

Response to CC1:

We appreciate the reviewer's feedback on the manuscript, and we carefully reviewed the comments and addressed each individually below, highlighting changes made in the revised manuscript.

General comments

In the context modeling demonstration of emission control effectiveness, the general understanding is that models and model inputs all have uncertainties. The work presented in the manuscript investigated two aspects of these: gas-phase chemical mechanisms and emissions inventories. To certain extent, other factors such as meteorological driving fields could cause even larger uncertainties.

We agree with the comment that uncertainties of other factors such as meteorological inputs can also lead to large differences in the model predictions. However, our WRF model performance for key surface parameters has been evaluated against observation data from NCDC, demonstrating good performance (Kang et al., 2021). It is beyond the scope of the study to consider meteorological inputs from multiple regional weather forecast models.

To address these uncertainties, some well-established approaches, such as those used in the United States for regulatory air quality modeling purposes, do not use model results in an absolute sense as done in this work. Rather, the emissions control effectiveness is assessed using the combination of observations and modeled relative changes. The appropriateness of the methodology in this work seems to be questionable.

We agree with the comment regarding how the models are used for regulatory applications, such as attainment demonstrations, in the United States. In fact, as discussed in section 2.4, for each emission/inventory combination, we conducted 5 NO_x reduction simulations (20, 40, 50, 60, and 80% reduction), 5 VOC reduction simulations, and 5 combined NO_x and VOC reduction simulations. Overall, a total of 80 simulations (15 simulations * 5 emission/inventory combinations + 5 base cases) were conducted to assess how the predictions response to relative emission changes.

Specific comments

Section 2.1: It will be good to add a table summarizing main features for CS07, S11, S18 mechanisms, for example, the number of species, the number of reactions, major updates.

Thanks for the suggestion. Table S1 in the Supplementary Materials summarizes the main features of the three mechanisms.

Line 159: CMAQ also includes SAPRC07 mechanism, is CS07 different from the SAPRC07 mechanism in the CMAQ model?

The CS07 is different from the standard SARPC07 mechanism in the CMAQ model. The CS07 is a condensed version of SAPRC-07 comparable in size to CB05. It incorporates the condensed, approximate peroxy radical lumped operator method used in SAPRC99, CB4, and CB05. CS07 provides predictions of ozone (O₃), total peroxy radicals (PANs), and hydroxyl (OH) radicals that closely resemble those of the uncondensed mechanism (Carter, 2010).

Line 165: Is there any reason why S18-MEIC is not simulated?

The MEIC only has five emission sectors, making it difficult to re-speciate the emissions for the S18 mechanism. Specifically, solvent utilization emissions are not represented in the public version of MEIC as a separate sector (Wang et al., 2018). Since solvent utilization accounts for a significant fraction of VOC emissions in urban areas and has very different emission characteristics than fuel combustion sources, re-speciating the five-sector MEIC VOC emissions will be inaccurate.

The above discussion is included in the revised manuscript on page 7, lines 180-186.

Section 2.4: Are emission reductions limited to anthropogenic emissions, or it also applies to biogenic and fire emissions? The last sentence in this section needs some clarifications.

Thanks for the question. The emission reductions are applied to the final combined emissions, which include anthropogenic, biogenic and fire emissions.

Page 7, first paragraph: Since inorganic aerosols are also investigated, in the emissions comparisons, particulate matter emissions should be included as well.

Thanks for the comment. The inorganic aerosols in the model are dominantly from secondary formation. The impact of differences in the primary particle emissions on secondary inorganic aerosol predictions is very small. Thus, we did not include the primary particle emissions in the comparison.

Section 3.1, model performance evaluation: is there any PM_{2.5} chemical component measurement that can be used to evaluate model performance for inorganic aerosols (i.e., ammonium sulfate and ammonium nitrate). Also how does the model perform for NO_x, SO₂, or selected VOCs (if possible)?

Unfortunately, we do not have enough data to assess the model performance of PM_{2.5} component concentrations. Since the current study focuses on how the relative changes on the predicts due to emission changes instead of determining the accuracy of the

emission inventories or chemical mechanisms, we did not perform model performance assessments for other species.

Line 242: The reference to the US EPA model performance criteria needs to be listed.

The reference to the US EPA performance criteria is revised to reference a more recent paper by Emery et al. (2017).

Figure 1: Why is O₃-8hr noticeably lower over the yellow sea when comparing S11-REAS to S11-MEIC?

Ozone formation sensitivity regime over the yellow sea is likely NO_x-limited as the VOC-sensitive urban plume is advected to the marine environment (Vermeuel et al., 2019). Furthermore, satellite observed HCHO/NO_x column over the Yellow Sea is greater than 6, clearing indicating a NO_x-limited regime (Li et al., 2021). Since the NO_x emissions in the upwind regions in the REAS inventory are significantly lower than these in the MEIC inventory, this leads to reduced O₃ formation in the S11-REAS results compared to the S11-MEIC results.

Line 274-286 and Figure 2: Model difference is presented for SIA (secondary inorganic aerosol). It is probably clearer if ammonium sulfate and ammonium nitrate are presented separately instead of being in a combined SIA. It will also help explain the impact of emission reductions on SIA concentrations.

The ISORROPIA aerosol thermodynamics model in the CMAQ model outputs only nitrate, sulfate, and ammonium ion concentrations. Depending on the pH of the aerosol aqueous solution, sulfate can be in the form of SO₄²⁻ or HSO₄⁻. Since these components are mostly dissolved and do not necessarily exist in solid form, we chose not to further break down SIA into ammonium sulfate and ammonium nitrate.

In addition, SIA is not clearly defined in the manuscript. What is included in SIA? How are the model primary inorganic aerosols separated from SIA?

SIA is defined in the original manuscript in the caption of Figure 2. In the revised manuscript, SIA is spelled out on page 10, line 298 as “secondary inorganic aerosol”.

Line 314, section 3.4: Is there any column concentrations data for HCHO that can be used to evaluate model performance for HCHO?

As the reviewer pointed out, the emissions control effectiveness is assessed using modeled relative changes, so we didn't compare the HCHO column concentrations with observations. Our predicted monthly average surface HCHO concentrations from S11-MEIC are similar to the surface observations made in China (Zhang et al., 2021). This is added to the revised manuscript on page 12, lines 355-356.

Figure 6: Why does Shenzhen seem to be least responsive to emission reductions?

Ozone concentration in Shenzhen is lower than other cities included in Figure 6 and most O₃ there is background O₃ on typical days (Kang et al., 2023). Thus, it is less responsive to emission reductions than other cities.

This discussion is included in the revised manuscript on page 13, lines 399-402.

Line 385-386: The explanation of the impact of VOC control on SIA concentrations seems to be too brief. More in-depth discussion will be helpful. For example, sulfate and nitrate probably need to be separated; maybe even PAN formation needs to be in the discussion.

Indeed, both sulfate and nitrate formation can be affected by the changes in the VOC emissions and sulfate and nitrate can be studied separately. However, from NO_x/VOC emission control perspective, both sulfate and nitrate concentrations are changed, leading to changes in the PM_{2.5} concentrations. It is not particularly useful to separate the two components. We agree with the reviewer that a more detailed study is needed to focus on the underlying mechanisms that lead to sulfate/nitrate increases when VOC emissions are reduced.

Line 460: Why NO₃ radical concentrations are much higher in northern China than southern China?

NO₃ radical is formed from the reaction of NO₂ + O₃. In northern China, O₃ and NO₂ concentrations are significantly higher, leading to high production rate of NO₃. In addition, NO₃ is lost due to photolysis reaction and reactions with NO₂. Due to higher PM concentrations northern China, the loss of NO₂ due to photolysis is slower. The reaction of NO₃ with NO₂ is temperature dependent. Lower temperatures in northern China allow a longer lifetime of NO₃. In summary, the faster formation rate and the slower loss rate of NO₃ in northern China led to higher NO₃ concentrations.

Section 3.5.4. HCHO concentration decreases due to NO_x emissions reductions are attributed to decrease of OH and NO₃ levels. A conclusion is drawn indicating that secondary formation is the dominant source of HCHO. But the reduction of VOC emissions lead to steeper decreasing of HCHO. The VOC emissions reductions also cause OH and NO₃ level increases as stated in the previous sections. These seem contradictory to the dominant secondary source conclusion.

We appreciate the comment. However, there is no contradiction. First of all, if the HCHO is mostly primary, NO_x reduction would not lead to significant decrease of HCHO as indicated in Figure 10. While Figure 10 shows reduction of HCHO with reduction of VOCs, this reduction is likely due to reduction of precursor VOCs, as HCHO is the oxidation product of most of the VOCs. Our results are also consistent with many previous studies

such as those in Houston (ref). Furthermore, as shown in Figure S15, the highest concentrations of HCHO occur in rural areas in Southern China with high biogenic emissions and low anthropogenic emissions, which confirms the secondary nature of HCHO. These secondary HCHO can also be transport to downwind urban areas.

The above discussion is included in the revised manuscript.

References

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