Ozone Pollution over China and India: Seasonality and Sources

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Abstract

A regional fully coupled meteorology-chemistry Weather Research and Forecasting model with Chemistry (WRF-Chem) was employed to study the seasonality of ozone ($O_3$) pollution and its sources in both China and India. Observations and model results suggest that $O_3$ in the North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD) and India exhibit distinctive seasonal features, which are linked to the influence of summer monsoons. Through a factor separation approach, we examined the sensitivity of $O_3$ to individual anthropogenic, biogenic, and biomass burning emissions. We found that summer $O_3$ formation is more sensitive to industrial sources than to other source sectors for China, while the transport vehicle sector is more important in all seasons for India. For India, in addition to transport, the residential sector also plays an important role in winter when $O_3$ concentrations peak. Tagged simulations suggest that sources in east China play an important role in the formation of the summer $O_3$ peak in the NCP, but sources from Northwest China should not be neglected to control summer $O_3$ in the NCP. For the YRD region, prevailing winds and cleaner air from the ocean in summer lead to reduced transport from polluted regions, and the major source region in addition to local sources is Southeast China. For the PRD region, the upwind region is replaced by contributions from polluted east China as autumn approaches, leading to an autumn peak. The major upwind regions in autumn for the PRD are YRD (11%) and Southeast China (10%). For India, sources in North India are more important than sources in the south. These analyses emphasize the relative importance of source sectors and regions as they change with seasons, providing important implications for $O_3$ control strategies.
1 Introduction

Tropospheric ozone (O₃) is the third most potent greenhouse gas in the atmosphere (Pachauri and Reisinger, 2007), an important surface air pollutant, and the major source of the hydroxyl radical (a key oxidant playing an essential role in atmospheric chemistry). With the rapid growth of industrialization, urbanization and transportation activities, emissions of O₃ precursors (nitrogen oxides and volatile organic compounds) in both China and India have increased significantly since 2000 (De Smedt et al., 2010; Duncan et al., 2014; Hilboll et al., 2013; Kurokawa et al., 2013; Ohara et al., 2007; Stavrakou et al., 2009; Zheng et al., 2018).

Increasing concentrations of O₃ precursors have led to emerging and far-flung O₃ pollution, threatening health and food security (Chameides et al., 1994; Malley et al., 2018). The decrease in crop yield resulting from the increase in surface O₃ would have been sufficient to feed 95 million people in India (Ghude et al., 2014).

Great efforts have been devoted to improving understanding of exceptionally high concentrations (Wang et al., 2006) and the increasing trend in O₃ for both China and India (Beig et al., 2007; Cheng et al., 2016; Ghude et al., 2008; Lu et al., 2018a; Ma et al., 2016; Saraf and Beig, 2004; Xu et al., 2008). Strong but distinctive seasonal variations of O₃ observed in India and China have been linked to higher emissions of precursor gases (Lal et al., 2000), stratospheric intrusions (Kumar et al., 2010), and the summer monsoon (Kumar et al., 2010; Lu et al., 2018b; Wang et al., 2017). The contributions of individual economic sectors and source regions were reported based on sensitivity simulations and source apportionment techniques (J. Gao et al., 2016; Li et al., 2008; Li et al., 2016; Li et al., 2012; Lu et al., 2019; Wang et al., 2019). With respect to the enhanced concentrations of O₃ over the past years, Sun et al. (2019) attributed this to elevated emissions of anthropogenic VOCs, while Li et al. (2019) argued that an inhibited aerosol sink for hydroperoxyl radicals induced by decreased PM₂.₅ over 2013-2017 played a more important role in the NCP.

Despite this progress, the seasonal behaviors of O₃ in different regions greatly differ, yet have not been intercompared and the underlying causes have not been comprehensively explored. In addition, previous source apportionment studies focused on specific regions or episodes, and the policy implications drawn from these studies might not be applicable for other regions and seasons. It is both of interest and of significance to understand the similarities and differences...
between O$_3$ pollution in China and India, the two most polluted and most populous countries in the world.

The present study uses a fully online coupled meteorology-chemistry model (WRF-Chem) to examine the general seasonal features of O$_3$ pollution, and its sources derived from economic sectors and regions over both China and India. Sect. 2 describes the air quality model and measurements. We examine then in Sect. 3 how the model captures the spatial and temporal variations of O$_3$ and relevant precursors. Sect. 4 presents general seasonal features of O$_3$ pollution, and the relative importance of both economic sectors and source regions. Results are summarized in Sect. 5.

2 Model and data

2.1 WRF-Chem model and configurations

The fully online coupled meteorology-chemistry model WRF-Chem (Grell et al., 2005) was employed in this study using the CBMZ (Carbon Bond Mechanism version Z, Zaveri and Peters, 1999) photochemical mechanism and the MOSAIC (Model for simulating aerosol interactions and chemistry, Zaveri et al., 2008) aerosol chemistry module. The model was configured with a horizontal grid spacing of 60km with 27 vertical layers (from the surface to 10 hPa), covering East and South Asia (Fig. 1). The selected physical parameterization schemes follow the settings documented in M. Gao et al. (2016). Meteorological initial and boundary conditions were obtained from the 6-hourly FNL (final analyses, NCEP, 2000) global analysis data with 1.0°×1.0° resolution. The four-dimensional data assimilation (FDDA) technique was applied to limit errors in simulated meteorology. Horizontal winds, temperature and moisture were nudged at all vertical levels. Chemical initial and boundary conditions were provided using MOZART-4 (Emmons et al., 2010) global simulations of chemical species.

Monthly anthropogenic emissions of SO$_2$, NO$_x$, CO, NMVOCs (Non-methane Volatile Organic Compounds), NH$_3$, PM$_{2.5}$, PM$_{10}$, BC (black carbon) and OC (organic carbon) were taken from the MIX 2010 inventory (Li et al., 2017), a mosaic Asian anthropogenic emission inventory covering both China and India. In this study, the emissions in China were updated with the MEIC (Multi-resolution Emission Inventory for China, http://www.meicmodel.org/) inventory for year 2012. The MIX inventory was prepared considering five economic sectors on a
0.25°×0.25° grid: power, industrial, residential (heating, combustion, solvent use, and waste disposal), transportation and agriculture. For India, SO₂, BC, OC, and power plant NOₓ emissions were taken from the inventory developed by the Argonne National Laboratory (ANL), with the REAS (Regional Emission inventory in Asia) inventory used to supplement for missing species. Speciation mapping of VOCs emissions follows the speciation framework documented in Li et al. (2014) and Gao et al. (2018). The MEGAN (Model for Emissions of Gases and Aerosols from Nature, Guenther et al., 2012) model version 2.1 was used to generate biogenic emissions online. Biomass burning emissions were obtained from the 4th generation global fire emissions database (GFED4, Giglio et al., 2013). For China, industrial and power sectors are the largest two contributors to NOₓ emissions, while industrial sector emits the largest amounts of NMVOCs (Li et al., 2017). For India, transportation and power sectors produce the largest amounts of NOₓ, while residential and transportation sectors are the largest two contributors to NMVOCs emissions (Li et al., 2017). China’s biogenic emissions of VOCs are estimated to be higher than anthropogenic sources (Li and Xie, 2014; Wei et al., 2011).

2.2 Ozone tagging method and setting of source regions
O₃ observed in a particular region is a mixture of O₃ formed by reactions of NOₓ with VOCs emitted at different locations and time. The O₃ tagging method has the capability to apportion contributions of different source regions to O₃ concentrations observed in particular regions. The present study adopted the ozone tagging method implemented in WRF-Chem by J. Gao et al. (2017), which is similar to the Ozone Source Apportionment Technology (OSAT, Yarwood et al., 1996) approach implemented in the Comprehensive Air Quality Model with extensions (CAMx). Both O₃ and its precursors from different source regions are tracked as independent variables. The ratio of formaldehyde to reactive nitrogen oxides (HCHO/NOₓ) was used as proposed by Sillman (1995) to decide whether the grid cell is under NOₓ or VOC limited conditions, and then different equations for these two conditions were selected to calculate total O₃ chemical production. A detailed description of the technique is provided in J. Gao et al. (2017).

The O₃ tagging method attributes production of O₃ and its precursors to individual geographic areas. We divided the entire modeling domain into 23 source regions, which were classified...
mainly using the administrative boundaries of provinces. In eastern China, each province was considered as a source region, while provinces in northeastern, northwestern, and southwestern China were lumped together (Fig. S1). India was divided into two source regions (north and south), and other countries were considered separately as a whole (Fig. S1). Additionally, the chemical boundaries provided by MOZART-4 were adopted to specify inputs of O\textsubscript{3}, and the initial condition was tracked also as an independent source. The names of all source groupings are indicated in Fig. S1.

### 2.3 Experiment design

To quantify the sectoral contributions to O\textsubscript{3}, a factor separation approach (FSA) was applied to differentiate two model simulations: one with all emission sources considered, and the other with some emission sources excluded. Table 1 summarizes the different sets of simulations conducted in this study. In addition to the control case, a series of sensitivity studies was performed, in which industrial, residential, transport, power, biogenic and fire emissions were separately excluded (Table 1). For each case, the entire year of 2013 was simulated.

### 2.4 Measurements

Surface air pollutants in China are measured and recorded by the Ministry of Environmental Protection (MEP), and the data are accessible on the China National Environmental Monitoring Center (CNEMC) website (http://106.37.208.233:20035/). This nationwide network was initiated in January 2013, and this dataset was used to evaluate model performance. This dataset has been extensively employed in previous studies to understand the spatial and temporal variations of air pollution in China (Hu et al., 2016; Lu et al., 2018), and to reduce uncertainties in estimates of health and climate effects (M. Gao et al., 2017). Measurements of air pollutants from the MAPAN network (Modeling of Atmospheric Pollution and Networking) set up by the Indian Institute of Tropical Meteorology (IITM) under project SAFAR (System of Air Quality and weather Forecasting And Research) (Beig et al., 2015) were used in the present study to evaluate the model performance over India. To further evaluate how the model performed in capturing the vertical distributions of O\textsubscript{3}, we used data from ozonesonde records obtained from the World Ozone and Ultraviolet Radiation Data Center website.

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Fig. 1 displays the locations of the relevant surface and ozonesonde observation sites. We evaluated also the spatial distribution of NO$_2$ columns using the KNMI-DOMINO products of tropospheric NO$_2$ column ([www.temis.nl](https://www.temis.nl)). We excluded pixels observed under cloudy conditions (cloud fractions greater than 0.2) in the comparison.

### 3 Model evaluation

We evaluated the spatial distribution of simulated seasonal mean (winter months include January, February and December; spring months include March, April and May; summer months include June, July and August; Autumn months include September, October, and November) O$_3$ concentrations by comparing model results with observations (filled circles in Fig. 2) for 62 cities in China and India. The model captures the spatiotemporal patterns of O$_3$ in east China, with lower values in fall (Fig. 2d) and winter (Fig. 2a), and enhanced levels in spring (Fig. 2b) and summer (Fig. 2c). However, O$_3$ concentrations are overestimated by the model in central, northwest and southwest China for all seasons (Fig. 2). *Hu et al. (2016)* reported also that their model tends to predict higher O$_3$ concentrations for these regions. Comparisons between simulated and observed diurnal variations suggest that nighttime O$_3$ concentrations inferred by the model are higher than observation (*Hu et al., 2016*). During nighttime, O$_3$ concentrations are depressed through reaction with NO (NO$_x$ titration). Fig. S2 indicates that modeled NO$_2$ column values in central, northwest and southwest China are not as high as observed, suggesting underestimation of NO$_x$ emissions and less nighttime consumption of O$_3$ by NO. The simulated magnitudes of O$_3$ in India are generally consistent with observations, though lower in central India.

We conducted a further site-by-site evaluation of monthly mean O$_3$ concentrations, and grouped stations into four major densely-populated regions, namely North China Plain (NCP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and India. The grouping categories follow the descriptions documented in *Hu et al. (2016)*. The seasonality of observed O$_3$ concentrations is reproduced well in these four regions (Fig. 3), although concentrations are underestimated in the NCP in spring. Stronger NO$_x$ titration (underestimation of O$_3$ during the night, figure not shown), as indicated by the overestimation of NO$_2$ column over the NCP in
spring (Fig. S2), is the most likely cause for these underestimations of O₃. The correlation coefficients between model and observations range between 0.84 and 0.98. Fig. 3 suggests also that the seasonal behavior of O₃ in these four major regions exhibits distinctive patterns, discussed in detail in Sect. 4.

In this work, ozonesonde measurements from the Hong Kong Observatory (HKO), Japan Meteorological Agency (JMA), and the Hydrometeorological Service of S.R. Vietnam (HSSRV) (locations marked in purple in Fig. 1) were used. Wintertime near-surface O₃ concentrations are overestimated for HKO (Fig. S3), while vertical variations are satisfactorily captured by the model.

Several issues are revealed through comparisons against measurements from multiple platforms. Comparisons of near-surface O₃ precursors suggest that CO concentrations are underestimated in all the regions, which could be explained by an underestimate of CO emissions (Wang et al., 2011). The coarse grid resolution of the model might provide another reason for this underestimation, as the observation sites in China are located mostly in urban areas. Underestimates of CO concentrations are reported also for many sites in India (Hakim et al., 2019). The effects of underestimated CO on O₃ were found to be small, but the underestimation of CO may lead to bias in methane lifetime (Strode et al., 2015), which is beyond the discussion of regional pollution in this study. Simulated NO₂ concentrations are slightly overestimated in the NCP but are underestimated in the PRD (Fig. S3), consistent with the comparison with satellite NO₂ columns (Fig. S2). However, the model still captures the seasonal behavior of O₃ in different regions, and we do not expect the model biases to change the major findings of the present study.

4 Seasonality, source sectors and source regions

4.1 Seasonality of surface O₃ in different regions

Comparisons between modeled and observed near-surface O₃ concentrations for different regions suggest distinctive seasonal patterns (Fig. 3). Over the NCP, near-surface O₃ exhibits an inverted V-shaped pattern, with maximum O₃ concentrations in summer, minimum in winter (Fig. 3). Over the YRD, O₃ presents a bridge shape, with relatively higher concentrations in spring, summer and autumn (Fig. 3). O₃ concentrations over the PRD peak in autumn, with a
minimum in summer (Fig. 3). Similarly, O$_3$ over India exhibits a minimum in summer, with highest concentrations in winter (Fig. 3). China and India are influenced largely by monsoonal climates (Wang et al., 2001), and the seasonality of O$_3$ in different regions is affected by wind pattern reversals related to the winter and summer monsoon systems (Lu et al., 2018). Various monsoon indices have been proposed to describe the major features of the Asian monsoon, based on pressure, temperature, and wind fields, etc. In the present study, we adopted the dynamical normalized seasonality monsoon index (DNSMI) developed by Li and Zeng (2002) to explore the influence of monsoon intensity on the seasonal behavior of O$_3$ in the boundary layer in different regions of China and India. DNSMI is defined as follows:

$$DNSMI = \frac{\|V_i - \bar{V}\|}{\bar{V}} - 2$$

in which $V_i$ and $\bar{V}$ represent the wind vectors in January, and wind vectors in month $i$, respectively. $\bar{V}$ denotes the mean of wind vectors in January and July. The norm of a given variable is defined as:

$$\|A\| = (\int \int |A|^2 dS)^{\frac{1}{2}}$$

where S represents the spatial area of each model grid cell. More detailed information on the definition is presented in Li and Zeng (2002).

This definition of monsoon focuses on wind vectors, representing the intensity of wind direction alternation from winter to summer. In winter, northwesterly winds are predominant, then higher DNSMI values indicate stronger alternation of wind directions. For example, DNSMI values are higher than 5 in coastal regions of South China and most environments in India (Fig. 4c), suggesting that these regions are influenced largely by the summer monsoon. Over the ocean, DNSMI increases as spring approaches, reaching a maximum in summer (Fig. 4). Over land, the magnitude of DNSMI decreases, and relatively large values are found only in coastal regions (Fig. 4). The alternation of wind vectors from winter to summer results also in changes in upwind areas, modulating the severity of O$_3$ pollution. In summer, the southerly winds containing clean maritime air masses, serve to reduce the intensity of pollution in regions that are affected largely by the summer monsoon (e.g., most regions over India, and coastal regions of China). Besides, cloudy and rainy conditions associated with the summer monsoon
are not conducive to photochemical production of O₃ (Tang et al., 2013).

North China is influenced less than South China and East China by the summer monsoon as suggested by DNSMI values lower than 0.5 as shown in Fig. 4c, and weather conditions favor O₃ formation in summer (higher temperature and stronger solar radiation). As a result, O₃ concentrations in the NCP peak in summer, exhibiting an inverted V-shaped pattern (Fig. 3a). The YRD region is affected moderately by the summer monsoon, with DNSMI values greater than 0.6 (Fig. 4c). The upwind sources for the YRD in summer include both polluted (south China) and clean (ocean) regions. Thus, the inhibition of O₃ formation in the YRD due to the summer monsoon does not lead to the annual minima in summer. Because of the favorable weather conditions (increasing temperature and solar radiation, and low precipitation) in spring and autumn, the seasonality of O₃ in the YRD exhibits a bridge shape, consistent with previous observations within this region (Tang et al., 2013). In addition, southerly winds might bring O₃ and its precursors from the YRD region in summer (Fig. 4c), which will be further quantified in Sect. 4.3. For India and the PRD region, the alternation of wind fields begins as spring approaches (Fig. 4). As a result, O₃ concentrations decline in response to input of cleaner air from the ocean. As summer arrives, the intensity of the monsoon reaches its maximum (Fig. 4c) and concentrations of O₃ in both India and South China decline to reach their annual minima (Fig. 3c and Fig. 3d). As wind direction changes over the east coast of China from summer to autumn, O₃ peaks in autumn in South China can be attributed also to the outflow of O₃ and its precursors from the NCP and YRD regions (Fig. 4d). This contribution will be discussed further also in Sect. 4.3.

### 4.2 O₃ sensitivity to emissions from individual source sectors

O₃ in the troposphere is formed through complex nonlinear processes involving emissions of NOₓ and VOCs from various anthropogenic, biogenic, and biomass burning sources. We illustrate in Fig. 5 the sensitivity of seasonal mean O₃ concentrations in both China and India to individual source sectors, patterns that offer important implications for seasonal O₃ control strategies in some highly polluted regions.

For China, summer O₃ formation is more sensitive to industrial sources than to other anthropogenic sources, including power, residential, and transport (Fig. 5c). Emissions from
the industrial sector are responsible for an enhancement of O$_3$ concentrations by more than 10 ppb in east China in summer (Fig. 5c). Using a similar approach, Li et al. (2017) reported that the contribution to O$_3$ from industrial sources exceeded 30 µg/m$^3$ (~15 ppb) in highly industrialized areas, including Hebei, Shandong, Zhejiang, etc. during an episode in May. Li et al. (2016) concluded that the industrial sector plays the most important role for O$_3$ formation in Shanghai, accounting for more than 35% of observed concentrations. Adopting a source-oriented chemical transport model, Wang et al., (2019) demonstrated that the industrial source contributes 36%, 46%, and 29% to non-background O$_3$ in Beijing, Shanghai and Guangdong, respectively.

In east China, O$_3$ formation in winter, spring, and autumn reflects negative sensitivity to the transport and power sectors (Fig. 5). These two sectors dominate emissions of NO$_x$ in China (Li et al., 2017). Removing these sectors would lead to increases in O$_3$ in VOC-limited regions of east China in winter, spring and fall (less biogenic emissions of VOCs in these seasons, Fu et al., 2012). Urban regions in China are still VOC-limited (Fu et al., 2012; Jin et al., 2017) in summer, leading to negligible or negative sensitivity to the transport and power sectors as shown in Fig. 5g and Fig 5o. In other regions of east China, removing transport and power sources would lead to an increase in O$_3$ concentrations by about 4 ppb in summer. The sensitivity of O$_3$ concentrations to the residential sector in spring and autumn displays appreciable magnitudes in the YRD, where O$_3$ peaks in autumn (Fig. 5).

Including biogenic emissions results in an increase in summer mean O$_3$ concentrations by more than 20 ppb in east China (Fig. 5s). Using a similar approach, Li et al. (2018) found that biogenic emissions contributed 8.2 ppb in urban Xi’an. Other source apportionment studies indicate that the contribution of biogenic emissions to O$_3$ formation is about 20% in China (Li et al., 2016; Wang et al., 2019). The enhancements due to biogenic emissions are larger over south China during winter, and the significantly impacted regions extend northwards in spring and autumn (Fig. 5q-5t). Biomass burning emissions lead to relatively lower O$_3$ enhancements over China in winter, but are responsible for an appreciable contribution to O$_3$ pollution (~7 ppb) in east China in summer (Fig. 5w). Li et al. (2016) suggested that biomass burning sources contribute about 4% to O$_3$ formation in the YRD region in summer. The enhancement due to biomass burning estimated by Lu et al. (2019) using a different model indicates lower values.
in east China.

For India, O₃ formation is most sensitive to the transport vehicle sector (~8 ppb) in all seasons, slightly higher than it is to the biogenic source (Fig. 5m-5p). Among other sectors, the sensitivity of O₃ formation to the residential sector is significant in winter as residential sector emits the largest amount of NMVOCs (Li et al., 2017), while the influence of biomass burning emissions is negligible.

Our results highlight the importance of industrial sources in O₃ formation in east China, consistent with the conclusions of Li et al. (2017). The significance of other sectors demonstrated by Li et al. (2017), especially transport and biogenic emissions, disagrees with the current finding. Conclusions from Li et al. (2017) rely on simulations of a one-week episode in May, while our results provide more information considering different seasons and different highly polluted regions.

4.3 O₃ contribution from individual source regions

The sensitivity of O₃ pollution to individual source sectors discussed in the previous section provides a quantitative understanding of the relative importance of individual source sectors. Additionally, information on the contribution of individual source regions to O₃ pollution should provide useful inputs for O₃ control strategies. Because of the large computational costs of sensitivity simulations, we employed the tagging method to examine contributions to O₃ pollution from individual source regions. Fig. 6 presents monthly mean concentrations of O₃ averaged over the NCP, YRD, PRD and India, with contributions from individual source regions.

The NCP region is influenced largely by sources outside China, especially in wintertime, which might be attributed to less local production and a longer O₃ lifetime in winter. In winter, sources outside China are responsible for more than 75% of O₃ formation in the NCP region. However, this contribution declines to about 50% as summer approaches. Using the tagged tracer method with a global chemical transport model, Nagashima et al. (2010) suggested that sources outside China contributed about 60% and 40% to surface O₃ in North China in spring and summer, respectively. Our estimate for the contributions of sources outside China in these two seasons suggests slightly higher values: 73% and 51% (Table 2). In summer, NCP local sources
contribute about 31%, with additional 8% from Northwestern China. For the YRD region, local emissions contribute 32% to O$_3$ formation in summer, but the contribution declines by 8% in spring and autumn (Table 2). The contribution of sources outside China decreases greatly in summer (46%), leading to a small summer O$_3$ trough. The source apportionment results in Nagashima et al. (2010) also indicated that the contribution of sources outside China to O$_3$ in the Yangtze River Basin decreases significantly from spring to summer (44% to 30%). The relatively lower contribution from sources outside China is associated with the prevailing winds and cleaner air from the ocean in summer (Fig. 4c). In addition to local sources, we further identified the major source region for O$_3$ in the YRD region is the NCP in winter, spring and autumn (14%, 6% and 8%, respectively). In summer, the major source region of O$_3$ in the YRD region is Southeast China (10%). J. Gao et al. (2016) concluded that YRD local emissions contribute 13.6%-20.6% to daytime O$_3$ under different wind conditions, and the contribution of super regional sources (Outside) ranges from 32 to 34% in May. In Hangzhou (a megacity within YRD), source apportionment results reveal that long-range transport contributes 36.5% to daily maximum O$_3$ with the overall contribution dominated by local sources (Li et al., 2016).

O$_3$ concentrations in the YRD region are influenced largely by the summer monsoon, and the prevailing winds from the ocean result in a minimum contribution from polluted regions. The estimated contribution of sources outside China declines to 46% in summer, which agrees well with the number 47% inferred from Nagashima et al. (2010). Li et al. (2012) applied the OSAT tool in the CAMx model to apportion O$_3$ sources in south China, and reported that super-regional sources contributed 55% and 71% to monthly mean O$_3$ in summer and autumn, respectively. They pointed out also that regional and local sources play more important roles in O$_3$ pollution episodes (Li et al., 2002). The contribution of local source peaks in summer (41%) exceeds the local contribution in the NCP and YRD regions. As discussed in Sect. 4.1, the outflow of O$_3$ and its precursors from the NCP and YRD regions might play important roles in peak autumn O$_3$ in the YRD (Fig. 4d), as wind direction switches from summer to autumn. We identified the major upwind regions for the PRD in autumn as YRD (11%) and Southeast China (10%). From summer to autumn, the contribution of YRD sources to the PRD increases from 2% to 11%. For India, O$_3$ concentrations are dominated by sources outside India, and
sources in North India (Fig. 6d). In winter, sources outside India contribute 49%, while sources in North India contribute 38%.

The estimated contributions of sources outside China to O$_3$ pollution in receptor regions exhibit slightly higher values than the values inferred from studies using global models (Nagashima et al., 2010; Wang et al., 2011). This might be related partly to the inconsistency between simulations from the applied regional model and boundary conditions from another global model. Global chemical transport models usually show better skills in simulating transboundary pollution. In addition, the current study focuses on seasonal mean (both daytime and nighttime) O$_3$ while many previous studies investigate sources of 8-h or daily maximum O$_3$. As illustrated in Li et al. (2016), the dominant contribution to nighttime O$_3$ is associated with long-range transport. All of these factors contribute to uncertainties in the results of source apportionment, but should not downplay the significance of current findings in terms of policy implications.

5 Summary

In this study, we used a fully coupled regional meteorology-chemistry model with a horizontal grid spacing of 60 km × 60 km to study the seasonality and characteristics of sources of O$_3$ pollution in highly polluted regions in both China and India. Both observations and model results indicate that O$_3$ in the NCP, YRD, PRD, and in India display distinctive seasonal features. Surface concentrations of O$_3$ peak in summer in the NCP, in spring in the YRD, in autumn in the PRD and in winter in India. These distinct seasonal features for different regions are linked to the intensity of the summer monsoon, to sources, and to atmospheric transport.

With confidence in the model’s ability to reproduce the major features of O$_3$ pollution, we examined the sensitivity of O$_3$ pollution to individual anthropogenic emission sectors, and to emissions from biogenic sources and from burning of biomass. We found that production of O$_3$ in summer is more sensitive to industrial sources than to other source sectors for China, while the transport vehicle sector is more important for all seasons in India. For India, in addition to transport, the residential sector also plays an important role in winter when O$_3$ concentrations peak. These differences in conditions between China and India suggest differences in control strategies on economic sectors should be implemented to minimize resulting pollution.
Tagged simulations suggest that sources in east China play an important role in the formation of the summer O₃ peak in the NCP, and sources from Northwest China should not be neglected to control summer O₃ in the NCP. For the YRD region, prevailing winds and cleaner air from the ocean in summer lead to reduced transport from polluted regions, and the major source region in addition to local sources is Southeast China. For the PRD region, the upwind region is replaced by contributions from polluted east China as autumn approaches, leading to an autumn peak. The major upwind regions in autumn for the PRD are YRD (11%) and Southeast China (10%). For India, sources in North India show larger contributions than sources in South India.

The focus of our analysis is on the seasonality of O₃ pollution and its sources in both China and India, with an emphasis on implications for O₃ control strategies. Most previous studies focused on the analysis of episodes or monthly means for a region, while the current study presents a more comprehensive picture. For the NCP region, O₃ concentrations peak in summer, during which industrial sources should be given higher priority. Besides local sources in the NCP, sources from Northwest China play also important roles. For the YRD region, O₃ concentrations in spring, summer and autumn are equally important, showing appreciable sensitivity to the industrial sources. In addition to local sources, sources from the NCP should be considered for control of O₃ in spring and autumn, while sources from Southeast China should be considered in summer. For the PRD region, O₃ concentrations peak in spring and autumn, during which reducing industrial and transport sources could be more effective. In both spring and autumn, sources from the YRD and Southeast China show appreciable contributions to O₃ pollution in the PRD. For India, O₃ pollution is more serious in winter, during which controlling residential and transport sources in North India could be more effective. Although large uncertainties remain in the tagged O₃ method, notably the inconsistency of transboundary simulation using regional models, the current findings are expected to provide useful insights on the relative importance of different source sectors and regions.
4 figures are listed in the supplement.

Author contribution
M.G. and M.B.M designed the study; M.G. performed model simulations and analyzed the data with the help from J. G., B. Z., R. K., X. L., S. S., Y. Z., P. W., P. S.; G. B., J.H., Q.Y., H.Z. provided measurements. M.G. and M.B.M. wrote the paper with inputs from all other authors.

Data availability
The measurements and model simulations data can be accessed through contacting the corresponding authors.

Competing interests
The authors declare that they have no conflict of interests.

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Reference


Ghude, S. D., Jena, C., Chate, D.M., Beig, G., Pfister, G.G., Kumar, R., Ramanathan, V.: Reductions


Table 1. Descriptions of simulations

<table>
<thead>
<tr>
<th>Simulations</th>
<th>Descriptions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>Anthropogenic, biogenic and fire emissions are considered;</td>
</tr>
<tr>
<td>Industrial</td>
<td>Same as control except industry sector in anthropogenic emissions is excluded;</td>
</tr>
<tr>
<td>Residential</td>
<td>Same as control except residential sector in anthropogenic emissions is excluded;</td>
</tr>
<tr>
<td>Transportation</td>
<td>Same as control except transportation sector in anthropogenic emissions is excluded;</td>
</tr>
<tr>
<td>Power</td>
<td>Same as control except power sector in anthropogenic emissions is excluded;</td>
</tr>
<tr>
<td>Biogenic</td>
<td>Same as control except biogenic emissions are excluded;</td>
</tr>
<tr>
<td>Fire</td>
<td>Same as control except fire emissions are excluded;</td>
</tr>
<tr>
<td>Season</td>
<td>NCP</td>
</tr>
<tr>
<td>---------</td>
<td>--------------</td>
</tr>
<tr>
<td>Winter</td>
<td>Outside: 81%</td>
</tr>
<tr>
<td></td>
<td>Local: 12%</td>
</tr>
<tr>
<td></td>
<td>NW China: 6%</td>
</tr>
<tr>
<td>Spring</td>
<td>Outside: 73%</td>
</tr>
<tr>
<td></td>
<td>Local: 17%</td>
</tr>
<tr>
<td></td>
<td>NW China: 5%</td>
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<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Summer</td>
<td>Outside: 51%</td>
</tr>
<tr>
<td></td>
<td>Local: 31%</td>
</tr>
<tr>
<td></td>
<td>NW China: 8%</td>
</tr>
<tr>
<td>Autumn</td>
<td>Outside: 69%</td>
</tr>
<tr>
<td></td>
<td>Local: 21%</td>
</tr>
<tr>
<td></td>
<td>NW China: 7%</td>
</tr>
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<td></td>
<td></td>
</tr>
</tbody>
</table>

(Outside sources include also transport from upper boundary of the model; NCP: Beijing, Tianjin, Hebei, Shandong, and Henan; YRD: Anhui, Jiangsu, Shanghai and Zhejiang; SE China: Jiangxi, Fujian and Taiwan; Central China: Hunan and Hubei; South China: Guangxi and Hainan)
Fig. 1. WRF-Chem domain setting with terrain height and the locations of surface ozone observations marked by solid red circles. Purple solid circles mark the location of ozonesonde observations.

Fig. 2. Spatial distribution of simulated and observed seasonal mean ozone concentrations for Winter (a), Spring (b), Summer (c) and Fall (d).
Fig. 3. Observed and simulated monthly mean O$_3$ concentrations averaged for the North China Plain (NCP) (a), Yangtze River Delta (YRD) (b), Pearl River Delta (PRD) (c), and India (d).
Fig. 4. Modeled mean near surface wind fields and the monsoon index in the boundary layer (0-1.5km) for winter (December, January, and February, a), spring (March, April, and May, b), summer (June, July and August, c), and autumn (September, October, and November, d)
Fig. 5. Distributions of the contributions to near-surface ozone averaged for winter, spring, summer and autumn from industry (a-d), power, (e-h), residential (i-l), transport (m-p), biogenic (q-t) and fire (u-x) emissions.
Fig. 6. Contributions to monthly mean ozone in NCP (a), YRD (b), PRD (c), and India (d) from different source regions (NCP: Beijing, Tianjin, Hebei, Shandong, and Henan; YRD: Anhui, Jiangsu, Shanghai and Zhejiang; SE China: Jiangxi, Fujian and Taiwan; Central China: Hunan and Hubei; South China: Guangxi and Hainan)