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Source contributions of surface ozone in China using an adjoint sensitivity analysis

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Abstract

Air pollution has become an adverse environmental problem in China, resulting in serious public health impacts. This study advanced and applied the CMAQ adjoint model to quantitatively assess the source-receptor relationships between surface ozone (O_3) changes over different receptor regions and precursor emissions across all locations in China. Five receptor regions were defined based on the administrative division, including northern China (NC), southern China (SC), Pearl River Delta region (PRD), Yangtze River Delta region (YRD), and Beijing-Tianjin-Hebei region (BTH). Our results identified the different influential pathways of atmospheric processes and emissions to O_3 pollution. We found that the atmospheric processes such as horizontal and vertical advection could offset the O_3 removal through chemical reactions in VOC-limited areas inside the receptor regions. In addition, O_3 pollution can be induced by transport of O_3 directly or its precursors. Our results of relative source contributions to O_3 show that transboundary O_3 pollution was significant in SC, NC and YRD, while the O_3 pollution in PRD and BTH were more contributed by local sources. Anhui, Hubei and Jiangsu provinces were the three largest source areas of NO_x and VOC emissions to O_3 in SC (>52%) and YRD (>69%). NO_x and VOC emissions from Tianjin and Beijing were the largest contributors to O_3 in NC (>34%) and BTH (>51%). PRD was the dominant source areas of NO_x (>89%) and VOC emissions (~98%) to its own regional O_3 .

Graphical Abstract

Appendix A. Supplementary data

Keywords

Surface ozone; Source-receptor relationships; Ozone-sensitive regime

1. Introduction

Ambient air pollution has become one of the most severe environmental issues in China due to its rapid industrialization and urbanization accompanied by increasing anthropogenic emissions (Ning et al., 2018; Xie et al., 2017; Yin et al., 2017). Ozone (O_3), as a strong oxidant, is the third most important greenhouse gas and is detrimental to human health, crop productivity and natural ecosystem (Feng et al., 2015; Gu et al., 2018; Karlsson et al., 2017). According to the national monitoring results, the mean daily 8-hour maximum O_3 concentrations in China increased from 69.5ppbv in 2013 to 75.0ppbv in 2015 (Wang et al., 2017). The three most developed regions, such as Beijing-Tianjin-Hebei region (BTH), Yangtze River Delta region (YRD) and Pearl River Delta region (PRD), have the highest anthropogenic emissions which have led to severe regional O_3 pollution (Ma et al., 2016; Shen et al., 2015; Wei et al., 2015). Quantitative estimation of O_3 source-receptor (S-R) relationships has become a critical issue in scientific understanding of O_3 pollution abatement in China.

The O_3 - NO_x -VOC sensitivity regimes, which describe the relationship between O_3 and its local precursors (NO_x and VOC), vary in urban and rural areas in China (Jin and Holloway, 2015). Classifying the photochemical regime of O_3 formation (i.e. VOC-limited and NO_x -limited) in an area helps determine which emissions should be targeted more to abate O_3 pollution. The major sources contributing to surface O_3 above background levels include transport of O_3 and its precursors (Sudo and Akimoto, 2007; Xue et al., 2014), photochemical production (Wang et al., 2010), or vertical transport (Wang et al., 2015). We note that the photochemical regime of O_3 formation only describes local ozone-precursor relationships, which does not account for the nonlocal contributions of atmospheric processes to local O_3 . To fully understand the O_3 generation and removal, it is necessary to identify the major influential atmospheric processes and emissions from local and nonlocal sources to O_3 in a receptor region, in particular to China which has significant domestic

transboundary air pollution (Luo et al., 2018; Hou et al., 2018; Yim et al., 2019; Gu and Yim, 2016).

The complexity in nonlinear chemical response makes it a challenge to quantify the local and transboundary source contributions of surface O₃ pollution (Fiore et al., 2012). Solving the nonlinear optimal problem of O₃ concentration-emission responses requires calculation of local sensitivities of O₃ concentrations (Hakami et al., 2007). It is noted that the chemical transport models (CTM) can determine the source sensitivities and apportionments of modeled chemical species to input variables. Previous studies apportioned source contributions of O₃ pollution in China based on ozone source apportionment technology (Gao et al., 2016; Li et al., 2012b), integrated process analysis (Fan et al., 2014; Li et al., 2012a), response surface modeling (Xing et al., 2011), or forward sensitivity analysis based on CTM (Itahashi et al., 2013; Wang et al., 2011). However, these methods are source-oriented and limited in cases of few specific input variables (Zhai et al., 2018). By contrast, adjoint method is receptor-oriented, and presents a unique advantage that it outputs the spatiotemporal variations of the sensitivities of the cost function to location-specific emission sources in one simulation. Therefore, adjoint method is computationally efficient than others and is used for source apportionment (Zhang et al., 2015).

This study advanced and applied the adjoint version of an air quality model to simulate O₃ sensitivity to NO_x and VOC emissions. Specifically, we investigated the surface O₃ response over a receptor region to emission changes at each grid cell. Our main objectives were to identify the local and transboundary influential pathways of emissions as well as atmospheric processes, and to quantify the source contributions of emissions to surface O₃ at intra-national as well as provincial levels in China. This study is expected to provide a fully scientific understanding of the S–R relationships of O₃ in China. The model configurations are described in Section 2. The details of adjoint sensitivity methods, model evaluation and validation are elaborated from TextS1 to TextS3 in the Supporting Information (SI). In Section 3.1, the patterns of O₃ concentration from baseline case in the study month are discussed. The S–R relationship and source contributions of O₃ are respectively provided in 3.2 Surface ozone source-receptor relationships, 3.3 Relative source contributions to O. The discussions and conclusions are provided in Section 4.

2. Material and methods

The Community Multiscale Air Quality Modeling System (CMAQ v4.7.1) model developed by U.S.EPA (Byun and Schere, 2006) was applied in this study. The input meteorological fields for CMAQ simulations were driven by the Weather Research and Forecasting Model (WRF v3.7) model (Skamarock et al., 2008). Both WRF and CMAQ models were constructed with the spatial resolution of 27km, centered at 37°N, 108.1°E on the Lambert conformal projection and covering the entire China region (Wang et al., 2019) (Fig. S1). Fig. S2 refers to the information of each province and municipal in China mentioned in this study. The boundary condition for CMAQ was provided by GEOS-Chem v8.3.2 model. CMAQ was configured with 10-day spin-up to minimize the influence of initial conditions. The Multi-resolution Emission Inventory for China (MEIC) in 2010 was used as the emission datasets (Li et al., 2017b) (<http://www.meicmodel.org/>). Monthly emissions with a

horizontal resolution at $0.25^{\circ} \times 0.25^{\circ}$ was firstly generated from MEIC and hourly emission data was further generated at a spatial resolution of 27km (Gu and Yim, 2016). The model configurations and evaluations of CMAQ are respectively described in TextS3 and TextS4 in SI. We integrated the adjoint calculation core of CMAQ ADJ v4.5.4 model developed for gas-phase process (Hakami et al., 2007) into the more updated version of CMAQ (v4.7.1). The details of integration and validations are fully presented in Text S3 in SI.

The adjoint model calculated the gradient of a pre-defined cost function with respect to model inputs at each location within model domain. The cost function was a concentration-related function which was defined as hourly O_3 concentration in this study. The details of adjoint method of CMAQ model are presented in Text S1. The gradients of cost function

with respect to NO_x ($\frac{\partial O_3}{\partial E_{NO_x}}$) and VOC ($\frac{\partial O_3}{\partial E_{voc}}$) emissions, were calculated by adjoint model

during the integration. The species of VOC (VOC=OLE+ALD2+ETH+FORM+ISOP+PAR+TOL+XYL) in CB4 chemical mechanism was considered in this study (Hogrefe et al., 2003). The values of gradients ($\frac{\partial O_3}{\partial E_{NO_x}}$) and ($\frac{\partial O_3}{\partial E_{voc}}$) were unitless, which would not

reflect the actual influence of NO_x and VOC emissions on O_3 concentrations. If an emission source with a relatively larger gradient had a small value of emission intensity, its actual influence would be negligible. Therefore, the semi-normalized sensitivity coefficients of O_3 with respect to NO_x emissions ($\phi_{O_3-E_{NO_x}}$) and VOC emissions ($\phi_{O_3-E_{voc}}$) were defined

(An et al., 2016) as follows:

$$\phi_{O_3-E_{NO_x}} = \frac{\partial O_3}{\partial E_{NO_x}} \times E_{NO_x} \quad (\text{Eq. 1})$$

$$\phi_{O_3-E_{voc}} = \frac{\partial O_3}{\partial E_{voc}} \times E_{voc} \quad (\text{Eq. 2})$$

where, E_{NO_x} and E_{voc} referred to NO_x and VOC emissions ($\mu\text{g}/\text{m}^3$), respectively.

In this way, both $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{voc}}$ had physical meanings with the same unit as emissions and could directly quantify how emissions from sources in different locations influence the overall O_3 concentrations in a receptor region.

This study focused on surface O_3 in five target regions: the southern China (SC), the northern China (NC), and three major metropolitan regions which have substantial anthropogenic emissions: PRD, YRD and BTH regions (Fig. S1). The SC and NC regions were separated as different receptor regions due to their different climate condition. The Huai River-Qin Mountains, a geographical dividing line acted as a barrier which led to differences in climate, air pollution and agricultural diversity, was used to geographically divide SC and NC (Li et al., 2017a; Lin, 2017; Wu et al., 2013; Xue et al., 2008). It was

known that mega cities and city clusters in China had severe O₃ pollution problems due to larger amounts of anthropogenic emission sources especially in BTH (Qu et al., 2014; Tang et al., 2012), YRD (Li et al., 2012a; Wang et al., 2006), and PRD (Liu et al., 2013; Shao et al., 2009). Therefore, the regions of SC, NC, PRD, YRD and BTH were designed as the receptor regions of O₃ concentration. Furthermore, in China, June is a typical month with relatively high overall O₃ concentration (Liu et al., 2018). June 2010, a typical summer O₃ episode without tropical cyclones, was thus selected in this study due to the relatively high O₃ levels in China.

3. Results

3.1. Baseline case

Our results show that the monthly mean O₃ concentrations in June were 74.5ppbv in China, 67.9ppbv in SC, 79.1ppbv in NC, 74.6ppbv in PRD, 60.5ppbv in YRD and 92.1ppbv in BTH. The spatial distributions of monthly mean surface O₃ show an explicit distinction between SC and NC (Fig. S8). In the NC plain, BTH, Taiyuan and Qinghai provinces had the highest O₃ concentration levels (>90ppbv). Most provinces in SC had relatively lower O₃ concentration levels (<40ppbv). The relatively low O₃ observed in Sichuan, Hubei, Hunan and Jiangxi provinces were related to their substantial NO_x emissions (Fig. S9(a)), indicating O₃ titration in these areas.

From 4 June in 2010, O₃ concentration in BTH was the highest among regions in China (Fig. S10). The O₃ concentration in NC was significantly higher than that in SC in June (Fig. S10). Previous studies have reported that a hot and dry environment provides favorable conditions for ozone production (Tong et al., 2018a, Tong et al., 2018b; Vautard et al., 2007; Zhao et al., 2015) as temperature is a critical factor to determine chemical reaction rates and thus largely affects O₃ variations (Pusede et al., 2015). O₃ concentrations in NC and BTH were thus relatively higher than the other regions in China due to the relatively high temperature (Fig. S11).

The relatively low O₃ concentration in SC was probably due to the persistent rainfalls, especially in Guangxi, Guangdong and Fujian provinces (Guo et al., 2016). Based on climate records from NCC, CMA (<http://cmdp.ncc-cma.net/cn/index.htm>), the monthly average precipitation was 95.0mm (close to normal) and temperature was 20.5°C (1.0°C above the normal) over entire China in June 2010. The cold air in the mid-latitude in East Asia was weakened, and the westerly jet moved southward (Fig. S12), which became a favorable circulation background for persistent rainfalls in SC (Fang et al., 2012). The monthly mean precipitation in NC especially in Northern China Plain (NCP), northeastern China, inner Mongolia and Shaanxi province, was lower than the long-term average. Barriopedro et al. (2012) reported that the northern China experienced severe drought in June of 2010.

The monthly mean O₃ concentration in eastern China was 34.02ppbv, which was within the range of 31.9±17.1ppbv as reported by Zhao et al. (2015). A positive correlation between O₃ over YRD and air temperature in June was found ($R=0.75$, $p=0.00000$), whereas O₃ was negatively correlated with air pressure ($R=-0.31$, $p<0.00001$). On the other hand,

O₃ concentration was negatively correlated with water vapor ratio ($R=-0.73$, $p<0.00001$) in PRD as well as in SC ($R=-0.61$, $p=0.00000$). The diurnal variation in O₃ over PRD in this study was closely correlated with regional mean daily precipitation from Fang et al. (2012).

3.2. Surface ozone source-receptor relationships

3.2.1. O₃ sensitivity in southern and northern China—Figure 1 shows that the values of $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{VOC}}$ in most of locations were positive within SC and NC.

The positive values indicate that NO_x and VOC emissions in these locations had the positive relationship with the overall O₃ concentration in SC and NC. Even though negative values of $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{VOC}}$ were observed, their magnitudes were insignificant. The

$\phi_{O_3-E_{NO_x}}$ values in the capitals of each province such as in Wuhan and Xi'an, and in highly urbanized regions and cities such as in Shanghai and Nanjing, where substantial NO_x emissions exist (Gao et al., 2017; Xie et al., 2016), were larger than 0.01 µg/m³, implying that a 10% reduction in NO_x emissions in any grid of these areas would lead to $>0.01 \times 10\% \mu\text{g}/\text{m}^3$ decrease in overall O₃ concentration in SC. It was noticeable that these areas also had relatively higher $\phi_{O_3-E_{VOC}}$ values due to substantial VOC emissions. The

magnitude of $\phi_{O_3-E_{VOC}}$ was relatively smaller than that of $\phi_{O_3-E_{NO_x}}$. his result indicates

that reducing the same amount of NO_x emissions in these areas would lead to a larger decrease in O₃ in SC than VOC emissions did. The most influential source areas outside SC were mainly located at its adjacent areas with substantial emissions, i.e. the northern part of Anhui, Xi'an and Zhengzhou, whereas influences of other areas outside to O₃ in all receptor regions were negligible. This pinpoints the significant local nature of the overall O₃ in SC.

To further explain the influential pathways of emissions from different source areas that affected O₃ in receptor regions, the relative contributions from transport and chemical reactions as well as O₃ chemical regimes were discussed. In this study, photochemical indicators such as H₂O₂/HNO₃ and O₃/NO_y ratios were used to identify O₃ photochemical production regimes (Sillman and He, 2002). Liu et al. (2010) suggested that the transition value of H₂O₂/HNO₃ was 1.6 in July in China. Ye et al. (2016) estimated the threshold value of O₃/NO_y which should be 10.2 in PRD. Fig. S13 suggests that the O₃ production regime was NO_x-limited in most of areas in China while mega-cities and urban areas were in a VOC-limited regime.

To diagnose the relative contributions of chemical reactions and physical processes, a process analysis was conducted in CMAQ, see Fig. S14. The process analysis implemented in CMAQ estimated the contribution of each process that affects the overall simulated O₃ in each synchronization time step (Li et al., 2012a). The contribution of each process was referred to the O₃ changes before and after the respective process and was compared with the overall O₃ changes in each synchronization time step. The typical VOC-limited areas within SC were PRD, YRD, and capitals, which had negative contributions from chemical reactions and positive contributions from transport process to local O₃ formation, and had

high positive $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{VOC}}$ values. It indicates that the transport process can offset the reduction in local O_3 concentration by chemistry.

Adjacent areas outside SC had relatively high positive $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{VOC}}$ values in source areas such as Xi'an and Lanzhou where substantial anthropogenic emissions exist. Emissions from source areas outside SC might affect the overall O_3 in SC through different influential pathways. In VOC-limited source areas outside SC, there were two possible major influential pathways: one was that O_3 precursors were transported to SC to increase the overall O_3 in SC through local chemical reactions; another pathway was that, during the transport of O_3 precursors, O_3 might be formed before reaching to receptor regions. For the NO_x -limited source areas outside SC, there were three possible major influential pathways: the first two pathways were same as those as discussed above; the third one might be that the locally formed O_3 in NO_x -limited source areas was directly transported to SC. In the VOC-limited source areas, O_3 decreased when increasing NO_x due to the reaction of hydroxyl radical and NO_2 (Yarwood et al., 2003). Thus, the third pathway might not be important in VOC-limited source areas outside SC.

The high positive $\phi_{O_3-E_{NO_x}}$ and $\phi_{O_3-E_{VOC}}$ values in NC also corresponded to source areas with substantial NO_x and VOC emissions such as in Beijing, Tianjin, Shijiazhuang and Jinan. The major influential pathways of the overall O_3 in NC were similar to those in SC that were thoroughly discussed above. Large amounts of NO_x emissions from industry, transportation and power plants coexisted in the southern Beijing, northern Tianjin, and southwestern Hebei, which led to relatively high $\phi_{O_3-E_{VOC}}$ and $\phi_{O_3-E_{VOC}}$ values.

Although biogenic emissions were high in Yan Mountains that located in southern BTH and Taihang Mountains that located in western BTH, the impacts on surface O_3 were low due to the lack of NO_x emissions there (Qu et al., 2014). It was noticeable that northern part of Anhui province was the major influential source areas for the overall O_3 in both SC and NC. The probable reasons were due to the rapid industrial growth in northern part of Anhui province (Qiu et al., 2017), and the conducive geographical condition that pollutants could be accumulated in the northern part of Anhui since it is the terminal of North China Plain (Yan and Wu, 2017). The most influential source areas outside NC were mainly located at its adjacent areas with substantial emissions, i.e. Nanjing and Wuhan, showing the similar significant local nature of the overall O_3 as that in SC.

3.2.2. O_3 sensitivity in three developed regions in China—In the other three receptor regions (PRD, YRD and BTH), the high positive $\phi_{O_3-E_{VOC}}$ and $\phi_{O_3-E_{VOC}}$ values were related to the source areas with substantial emissions (Fig. 2). The source contributions from emissions to O_3 in PRD and BTH were more concentrated in local areas within the receptor regions. The most influential source areas in PRD were the urban areas in Guangzhou, Dongguan and Shenzhen. In BTH, the major source contributions of emissions were from southern Beijing, northern Tianjin and southwestern Hebei. The most influential source areas to the overall O_3 in YRD were not only from local areas inside YRD but also

from large areas outside YRD such as the northern part of Anhui province, Wuhan and Nanchang. Emissions from outside YRD made comparable contributions as local emissions did, indicating the transboundary nature of O₃ pollution in YRD. The significant transboundary O₃ may be due to the prevailing southwesterly (~19.5% in frequency) and southerly (~17.9% in frequency) surface wind in June (Fig. S11). For example, the non-negligible contributions from emissions in PRD to overall O₃ in YRD are obviously showed in Fig. 2. The prevailing wind direction was conducive to the pollutant transport from PRD to YRD.

3.3. Relative source contributions to O₃ from different source areas and provinces

This section assesses the relative source contributions to the overall O₃ concentration in five receptor regions from their NO_x and VOC emissions (Fig. 3). A source contribution was referred to semi-normalized adjoint sensitivity coefficients $\phi_{O_3-E_{VOC}}$ and $\phi_{O_3-E_{NO_x}}$ (adjoint sensitivity × emissions). To calculate a relative source contribution, we first calculated the contribution of each source area by summing up the values of $\phi_{O_3-E_{VOC}}$ and $\phi_{O_3-E_{NO_x}}$ over all the grid points in the receptive source area. The relative source contribution was then calculated based on the relative ratio of all source areas including SC, NC, PRD, YRD and BTH. We note that the source contributions of O₃ in PRD, YRD and BTH regions were assessed individually so that YRD and PRD were excluded from SC, and BTH was excluded from NC. Each column in Fig. 3 refers to the apportionment of relative contributions by NO_x or VOC emissions to their total contributions to the overall O₃ concentration in each receptor region. For example, when summing up all relative source contributions of NO_x and VOC emissions to O₃ in NC, the total percentage values were 100%.

The transboundary nature of O₃ pollution was found in SC, NC and YRD. Fig. 3 clearly shows the significant contributions of nonlocal emissions to O₃ over SC, NC and YRD. In addition, the total relative source contributions from nonlocal emissions cannot be neglected. For example, nonlocal NO_x emissions contributed to around 20% O₃ in both NC and SC, and around 55% O₃ in YRD. In addition, nonlocal VOC emissions contributed to around 11% O₃ in YRD.

The contributions of different source areas to O₃ in PRD and BTH were more localized. For example, local NO_x and VOC emissions in PRD contributed 61.3% and 32.1% O₃ in PRD, respectively. 71.9% and 13.5% O₃ in BTH were from local NO_x and VOC emissions.

When comparing the source contributions from different emission species, NO_x emissions were the key contributor to O₃ concentrations in all receptor regions. The NO_x emissions from all source areas contributed around 80% O₃ in receptor regions except YRD in which NO_x emissions contributed around 67% O₃.

The relative source contributions of NO_x and VOC emissions from each province in China to the overall O₃ in the five receptor regions were quantified. Table S3 shows the relative contributions of provinces from NO_x or VOC emissions that accounted for >95% of total

contributions in each receptor region. For NO_x emissions, Anhui, Hubei, and Jiangsu provinces were the three largest contributors (>16%) to O_3 in SC and YRD. For VOC emissions, the major source areas of O_3 in SC (>20%) were Guangdong and Hubei provinces, whereas those of O_3 in YRD (>20%) were Hubei, Jiangsu provinces and Shanghai municipality. The two major source areas of O_3 in BTH and NC were Beijing (>30%) and Tianjin (>50%) municipalities. NO_x (~90%) and VOC (~99%) emissions from Guangdong province were dominant contributors to O_3 in PRD. Most of the largest contributors of both NO_x and VOC emissions were from the same province to O_3 in receptor regions except for receptor YRD. The largest source contributions of NO_x emissions were from Anhui (~36%) and of VOC emissions were from Jiangsu (~25%) to O_3 in YRD. The NO_x emissions in June in Anhui provinces were significantly high due to the biomass burning in the month (Ding et al., 2017). Jiangsu province was one of the three largest contributors of VOC emission in China (Wu et al., 2015).

4. Discussion

Emission control efficiency largely depends on the photochemical regimes of O_3 production (i.e. VOC-limited or NO_x -limited regimes). Indicators such as $\text{H}_2\text{O}_2/\text{HNO}_3$ and O_3/NO_y ratios have been widely used to identify the O_3 photochemical production regimes in China based on measurements (Xue et al., 2014), OMI satellite products (Jin and Holloway, 2015), and modeling simulations (Xie et al., 2014). Our results were consistent with previous studies that the megacities such as Beijing and Shanghai, and city clusters such as PRD and YRD regions were typical VOC-limited regimes in June. In our study, the indicators were used as the proxy for chemical regimes of O_3 to help investigate the most probable influential pathways of emissions outside receptor regions to O_3 within different receptors.

The main S-R relationships of O_3 were investigated in previous studies which mostly were conducted based on site observations (Liu et al., 2010; Xu et al., 2008) and city clusters such as PRD (Wang et al., 2010). Our study focused on the S-R relationships over entire China including the PRD, YRD and BTH regions. Although we found that reducing both NO_x and VOC emissions were effective for O_3 control in China, reducing NO_x emissions was more important.

While previous studies identified the source contributions of emissions to regional O_3 in BTH (Wang et al., 2009), YRD (Gao et al., 2016) and PRD (Li et al., 2012b) during an O_3 episode, our study further investigated the monthly relative source contributions of NO_x and VOC emissions from different source areas to O_3 in receptor regions in China. June, a typical summer O_3 episode, was selected as the study period so that our study can investigate more information of O_3 pollution in China based on prior knowledge from previous studies. Our results show that the major local source areas were those that had substantial anthropogenic emissions such as capitals in each province, PRD and YRD. The nonlocal source areas had different influential pathways to O_3 which had been discussed in the results section.

The transboundary O_3 pollution in China had been found by many studies. For example, the transport of O_3 from city regions to surrounding rural areas was found by Li et al. (2012a) in

August in YRD; the transport of O₃ precursors from central PRD to downwind rural areas was found by Wang et al. (2010) in October in PRD; the transport of O₃ and its precursors from Tianjin and the south of Hebei to Beijing urban areas was found by Wang et al. (2009) during 26 June to 2 July. In our study, the relative contributions from individual chemistry and transport process were assessed as the supplementary information to understand the influential pathways of emissions to O₃ in receptor regions. In addition, the relative source contributions of emissions from source areas outside receptor to O₃ in receptor to highlight the transboundary nature of O₃ pollution were also quantified. In SC, NC and YRD receptors, the transboundary nature of O₃ was found. The relative contributions of NO_x and VOC emissions from each province were quantified separately to provide provincial information for O₃ pollution control.

5. Conclusions

This study advanced and applied the CMAQ adjoint model to calculate the O₃ sensitivity with respect to emissions in China. The aim of this study was to investigate the source-receptor relationship between O₃ changes over different receptor regions and its emission precursors at each grid cell, with a special focus on relative source contributions to O₃ at different geographical levels such as at different source areas and at provincial level. This effort should provide a fully scientific understanding of O₃ generation and removal, and its most influential pathways of emission and atmospheric processes in China. We found that the atmospheric processes such as horizontal and vertical transport can offset the O₃ removal through chemical reactions in VOC-limited areas inside the receptor regions. In addition, O₃ pollution can be induced by transport of O₃ directly or its precursors. Our results found that the reducing NO_x and VOC emissions from local source areas would be effective for O₃ pollution control in all receptor regions especially in PRD and BTH. The transboundary O₃ pollution in SC, NC and YRD should also be paid attention since the large source contributions from nonlocal emissions were found. Therefore, reducing local emissions and nonlocal emissions were both important for O₃ pollution control in SC, NC and YRD. Anhui, Hubei, and Jiangsu provinces were the three largest source areas of NO_x emissions to O₃ in SC and YRD. The largest contributors of VOC emission sources to O₃ in SC were Guangdong and Hubei provinces, whereas those of O₃ in YRD were Hubei and Jiangsu provinces, and Shanghai. The top two largest contributors of O₃ in BTH and NC were emissions from Tianjin and Beijing. NO_x and VOC emissions from Guangdong province were dominant contributors (>89%) to O₃ in PRD. The largest source areas of NO_x emission were Anhui and those of VOC emissions were Jiangsu to O₃ in YRD. We should pay more attention to emission reductions in these provinces and municipalities to develop the most expeditious and cost-effective emission control strategies in China.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1. Spatial distributions of monthly mean sensitivity coefficients of O_3 in (a) NC and (b) SC with respect to NO_x emissions ($\phi_{O_3-E_{NO_x}}$) ($\mu g/m^3$), and those in (c) NC and (d) SC with respect to VOC emissions ($\phi_{O_3-E_{VOC}}$) ($\mu g/m^3$). Red boundaries refer to NC in (a) and (c), and SC in (b) and (d).

Figure 2. Spatial distributions of the sensitivity of the sensitivity coefficients of O_3 in (a) PRD, (b) YRD and (c) BTH with respect to NO_x emissions ($\phi_{O_3 - E_{NO_x}}$) ($\mu g/m^3$), and those in (d) PRD, (e) YRD and (f) BTH with respect to VOC emissions ($\phi_{O_3 - E_{VOC}}$) ($\mu g/m^3$). Red boundaries refer to PRD in (a) and (d), YRD in (b) and (e), and BTH in (c) and (f). PRD, YRD and BTH are zoomed and displayed in the left corner of each sub-figure.

Fig 3. Relative source contributions (%) of NO_x and VOC emissions from different source regions to the overall O₃ in receptor regions. YRD and PRD were excluded from SC, and BTH was excluded from NC.