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Geophysical Research Letters

RESEARCH LETTER
10.1029/2021GL092816

Chinese Regulations Are Working—Why Is Surface Ozone Over Industrialized Areas Still High? Applying Lessons From Northeast US Air Quality Evolution

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Key Points:

- Reductions of nitrogen oxides (NO_x) led to increase of surface ozone (O_3) in North China Plain and Yangtze River Delta
- Shift of O_3 chemical regimes with turning points between NO_x- and volatile organic compound-limited regimes around 2019
- The impacts of high fine particles ($PM_{2.5}$) on O_3 formation has declined with reduction of $PM_{2.5}$ concentrations

Supporting Information:

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

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Citation:

Chen, X., Jiang, Z., Shen, Y., Li, R., Fu, Y., Liu, J., et al. (2021). Chinese regulations are working—Why is surface ozone over industrialized areas still high? Applying lessons from northeast US air quality evolution. *Geophysical Research Letters*, *48*, e2021GL092816. https://doi.org/10.1029/2021GL092816

Received 6 FEB 2021
Accepted 28 JUN 2021

Abstract

Observational data indicate increasing trends of surface ozone (O_3) in China, despite emission controls that have resulted in reductions of precursor emissions. Here, we explore the cause of this contradiction, through analysis of surface observations (2014–2019) in China and historical observation record in the United States (US, 1990–2019). Our observation-based analysis indicates that the reductions of nitrogen oxides (NO_x) emissions led to increase of surface O_3 in North China Plain (NCP) and Yangtze River Delta (YRD) of around 8 ppb. However, NO_x controls resulted in shift of O_3 chemical regimes over NCP and YRD, with turning points between NO_x- and volatile organic compound (VOC)-limited regimes around 2019, while model simulations suggest transitional or NO_x-limited regimes over the rest of China. The impacts of high fine particles ($PM_{2.5}$) on O_3 formation has declined because of the reduction of $PM_{2.5}$ concentrations. Stricter NO_x controls can mitigate O_3 pollutions over industrialized areas in China.

Plain Language Summary

Ozone (O_3) in surface air is an important pollutant with adverse effects on human health and vegetation growth. Here, we explore the sustainable pathway to control O_3 pollution in China through analysis of observations in China (2014–2019) and the United States (US, 1990–2019). We find that the reductions of nitrogen oxides (NO_x) emissions have led to increase of surface O_3 in North China Plain (NCP) and Yangtze River Delta (YRD) by about 8 ppb. However, the severe O_3 pollution in China can be mitigated because NO_x controls led to the shift of O_3 chemical regimes over NCP and YRD to transitional regime around 2019. In addition, the impacts of high fine particles ($PM_{2.5}$) on O_3 formation has declined because of the reduction of $PM_{2.5}$ concentrations.

1. Introduction

The large fuel consumption associated with rapid development of the Chinese economy has resulted in dramatic deterioration in air quality. To mitigate air pollution, the Chinese government has implemented a series of measures that curb emissions from industry, vehicle, and power generation (China State Council [CSC], 2013, 2016). These controls have resulted in significant reductions in many of the major atmospheric pollutants such as NO_x (Itahashi et al., 2019; Zheng et al., 2018) and high fine particles ($PM_{2.5}$; Zhang et al., 2019; Zhao et al., 2018). However, surface O_3 concentrations have continued to increase (Sun et al., 2019; Wang et al., 2020). The opposite changes in surface O_3 and its precursors have attracted substantial attention from scientific and economical perspectives. It is suggested that the changes in O_3 may be attributed to the nonlinear O_3-NO_x responses in 2013–2017 (Liu & Wang, 2020), increases in anthropogenic volatile organic compounds (VOCs) emissions (Sun et al., 2019), or possible missed $PM_{2.5}$ chemical processes in the model simulations, for example, the decreases of $PM_{2.5}$ may lead to stronger O_3 formation (Li, Jacob, Liao, Shen, et al., 2019; Li, Jacob, Liao, Zhu, 2019).

Despite the above advances, it remains a challenge to understand O_3 changes in China. For example, K. Li et al. (2020) suggests that the about 10% increase of surface O_3 over North China Plain (NCP) in 2019 is mainly driven by the reduction of $PM_{2.5}$; Tan et al. (2020) suggests that the reduced $PM_{2.5}$ is unlikely the reason for the increasing number of O_3 pollution events over NCP. Accounting for the influence of variable meteorological conditions presents additional complexity (Dang et al., 2021; Han et al., 2020; K. Li et al., 2020). While long-term O_3 changes likely depend more on the variation in anthropogenic emissions, the short-term fluctuations may be strongly affected by non-anthropogenic processes (Barnes et al., 2016; Shen et al., 2015; Young et al., 2018). However, the limited observation records (starting in 2013) from the China Ministry of Ecology and Environment (MEE) monitoring network poses a significant barrier to distinguishing anthropogenic and non-anthropogenic effects in China.

Effective emission controls have been implemented in the United States (US) since the 1970s (Environmental Protection Agency [EPA], 2017) with noticeable decreases in tropospheric NO_2 and O_3 concentrations (Chang et al., 2017; Miyazaki et al., 2017; Strode et al., 2015). The successful control of O_3 pollution in the US over the past decades may provide insights to understand both anthropogenic and non-anthropogenic influences on O_3 pollution. As well, it could offer guidance for sustainable O_3 control strategies in countries more recently applying air quality regulations, such as China. In this study, we perform a comparative analysis between recent O_3 changes in China (2014–2019) and the 30-years historical observation record over northeast US (1990–2019) to provide an observation-based diagnosis for the sources of surface O_3 changes in China. GEOS-Chem model simulations (www.geos-chem.org) are performed for China (in 2019) and northeast US (in 1997 and 2019) cases to interpret the observed O_3-NO_2-VOCs relationships.

2. Results

2.1. Observation-Based O_3-NO_2 and O_3-HCHO Relationships

Unlike west or southeast US, where changes in surface O_3 are strongly affected by background or natural sources (Fu et al., 2015; Zhang et al., 2014), the O_3 changes over northeast US are dominated by local anthropogenic sources (Dunker et al., 2017), similar to the evolution of surface O_3 in China. We thus use O_3 data in northeast US to investigate the influence of anthropogenic emission controls on O_3 pollution. Figures 1a and 1b show the summertime NO_2 and O_3 concentrations (1990–2019) at US Air Quality System (AQS) stations over northeast US. The successful emission controls led to continuous decreases of surface NO_2 concentrations from about 16 to 3 ppb, and surface O_3 concentrations from about 60 to 45 ppb. Figures 1c–1h show the summertime NO_2 and O_3 concentrations (2014–2019) for three domains in China including NCP, Yangtze River Delta (YRD), and Pearl River Delta (PRD) from the MEE stations. The domain definitions are shown in Figure 3. With contribution by over 40% of Chinese Gross Domestic Product, NCP, YRD, and PRD have large populations and experience severe air pollution, and thus, are the major targets for air quality controls in China. We find noticeable decreases of NO_2 concentrations, as well as increases of surface O_3 concentrations over NCP and YRD. The changes of NO_2 and O_3 concentrations over PRD are insignificant.

Figure 2a shows the summertime O_3-NO_2 relationship over northeast US at the AQS stations for the period of 1990–1999. The data (orange dots) are regional averages of daily mean O_3 and NO_2 concentrations, binned into 1 ppb NO_2 increments. Schroeder et al. (2017) demonstrate that an O_3-NO_2 relationship driven by nonlinear O_3-NO_x chemistry follows a lognormal distribution when VOCs concentrations are stable. As suggested by Schroeder et al. (2017), the lognormal fit (blue line) in Figure 2a demonstrates a nonlinear O_3-NO_2 relationship with a turning point between NO_x- and VOC-limited regimes around 12 ppb, from which O_3 increases (decreases) with NO_2 on the left (right). While the emission controls resulted in a noticeable decrease of observed NO_2 concentrations from about 16 ppb (in 1990) to 12 ppb (in 1999), these observations were the right side of the turning point and corresponded to an insignificant change in surface O_3 over 1990–1999. However, over 2010–2019, the observed NO_2 concentrations, which continued to decrease, are on the left side of the turning point (Figure 2b), thus emission controls in NO_x led to effective reduction of O_3 concentrations.

Figures 2c–2e show the summertime O_3-NO_2 relationship (2014–2019) in three Chinese domains at the MEE stations. Like the US cases (Figures 2a and 2b), the fitted lines with lognormal distribution in Figures 2c–2e

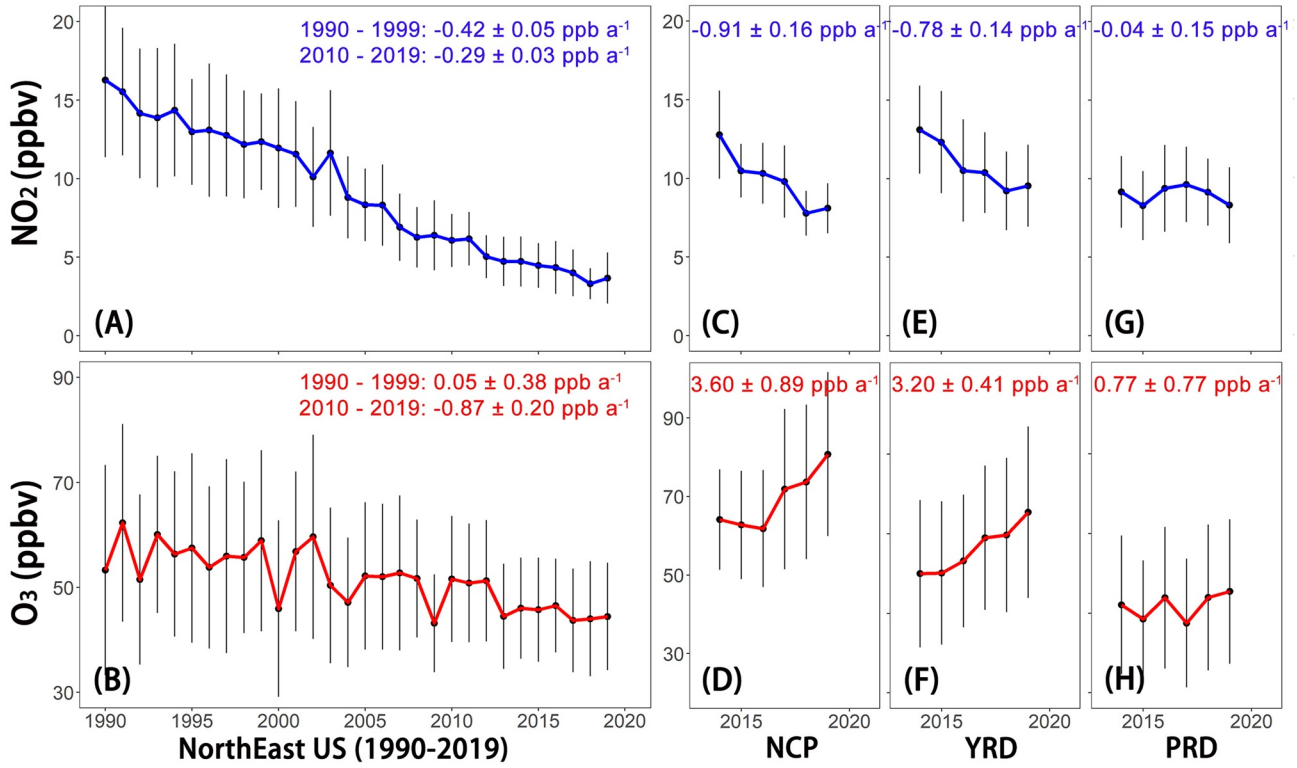

Figure 1. Regional averages of daily mean O_3 and NO_2 concentrations (12:00–19:00 local time in June–Aug) from US AQS and Chinese MEE stations with both O_3 and NO_2 measurements. The error bars represent standard deviation. The domain definitions are shown in Figure 3. The locations of AQS and MEE stations are shown in Figure S5.

exhibit good agreement with surface observations, confirming the important role of nonlinear O_3-NO_x chemistry in the derived O_3-NO_2 relationships. We find similar fitted lines in NCP/YRD/PRD (Figures 2c–2e) and northeast US in the 1990s (Figure 2a), that is, high O_3 with medium NO_2 and low O_3 with high or low NO_2, as well as large discrepancy with northeast US in the 2010s (Figure 2b), that is, high O_3 in the high NO_2 side, suggesting similar (different) O_3 chemical regimes between NCP/YRD/PRD in 2014–2019 and northeast US in the 1990s (2010s). As for the historical record in the US, the decline of anthropogenic NO_x emissions has led to a shift in O_3 chemical regimes over NCP and YRD, meeting the turning points between NO_x- and VOC-limited regimes around 2019 (Figures 2c and 2d). Consistent with the modeled 2013–2017 O_3 change by Liu and Wang (2020), our observation-based analysis indicates the reductions of NO_2 concentrations led to increases of surface O_3 over NCP and YRD in 2014–2019.

The remotely sensed measurements of OMI formaldehyde (HCHO) column data have been widely used to constrain anthropogenic and biogenic VOCs emissions (Cao et al., 2018; Stavrakou et al., 2018). Here we use the OMI HCHO column data as a proxy for the surface VOCs concentrations to investigate the relations between O_3 and VOCs. As shown in Figures 2f–2i, we find linear correlations between changes in O_3 and HCHO, which include effects such as O_3-VOCs, VOCs-temperature and O_3-temperature. The reported increase of anthropogenic VOCs emissions may have a noticeable influence on surface O_3 concentrations in 2003–2015 in China (Sun et al., 2019). Furthermore, M. Li et al. (2019) suggests limited change of anthropogenic VOCs emissions in China since 2013: VOCs emissions increased by about 3% in 2013–2017. It is consistent with the observed HCHO changes: we did not find noticeable trends in OMI HCHO in 2014–2019 in Figures 2g–2i.

2.2. Model-Based Responses of O_3 to NO_x and VOCs Emissions

Here we interpret the observed O_3-NO_2 and O_3-HCHO relationships using the GEOS-Chem chemical transport model with 0.5° × 0.625° horizontal resolution. Figure S5 shows modeled summertime surface O_3

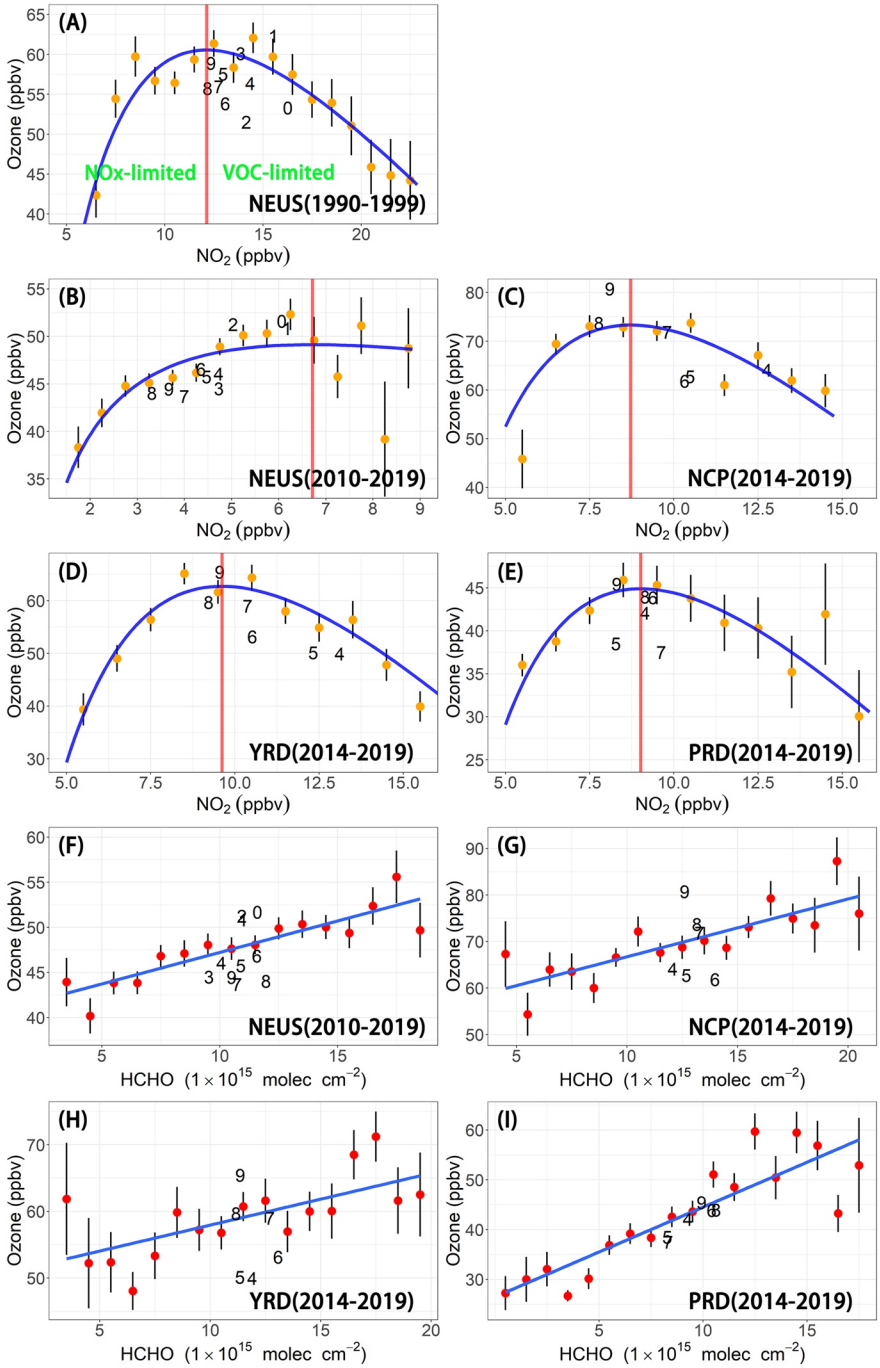

Figure 2. (a–e) Observed O_3-NO_2 relationships from US AQS and Chinese MEE stations with both O_3 and NO_2 measurements (June–August). The orange dots represent regional averages of daily mean O_3 and NO_2 concentrations (12:00–19:00 local time), binned into 1 ppb NO_2 increments (0.5 ppb for panel (b). The blue line is the lognormal fitting line. (f–i) Observed O_3-HCHO relationships from US AQS and Chinese MEE stations and regional averages of OMI HCHO data. The red dots represent regional averages of daily mean O_3 (12:00–19:00 local time) and HCHO column abundances, binned into 1e15 mol/cm^2 HCHO increments. The blue line is the linear fitting line. The error bars represent standard error. The numbers (0–9) represent the summertime mean O_3 and NO_2 abundances, and a number itself corresponds a year with the year's last digit during the corresponding period. All daily data (rather than only binned data) are used to produce the fitted lines.

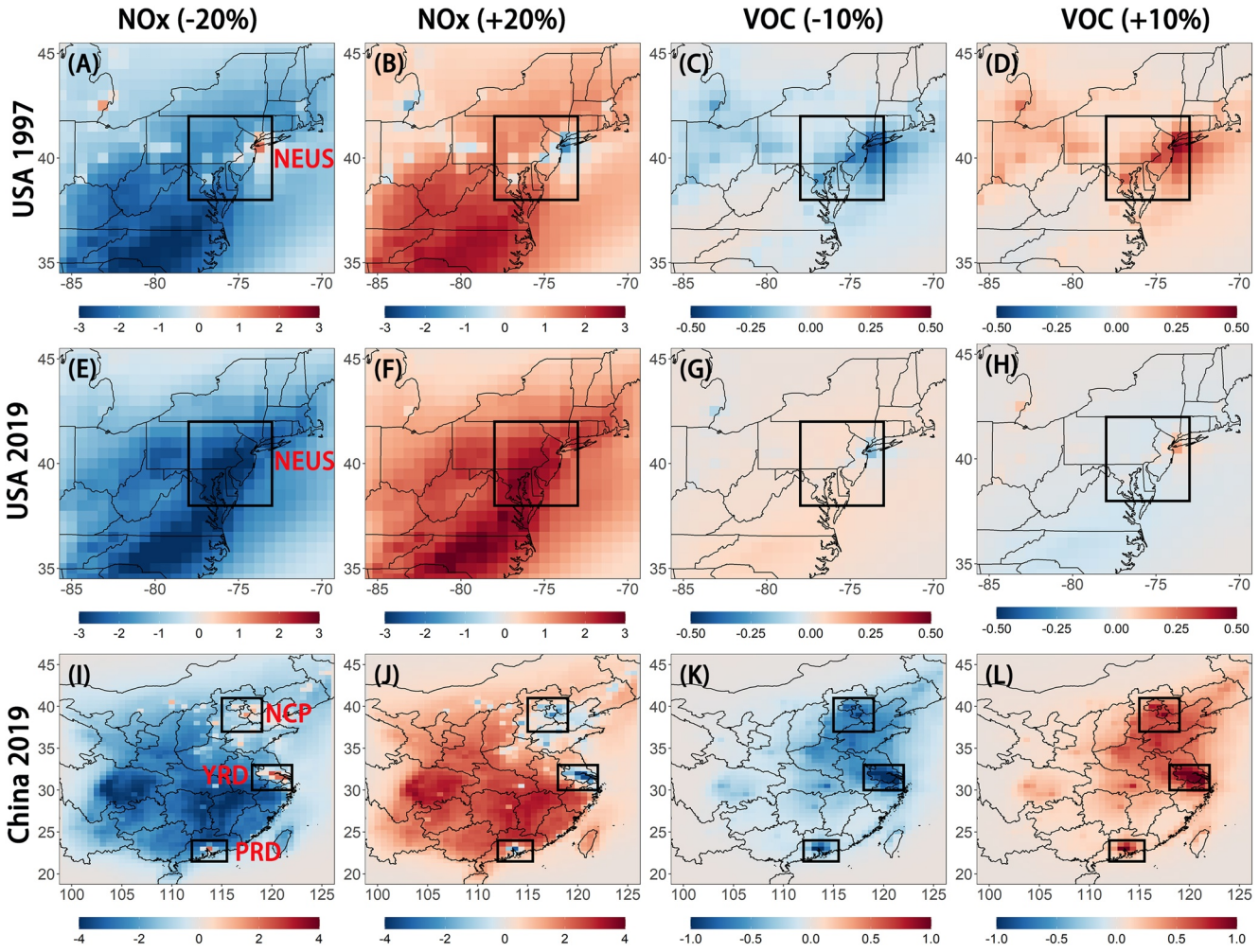

Figure 3. Modeled responses of O_3 to NO_x and VOCs emissions. Simulations are performed in 1997 (US) and 2019 (US and China) by perturbing anthropogenic NO_x or VOCs emissions in GEOS-Chem model. The black boxes define the domains of NEUS (38°–42° N, 78°–73° W), NCP (36°–40.5° N, 114°–120° E), YRD (30°–33° N, 118°–122° E) and PRD (21.5°–24° N, 112°–115.5° E).

concentrations over northeast US (in 1997 and 2019) and east China in 2019. The simulation timeframes (i.e., 1997 and 2019) are chosen because they are close to the observed turning points (Figure 2). The modeled surface O_3 distributions provided by GEOS-Chem demonstrate broadly good agreement with surface measurements, as well as the decline of surface O_3 over the US from 1997 to 2019 (Figure S5), supporting the application of the model simulations in explaining the observed O_3 changes.

Figures 3a–3d show the modeled responses of surface O_3 to perturbations in anthropogenic NO_x and VOCs emissions over northeast US in 1997. Over polluted regions such as New York, the modeled responses of O_3 to NO_2 are negative; responses of O_3 to VOCs are positive, suggesting a VOC-limited regime, whereas the rest of northeast US is NO_x-limited. By contrast, the modeled responses of O_3 to NO_2 are positive; responses of O_3 to VOCs are insensitive in 2019, suggesting that O_3 formation is NO_x-limited (Figures 3e–3h). The widespread distribution of a NO_x-limited regime over the northeast US, as well as the shift of the O_3 regime in the past decades is consistent with reported results (He et al., 2020; Jin et al., 2020). Figures 3i–3l suggest transitional or weak VOC-limited regimes in NCP, YRD, and PRD in 2019, consistent with the reported O_3 regime (M. Jiang et al., 2018; Jin et al., 2017). Our model simulations show a widespread distribution of transitional or NO_x-limited regimes over rest of China, consistent with the reported distribution of O_3 regime in China (Wang et al., 2019).

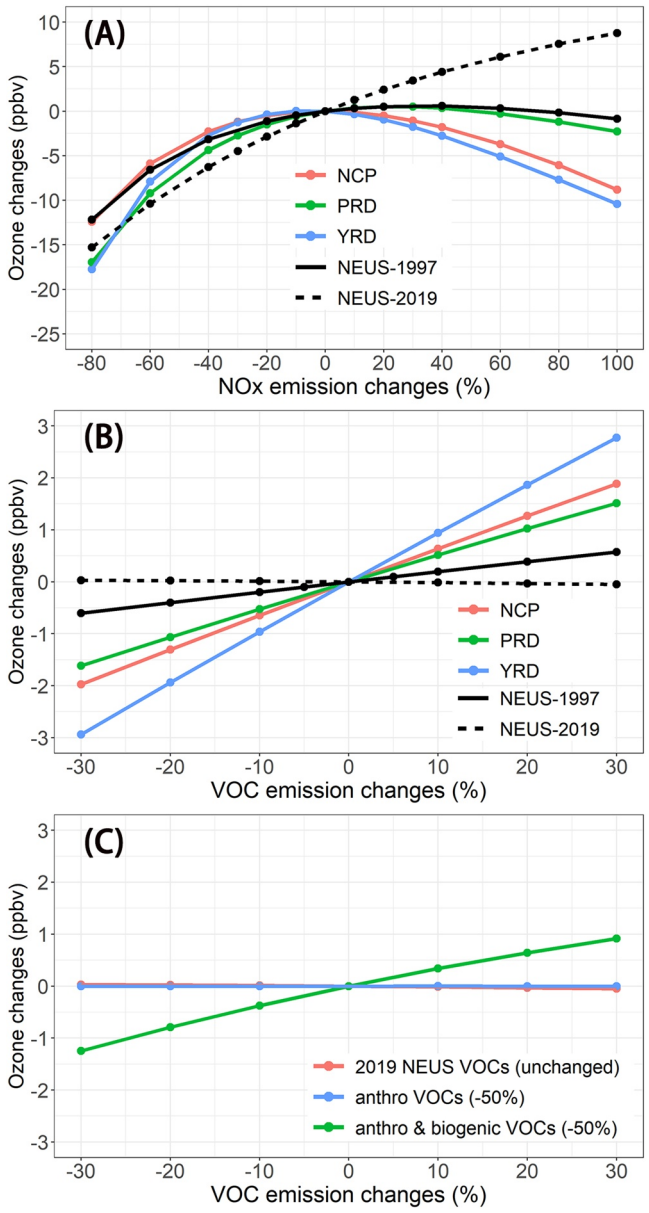

Figure 4. Modeled responses (normalized) of O_3 to NO_x and VOCs emissions. Simulations are performed in 1997 (US) and 2019 (US and China) in GEOS-Chem model (a) perturb anthropogenic NO_x emissions; (b) perturb anthropogenic VOCs emissions; (c) US 2019: perturb anthropogenic VOCs emissions (orange), reduce total anthropogenic VOCs emissions by 50% and then perturb anthropogenic VOCs emissions (blue), reduce total anthropogenic and biogenic VOCs emissions by 50% and then perturb anthropogenic and biogenic VOCs emissions (green).

As shown in Figure 3, the modeled responses of surface O_3 to perturbations in anthropogenic NO_x and VOCs emissions are similar between NCP/YRD/PRD in 2019 and northeast US (particularly, New York) in 1997. The model simulations (normalized in 2019 or 1997, Figures 4a and 4b) exhibit noticeable turning points in O_3-NO_x relationships in NCP, YRD, PRD, and northeast US (in 1997), as well as linear correlations between O_3 and anthropogenic VOCs emissions. The consistency between observation- and model-based responses of surface O_3 to precursors supports the observation-based analysis, that is, the similarity in O_3 chemical regimes between eastern China industrialized areas in 2014–2019 and northeast US in the 1990s with turning points between NO_x- and VOC-limited regimes around 2019 in China. Furthermore, Figure 3 shows noticeable urban and rural discrepancies, particularly, in YRD and PRD, suggesting large difference in O_3 chemical regimes between urban and rural areas.

The observations suggest the northeast US is in a weak NO_x-limited regime in 2019: there is turning point in the observation-based O_3-NO_2 relationship (Figure 2b); the O_3-HCHO relationship shows positive correlation (Figure 2f). By contrast, the model suggests a strong NO_x-limited regime: there is no turning point in the model-based O_3-NO_x relationship (Figure 4a); the O_3-VOCs response is flat (Figure 4b). This discrepancy suggests possible underestimation in the modeled NO_2/VOCs ratio in the 2010s. As shown in Figure 4c, the modeled O_3-VOCs relationship with reduced anthropogenic and biogenic VOCs emissions matches better with observations. It could be associated with the reported overestimation of biogenic VOCs emissions (MEGAN 2.1) in the model simulations (Kaiser et al., 2018; Wang et al., 2017), as well as the reported overestimation of NO_x emission reductions (Z. Jiang et al., 2018).

2.3. Impacts of $PM_{2.5}$ and Meteorology Changes

Following Li, Jacob, Liao, Zhu (2019), here we assess the impact of $PM_{2.5}$ controls on O_3 formation. In order to consider the heterogeneous uptake of atmospheric radicals and radiation attenuation that would influence O_3 formation, Figure 5 shows the summertime O_3 (12:00–19:00 local time) and $PM_{2.5}$ (24-h average) relationship. The trends for both O_3 and $PM_{2.5}$ are removed following Li, Jacob, Liao, Zhu (2019). The common dependence on meteorology resulted in positive correlations between O_3 and $PM_{2.5}$ changes, followed by flat correlations when $PM_{2.5}$ is high (>60 μg/m^3) in NCP and YRD. As suggested by Li, Jacob, Liao, Zhu (2019), the flat correlation reflects the suppression of high $PM_{2.5}$ on O_3 formation. However, the effective controls of $PM_{2.5}$ have resulted in dramatic reductions of $PM_{2.5}$ concentrations from about 70 μg/m^3 in NCP in 2014 to about 30 μg/m^3 in 2019, which is lower than 60 μg/m^3. As shown in Li, Jacob, Liao, Zhu (2019), reductions of $PM_{2.5}$ are expected to lead to increase of surface O_3 by up to 20 ppb, when $PM_{2.5}$ is about 70 μg/m^3; and by up to 5 ppb, when $PM_{2.5}$ is about 30 μg/m^3. Therefore, the impact of $PM_{2.5}$ controls on O_3 formation in China is expected to be weaker in 2019 than that in 2014. Furthermore, Figure 5b exhibits flat correlation over NCP with $PM_{2.5}$ < 30 μg/m^3, which is different with the positive correlations over other domains.

Besides the anthropogenic factors (NO_x, VOCs, and $PM_{2.5}$), the observed short-term variability in surface O_3 is also affected by natural processes such as changes in meteorology conditions (Dang et al., 2021; Han et al., 2020; Li et al., 2019). Following Han et al. (2020), we analyze the local and synoptic meteorological

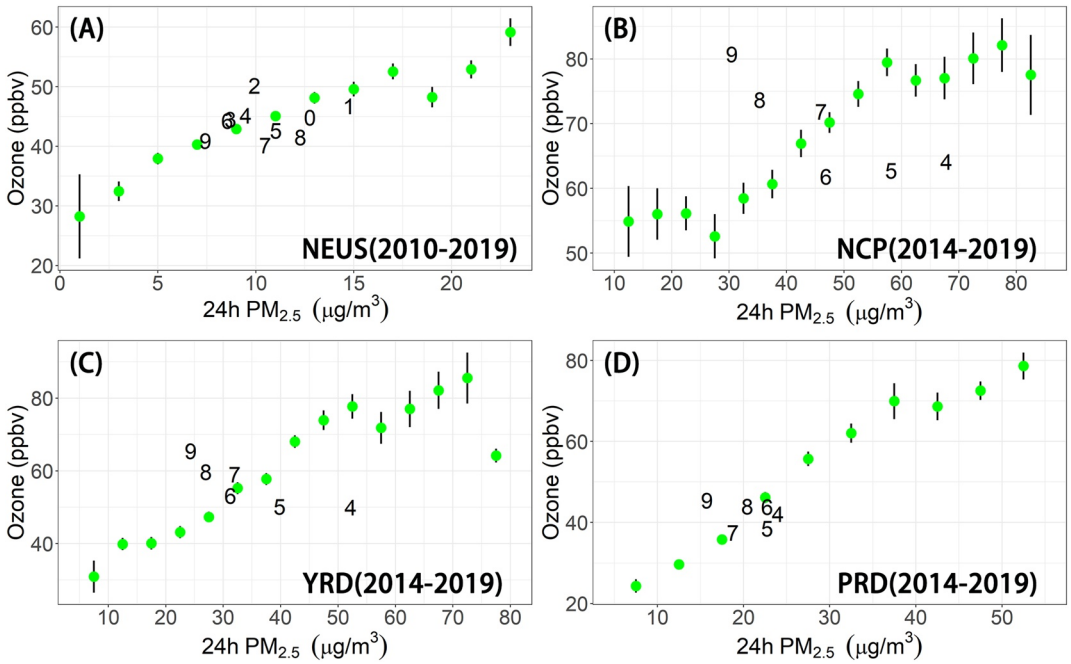

Figure 5. Observed O_3-$PM_{2.5}$ relationships from US AQS and Chinese MEE stations with both O_3 and $PM_{2.5}$ measurements. The green dots represent regional averages of daily mean O_3 (12:00–19:00 local time) and $PM_{2.5}$ (24-h average) abundances, binned into 5 μg/m^3 $PM_{2.5}$ increments (2 μg/m^3 for panel a). The error bars represent standard error. The numbers (0–9) represent the summertime mean O_3 and $PM_{2.5}$ abundances, and a number itself corresponds a year with the year's last digit during the corresponding period. Following Li, Jacob, Liao, Zhu (2019), multi-year trends of O_3 and $PM_{2.5}$ (green dots) were removed.

influences on surface O_3 concentrations in China with multiple linear regression by considering 10 local meteorological variables (relative humidity, cloud fraction, temperature, planetary boundary layer height, wind speeds/geopotential height, and sea level pressure), and two synoptic weather factors identified through the singular value decomposition (SVD) analysis of spatial correlations. As shown in Figure S2, we find the contribution from changes in meteorological conditions to the increase of surface O_3 in 2014–2019 over NCP is about 17%, which is lower than the 42% reported by K. Li et al. (2020) and 49% reported by Dang et al. (2021). Despite uncertainties in magnitude, our analysis agrees with the previous studies that changes in meteorological conditions are an important influence on surface O_3 concentrations.

3. Conclusions

The evolution of O_3 nonlinear chemistry in the US over the past three decades illustrates the effects of emission controls on O_3 pollution in regions dominated by anthropogenic emission, which allows us to predict the evolution of O_3 pollution in analogous regions of China. The similarity in the O_3 chemical regimes between eastern China industrialized areas in 2014–2019 and northeast US in the 1990s indicates that initial reductions of NO_2 concentrations have led to higher surface O_3 concentrations. As shown in Figures 2c and 2d, the fitted lines suggest about 8 ppb increase of surface O_3 over NCP and YRD due to NO_2 decreases, contributed to about 40% of observed O_3 increases. The deterioration in O_3 pollution in China in 2014–2019 is driven by the combined effects of NO_x emission controls, as well as changes in VOCs, $PM_{2.5}$ and meteorological conditions. Similar to the US in the 2010s, the severe O_3 pollution in China can be mitigated because of the shift of O_3 chemical regimes with turning points between NO_x- and VOC-limited regimes around 2019, and the weakened impacts of $PM_{2.5}$ decline on O_3 formation enhancement due to $PM_{2.5}$ controls.

Stricter controls of NO_x emissions, guided by monitoring O_3 nonlinear chemistry evolution as shown in this work, are expected to lead to an effective decrease in surface O_3 concentrations over industrialized areas. While the prospect for controlling O_3 pollution in China is optimistic, the historical experience of the US in 1990–2019 suggests that significant improvements in O_3 pollution may not be reached immediately

because the effects of NO_x emission controls can be diminished by VOCs and meteorology changes. We advise further studies into the effects of changing $PM_{2.5}$ on O_3 formation in China, particularly, the O_3-$PM_{2.5}$ relationship with $PM_{2.5} < 30$ μg/m^3 (i.e., daily mean summertime $PM_{2.5}$ in NCP in 2019). We also suggest further exploration of the underestimated NO_2/VOCs ratios over the northeast US, with associated impacts on O_3 trends.

Data Availability Statement

We thank the China Ministry of Ecology and Environment and the United States Environmental Protection Agency for providing the surface O_3, NO_2 and $PM_{2.5}$ measurements (from https://aqs.epa.gov/aqsweb/airdata/download_files.html#Row: Tables of Hourly Data). We thank the providers of the OMI tropospheric HCHO column data (from https://disc.gsfc.nasa.gov/datasets/OMHCHOd_003/summary). The meteorological variable data are downloaded from https://rda.ucar.edu/datasets/ds083.2. The source code, run directories for model simulations and MEE surface measurements can be downloaded from https://doi.org/10.5281/zenodo.5030857.

Acknowledgments
The numerical calculations in this paper have been done on the supercomputing system in the Supercomputing Center of University of Science and Technology of China. This work was supported by the Hundred Talents Program of Chinese Academy of Science, the Fundamental Research Funds for the Central Universities, and National Natural Science Foundation of China (41721002).

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