Large discrepancy between observations and simulations: Implications for urban air quality in China

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Abstract

Chemical transport models (CTMs) have been widely used to provide instructions for the control of ozone (O₃) pollution. However, we find large discrepancies between observation- and model-based urban O₃ chemical regimes: volatile organic compound (VOC)-limited regimes over N. China and weak nitrogen oxides (NOₓ)-limited regimes over S. China in observations, in contrast to simulations with widespread distributions of strong NOₓ-limited regimes. The conflicting O₃ evolutions are caused by underestimated urban NOₓ concentrations and the possible overestimation of biogenic VOC emissions. Reductions in NOₓ emissions, in response to regulations, have thus led to an unintended deterioration of O₃ pollution over N. China provinces,
for example, an increase in surface O_3 by approximately 7 ppb over the Sichuan Basin (SCB) in 2014-2020. The NO_x-induced urban O_3 changes resulted in an increase in premature mortality by approximately 3000 cases in 2015-2020.

1. Introduction

Emission regulations have led to a significant decline in NO_x emissions in China [Jiang et al., 2022; Zheng et al., 2018], accompanied by dramatic increases in surface O_3 concentrations [Ma et al., 2019; Sun et al., 2019; Y-H Wang et al., 2020]. There are currently many efforts focusing on the opposite changes in surface O_3 and its precursors, with explanations including nonlinear O_3-NO_x response [Y Liu and Wang, 2020; Z Liu et al., 2021], the contribution from soil NO_x emissions [X. Lu et al., 2021], as well as changes in meteorological and fine particle (PM_{2.5}) conditions [Dang et al., 2021; K Li et al., 2019a; K Li et al., 2019b]. A recent study [Chen et al., 2021] further predicts mitigation of O_3 pollution over the highly industrialized North China Plain (NCP) and Yangtze River Delta (YRD) because NO_x controls have led to a shift of O_3 chemical regimes from a VOC-limited to a transitional regime, and thus, stricter controls of NO_x emissions are expected to lead to a decrease in O_3 pollution.

In contrast to highly industrialized E. China coast provinces, model studies suggested strong NO_x-limited regimes over inland China. Sustainable controls of NO_x emissions are suggested to lead to decreases in O_3 pollution in recent years and are expected to lead to further decreases in O_3 pollution in the future. For example, K Li et al. [2019a] suggested that NO_x reductions led to surface O_3 decrease over inland China by approximately 1 ppb/y in 2013-2017; Y Liu and Wang [2020] suggested that NO_x reductions led to a surface O_3 decrease over inland China by approximately 3 ppb in 2013-2017; Chen et al. [2021] suggested that a 20% reduction in NO_x emissions...
emissions in 2019 will lead to an approximately 2-4 ppb decrease in surface O$_3$ over inland China; Z Liu et al. [2021] suggested that a 20% reduction in NO$_x$ emissions in 2016 will cause an up to 8 ppb decrease in surface O$_3$ over inland China. However, the hypothesis of strong NO$_x$-limited regimes over inland China conflicts with the observed O$_3$ increase.

The difficulty in explaining the observed surface O$_3$ changes over inland China implies possible deficiencies in our understanding of O$_3$ evolution, which poses a potential barrier to making effective regulatory policies to control O$_3$ pollution. In this study, we provide a comparative analysis of the observation- and model-based responses of urban O$_3$ concentrations to changes in NO$_x$ emissions in China in 2014-2020, by integrating surface in situ observations of NO$_2$, VOC and O$_3$, the GEOS-Chem chemical transport model (with 0.5°×0.625° horizontal resolution), and a photochemical box model. The objective of this work is to explore the causes for the observed O$_3$ changes and to evaluate the capability of state-of-the-art CTM to capture air quality evolution. The impact of long-term O$_3$ exposure is further analyzed based on the exposure-response function to understand their health effects.

2. Methods

2.1 MEE surface NO$_2$ and O$_3$ measurements

We use surface in situ NO$_2$ and O$_3$ concentration data from the China Ministry of Ecology and Environment (MEE) monitoring network for the period of 2014-2020. These real-time monitoring stations can report hourly concentrations of criteria pollutants from over 360 cities in 2019. Concentrations were reported by the MEE in units of ug/m$^3$ under standard temperature (273 K) until 31 August 2018. This reference state was changed on 1 September 2018 to 298 K. We
converted the O₃ and NO₂ concentrations to ppb, and rescaled the post-August 2018 concentrations to the standard temperature (273 K) to maintain the consistency in the trend analysis.

2.2 GEOS-Chem model simulations

The GEOS-Chem chemical transport model (http://www.geos-chem.org, version 12-8-1) is driven by assimilated meteorological data of MERRA-2 with nested 0.5°×0.625° horizontal resolution. The GEOS-Chem model includes fully coupled O₃-NOₓ-VOC-halogen-aerosol chemistry. The chemical boundary conditions are updated every 3 hours from a global simulation with 4°×5° resolution. Emissions in GEOS-Chem are computed by the Harvard-NASA Emission Component (HEMCO). We refer the reader to Chen et al. [2021] for the details of the model configurations.

2.3 Photochemical box model

The photochemical box model (OBM) is configured with master chemical mechanisms (MCM v3.3.11; http://mcm.york.ac.uk/home.htt). The MCM-OBM model was designed to investigate the atmospheric oxidation processes of VOC species [X Liu et al., 2019; Xue et al., 2013; Xue et al., 2016]. The concentrations of sulfur dioxides (SO₂), carbon monoxide (CO), NOₓ and VOC, as well as meteorological parameters (atmospheric pressure, temperature, and relative humidity) from two monitoring sites in Chengdu city (in the SCB), were used as constraining parameters in the model. The MCM-OBM model simulations start at 12:00 local time for 8 hours, by inputting the observed O₃ concentration at the initial time. MCM-OBM simulations have been widely used to calculate the relative incremental reactivity to describe the response of O₃ to individual precursors [He et al., 2019; J Li et al., 2018; Tan et al., 2018; M Wang et al., 2020].

2.4 Long-term health impacts of surface O₃ change
We applied the following function to assess the mortality attributed to long-term O₃ exposure:  

\[ \Delta Health\ Impacts = y_0 \times (1 - e^{-\beta \Delta Exposure}) \times Population \]

where \( \Delta Health\ Impacts \) is the mortality change caused by the change in O₃ concentration in 2015-2020. \( y_0 \) is the baseline rates of specific diseases for various age groups, which we downloaded from the recent global burden of disease for 2019. \( \beta \) is the exposure-response function for the unit of exposure. The relative risk (RR) value was set to 1.08 (90% confidence interval: 1.06, 1.11) based on previous studies [Xiao Lu et al., 2020; Turner et al., 2016; Zhang et al., 2021]. In addition, we used the change in summertime MDA8 O₃ concentration in 2015-2020 (Fig. 4A) to represent \( \Delta Exposure \). Population is the number of people in different age groups living in the same grid box (0.1° × 0.1°) obtained from the Gridded Population of the World database [2018] for 2019, and then re-gridded to 1°×1° resolution.

3. Results and Discussions

3.1 Observation-based O₃-NO₂ relationships

Fig. 1 shows the summertime maximum daily 8-hour average (MDA8) O₃ from the China Ministry of Ecology and Environment (MEE) monitoring network in 2019. Only urban stations are considered in this work, and background stations were excluded. The station-based measurements are averaged and regrided to 1°×1° resolution. The observed surface O₃ exhibits latitude dependence, i.e., higher O₃ in N. China and lower O₃ in S. China. Fig. 2A-B show the summertime O₃-NO₂ relationship over four domains in China at the MEE stations in the period of 2014-2020. The data (dots) are regional averages of MDA8 O₃ and NO₂ concentrations, binned into 1 ppb NO₂ increments. The lognormal fitting lines in Fig. 2A demonstrate nonlinear O₃-NO₂ relationships with turning points between NOₓ- and VOC-limited regimes of approximately 9 ppb.
over the NCP, 10 ppb over the YRD, and 11 ppb over the PRD. We find shifts in O₃ chemical regimes from VOC-limited to transitional regimes over the NCP and YRD in 2014-2020. Our analysis suggests a NOₓ-limited regime over the PRD in 2020 (Fig. 2A) and a VOC-limited regime over the SCB (Fig. 2B).

An observation-based photochemical box model (OBM) was further employed to evaluate the O₃ regime in the SCB. As shown in Fig. 2C, the OBM model indicates increases in O₃ with 60-80% reductions in NOₓ concentrations in Chengdu city (in the SCB) in 2017 and 2018. Similarly, Fig. 2B suggests an increase in O₃ with 40% reductions in NO₂ concentrations in the urban areas in the SCB in 2017 and 2018. The consistent VOC-limited regimes in the two observation-based approaches (Fig. 2B and Fig. 2C) support the feasibility of our approach (Fig. 2A-B) to analyze the O₃ chemical regime. The discrepancy between the two observation-based approaches (i.e., 60-80% and 40%) may reflect the difference in O₃ regimes among different cities in the SCB. As shown in Fig. 2B, reductions in NOₓ emissions have led to an increase in surface O₃ in the SCB by approximately 7 ppb in 2014-2020 (based on the lognormal fitting line). Stricter controls of NOₓ emissions and regardless of VOC are therefore expected to lead to even more O₃ pollution in the SCB.

3.2 Unintended deterioration of O₃ pollution over inland China

As shown in Fig. 2B, the lognormal fitting approach allows us to approximately predict the response of O₃ to NO₂ change. For example, the difference in the O₃ concentrations along the lognormal fitting line in Fig. 2B represents the predicted change in O₃ concentration (dO₃) in response to the increase in NO₂ concentrations (dNO₂) over the SCB. Furthermore, Fig. 3A shows the summertime NO₂ concentrations from the MEE stations in 2019. The 8-hour range of surface NO₂ measurements is selected according to the time range of MDA8 O₃, and then averaged and
regrided to 1°×1° resolution. Following the prediction approach shown in Fig. 2B, lognormal fitting lines are produced for each 1°×1° grid in Fig. 3A, which allows us to predict grid-based O3 changes in response to NO2 in 2019.

Fig. 3B exhibits the response of surface O3 to 20% increases in NO2 concentrations in 2019. According to Jiang et al. [2022], we assume a 20% decline in anthropogenic NOx emissions in 2015-2019, and thus, Fig. 3B represents the observation-based O3 changes driven by NOx changes in China in 2015-2019, approximately. We find widespread decreases in surface O3 over N. China inland provinces in 2019 in response to 20% increases in NO2 concentrations. In contrast to the hypothesis of strong NOx-limited regimes [Chen et al., 2021; K Li et al., 2019a; Y Liu and Wang, 2020; Z Liu et al., 2021], the observation-based analysis (Fig. 3B) indicates that NOx controls have led to increases in surface O3 over N. China inland provinces. The 2018–2020 Chinese Clean Air Action plan called for a 9% decrease in NOx emissions [CSC, 2018], and thus, Fig. 3C further predicts surface O3 changes driven by a 10% decrease in NO2 concentrations. Continuous NOx controls, as shown in Fig. 3C, are thus predicted to result in the deterioration of O3 pollution over N. China inland provinces.

3.3 Discrepancy between observation- and model-based analyses

Here we further investigate the O3-NO2 responses using the GEOS-Chem chemical transport model at 0.5°×0.625° horizontal resolution. Fig. 3D shows the summertime NO2 concentrations from GEOS-Chem in 2019; Fig. 3E-F exhibit the modeled responses of surface O3 to perturbations in anthropogenic NOx emissions (Run 1, See Table1). The modeled NO2 and O3 are sampled at the locations and times of the MEE surface measurements, and then averaged and regrided to 1°×1° resolution. Because only urban stations are considered in this work, the sampled simulations represent modeled urban NO2 concentrations (Fig. 3D) and O3 responses (Fig. 3E-F). The MEE
NO$_2$ observations are provided by chemiluminescence analyzers, in which NO$_2$ is catalytically transformed into nitrogen oxide (NO) by a molybdenum converter. Following F Liu et al. [2018], the modeled NO$_2$ concentrations in Fig. 3D are adjusted using the ratios of NO$_y$/NO$_2$. Here $NO_y = NO_2 + \Sigma AN + 0.95 \times PAN + 0.35 \times HNO_3$, where $\Sigma AN$ is the sum of all alkyl nitrate concentrations.

We find large discrepancies between the observations and simulations: 1) the sampled NO$_2$ concentrations (Fig. 3D) are comparable with surface NO$_2$ observations (Fig. 3A) over the industrialized NCP and YRD but lower in the rest of China. F Liu et al. [2018] found that modeled NO$_2$ concentrations are higher than surface NO$_2$ observations over industrialized NCP and YRD, and broadly lower in the rest of China. Different station types, i.e., all stations in F Liu et al. [2018] and urban stations in this work, as well as different periods, i.e., daily averages in F Liu et al. [2018] and MDA8 (based on O$_3$) in this work may affect the comparison between observations and simulations. For example, Fig. S1 shows large differences in the ratios of NO$_2$/NO$_y$ between different periods. 2) conflicting responses of O$_3$ to NO$_2$ changes between observation-based and model-based analyses in the rest of China: VOC-limited regimes (blue in Fig. 3B) over N. China and weak NO$_x$-limited regimes (slight red in Fig. 3B) over S. China in observations, in contrast to widespread distributions of strong NO$_x$-limited regimes in simulations (red in Fig. 3E).

3.4 Impacts of biased urban NO$_2$ and VOC concentrations

The underestimated urban NO$_2$ concentrations in Fig. 3D pose a significant barrier to simulating urban O$_3$ evolution. GEOS-Chem simulations (Run 2) adjust anthropogenic and soil NO$_x$ emissions over urban grids (i.e., grids have MEE urban stations) using the ratios of MEE-modeled NO$_2$. Emissions over the highly industrialized NCP, YRD and PRD are not adjusted because the modeled O$_3$ evolutions match well with observations in these areas [Chen et al., 2021].
The adjustment of NO\textsubscript{x} emissions over urban grids led to enhancements of sampled NO\textsubscript{2} concentrations in Fig. 3G; however, they are still noticeably lower than observations (Fig. 3A), indicating influences from strong regional transport. Consequently, we further adjust regional background NO\textsubscript{x} emissions based on the ratios of averaged MEE/modeled NO\textsubscript{2} within neighboring grids (Run 3). As shown in Fig. 3J, the sampled NO\textsubscript{2} concentrations in Run 3 match well with the observed urban NO\textsubscript{2} concentrations (Fig. 3A). It should be noted that the adjustments of NO\textsubscript{x} emissions are designed to cover the influences of coarse model resolutions and strong regional transport on urban air quality simulations, which cannot be explained as underestimation in NO\textsubscript{x} emissions.

As shown in Fig. 3, the consistent NO\textsubscript{2} concentrations between the observations and simulations lead to significant improvements in the modeled urban O\textsubscript{3} evolution. For example, the observation-based analysis predicts a -1.8 ppb decrease in surface O\textsubscript{3} due to a 20% increase in anthropogenic NO\textsubscript{x} emissions in the SCB in 2019 (Fig. 3B); in contrast, the modeled responses are increases in surface O\textsubscript{3} by 2.8 ppb (Fig. 3E), 1.4 ppb (Fig. 3H) and 0.0 ppb (Fig. 3K). In a recent study, Chen et al. [2021] found that a 50% decrease in biogenic VOC emissions can improve surface O\textsubscript{3} simulations over the United States because of the reported overestimation of biogenic VOC emissions (MEGAN 2.1) in GEOS-Chem simulations [Kaiser et al., 2018; Wang et al., 2017]. Following Chen et al. [2021], GEOS-Chem simulations (Run 4) further reduce biogenic VOC emissions in China by 50%. The reductions in biogenic VOC emissions further improved the agreement between observations and simulations: the responses of surface O\textsubscript{3} to a 20% increase in anthropogenic NO\textsubscript{x} emissions are: -1.8 ppb (Fig. 3B) and -1.4 ppb (Fig. 3N) in the SCB.

Finally, Fig. 4A shows the predicted responses of O\textsubscript{3} concentrations (\textit{d}O\textsubscript{3}) to observed NO\textsubscript{2} changes (\textit{d}NO\textsubscript{2}) in 2015-2020. Changes in NO\textsubscript{2} concentrations have led to an increase of surface
O$_3$ over provinces such as Anhui, Shanxi and Hebei (Fig. 4B). We then calculated the health impacts of long-term O$_3$ exposure. As shown in Fig. 4C, we find a noticeable increase in urban premature mortality burdens, by approximately 1500 cases in Anhui, and 500 cases in Zhe Jiang and Hebei Provinces. In contrast, changes in O$_3$ pollution led to a decrease in urban premature mortality burdens by approximately 1500 cases in Guangdong Province. It should be noted that our analysis is designed to predict urban O$_3$ pollution and health impacts (i.e., grids with urban MEE stations). The premature mortality burdens in this work are thus expected to be lower than those in the literature based on national total populations.

4. Conclusion

Chemical transport models, as powerful tools, have been widely used to provide instructions for the control of worldwide O$_3$ pollution. However, the comparative analysis in this work demonstrates large discrepancies between observation- and model-based urban O$_3$ chemical regimes: VOC-limited regimes over N. China; weak NO$_x$-limited regimes over S. China in the observations, in contrast to widespread distributions of strong NO$_x$-limited regimes in the simulations. The conflicting O$_3$ evolutions between observations and simulations are caused by underestimated urban NO$_x$ concentrations associated with coarse model resolutions and strong regional transport, as well as possible overestimation of biogenic VOC emissions. Different from the hypothesized decreases in surface O$_3$ driven by NO$_x$ control [Chen et al., 2021; K Li et al., 2019a; Y Liu and Wang, 2020; Z Liu et al., 2021], reductions in NO$_x$ emissions have led to an unintended deterioration of O$_3$ pollution in 2014-2020 over N. China inland provinces. The NO$_2$-induced O$_3$ pollution changes resulted in an increase in premature mortality by approximately 3000 cases in 2015-2020. While NO$_x$ control is an effective pathway to mitigate O$_3$ pollution over S. China and E. coast provinces, our analysis highlights the importance of VOC controls for O$_3$
pollution mitigation over N. China inland provinces.

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Tables and Figures

**Table. 1.** GEOS-Chem standard (Run 1) and sensitivity (Runs 2-4) simulations with 0.5°×0.625° horizontal resolution in Jun-Aug 2019. The scaling factors (A1) to enhance anthropogenic and soil NOₓ emissions over urban grids (i.e., grids have MEE urban stations) and urban grids+ regional background, based on the ratios between MEE and sampled NO₂ concentrations, are shown in Fig. S2. NOₓ (A1) and VOC (A3) emissions over highly industrialized NCP, YRD and PRD are not adjusted.

**Fig. 1.** Summertime MDA8 O₃ (with unit ppb) in 2019 from the MEE stations. The station-based measurements are averaged and re-grided to 1°×1° resolution. Only urban stations are considered. The black boxes define the domains used in this work. The star represents the location of Chengdu City in SCB.

**Fig. 2.** (A-B) Observed summertime O₃-NO₂ relationships from MEE stations with both O₃ and NO₂ measurements. The dots represent regional averages of MDA8 O₃ and NO₂ concentrations, binned into 1 ppb NO₂ increments. The 8-hour range of surface NO₂ measurements is selected according to the time range of MDA8 O₃. The lines are lognormal fitting lines. 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 2014-2020. (C) 8-hour averaged responses of O$_3$ to NO$_x$ changes at the SL (103.93°N, 30.58°E, 20170801-20170815) and JPJ (104.05°N, 30.66°E, 20180601-20180610) sites in Chengdu, Sichuan Province. The simulations are performed using a photochemical box model with inputs of SO$_2$, CO, NO$_x$ and VOC concentrations and meteorological parameters from observations. Positive responses represent increases of O$_3$ within 8 hours (12-19 local time) due to a decrease of NO$_x$, indicating a VOC-limited regime.

**Fig. 3.** (A) Observed surface NO$_2$ concentrations from the MEE stations in Jun-Aug 2019. The station-based measurements are averaged and re-gridded to 1°×1° resolution. (B-C) Predicted responses of MDA8 O$_3$ in 2019 to NO$_2$ changes based on the lognormal fitting lines. (D) Modeled surface NO$_2$ concentrations (Run1). The modeled NO$_2$ in panel D are adjusted using the ratios of $NO_y/NO_2$ to consider the influences from reactive oxidized nitrogen compounds in the chemiluminescence analyzers. (E-F) Modeled responses of MDA8 O$_3$ in 2019 to NO$_x$ emission changes. (G-O) Similar to panels D-F, but for sensitivity simulations (Runs 2-4). The modeled NO$_2$ and O$_3$ are sampled at the locations and times of MEE surface measurements, and then averaged and re-gridded to 1°×1° resolution. The 8-hour range of surface NO$_2$ measurements is selected according to the time range of MDA8 O$_3$. The unit is ppb.

**Fig. 4.** (A-B) NO$_2$-induced urban O$_3$ changes in 2015-2020 based on the lognormal fitting method with unit ppb. (C) Changes in urban premature mortality burdens due to predicted O$_3$ changes.

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Table 1. GEOS-Chem standard (Run 1) and sensitivity (Runs 2–4) simulations with 0.5°×0.625° horizontal resolution in Jun-Aug 2019. The scaling factors (A1) to enhance anthropogenic and soil NOx emissions over urban grids (i.e., grids have MEE urban stations) and urban grids+regional background, based on the ratios between MEE and sampled NO2 concentrations, are shown in Fig. S2. NOx (A1) and VOC (A3) emissions over highly industrialized NCP, YRD and PRD are not adjusted.

|       | Enhanced anthro + soil NOx (A1) | anthro NOx Factors (A2) | biogenic VOC Factors (A3) |
|-------|----------------------------------|-------------------------|---------------------------|
| Run1  | #1 (Base)                        | N/A                     | 1.0                       | 1.0                       |
|       | #2                               | 1.2                     | 1.0                       | 1.0                       |
|       | #3                               | 0.9                     | 1.0                       | 1.0                       |
| Run2  | #1 (Base)                        | urban grids             | 1.0                       | 1.0                       |
|       | #2                               |                         | 1.2                       | 1.0                       |
|       | #3                               |                         | 0.9                       | 1.0                       |
| Run3  | #1 (Base)                        | urban grids + regional backgrounds | 1.0 | 1.0 |
|       | #2                               |                         | 1.2                       | 1.0                       |
|       | #3                               |                         | 0.9                       | 1.0                       |
| Run4  | #1 (Base)                        | urban grids + regional backgrounds | 1.0 | 0.5 |
|       | #2                               |                         | 1.2                       | 0.5                       |
|       | #3                               |                         | 0.9                       | 0.5                       |

Fig. 1. Summertime MDA8 O3 (with unit ppb) in 2019 from the MEE stations. The station-based measurements are averaged and re-grided to 1°×1° resolution. Only urban stations are considered. The black boxes define the domains used in this work. The star represents the location of Chengdu City in SCB.
Fig. 2. (A-B) Observed summertime O₃-NO₂ relationships from MEE stations with both O₃ and NO₂ measurements. The dots represent regional averages of MDA8 O₃ and NO₂ concentrations, binned into 1 ppb NO₂ increments. The 8-hour range of surface NO₂ measurements is selected according to the time range of MDA8 O₃. The lines are lognormal fitting lines. The error bars represent standard error. The numbers (0 - 9) represent the summertime mean O₃ and NO₂ abundances, and a number itself corresponds a year with the year’s last digit during 2014-2020. (C) 8-hour averaged responses of O₃ to NOₓ changes at the SL (103.93°N, 30.58°E, 20170801-20170815) and JPJ (104.05°N, 30.66°E, 20180601-20180610) sites in Chengdu, Sichuan Province. The simulations are performed using a photochemical box model with inputs of SO₂, CO, NOₓ and VOC concentrations and meteorological parameters from observations. Positive responses represent increases of O₃ within 8 hours (12-19 local time) due to a decrease of NOₓ, indicating a VOC-limited regime.
Fig. 3. (A) Observed surface NO$_2$ concentrations from the MEE stations in Jun-Aug 2019. The station-based measurements are averaged and re-gridded to 1°×1° resolution. (B-C) Predicted responses of MDA8 O$_3$ in 2019 to NO$_2$ changes based on the lognormal fitting lines. (D) Modeled surface NO$_2$ concentrations (Run1). The modeled NO$_2$ in panel D are adjusted using the ratios of $NO_y/NO_2$ to consider the influences from reactive oxidized nitrogen compounds in the chemiluminescence analyzers. (E-F) Modeled responses of MDA8 O$_3$ in 2019 to NO$_x$ emission changes. (G-O) Similar to panels D-F, but for sensitivity simulations (Runs 2-4). The modeled NO$_2$ and O$_3$ are sampled at the locations and times of MEE surface measurements, and then averaged and re-gridded to 1°×1° resolution. The 8-hour range of surface NO$_2$ measurements is selected according to the time range of MDA8 O$_3$. The unit is ppb.
Fig. 4. (A-B) NO$_2$-induced urban O$_3$ changes in 2015-2020 based on the lognormal fitting method with unit ppb. (C) Changes in urban premature mortality burdens due to predicted O$_3$ changes.
Supplemental Information

Fig. S1. Ratios of $\frac{NO_2}{NO_y}$ in GEOS-Chem simulations in Jun-Aug 2019. Here $NO_y = NO_2 + \Sigma AN + 0.95 \times PAN + 0.35 \times HNO_3$, where $\Sigma AN$ is the sum of all alkyl nitrate concentrations. (A) MDA8 based on $O_3$; (B) Daily averages.

Fig. S2. (A) Ratios of MEE/Modeled NO$_2$ (0.5°×0.625°) over urban grids (i.e., grids have MEE urban stations). The modeled NO$_2$ are adjusted using the ratios of $\frac{NO_y}{NO_2}$. (B) Besides urban grids, adjusting regional background NO$_2$ based on the averaged MEE/Modeled NO$_2$ within neighboring ±1 grid domain (1.5°×1.875°); if no neighboring MEE station was found, then searching neighboring ±2 grid domain (2.5°×3.125°); the search stopped at neighboring ±4 grid domain (4.5°×5.625°).