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Large Modeling Uncertainty in Projecting Decadal Surface Ozone Changes Over City Clusters of China

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Abstract Climate policies will affect future surface ozone pollution in China. Here, we simulate changes in summertime ozone across China by 2030 under four emission scenarios reflecting different levels of climate action. We also contrast results obtained with two different chemical mechanisms employed in the chemical transport model (WRF-Chem). With emission reductions in ozone precursors introduced by climate policies, both mechanisms show promising ozone mitigation for most parts of China. However, they disagree starkly in China's three main city clusters, where one mechanism projects worsening ozone pollution by 2030 despite the emission reductions. We analyze possible drivers of this important discrepancy, in particular the role of varying ozone chemical regimes affecting its sensitivity to emission changes. We recommend an intercomparison project to examine this critical modeling uncertainty among other models/mechanisms, which would be invaluable for informing local and regional emission control strategies that are based on single-model results.

Plain Language Summary Surface ozone pollution is harmful to both human health and ecosystems. Reducing ozone formation through effective emission control strategies has therefore been identified as a pressing need. Chemical transport models (CTMs) are important tools that can help scientists and policymakers assess how effectively the emission reductions may alleviate ozone pollution. However, we show that the predicted effectiveness of emission control strategies for ozone mitigation in areas within the three city clusters of China are strongly dependent on the choice of chemical mechanism commonly employed in CTMs. For example, given emission reductions driven by ambitious climate action, we find that projected ozone pollution in these regions could be improved or worsened by the year 2030 depending on the model mechanism used. Our work underlines the importance of considering and understanding this disagreement when it comes to projecting even near-term emission-control strategies. Furthermore, we highlight the potential benefits of conducting a multi-model/mechanism intercomparison project to better understand how and why different models/mechanisms disagree on the simulated ozone response to emission changes, as to produce more robust mitigation scenario assessments.

Key Points:

- Future emission pathways driven by climate actions are projected to alleviate surface ozone pollution in most parts of China by 2030
- However, for China's three main city clusters, model projections disagree strongly for two widely used chemical mechanisms
- This modeling uncertainty may arise from the inconsistency of categorizing ozone chemical regimes by different chemical mechanisms

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Surface ozone pollution is one of the key environmental concerns in China. In contrast to the remarkable reduction in fine particle ($PM_{2.5}$) pollution driven by clean air policies (Zhang et al., 2019), many studies report a worsening of ozone pollution in urban regions of China over the last decade (Li et al., 2020; Y. Liu & Wang, 2020a; X. Lu et al., 2018, 2020; Weng et al., 2022). Earlier worsening ozone pollution, from 2013 to 2017, have been attributed to ozone's nonlinear response to large reductions in nitrogen oxides (NO_x) emissions without equivalent reductions in volatile organic compounds (VOCs) emissions (Y. Liu & Wang, 2020b; N. Wang et al., 2019; T. Wang et al., 2017), particularly in many urban regions that were within the NO_x-saturated regime (H. Lu et al., 2019; Ou et al., 2016). Nevertheless, with the continuous reductions in NO_x, the sensitivity of summertime ozone production to VOCs in some urban areas of China may have gradually weakened (W. Wang et al., 2021, 2022). Furthermore, it has been suggested that the turning point between NO_x-saturated and NO_x-limited regimes in some densely populated urban areas of China was reached in 2019 (Chen et al., 2021).

A key question is how ozone pollution in China can be effectively mitigated through future emission controls, especially considering potential co-benefits from policy measures aimed at climate change mitigation. For

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instance, in 2016, China committed its Nationally Determined Contribution (NDC) pledges within the context of Paris Agreement; and further scaled up its commitment in 2020, by proposing the goal of achieving carbon neutrality (Cheng et al., 2021; Z. Liu et al., 2022). This ambitious climate action may bring large emission reductions in CO_2 accompanied by reductions in emissions of other pollutants such as $PM_{2.5}$ and ozone precursors, which may lead to air quality improvements in the near future. For example, Cheng et al. (2021) suggested that by 2030, following the emission reduction plans in NDC pledges, the majority of the Chinese population may be exposed to less $PM_{2.5}$ pollution (<35 $\mu g \cdot m^{-3}$). For surface ozone, long-term projections under the Representative Concentration Pathways (RCPs) using chemical transport models (CTMs) have been conducted by previous studies (Hong et al., 2019; Y. Wang, Hu, et al., 2021; Zhu & Liao, 2016) but fewer studies (e.g., Shi et al., 2021) have examined the effectiveness of emission controls on ozone mitigation following recent carbon neutrality plan which reflects China's up-to-date emission control strategies. Moreover, projections by these studies were often heavily dependent on the results from a single CTM. This presents a limiting factor as varying uncertainties and configurations in model setup can produce inconsistent predictions (Gilliam et al., 2015; Thomas et al., 2019). For instance, it has been well documented that the choice of chemical mechanism in a model can lead to discrepancies in simulated gaseous species and aerosols (Archibald et al., 2020; Balzarini et al., 2015; Crippa et al., 2019; Mar et al., 2016; Visser et al., 2019; Yang et al., 2018). Consequently, disagreement in ozone projections from different chemical mechanisms can ultimately affect policymaking for ozone mitigation. Nevertheless, this issue has not been considered in sufficient detail in recent CTM studies that assess the effectiveness of current or future nationwide ozone mitigation strategies in China.

Therefore, we here conduct simulations with two widely used chemical mechanisms employed in a state-of-art CTM. We highlight and discuss noticeable discrepancies in ozone projections when considering near-future emission reductions driven by climate actions.

2. Materials and Methods

2.1. Model Setup

We use the Weather Research and Forecasting Model with Chemistry (WRF-Chem; Grell et al., 2005) standard version 4.1.5 to conduct simulations for the boreal summer period (i.e., June, July, and August). The simulation domain covers the entirety of China (see Figure S1 in Supporting Information S1). The model setup is based on the settings of Silver et al. (2020). The details of the setup are documented in Text S1 of Supporting Information S1.

To simulate gas phase chemistry, we use WRF-Chem in two configurations (i.e., chemical mechanisms) with (a) the Model for Ozone and Related Chemical Tracers (MOZART; Emmons et al., 2010) and (b) the Carbon Bond Mechanism Z (CBMZ; Zaveri & Peters, 1999). Aerosols are simulated using a 4-bin Model for Simulating Aerosol Interactions and Chemistry (MOSAIC; Zaveri et al., 2008) which is coupled to these two gas-phase mechanisms (i.e., MOZART and CBMZ) in WRF-Chem. Biogenic emissions for both mechanisms are calculated online by the Model of Emissions of Gases and Aerosol from Nature (MEGAN; Guenther et al., 2000). Biomass burning emission is not included in all the simulation runs, following Crippa et al. (2019), in order to avoid the inconsistence in processing fire emission between MOZART and CBMZ. Simulations by both chemical mechanisms share the same settings of physics and dynamics.

2.2. Simulations of Emission Scenarios

A total of five scenarios are simulated. These include a base simulation for which emissions are set at the level of summer 2017 to represent a real-world scenario. The remaining four simulations project ozone under different emission pathways for the summer of 2030. All scenarios are run for both chemical mechanisms.

The emission data used in the base simulation are from Multi-resolution Emission Inventory for China (MEIC) version 1.3, and we denote the base simulation as "Base-2017" hereafter. For model evaluation, we compare the simulated pollutants from Base-2017 with observational data (see Text S2 in Supporting Information S1). For simulations with emissions projected for summer 2030, data from the Dynamic Projection model for Emissions in China (DPEC) version 1.1 (Cheng et al., 2021) are used. The development of DPEC is based on MEIC (Tong et al., 2020).

Table 1
Summary of the Emissions Scenarios and Their Corresponding Labels

Emission scenario	Change of NO_x emissions relative to 2017	Change of NMVOC emissions relative to 2017	Scenario label
MEIC emissions in 2017	–	–	Base-2017
Baseline emissions in 2030	41.0%	12.4%	Limited-controls-2030
Current-goals emissions in 2030	−41.5%	−23.2%	Current-goals-2030
Ambitious-pollution-Neutral-goals emissions in 2030	−60.2%	−28.9%	Neutral-goals-2030
Ambitious-pollution-1.5°C-goals emissions in 2030	−60.4%	−30.4%	1.5°C-goals-2030

For the future projections, we consider four emission pathways from DPEC. These include three projected emission reduction pathways from a moderate reduction scenario considering current released and upcoming emission control policies (i.e., "Current-goals") to the other two ambitious scenarios for the pursuit of carbon neutrality ("Ambitious-pollution-Neutral-goals") and 1.5°C temperature limit ("Ambitious-pollution-1.5°C-goals"). Besides, we also include an additional pathway named "Baseline" by Cheng et al. (2021), which has overall increases of emissions compared to 2017 levels as it represents limited actions in emission controls. To avoid confusion with "Base-2017" (i.e., the real-world scenario), we term simulations of "Baseline" as "Limited-controls-2030" herein. A more detailed description of all these emission pathways is presented in Text S3 in Supporting Information S1 and can be referred to Cheng et al. (2021). In Table 1, we list all scenarios and their labels together with their relative-to-2017 changes of summertime NO_x and non-methane VOC (NMVOC) total emissions in China. Overall, relative to 2017, the sign of emission changes (i.e., increases or decreases) are spatially consistent over eastern China in each 2030 scenario (Figure S8 in Supporting Information S1).

We compare results for each of the projected emission scenarios in 2030 with the Base-2017 levels. In particular, we are interested in how future scenarios may induce changes of summertime maximum daily 8 hr average (MDA8) ozone pollution relative to 2017 levels across China. To account for emissions from outside of China, EDGAR version 5.0 at 2015 levels (Mogno & Marvin, 2022) is used. These emissions from outside China, as well as other settings of WRF-Chem, initial and boundary conditions for both meteorology and chemistry (set at the summer of 2017, see Text S1 in Supporting Information S1 for details) and biogenic emissions, remain fixed for all of the simulations. This setup reflects our objective to isolate the effect of emission changes within China on its own ozone levels by 2030.

3. Results and Discussion

3.1. Projected Ozone Changes

Figure 1 shows projections of summertime ozone changes for 2030, relative-to-2017, for the two different chemical mechanisms.

Both mechanisms show overall agreement in predicting the sign of ozone changes in areas outside of the three highly populated city clusters which are Beijing–Tianjin–Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD). Specifically, due to the lax control policies in Limited-controls-2030, ozone increases occur in these areas; whereas decreases in ozone are found under the three emission reduction pathways (Current-goals-2030, Neutral-goals-2030 and 1.5°C-goals-2030). As the ozone sensitivity in these areas have been mostly classified as NO_x-limited and transitional regime (e.g., Ren et al., 2022; W. Wang et al., 2021), it is not unexpected that ozone changes over these areas follow a similar trend as the changes in NO_x emissions (see Table 1 and Figure S8 in Supporting Information S1): lower NO_x emissions lead to ozone decreases and vice versa.

In contrast, in regions within the three city clusters where ozone sensitivity has been classified to be mostly in the NO_x-saturated regime (Li et al., 2019; Y. Liu & Wang, 2020b; Ren et al., 2022), even whether ozone may increase or decrease is disagreed by the two mechanisms. For example, in Current-goals-2030, despite the emissions of NO_x and NMVOC in these three city clusters being reduced when compared with the Base-2017 emissions (Figure S8 in Supporting Information S1), the surface ozone levels in some of these areas are predicted to be enhanced by 4–10 ppbv by MOZART (Figure 1b), lifting pollution levels further above the threshold of air quality guideline for short-term ozone exposure (100 μg m^{-3} for MDA8 ozone, roughly 50 ppbv) proposed by World

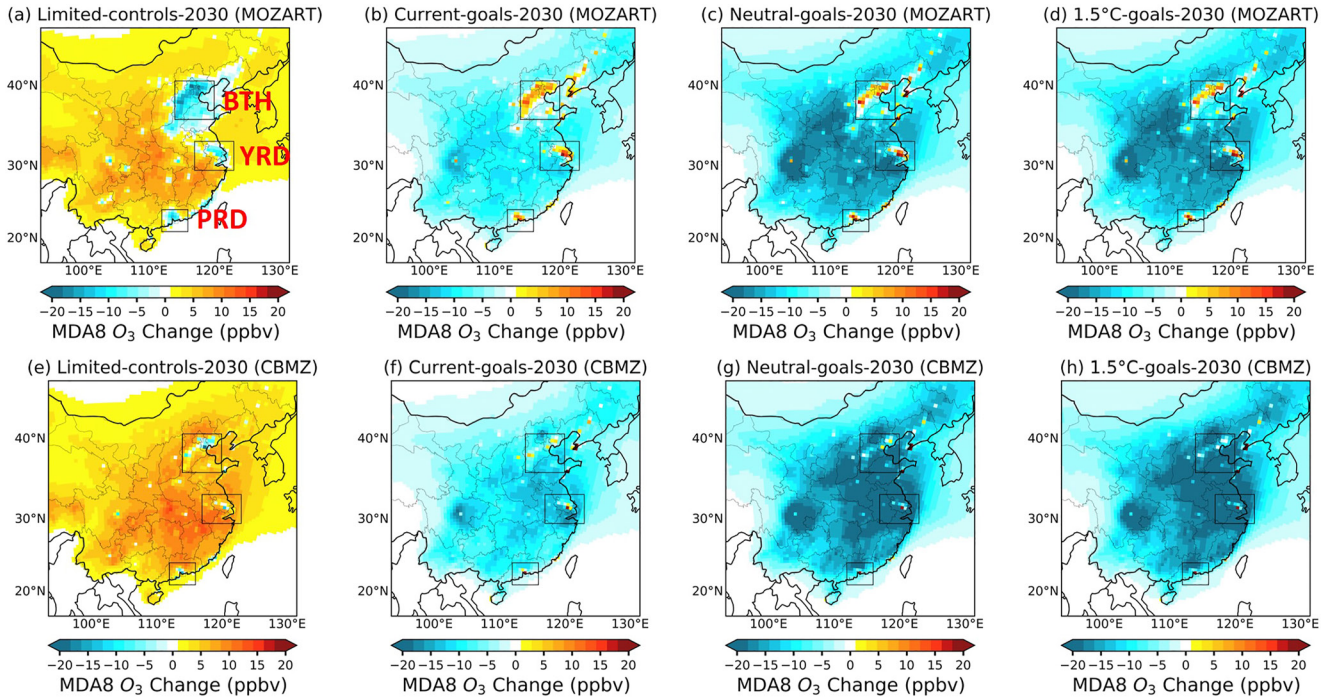

Figure 1. Simulated summertime average maximum daily 8 hr average ozone changes for each emission scenario for the year 2030, relative to Base-2017 levels. Results are shown for the two WRF-Chem configurations, with the chemical mechanisms Model for Ozone and Related Chemical Tracers (a–d) and Carbon Bond Mechanism Z (e–h), respectively. The boundaries for the three city clusters, where ozone changes mostly do not agree on the sign, are highlighted and labeled in (a).

Health Organization. Moreover, most of these simulated increases persist despite stronger emission reductions in Neutral-goals-2030 and 1.5°C-goals-2030 (Figures 1c and 1d). In stark contrast, CBMZ predicts that ozone concentrations in these regions decrease or remain close to the 2017 levels under the three emission reduction pathways (Figures 1f–1h). Crucially, this would imply different requirements for policymakers to further control emissions of ozone precursors on top of emission reductions aimed at achieving already ambitious climate policy goals. To quantitatively summarize these results, we compare averaged ozone changes for the polluted urban and industrial regions within the three city clusters and the rest of China in Figure 2.

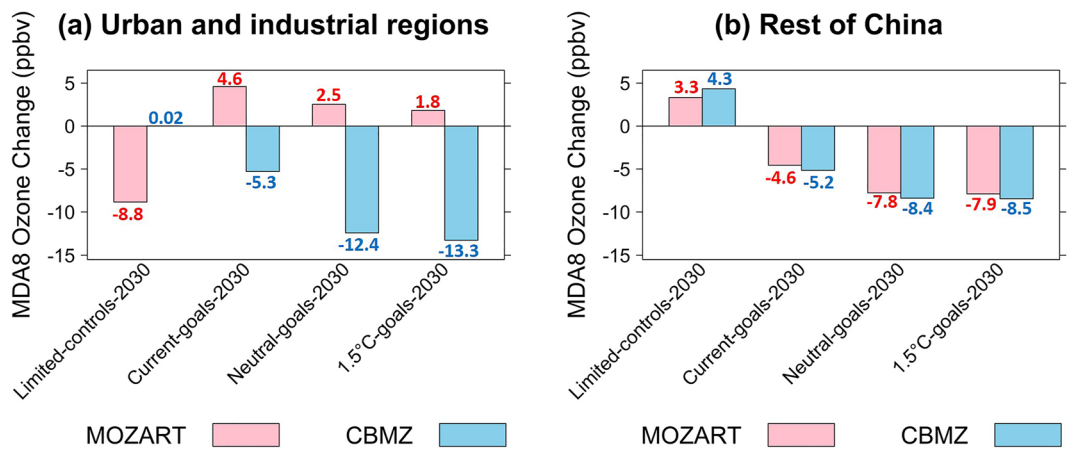

Figure 2. Simulated summertime average maximum daily 8 hr average ozone changes for each 2030 emission scenario, relative to Base-2017, by Model for Ozone and Related Chemical Tracers and Carbon Bond Mechanism Z. Shown are differences for the polluted urban and industrial regions (a) and the rest of China (b). Areas with emissions of NO_x higher than the 80th percentile in each city cluster are defined as the polluted urban and industrial regions. Areas excluding these regions within China are then defined as rest of China.

Overall, this discrepancy highlights major modeling uncertainty in projecting future ozone changes over urban and industrial regions of China, with important implications for policymakers. Using CBMZ, one might conclude that emission controls following the three emission reduction pathways are promising for mitigating ozone pollution in most regions, including those within the city clusters. This agrees well with previous findings of Chen et al. (2021) and Kang et al. (2021). Using MOZART, however, would indicate that these emission reductions might remain insufficient in some of the most highly populated areas of China, where increasing ozone pollution would negatively affect a large portion of the Chinese population.

3.2. Discrepancy in Ozone Sensitivity

The striking discrepancies in the projected ozone changes may be mainly the result of differences in gas-phase chemistry for ozone production in MOZART and CBMZ, rather than being driven by aerosol effects (Text S4 in Supporting Information S1). A most likely cause is their different chemical sensitivities to NO_x and VOC emission changes. To illustrate this, we conducted a set of test simulations for July 2017 in which the emissions across China of NO_x and VOCs were reduced by 10%, 30%, and 50%, respectively.

In essence, these simulations underline the important differences in how ozone responds within the three city clusters to a given change in emissions, in particular for reduced NO_x emissions (Figure 3). While both mechanisms show decreases in ozone when VOC emissions are reduced, consistent with the simulations by Kang et al. (2021), ozone in NO_x-saturated urban and industrial regions exhibits strikingly different responses to the perturbation of NO_x emissions. For instance, MOZART shows that the MDA8 ozone increases on average by over 5 ppbv in the urban and industrial regions when NO_x emissions are reduced by 30% or even 50% (Figure 3a). However, for CBMZ, decreases in ozone are predicted given the same NO_x emission reductions. In Addition, the ozone increases following 30% NO_x emission reductions are more pronounced and more spatially extended in MOZART (Figure 3c) when compared to CBMZ (Figure 3e), analogous to the results for emission reductions in 2030 (Figure 1). This suggests that the difference in how ozone responds to NO_x reductions is the key factor to explain the mechanism dependency.

Our tests here also indicate a disagreement in categorizing ozone chemical regimes in urban and industrial regions: the results are consistent with MOZART tending toward a more NO_x-saturated regime whereas a transitional or NO_x-limited regime may be suggested by CBMZ. Similarly, an earlier study by Knote et al. (2015) showed that simulations using MOZART tends to predict more air quality stations within NO_x-saturated regime in the North America than using CBMZ. Our results indicate that this phenomenon may also occur in China, where alleviating ozone pollution is currently pivotal for promoting public health. Such important modeling uncertainties, and their implications, should ideally be considered during the decision-making and design of future ozone control strategies.

3.3. Possible Causes of the Chemical Regime Discrepancy

Increasing ozone pollution given reductions in NO_x emissions is not unexpected in a NO_x-saturated regime. First, ozone can be consumed through its reaction with NO, which is known as NO_x-titration (i.e., $NO + O_3 \rightarrow NO_2 + O_2$) (Sillman, 1999). With reductions in NO_x emissions, NO_x-titration can be weakened, thereby leading to less ozone suppression. Second, reducing NO_x emissions in an NO_x-polluted environment can weaken the formation of nitric acid (HNO_3) that can be formed through a chain terminating reaction with the hydroxyl radical (OH) ($OH + NO_2 \rightarrow HNO_3$). In this scenario, this reaction becomes a major sink of HO_x ($HO_x \equiv OH + H +$ preoxy radicals; Jacob, 2000), which affects VOCs oxidation, thus suppressing subsequent net ozone production. Reducing NO_x emissions in such a scenario leads to more available OH for atmospheric oxidation and can ultimately enhance ozone formation (H. Lu et al., 2019; Sillman, 1999).

Mar et al. (2016) showed that inconsistencies in rate constants of inorganic gas phase reactions among different mechanisms can cause a large discrepancy in simulated ozone concentrations, which may also result in different ozone sensitivity regimes. We indeed find that the MOZART mechanism has higher rate constants for both NO_x-titration and HNO_3 formation under usual atmospheric conditions (see Text S5 in Supporting Information S1). This suggests that MOZART may have a faster suppression of net ozone formation. In turn, ozone increases may also be more responsive to NO_x reductions than for CBMZ. We tested this hypothesis by setting the rate constants for NO_x-titration and HNO_3 formation (or rate constants for most of the other inorganic reactions that are listed by Mar et al. (2016)) the same for both mechanisms. However, the discrepancy in ozone changes

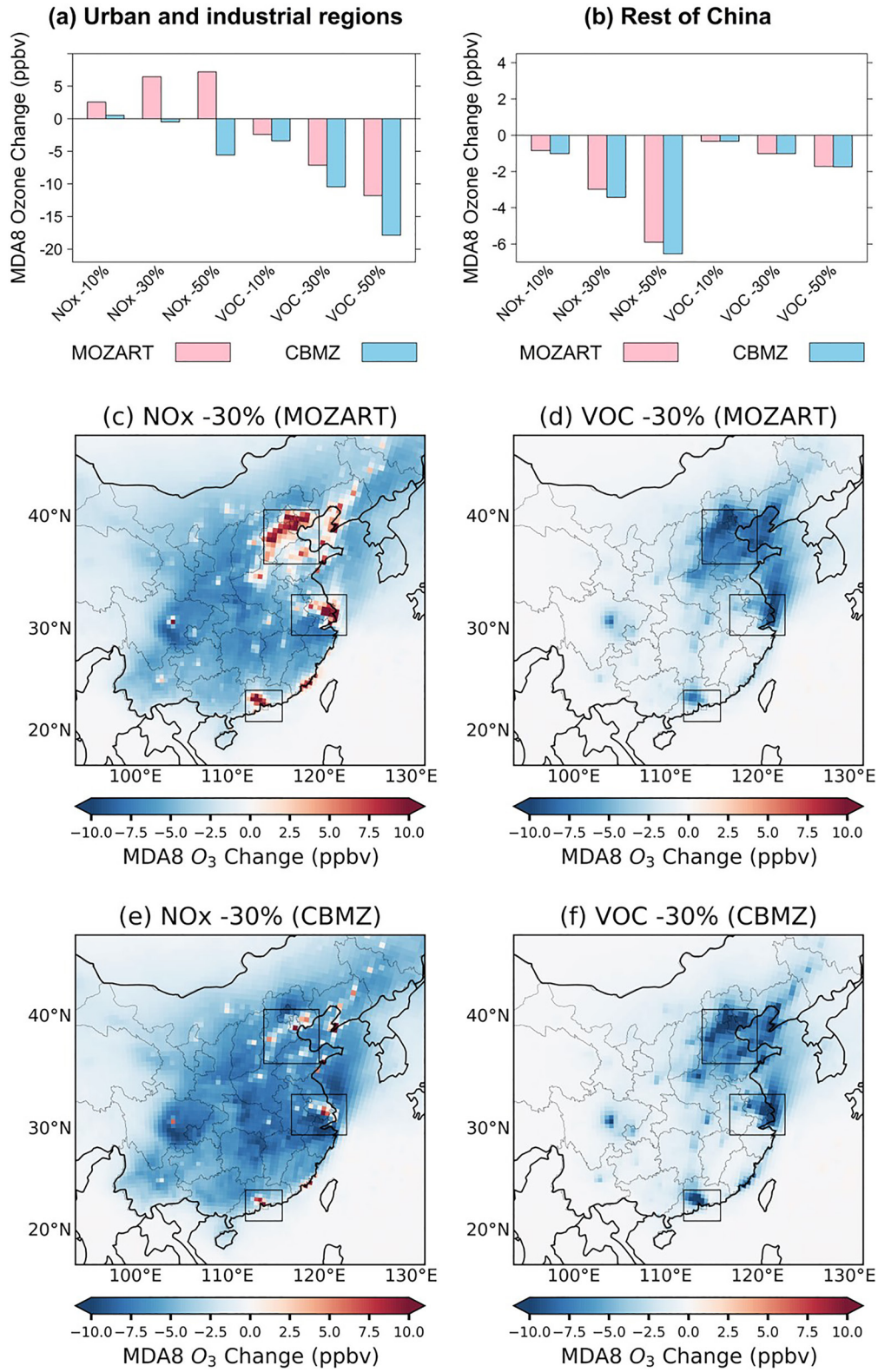

Figure 3. Averaged maximum daily 8 hr average ozone changes in response to the emission reductions of NO_x and volatile organic compounds (VOCs) during July 2017 in urban and industrial regions within the three city clusters (a) and the rest of China (b). Spatial patterns of ozone changes in Model for Ozone and Related Chemical Tracers given NO_x (c) and VOCs (d) emissions reductions of 30%. The same, but for Carbon Bond Mechanism Z (e, f).

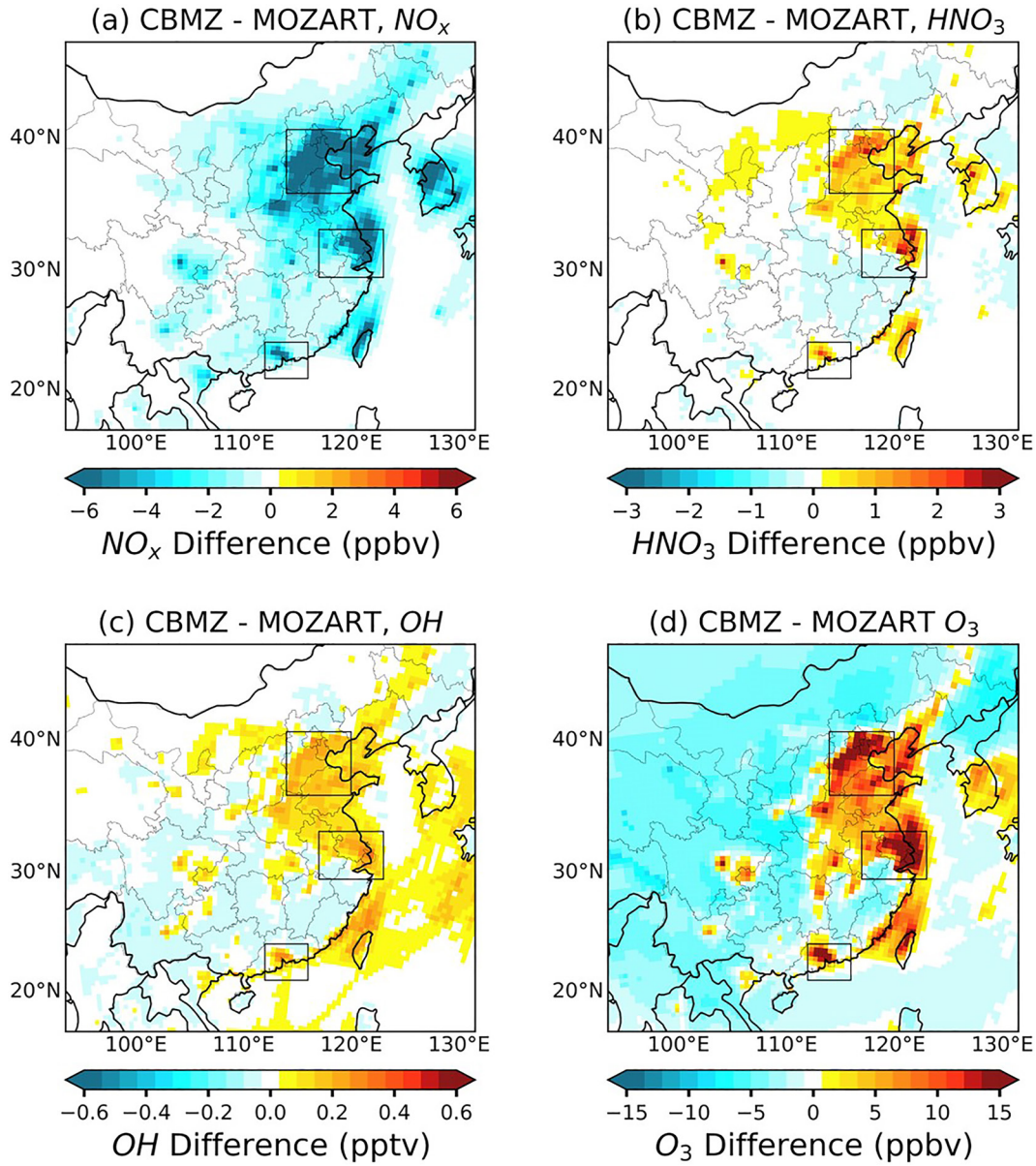

Figure 4. Differences of daytime (06:00–18:00) average concentrations of NO_x (a), HNO_3 (b), OH (c), and O_3 (d) during July in the Base-2017 scenario between Carbon Bond Mechanism Z (CBMZ) and Model for Ozone and Related Chemical Tracers (MOZART) (i.e., predicted concentration levels in CBMZ subtracted by MOZART).

persisted, suggesting that the differences in these rate constants alone cannot explain the predicted discrepancy. Clearly, rates of chemical reactions are not only determined by the rate constant but will also depend on the reactant concentrations. The simulated concentrations of key chemical compounds involved in NO_x-titration and HNO_3 formation indeed also differ significantly for the two mechanisms. For example, in Base-2017, the simulated NO_x are lower in CBMZ (Figure 4a) despite the fact that both of the mechanisms have the same emissions of NO_x, suggesting that there is a stronger sink for NO_x (e.g., HNO_3 formation) in CBMZ. This is also consistent with the higher concentrations of simulated HNO_3 when using the CBMZ mechanism (Figure 4b). It is likely that the higher simulated abundance of OH in CBMZ (Figure 4c) may be an important contributing factor that leads to a stronger formation of HNO_3, even when considering its lower reaction rate constant than the one in MOZART. Higher OH can therefore lead to higher ozone concentrations (Figure 4d) and vice versa.

In summary, with respect to NO_x-titration, MOZART may have an overall stronger NO_x-titration as it has higher simulated NO_x (i.e., weaker sink for NO_x; see Figure 4a) and a higher rate constant for NO_x-titration (Text S5 in

Supporting Information S1). Given this, the simulated ozone increases in MOZART may be more sensitive to a reduction in NO_x emission. Furthermore, the higher levels of simulated OH (Figure 4c) in CBMZ may already be sufficient to drive more ozone formation (Figure 4d), and higher ozone also leads to more OH. If this is the case, then the addition of OH from the reduced HNO_3 formation induced by NO_x reduction might not have a large effect on ozone increases in CBMZ. Therefore, with the combined effect of all these factors, simulated ozone increases may be less pronounced in CBMZ. We further hypothesize that ozone changes in CBMZ might move closer to the simulated increases by MOZART, if additional heterogenous reactions or aerosol uptakes of HO_x and ozone (e.g., Y. Liu & Wang, 2020a) are included.

Overall, our results suggest that a more thorough intercomparison of NO_x-titration, HNO_3 formation along with their corresponding chemical species across several chemical mechanisms is necessary for elucidating the discrepancy in simulated ozone responses to near-term or future emission changes. We further note that soil NO_x emissions could also be addressed in future intercomparisons, because such emissions may affect ozone responses to anthropogenic emissions changes (X. Lu et al., 2021; Y. Wang, Fu, et al., 2021).

4. Conclusions

Climate-action-driven emission controls will substantially change surface ozone pollution in China. Reliable projections of ozone changes given emission reductions in ozone precursors are key to assess which control strategy may be optimal for future ozone mitigation. Here, we show that two widely used chemical mechanisms from a CTM could produce highly inconsistent conclusions on the efficacy of planned control measures. Specifically, we project ozone changes under four climate-action-driven emission pathways by summer 2030 using two chemical mechanisms (i.e., MOZART and CBMZ) in WRF-Chem. Although both mechanisms agree that summertime ozone in most parts of China can be mitigated to lower-than-2017 levels given emission reductions, we find marked discrepancies in major populated city clusters of China. In particular, MOZART simulates worsening ozone pollution in some of these areas by 2030 despite the ambitious emission reductions in ozone precursors as part of the proposed climate actions. In contrast, CBMZ typically shows reduced ozone pollution for the same scenarios and areas. We propose that this opposite response can mainly be attributed to differences in the simulated ozone chemical regimes. Policy-making aimed at ozone mitigation that often relies on model simulations should be aware of such major discrepancies, because it can lead to inconsistent conclusions regarding the effectiveness of emission control strategies. Therefore, we see an urgent need for a multi-model intercomparison project involving various chemical mechanisms from CTMs in order to achieve a more thorough understanding of how and why simulated ozone in different mechanisms/models responds differently to emission changes of its precursors.

Data Availability Statement

The source code of WRF-Chem version 4.1.5 is available at https://github.com/wrf-model/WRF/releases/tag/v4.1.5. MEIC (version 1.3) emission data can be accessed from http://meicmodel.org.cn/?page_id=541&lang=en, and DPEC emission data (version 1.1) are available at http://meicmodel.org.cn/?page_id=1918&lang=en. EDGAR emission version 5.0 can be downloaded from https://zenodo.org/record/6130621. Meteorological data from National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS)/final analysis (FNL) are provided at https://rda.ucar.edu/datasets/ds083.3/. Simulations by Community Atmosphere Model with Chemistry (CAM-chem) can be accessed from https://www.acom.ucar.edu/cam-chem/cam-chem.shtml. Surface hourly measurement data of O_3, NO_2 and $PM_{2.5}$ provided by the Chinese Ministry of Ecology and Environment (MEE) are available at https://zenodo.org/record/7629985. Model results and configuration files for WRF-Chem simulations can be downloaded from https://zenodo.org/record/7625666. WRF-Chem preprocessing tools mozbc, anthro_emiss and bio_emiss can be downloaded from https://www.acom.ucar.edu/wrf-chem/download.shtml.

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