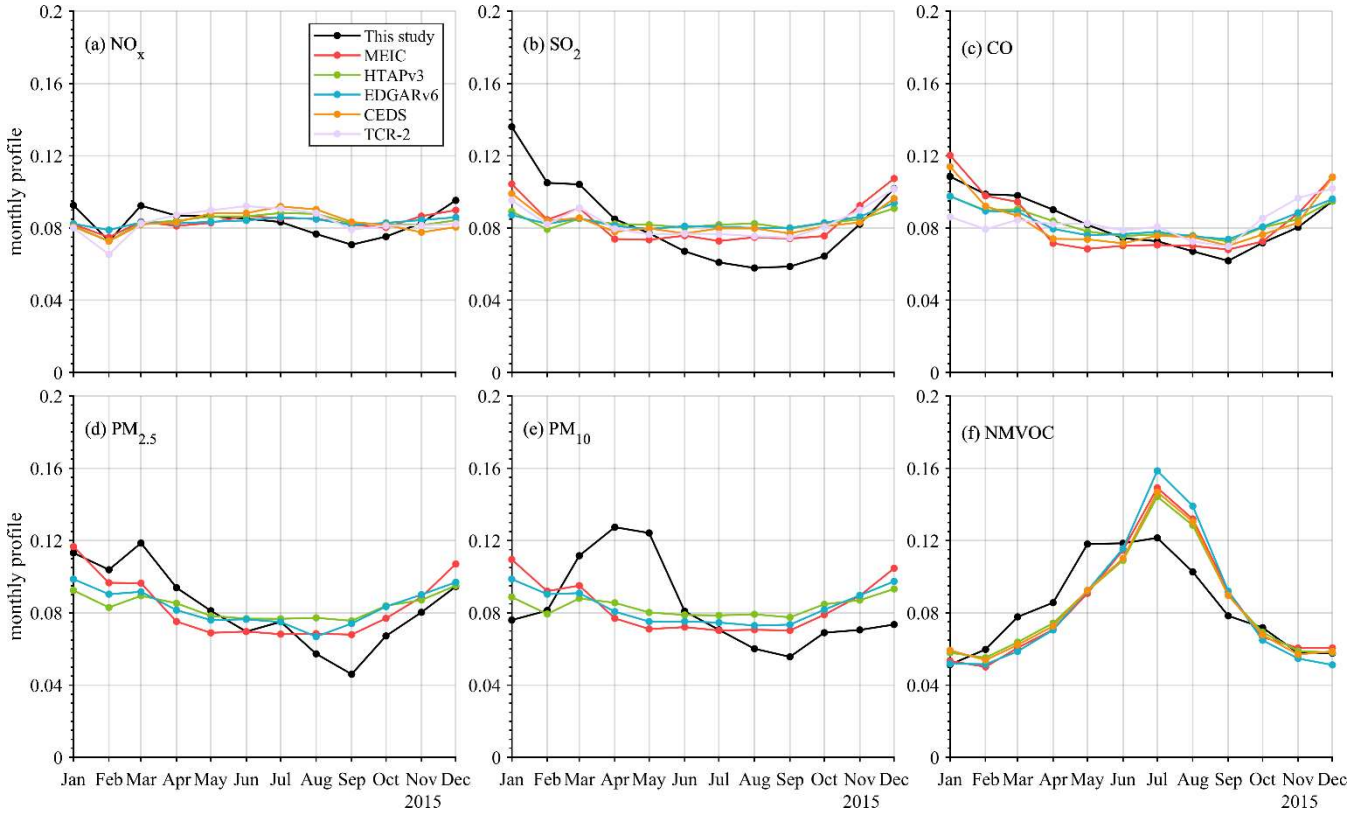

Figure 12: Comparisons of the monthly profiles of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over China averaged from~~in~~ 2015 to 2018 between CAQIEI and previous inventories added with natural sources.

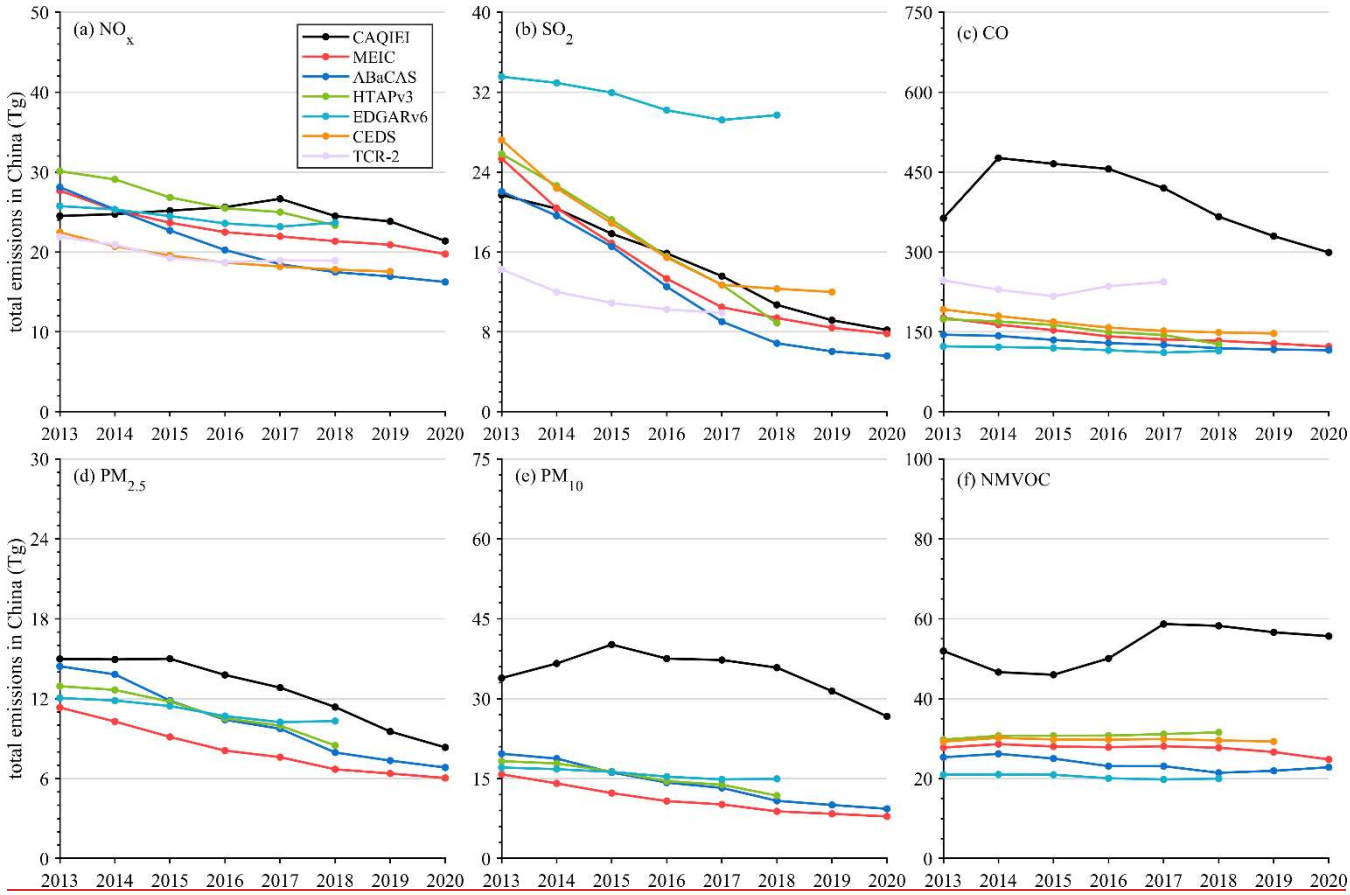

Figure 13: Time series of annual emissions of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10} and (f) NMVOC over China from 2013 to 2020 obtained from CAQIEI and previous inventories. Note that the natural sources were not included in the previous inventories in this figure.

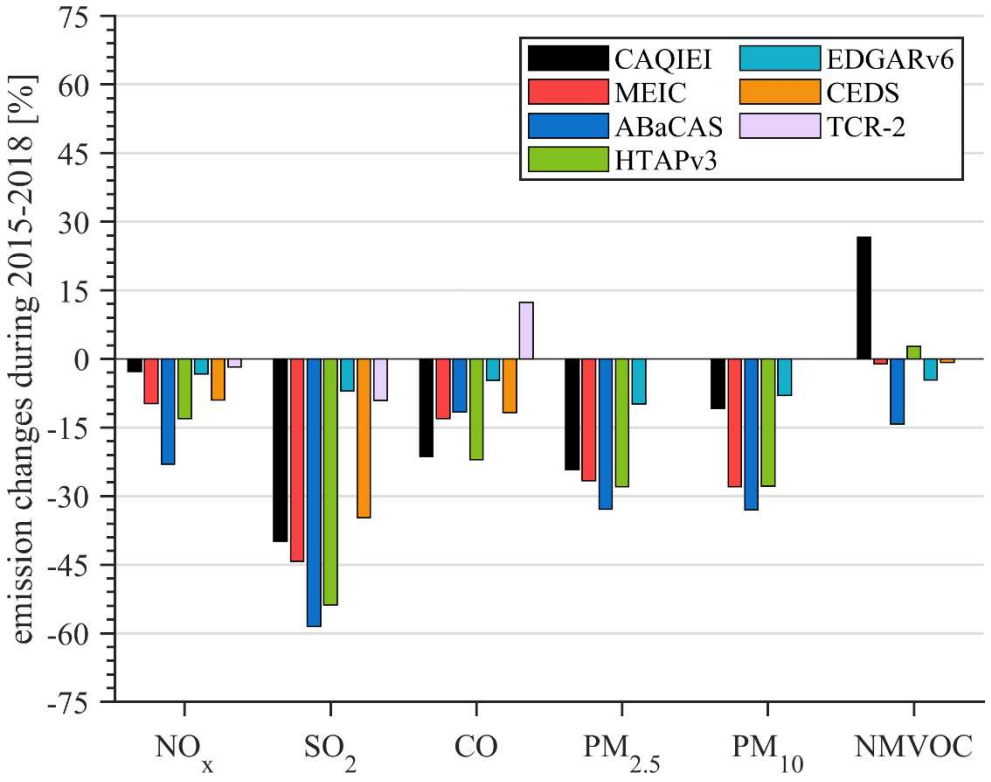

Figure 143: Comparisons of the calculated emission changes of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over China from 2015 to 2018 between CAQIEI and previous inventories.

Author contributions

X.T., Z.W., and J.Z. conceived and designed the project; L.K., H.W., X.T., and L.W. established the data assimilation system; Q.W. and L.K. performed the meteorology simulations; L.K., H.C., and J.L. conducted the ensemble simulation with the NAQPMS model; J.L., L.Z., W.W., B.L., Q.W., D.C. and Y.P. provided the air quality monitoring data; H.W. performed the quality control of the observation data; and L.K. performed the inversion estimation, generated the figures, and wrote the paper, with comments provided by G.R.C.

Competing interests

The authors declare no competing financial interest.

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Supplementary Material

Text S1: Assessment of the influences of site differences on the emission inversions

The emission increments at the observation sites (a posteriori minus a priori) for different species in China from 2013 to 2015 under the scenarios of fixed observation sites (blue lines) and varying observation sites (orange) were calculated to assess the influences of the site differences on the emission inversions (Fig. S2). 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 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 the emission increments can be used to approximate the temporal variations of the a posterior emissions. It can be clearly seen that 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. S2, 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 PM_{10} 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. 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 PM_{10} 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 SO_2 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 SO_2 is generally negative over the most sites, thus the increased observation sites would lead to larger decreasing trend in the inversed emissions of SO_2. To date, these results highlighted the significant influences of the site differences on the estimated emissions and their trends. Therefore, we recommend not to use the emission in 2013 and 2014 when analyze the trends of the emissions.

Table S1 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)
NMVOC	1.4 (0.6)	4.5 (4.4)	1.6 (0.5)	8.1 (8.6)	6.5 (5.8)	8.1 (10.1)

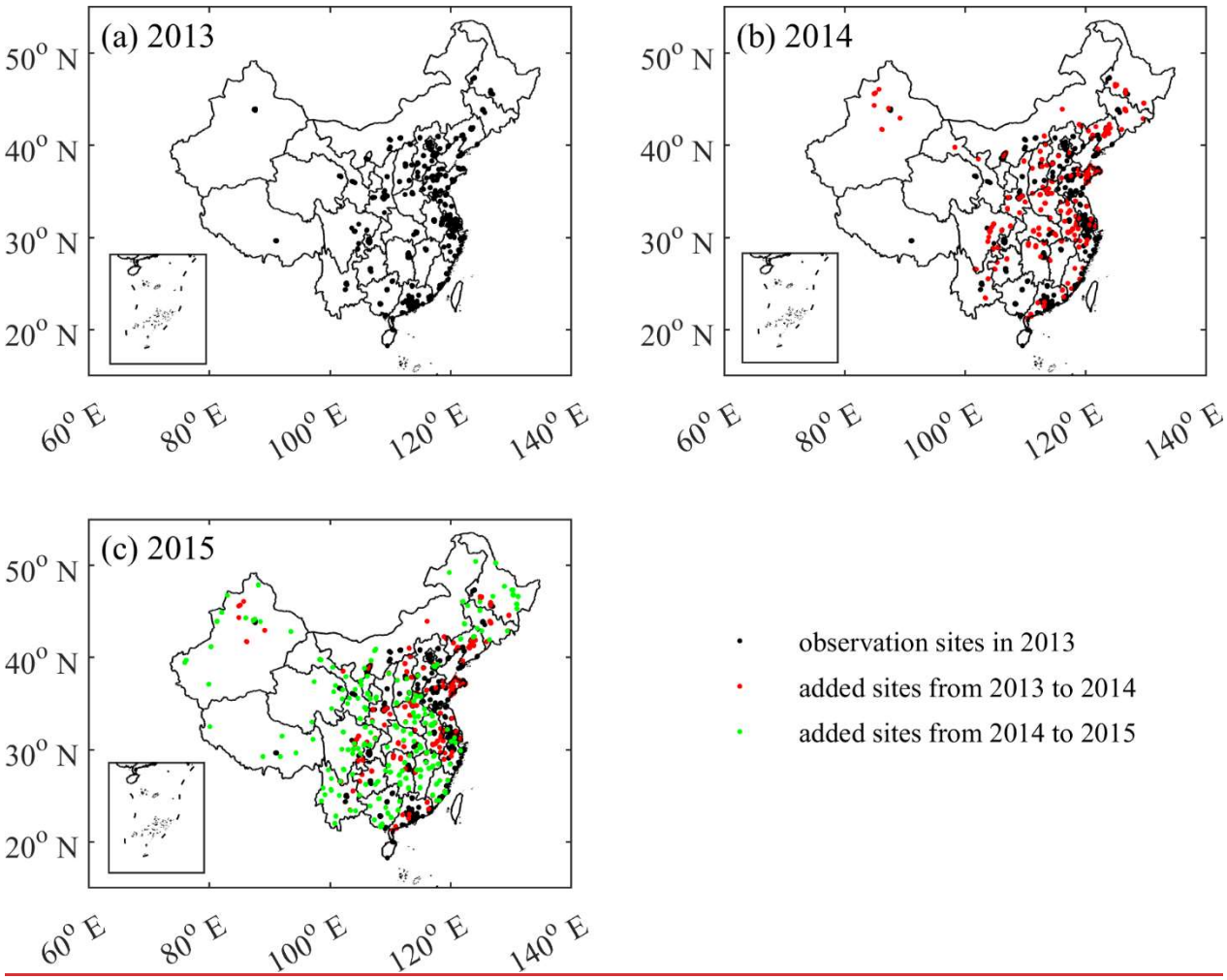

Figure S1: Spatial distributions of observation sites in (a) 2013, (b) 2014 and (c) 2015. The observation sites in 2013 were marked as black dots, while the added observation sites from 2013 to 2014 and those from 2014 to 2015 were marked as red and green dots, respectively.

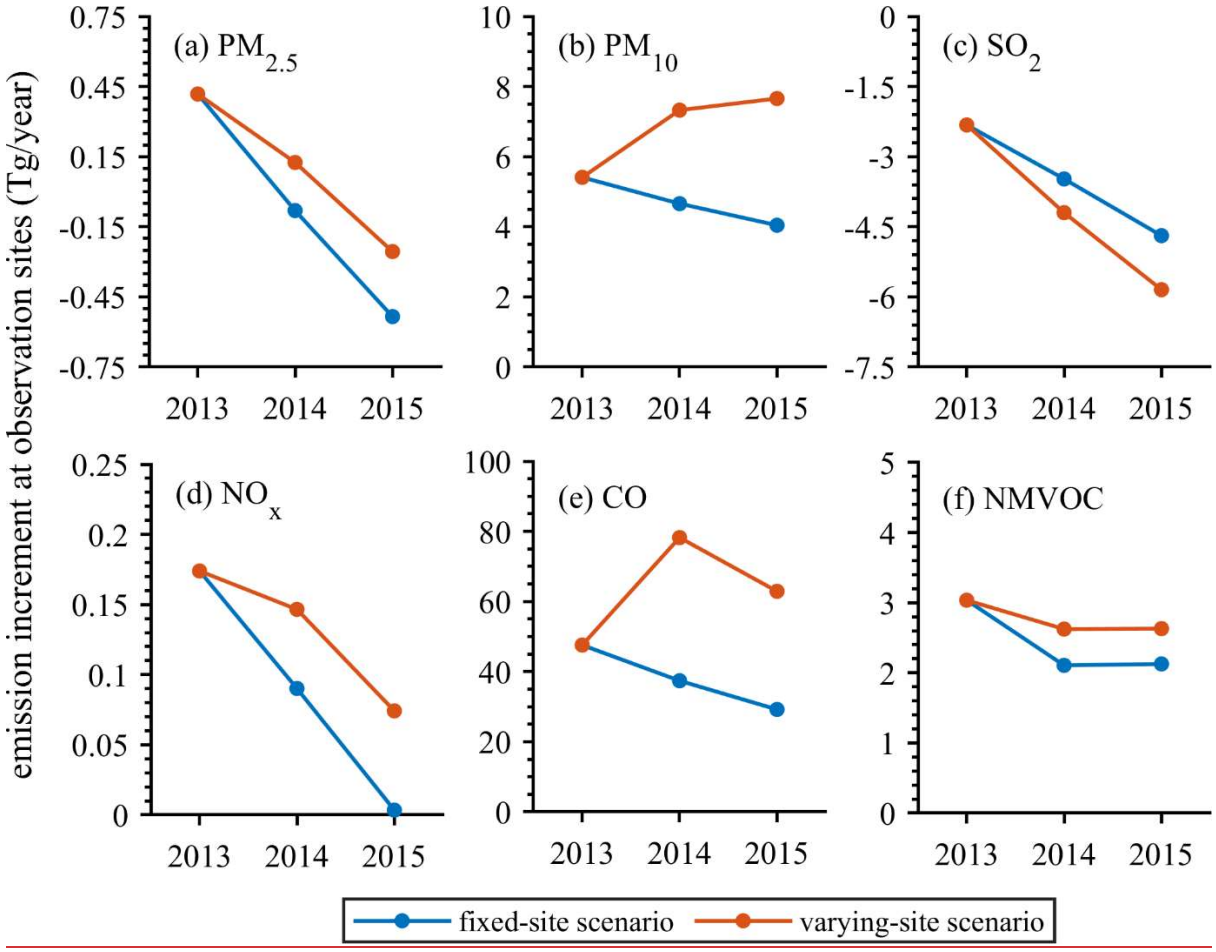

Figure S2: The calculated total emission increments at the observation sites for different species under the fixed-site scenario and varying-site scenario.

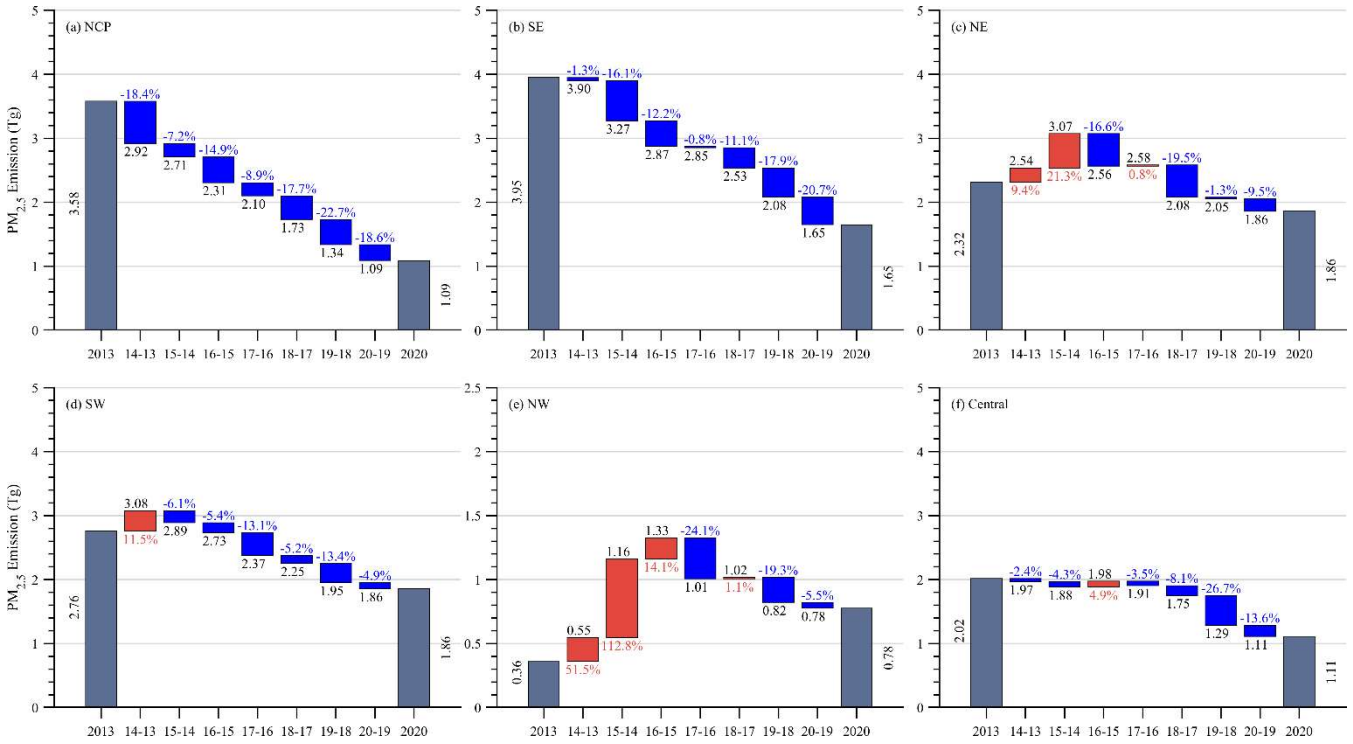

Figure S3: Emission changes of $PM_{2.5}$ from 2013 to 2020 over different regions of China obtained from CAQIEI.

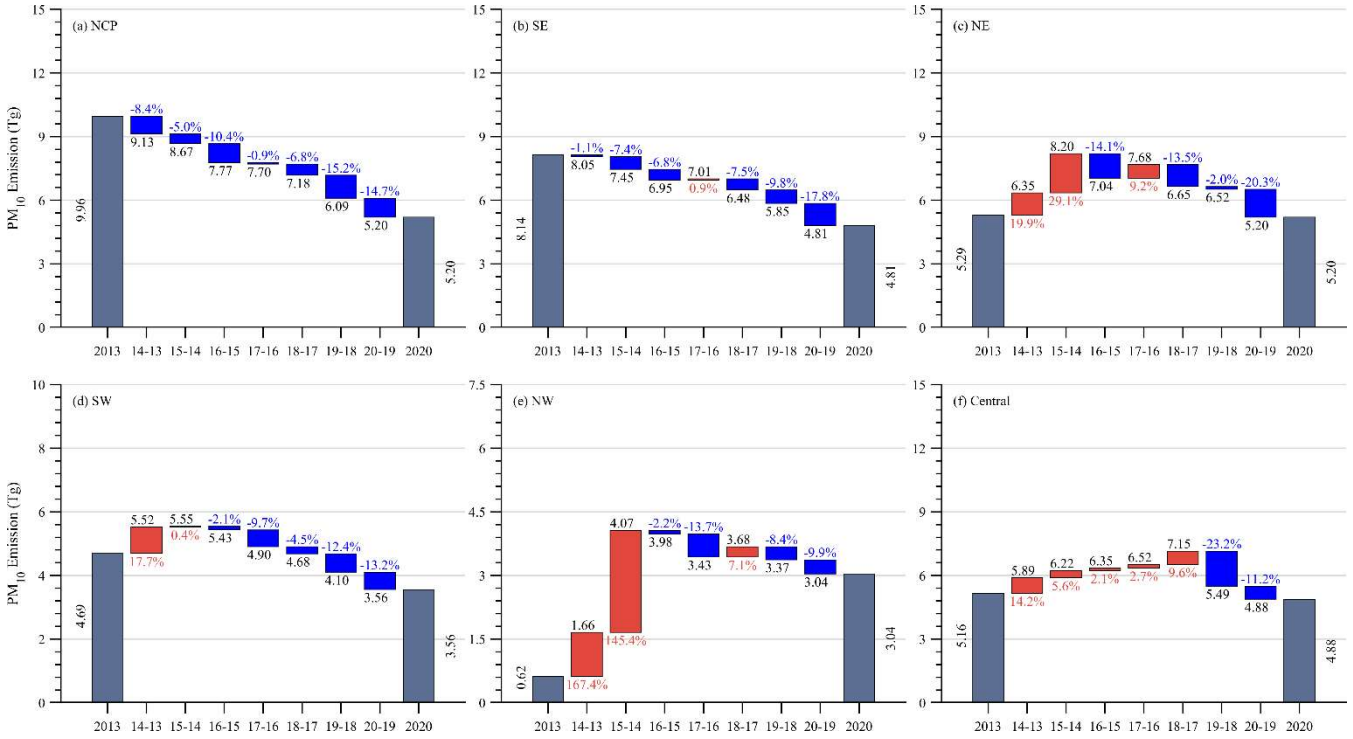

Figure S4: Same as Fig. S3 but for PM_{10}.

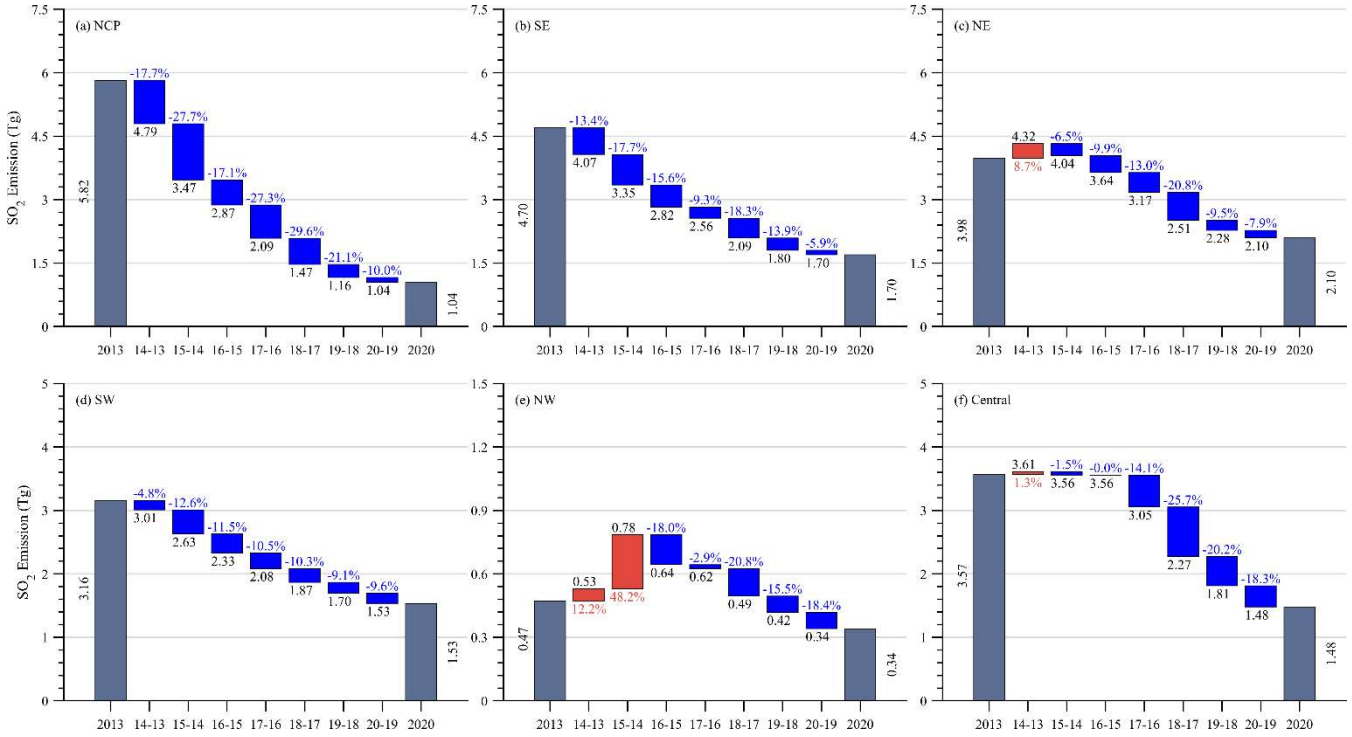

Figure S53: Same as Fig. S31 but for SO_2.

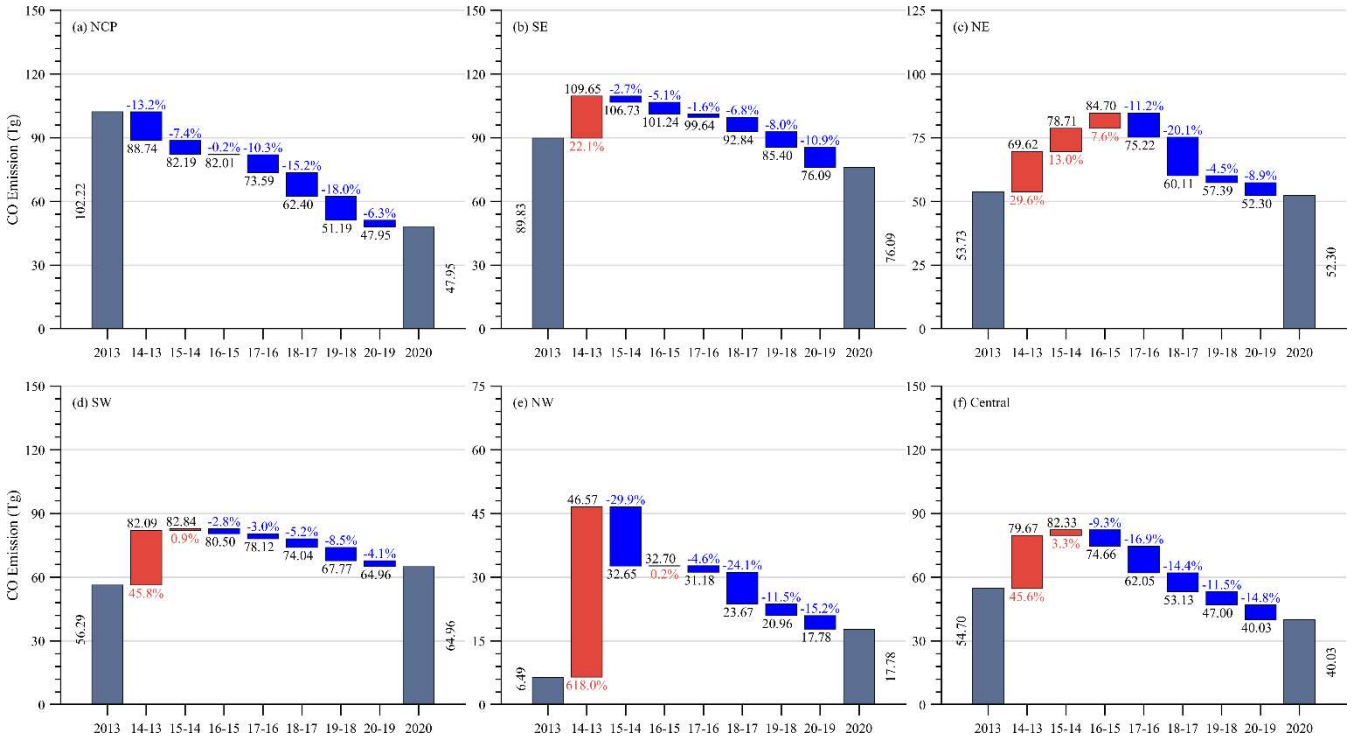

Figure S64: Same as Fig. S31 but for CO.

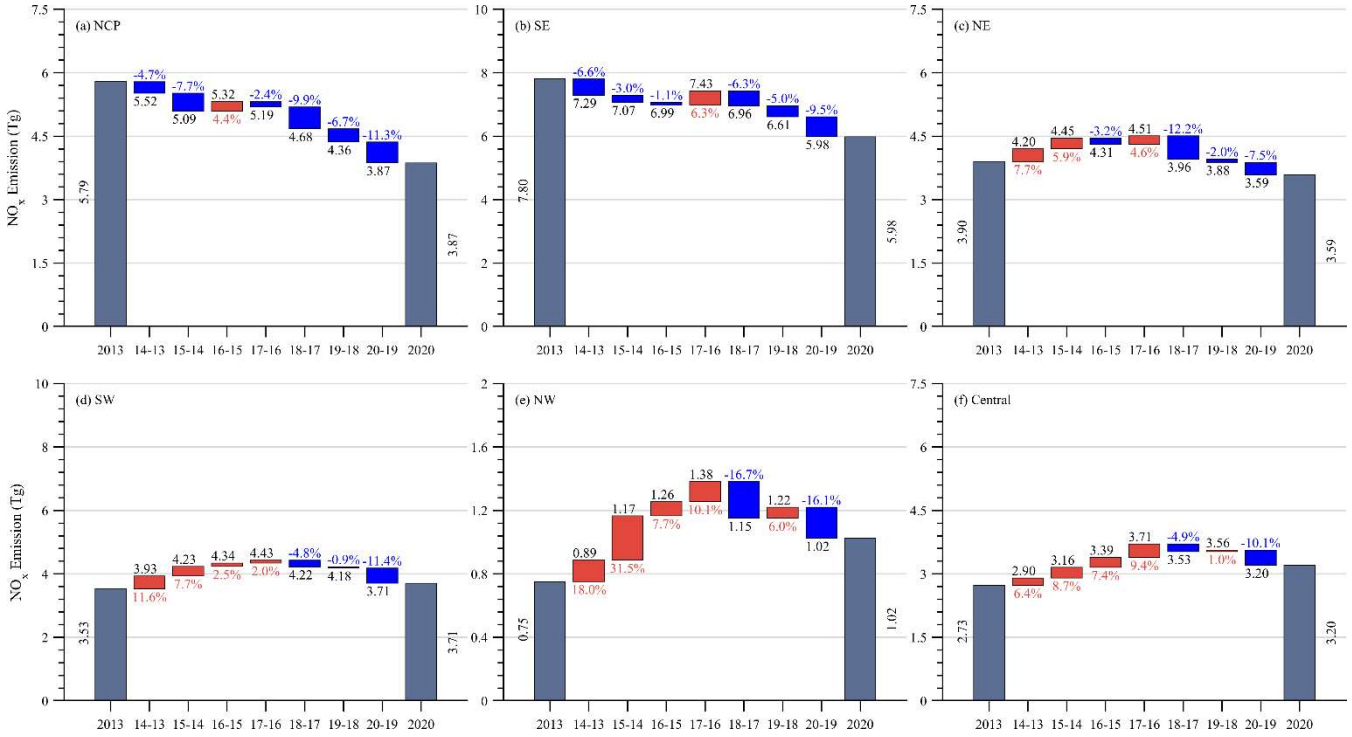

Figure S75: Same as Fig. S31 but for NO_x.

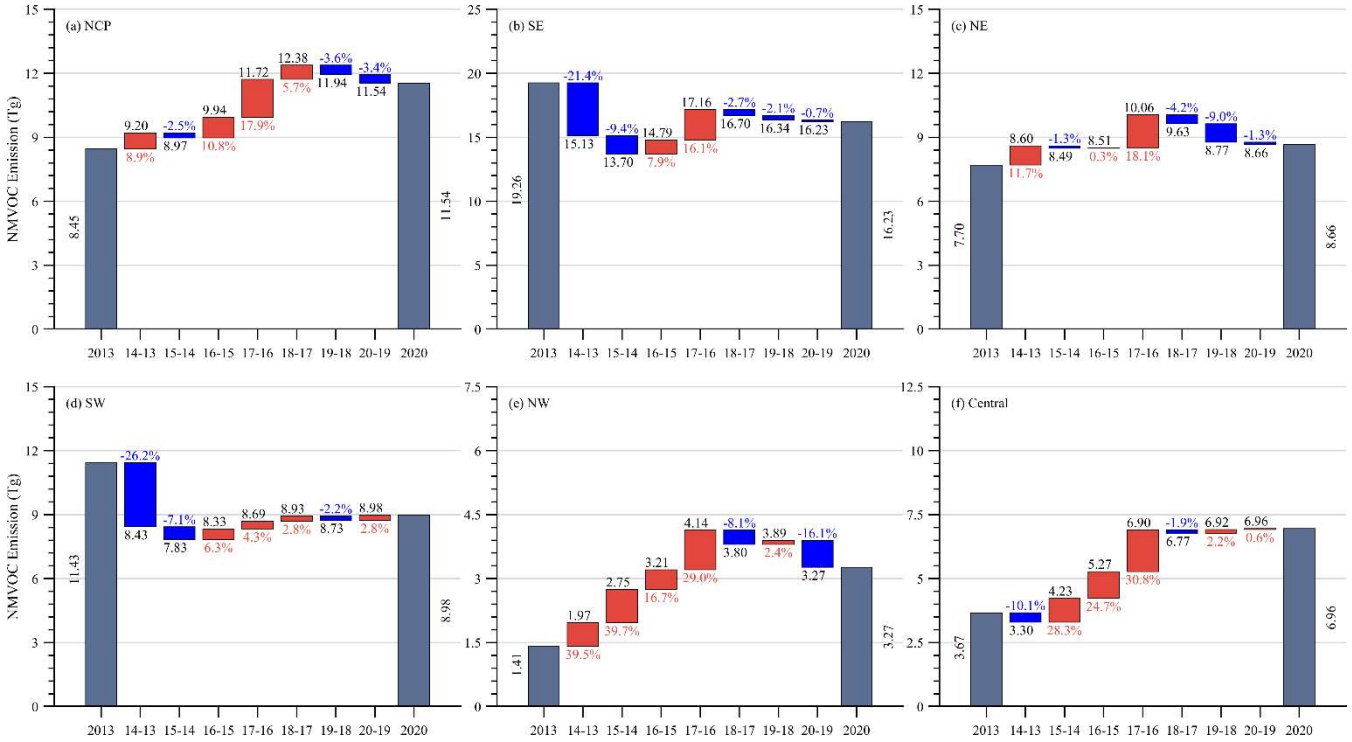

Figure S86: Same as Fig. S31 but for NMVOC.

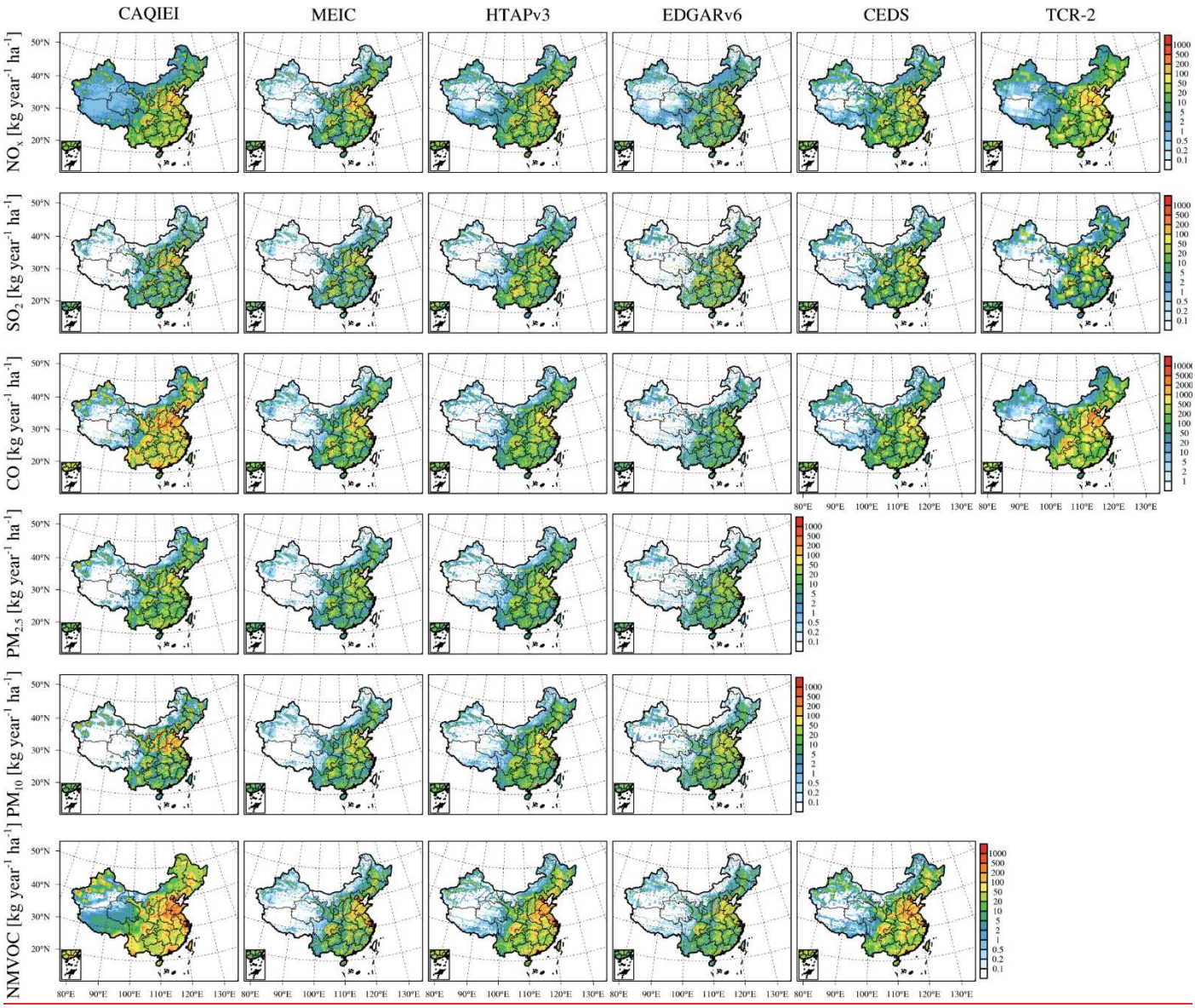

Figure S9: Spatial distributions of the averaged emissions of different air pollutants in China during 2015–2018 obtained from CAQIEI, MEIC, HTAPv3, EDGARv6, CEDS and TCR-2. Note the due to absence of gridded products of the ABaCAS inventory, we did not provide its spatial distributions. Also, the natural sources were not added to the previous emission inventories in this figure because of the different spatial resolutions among these inventories.

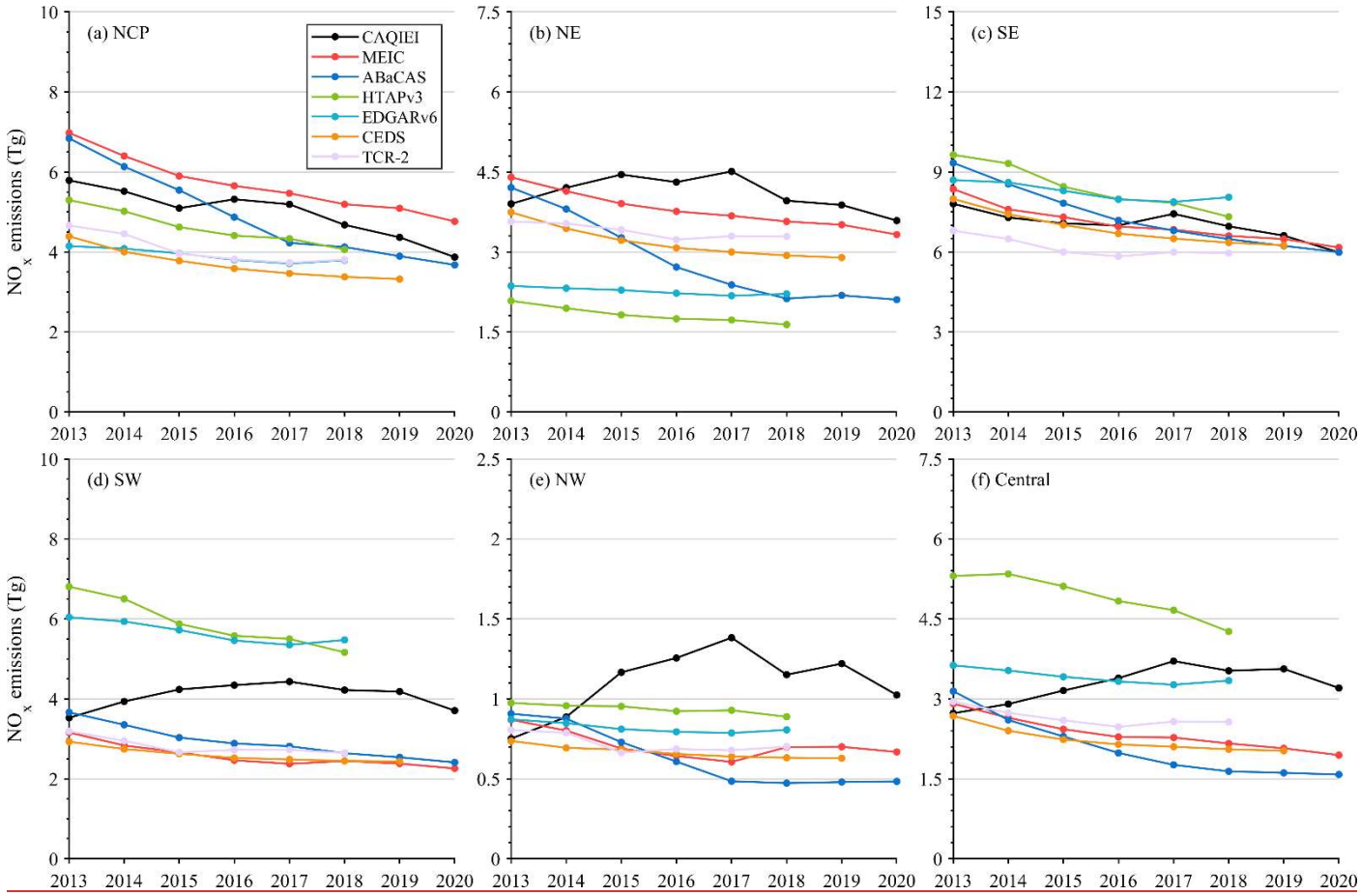

Figure S10: Time series of annual NO_x emissions over of different regions of China: (a) NCP, (b) NE, (c) SE, (d) SW, (e) NW and (f) Central from 2013 to 2020 obtained from CAQIEI and previous inventories. Note that the natural sources were not included in the previous inventories in this figure.

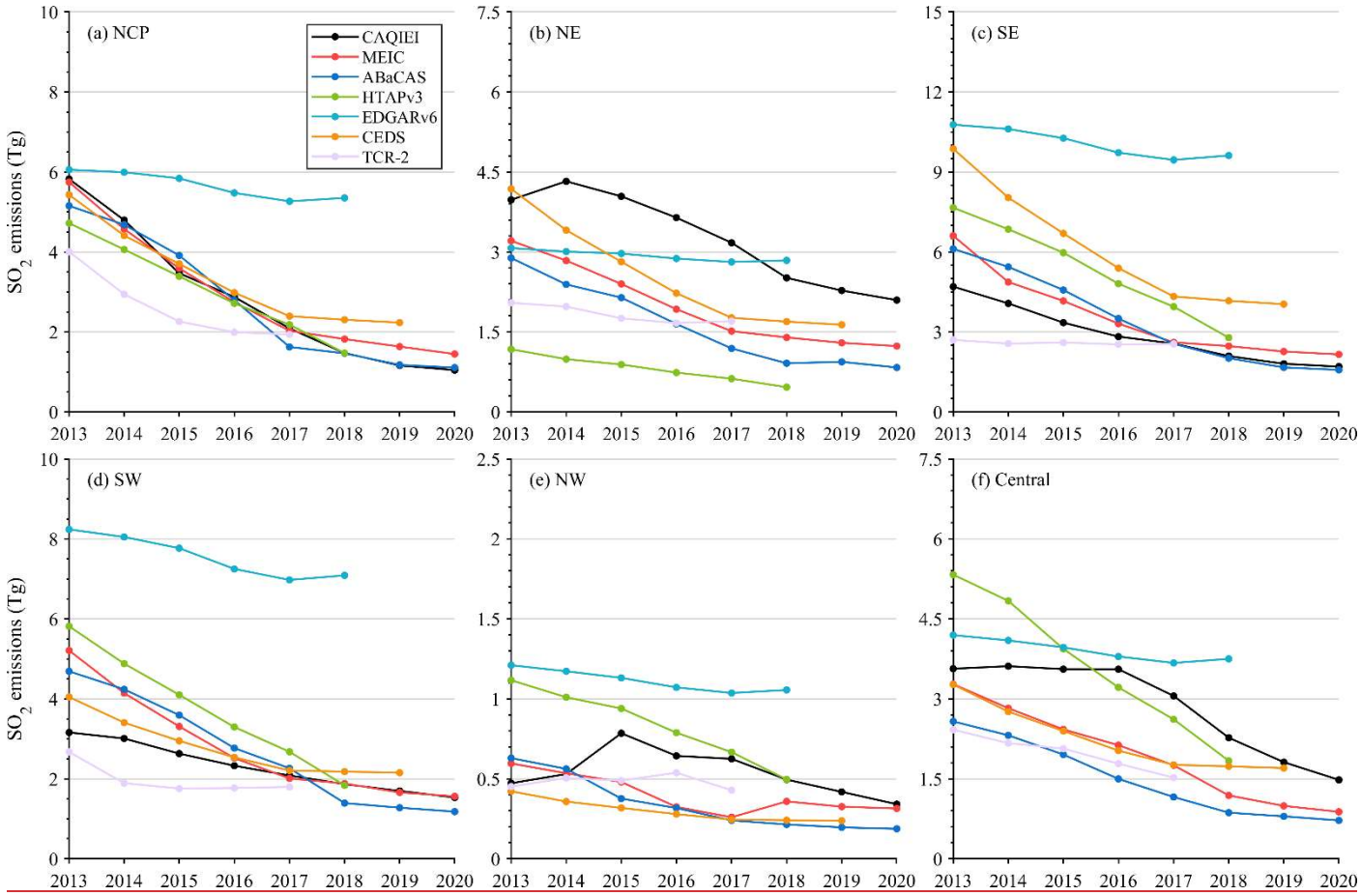

Figure S11: Same as Fig. S10 but for SO_2.

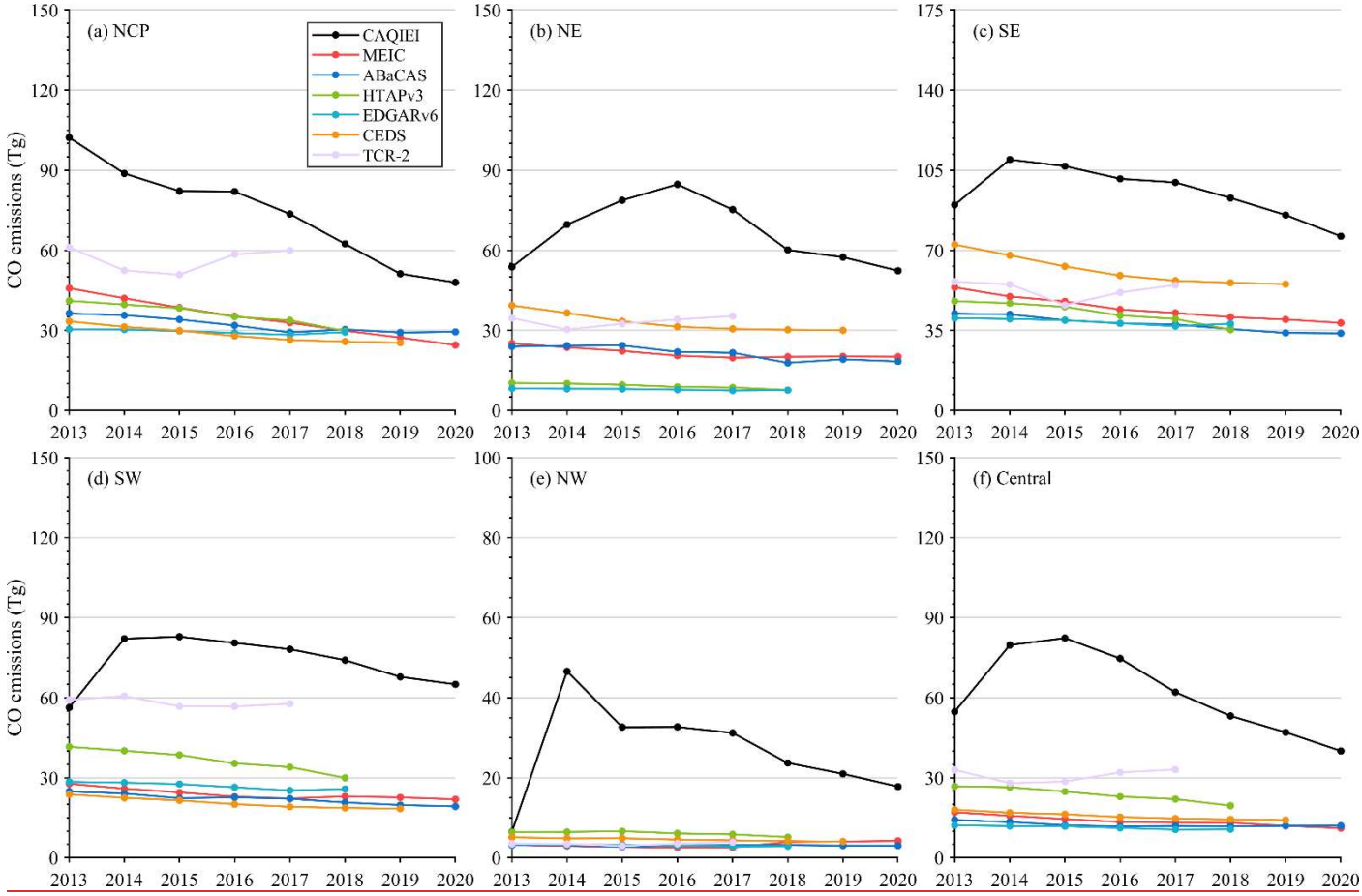

Figure S12: Same as Fig. S10 but for CO.

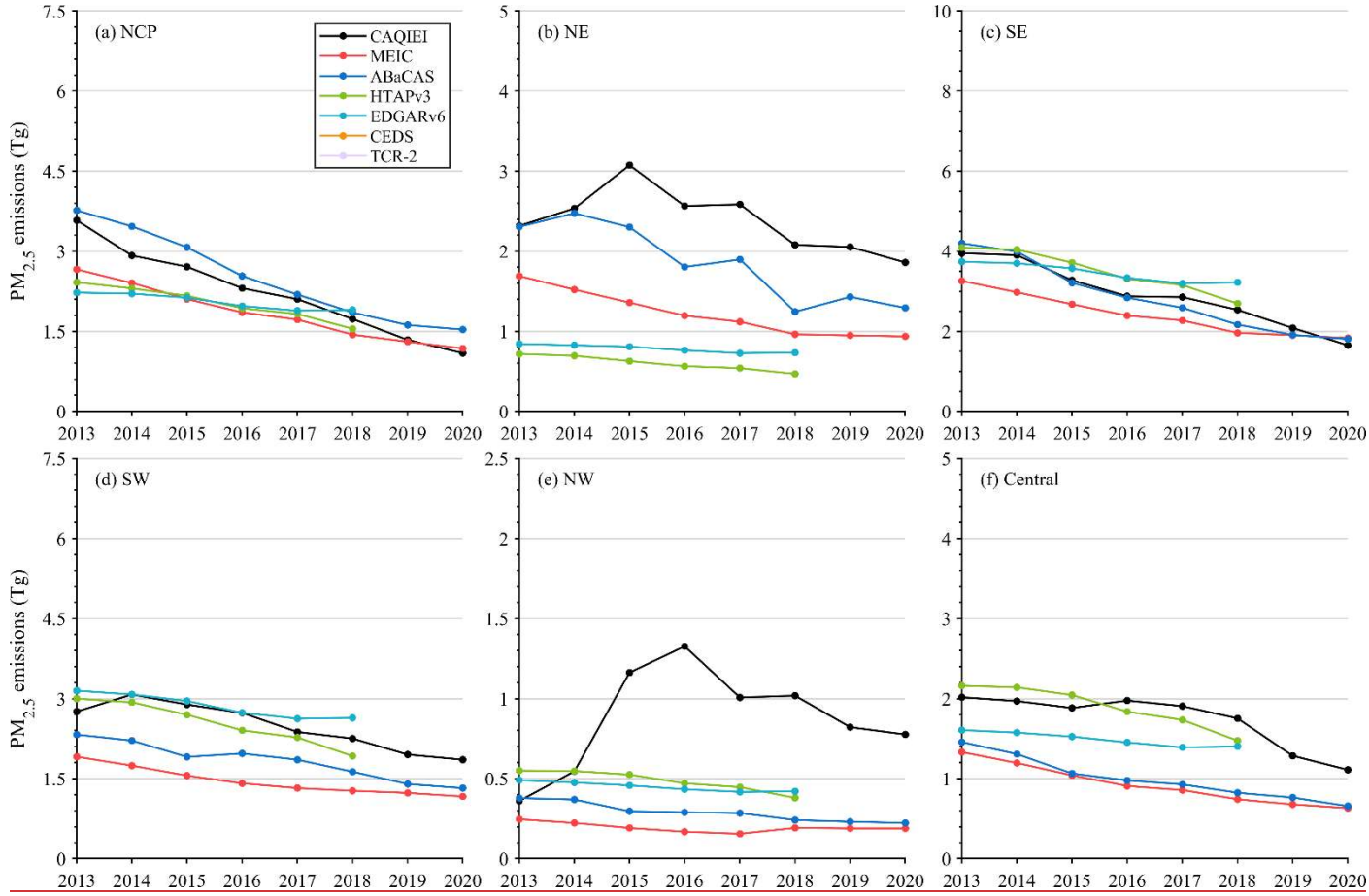

Figure S13: Same as Fig. S10 but for $PM_{2.5}$.

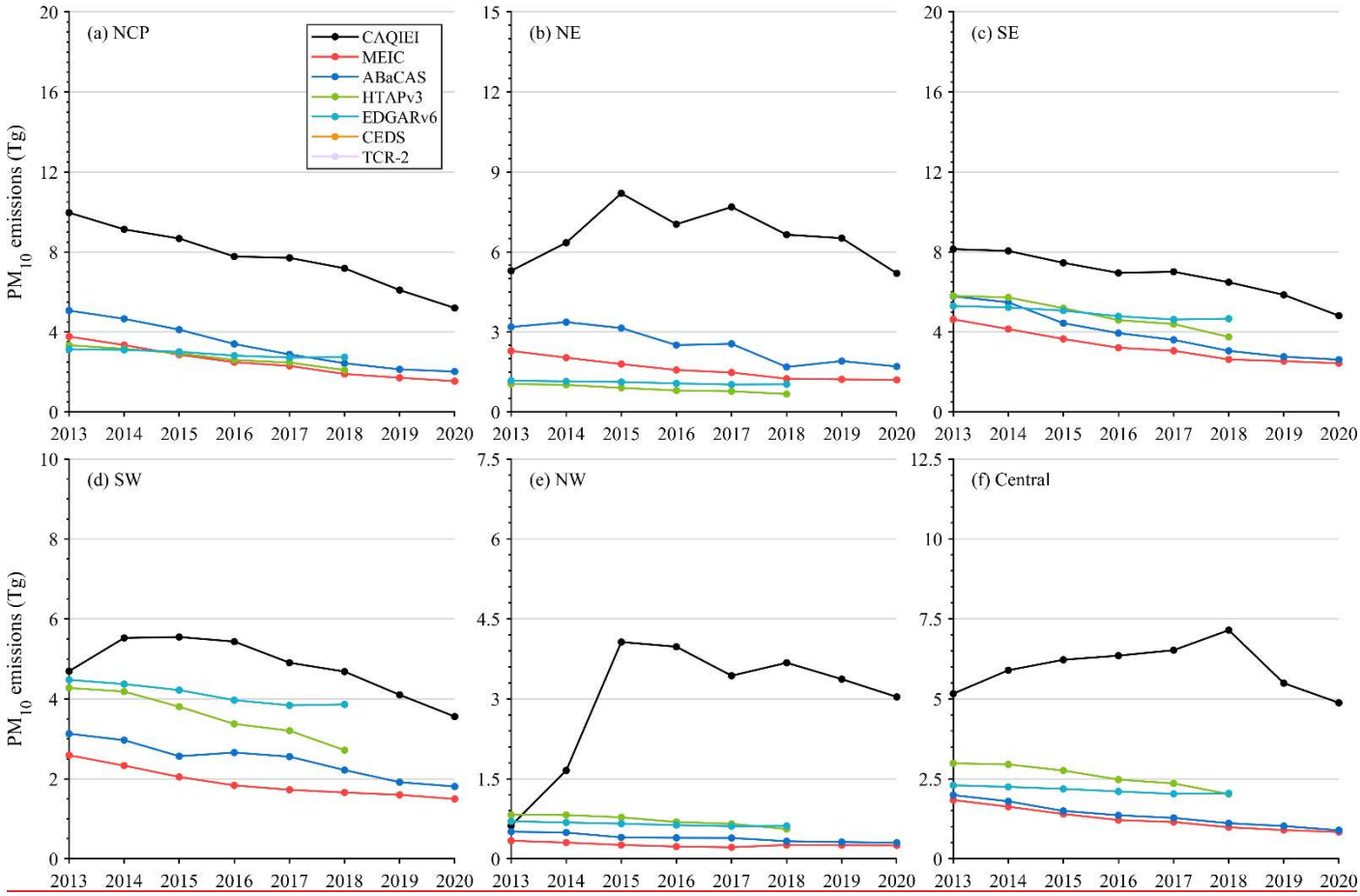

Figure S14: Same as Fig. S10 but for PM_{10}

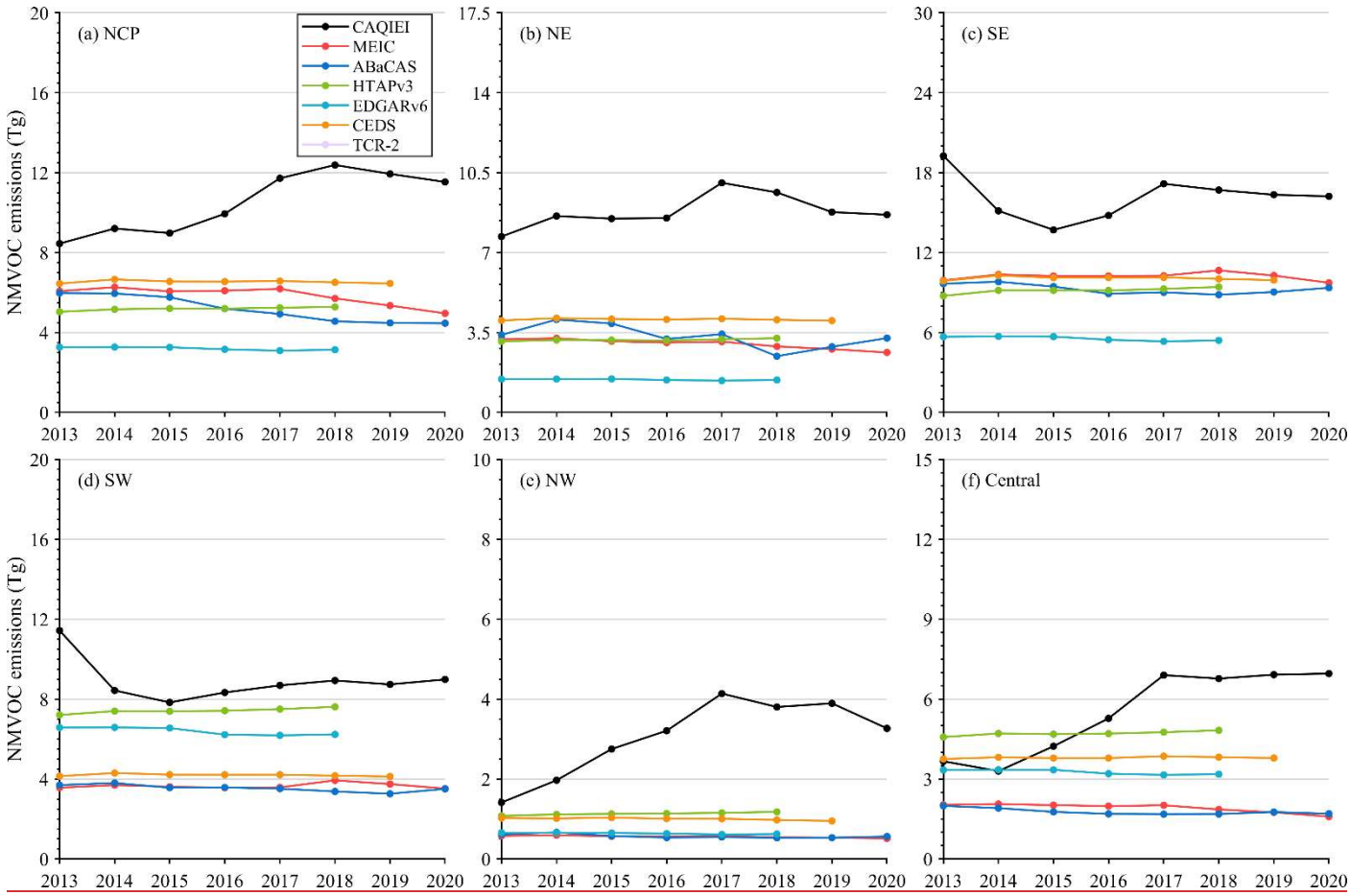

Figure S15: Same as Fig. S10 but for NMVOC.

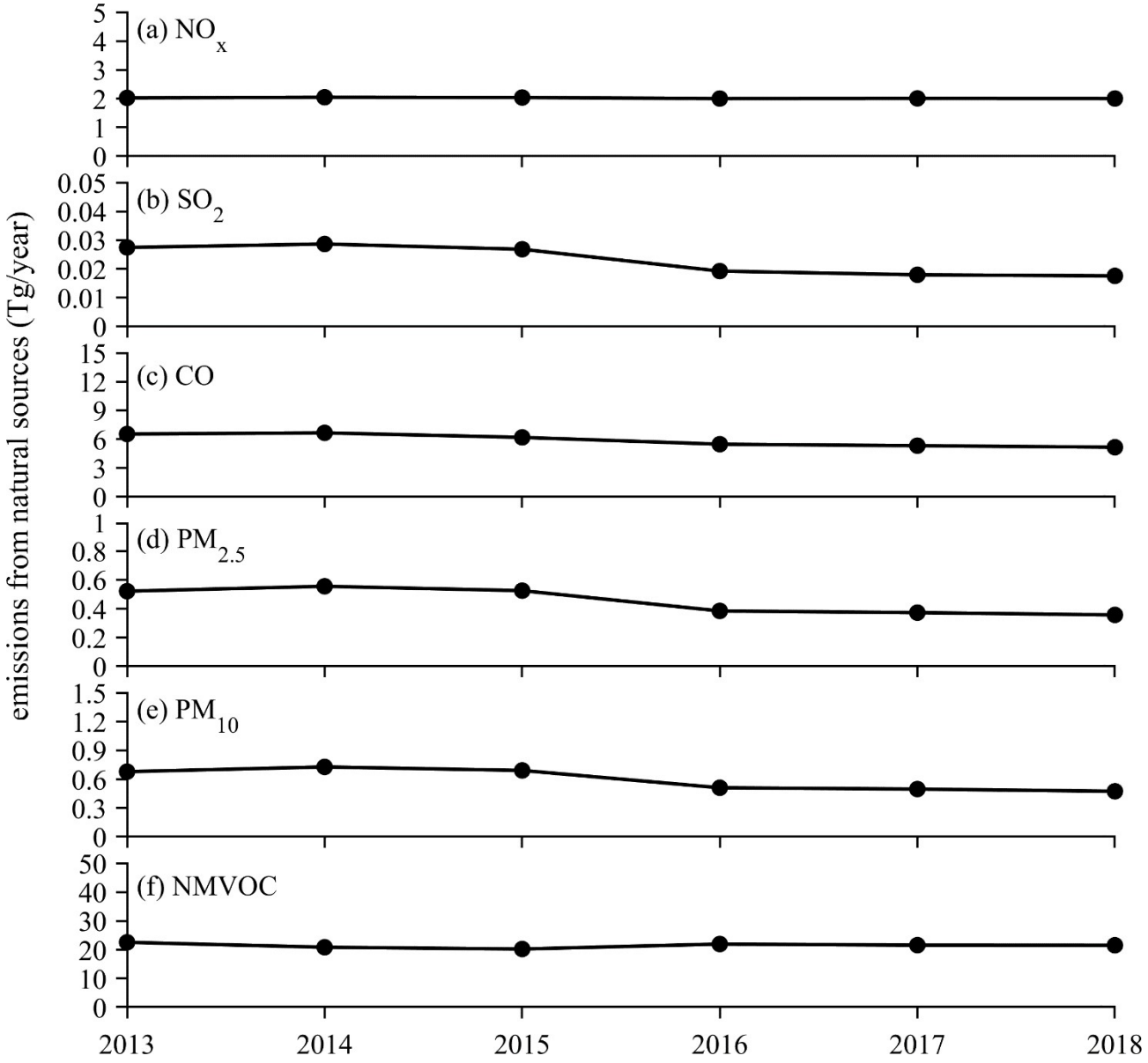

Figure S167: Time series of annual natural emissions of (a) NOx, (b) SO2, (c) CO, (d) PM2.5, (e) PM10 and (f) NMVOC in China from 2013 to 2018. The considered natural sources includes the biogenic, biomass burning and soil emissions.

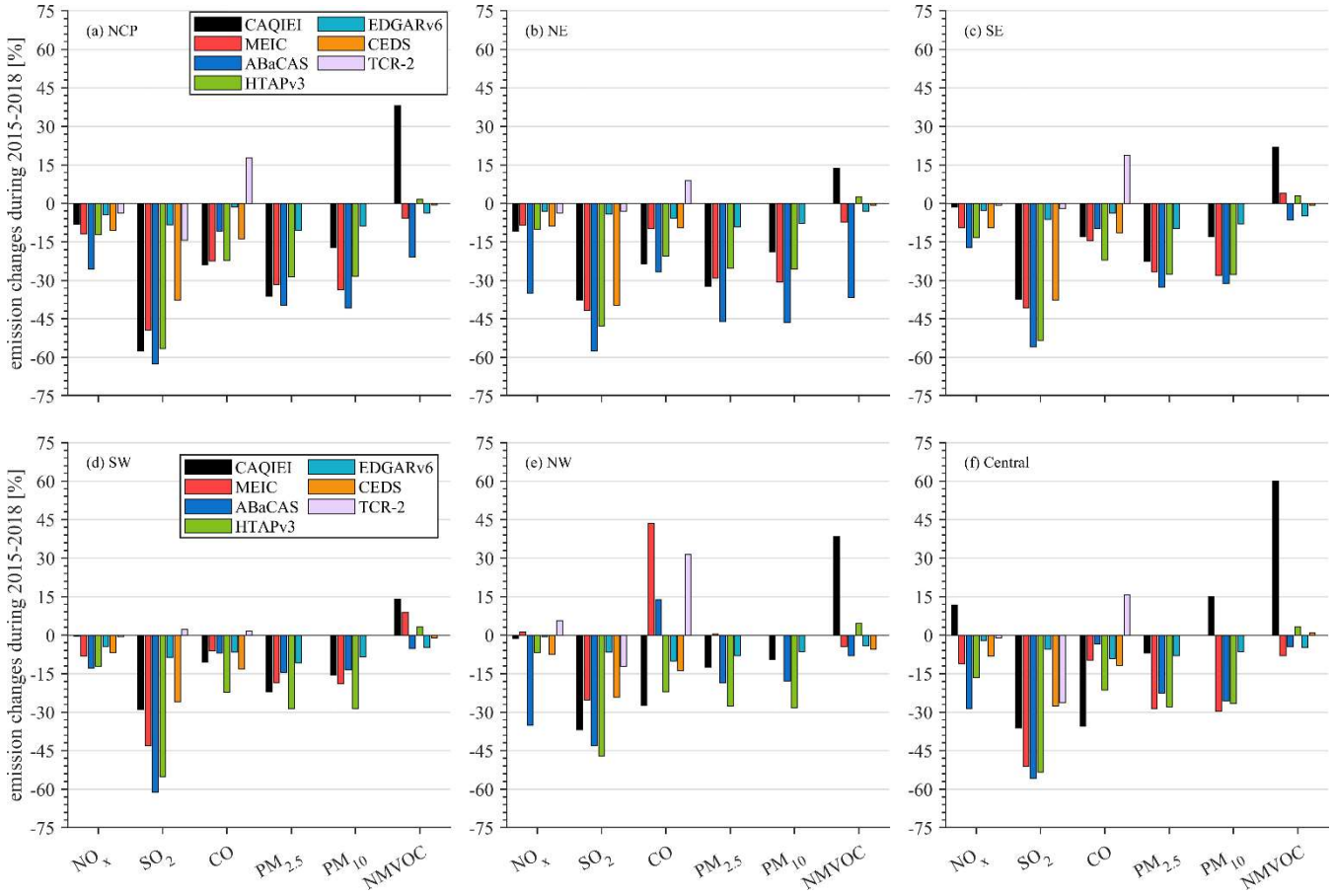

Figure S178: Comparisons of the calculated emission changes of (a) NO_x, (b) SO_2, (c) CO, (d) $PM_{2.5}$, (e) PM_{10}, and (f) NMVOCs over different regions of China from 2015 to 2018 between CAQIEI and previous inventories.