Interannual and Decadal Changes in Tropospheric Ozone in China and the Associated Chemistry–Climate Interactions: A Review

Yu FU[1], Hong LIAO[*2], and Yang YANG[2]

[1]*Climate Change Research Center, Chinese Academy of Sciences, Beijing* 100029*, China*

[2]*Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing* 210044*, China*

ABSTRACT

China has been experiencing widespread air pollution due to rapid industrialization and urbanization in recent decades. The two major concerns of ambient air quality in China are particulate matter (PM) and tropospheric ozone (O_3). With the implementation of air pollution prevention and control actions in the last five years, the PM pollution in China has been substantially reduced. In contrast, under the conditions of the urban air pollution complex, the elevated O_3 levels in city clusters of eastern China, especially in warm seasons, have drawn increasing attention. Emissions of air pollutants and their precursors not only contribute to regional air quality, but also alter climate. Climate change in turn can change chemical processes, long-range transport, and local meteorology that influence air pollution. Compared to PM, less is known about O_3 pollution and its climate effects over China. Here, we present a review of the main findings from the literature over the period 2011–18 with regard to the characteristics of O_3 concentrations in China and the mechanisms that drive its interannual to decadal variations, aiming to identify robust conclusions that may guide decision-making for emissions control and to highlight critical knowledge gaps. We also review regional and global modeling studies that have investigated the impacts of tropospheric O_3 on climate, as well as the projections of future tropospheric O_3 owing to climate and/or emission changes.

Key words: tropospheric ozone, chemistry–climate interactions, interannual and decadal variations, China

Article Highlights:

- Available data reveal an increasing trend of tropospheric O_3 in China with more frequent pollution episodes in the last five years.
- An integrated literature review on the mechanisms of interannual to decadal variations in tropospheric O_3 over China is provided.
- Future directions of O_3 air quality research in China are suggested.

---

1. Introduction

Ozone (O_3) pollution in North America and Europe has been alleviated as a result of the rigorous emission controls since the 1990s, whereas the O_3 levels in East Asia have been increasing (Cooper et al., 2014; Chang et al., 2017; Gaudel et al., 2018), especially in the rapidly developing city clusters [e.g., the Yangtze River Delta (YRD), the Pearl River Delta (PRD), and the North China Plain (NCP)]. Observed concentrations from 74 Chinese cities showed that the mean value of daily maximum 8-h average O_3 concentrations (MDA8) for warm seasons (April–September) increased by approximately 3% yr^{-1} over 2013–17 (Lu et al., 2018) and the mean value of the 90th percentile of MDA8 O_3 increased from 139 μg m^{-3} (~65 ppbv[a]) in 2013 to 167 μg m^{-3} (~78 ppbv) in 2017 (Fig. 1). High O_3 concentrations at the ground level have been shown to pose major threats to human health and the ecosystem, as well as crop production, thereby having ramifications for future food security (Lim et al., 2012; Feng

---

* Corresponding author: Hong LIAO

  Email: hongliao@nuist.edu.cn

[a]The atmospheric conditions for temperature (273.15K) and pressure (1013 hPa) are applied for unit conversion from μg m^{-3} to mixing ratio (ppbv) in this paper.

et al., 2015; Wang et al., 2017a). Furthermore, tropospheric O_3 plays a key role in determining the oxidation capacity of the atmosphere as a photochemical precursor of the hydroxyl radical (OH), and thus impacts the formation of other secondary pollutants, such as sulfate and secondary organic aerosol (Jacob, 2000). In addition, tropospheric O_3 has received continuous attention from both the scientific and regulatory communities because of its effect on climate. Tropospheric O_3 is estimated to have the third-highest positive radiative forcing after carbon dioxide (CO_2) and methane (CH_4) (Myhre et al., 2013).

Tropospheric O_3 is a secondary air pollutant produced by complex photochemical reactions of nitrogen oxides [NO_x = nitric oxide (NO) + nitrogen dioxide (NO_2)], carbon monoxide (CO), CH_4, and non-methane volatile organic compounds (VOCs). The downward transport of O_3 from the stratosphere is also an important source of tropospheric O_3. The concentration of O_3 in the troposphere is influenced by changes in the source emissions of its precursors (e.g., VOCs and NO_x). Most precursors are from anthropogenic sources, and some precursors can come from natural sources, such as biogenic VOCs (BVOCs) and soil and lightning NO_x. The relationships between O_3 and its precursors are complex. In general, O_3 formation is nonlinearly dependent on its precursors, and can typically be summarized into two categories of chemical regimes—"NO_x-limited" and "VOC-limited"—which are determined by the VOCs/NO_x ratio, VOC reactivity, photochemical aging, and meteorological conditions (Seinfeld and Pandis, 2006). In the NO_x-limited regime, increase in NO_x leads to increased O_3 concentration, with a weak sensitivity to VOC concentration. In the VOC-limited regime (the usual case in polluted regions in China), in contrast, increase

in VOCs leads to increased O_3 with a weak or even negative sensitivity to NO_x concentration. The transitional conditions of dual sensitivities also occur. Therefore, identifying the O_3 formation regime is essential for the regulation of O_3 air pollution regionally (Xing et al., 2011; Ma et al., 2012). In addition to emissions, changes in O_3 concentrations are also influenced by changes in meteorological conditions. Previous studies have extensively suggested that high O_3 events are usually associated with conducive meteorological factors (e.g., high temperature, low wind speed and relative humidity) and typically synoptic circulation patterns (e.g., anticyclones), which can enhance chemical production and/or accumulation of O_3 in the boundary layer (Xu et al., 2011b; Shen et al., 2015; Zhao et al., 2016). Moreover, meteorological conditions can alter the stratosphere–troposphere exchange of O_3, which influences the lower tropospheric O_3 (Stohl et al., 2003; Li et al., 2015; Lin et al., 2015). O_3 is mainly removed from the troposphere by chemical reactions. Dry deposition onto the surface, in particular to vegetation, is also a dominant sink of tropospheric O_3. These removal processes are also affected by meteorological conditions.

In recent decades, the observed aerosol concentrations have been extremely high in China (Zhang et al., 2012b). These aerosols have large impacts on O_3 concentrations by heterogeneous reactions and by altering photolysis rates (Li et al., 2011; Lou et al., 2014; Feng et al., 2016). With the implementation of air pollution prevention and control actions in the last five years, assessing the potential changes in O_3 concentration under the conditions of decreases in aerosols has become an emerging scientific topic in China.

In this paper, we review the current understanding of the spatial and temporal variations of O_3 concentrations in China,
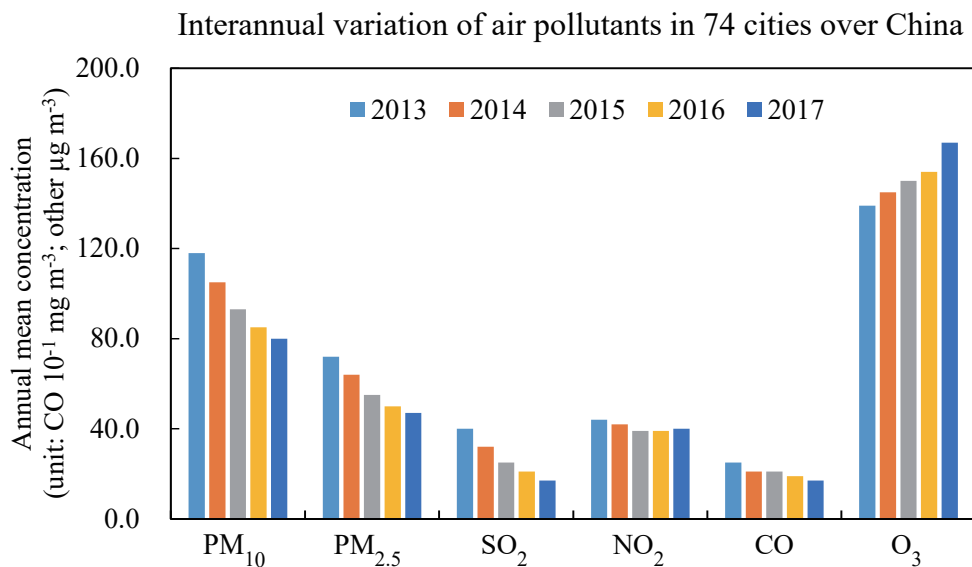

Fig. 1. Interannual variations in annual mean concentrations of PM_{10}, $PM_{2.5}$, SO_2, NO_2, CO, and O_3 based on the monitoring results from 74 Chinese cities over 2013–17. The O_3 values represent the mean value of the 90th percentile of MDA8 O_3. The units for CO are mg m^{-3}, while those for other species are µg m^{-3}. For clarity, the CO values are scaled by a factor of 10. All the data were obtained from the China Environment Report, 2013–17, available at http://www.mep.gov.cn, in Chinese.

the factors or processes that influence O_3 formation and distribution, and the interactions between tropospheric O_3 and climate in China, by summarizing the findings from the literature over the period 2011–18. Section 2 reviews the spatial and temporal variations of tropospheric O_3 based on observations; section 3 summarizes the impacts of changes in anthropogenic emissions, climate, land cover and land use, as well as aerosols, on O_3 concentrations; section 4 reviews the effects of O_3 on the climate over China; and section 5 provides a summary and recommendations for future studies.

2. Characteristics of observed O_3 in China

Field measurements of O_3 are important for understanding the characteristics of O_3 concentrations. They also provide datasets for evaluating model simulations and improving the representation of chemical processes in models. Although numerous long-term monitoring sites have been established worldwide [mostly focusing on North America, Europe, and East Asia (Japan and Korea)] (Sofen et al., 2016; Schnell and Prather, 2017; Schultz et al., 2017; Gaudel et al., 2018; Yan et al., 2018), O_3 monitoring was very limited in mainland China before the mid-2000s, partly because most studies and control efforts in China were focused on particulate matter (PM) air quality in the early years. In 2012, the Chinese government revised the national ambient air quality standards, setting O_3 pollution standards for urban and industrial areas as an hourly concentration of 200 μg m^{-3} or MDA8 of 160 μg m^{-3} [GB3095-2012, National Ambient Air Quality Standards (NAAQS), in Chinese, http://kjs.mep.gov.cn/hjbhbz/bzwb/dqhjbh/dqhjzlbz/201203/t20120302_224165.htm, accessed on 31 August 2018]. Meanwhile, the surface O_3 be-

came one of the species regularly monitored by the National Air Quality Monitoring Network, which became operational in 2013. The network covered 74 major cities in 2013, and, by the end of 2017, had nearly 1436 monitoring stations in 338 Chinese cities (Fig. 2). This monitoring network of the Ministry of Ecology and Environment of China reports hourly air quality data including concentrations of O_3, CO, NO_2, sulfur dioxide (SO_2), and $PM_{2.5}$ (PM with a diameter less than 2.5 μm) (http://106.37.208.233:20035/).

2.1. Seasonal variation of O_3 concentrations

Numerous studies have reported severe O_3 episodes observed in the most developed regions of China, where O_3 concentrations are influenced by large anthropogenic emissions of O_3 precursors. During severe O_3 pollution episodes, the maximum 1-h O_3 mixing ratio reached 170–270 ppbv in the Beijing–Tianjing–Hebei (BTH) region (Xu et al., 2011a; Wei et al., 2015), 130–180 ppbv in the YRD (Ding et al., 2013; An et al., 2015; Shi et al., 2015), and 200–220 ppbv in the PRD (Lu et al., 2012; Ou et al., 2016; Wang et al., 2016b). Remarkably, Li et al. (2017a) reported that, in April–September of 2015, the maximum hourly O_3 concentrations exceeded 400 μg m^{-3} (~200 ppbv) in more than 60 cities in eastern China, indicating widespread O_3 pollution in China.

Tables 1 summarize the observational studies of O_3 in China published over the period of 2011–18. Most of these studies were focused on the spatial and temporal variations of surface-layer O_3 and precursors of O_3. In general, the seasonal mean surface-layer O_3 concentrations were within the range of 20–60 ppbv, with the highest concentrations occurring in warm seasons in eastern China (Xu et al., 2011a; Tang et al., 2012; Wang et al., 2012, 2013a, 2017b; Ding et al.,
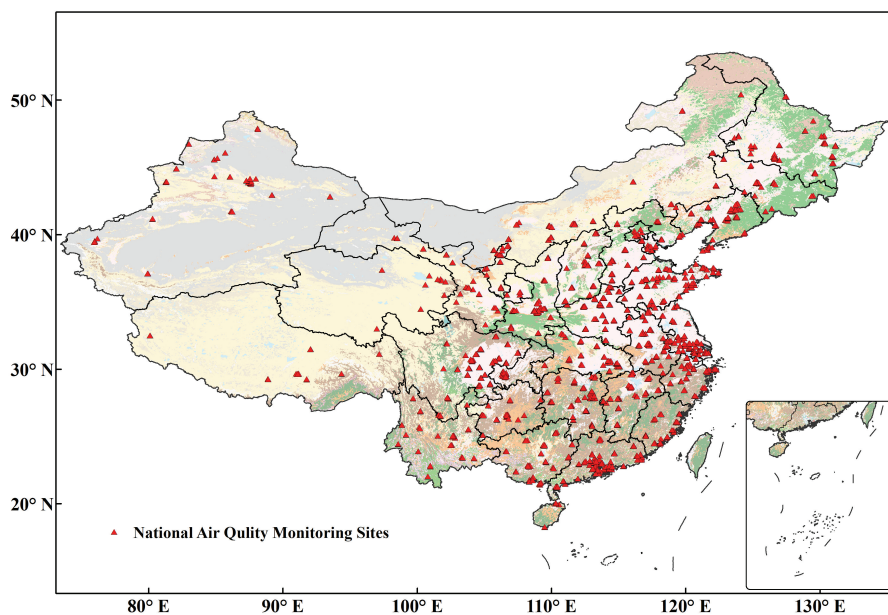

Fig. 2. The National Air Quality Monitoring Network of China in 2017, featuring 1436 monitoring sites in 338 cities, developed by the China National Environmental Monitoring Center. Data source: http://www.cnemc.cn/ and http://beijingair.sinaapp.com/.

Table 1. Summary of field measurements of O$_3$ in China based on the literature during 2011–18.

(a) NCP region					
			Concentration (ppbv)		
Location	Measurement period		Mean	Maximum	Reference
Shangdianzi (SDZ), Beijing	2004–15	Annual	49–60	120–165	Ma et al. (2016)
SDZ, Beijing	Jun–Sep 2007		58	189	Xu et al. (2011a)
Fengtai, Beijing	Jun–Sep 2007		36	171	Xu et al. (2011a)
Baolian, Beijing	Jun–Sep 2007		47	275	Xu et al. (2011a)
Shunyi, Beijing	Jun–Sep 2007		40	200	Xu et al. (2011a)
Wuqing, Tianjin	Jul 2009–Jan 2010	Jul–Aug	45	194	Xu et al. (2011b)
Wuqing, Tianjin	Jul 2009–Jan 2010	Sep–Oct–Nov	16	88	Xu et al. (2011b)
Wuqing, Tianjin	Jul 2009–Jan 2010	Dec–Jan	10	39	Xu et al. (2011b)
Xinglong, Hebei	Aug 2009–Jun 2010		47.3 ± 11.5	87.2 (Jun)	Wang et al. (2013a)
Gucheng, Hebei	Aug 2009–Jun 2010		19.4 ± 7.9	44.3 (Jun)	Wang et al. (2013a)
Longtanhu, Beijing	Aug 2009–Jun 2010		19.8 ± 8.7	45.1 (Jun)	Wang et al. (2013a)
Yanshan, Beijing	2010–11	Jul–Aug	63.5	120.7	Wei et al. (2015)[*]
Multiple sites, Beijing	Dec 2012–Nov 2013	Annual	23.1 ± 25.6		Wang et al. (2015)[*]
Multiple sites, Beijing–Tianjin–Hebei	2013–15	Annual	~ 42		Wang et al. (2017b)[*]
Changping, Beijing	21 Jun–31 Jul 2005			286	Xue et al. (2014)

(b) YRD region					
			Concentration (ppbv)		
Location	Measurement period		Mean	Maximum	Reference
Caochangmen, Nanjing	Jan–Dec 2008	Annual	28 ± 20		Xie et al. (2016)
Pukou, Nanjing	Jan–Dec 2008	Annual	31 ± 11		Xie et al. (2016)
Nanjing	Jun–Aug 2013	Daily		110–135	An et al. (2015)
Jiangdu, Jiangsu	Mar 2007–Dec 2011		54.6 ± 25.3 (Jun)	144	Tang et al. (2013)
Jiangdu, Jiangsu	Mar 2007–Dec 2011		39.4 ± 18.0 (Oct)	144	Tang et al. (2013)
Xianlin, Nanjing	Aug 2011–Jul 2012		8–31	~ 136	Ding et al. (2013)[*]
Fudan University, Shanghai	Jul–Aug 2013		61–117	~ 182	Shi et al. (2015)[*]
Multiple sites, YRD	2013–15	Annual	~ 46		Wang et al. (2017b)[*]
Sites in Nanjing, Shanghai and Hangzhou	Jun–Aug 2013			160.5	Pu et al. (2017)
Taicang, Shanghai	May–Jun 2005			127	Xue et al. (2014)

(c) PRD region					
			Concentration (ppbv)		
Location	Measurement period		Mean	Maximum	Reference
Panyu, Guangzhou	Jun 2011–May 2012	Spring	17.7		Zou et al. (2015)
Panyu, Guangzhou	Jun 2011–May 2012	Summer	26		Zou et al. (2015)
Panyu, Guangzhou	Jun 2011–May 2012	Autumn	25.6		Zou et al. (2015)
Panyu, Guangzhou	Jun 2011–May 2012	Winter	13.2		Zou et al. (2015)
Multiple sites, Guangzhou	2010		17–50	178–198	Liu et al. (2013a)[*]
Multiple sites, PRD	2013–15	Annual	~ 44		Wang et al. (2017b)[*]
Wuqingsha, Guangzhou	Apr–May 2004			178	Xue et al. (2014)
PRD regional air quality monitoring network	2006–12			162–217	Li et al. (2013)

(d) Other regions in China					
			Concentration (ppbv)		
Location	Measurement period		Mean	Maximum	Reference
Xi'an	Mar 2008–Jan 2009	Spring	21.8	139.7	Wang et al. (2012)
Xi'an	Mar 2008–Jan 2009	Summer	32.5	131.5	Wang et al. (2012)
Xi'an	Mar 2008–Jan 2009	Autumn	8.8	106.5	Wang et al. (2012)
Xi'an	Mar 2008–Jan 2009	Winter	3.0	22.6	Wang et al. (2012)
Xi'an	Jun–Aug 2013		~ 46–61	~ 87–107	Feng et al. (2016)[*]
Renshoushan Park, Lanzhou	19 Jun–16 Jul 2006			143	Xue et al. (2014)

[*] Values of the observation are approximately converted from μg m^{-3} to ppbv under 298 K, 1013 hPa.

2013; Chen et al., 2015). For example, Tang et al. (2012) reported highest O_3 concentrations in June and the lowest values in December in Beijing, which were attributed to local meteorological conditions. Ding et al. (2013) found that O_3 levels over the YRD exhibited a maximum in July, a second maximum in September, and a minimum in November, on the basis of the observations from August 2011 to July 2012. Tang et al. (2013) also found a bimodal seasonal pattern of O_3 at an agricultural site in the northern YRD, which were caused by winds associated with monsoon and open crop residue burning. In comparison, O_3 concentrations were found to be minimum in summer and maximum in autumn over the PRD region (Hu et al., 2016; Liu et al., 2017). In summer, prevailing southerlies with clean air from the oceans and the large rainfall associated with the East Asian monsoon leads to the decreases in O_3 levels. In autumn, the continental flow from the north with polluted air, in conjunction with the stable and warm weather, contributes to the high O_3 concentrations (Newell and Evans, 2000; Wang et al., 2009; Zheng et al., 2010; Wang et al., 2017c). By analyzing 16 ozonesonde observations during a field campaign in April–May of 2005, Huang et al. (2015) found that the observed O_3 enhancement in the lower troposphere in Beijing in the late spring and early summer was attributed to an earlier onset of the East Asian summer monsoon (EASM).

2.2. *Trends of O_3 in recent decades*

Recent studies in the framework of the Tropospheric Ozone Assessment Report, supported by IGAC (International Global Atmospheric Chemistry), showed that observed monthly mean MDA8 O_3 levels in South Korea and Japan were increasing with a regional trend of 0.35 ppbv yr^{-1} over the 2000–14 period (Chang et al., 2017; Gaudel et al., 2018). In China, knowledge of decadal variation of O_3 on a nationwide scale is limited due to the lack of continuous measurements. Only a small number of studies have conducted long-term measurements of O_3 (Zhou et al., 2013; Li et al., 2014; Ma et al., 2016; Sun et al., 2016; Xu et al., 2018). In the NCP, based on the O_3 profiles derived from the MOZAIC (Measurement of Ozone and Water Vapor on Airbus In-service Aircraft) and IAGOS (In-service Aircraft for the Global Observing System) programs, Ding et al. (2008) showed an increasing trend of O_3 of 1.0 ppbv yr^{-1} in the lower troposphere during 1995–2005. Zhang et al. (2014) found that the averaged O_3 in summer in Beijing increased by 2.6 ppbv yr^{-1} (5% yr^{-1}) during 2005–11, although NO_x and total VOC mixing ratios decreased by 1.4 ppbv yr^{-1} and 1.6 ppbv yr^{-1}, respectively. They explained that such increases in O_3 mainly resulted from the decreased NO titration and elevated regional O_3 background concentration in the NCP, suggesting that the reduction of VOCs should be coordinated with NO_x emission controls in urban areas. Ma et al. (2016) and Sun et al. (2016) reported that the annual mean surface O_3 concentration increased significantly at two background sites in the NCP over 2003–15, with an average rate of 1.13 ± 0.01 ppbv yr^{-1} at Shangdianzi and 2.1 ppbv yr^{-1} at Mount Tai. Recent analyses by Zhang et al. (2016) and Gaudel et al. (2018) showed by using IAGOS data that O_3 in the NCP increased largely in the boundary layer, with the summertime concentrations increased by 15% from 1995 to 2014. In the PRD region, Li et al. (2014) observed an increase in O_3 of 2.0 ppbv yr^{-1} and a decrease in NO_2 of 1.2 ppbv yr^{-1} from 2006 to 2011 in suburban/urban areas, implying that photochemical O_3 production in the PRD was mainly VOC-controlled. Zhang et al. (2013) reported increases in O_3 concentrations in Hong Kong during 1999–2011 and suggested that the changes in meteorology contributed 36% to the observed increases. In western China, observed O_3 concentrations in spring and autumn at Mount Waliguan of the Tibetan Plateau also showed an increasing trend, of 0.2–0.3 ppbv yr^{-1}, over 1994–2013, which was caused by the increased springtime transport from the stratosphere to the troposphere and was influenced by polluted air from Southeast Asia in autumn (Xu et al., 2016, 2018).

Satellite remote sensing is a useful approach to investigate tropospheric O_3 over large regions, although retrieving tropospheric O_3 from satellite observations is still challenging, as tropospheric O_3 contributes only approximately 10% to the total atmospheric column O_3 (Seinfeld and Pandis, 2006). Currently, there are several commonly used tropospheric column ozone products retrieved from the new generation of nadir-looking instruments onboard polar-orbiting satellites, such as IASI (Infrared Atmospheric Sounding Interferometer)-FORLI (Fast Optimal Retrievals on Layers), IASI-SOFRID (Software for a Fast Retrieval of IASI Data), IASI-LISA (Laboratoire Interuniversitaire des Systèmes Atmosphériques), GOME (Global Ozone Monitoring Experiment), IASI+GOME-2, OMI (Ozone Monitoring Instrument)/MLS (Microwave Limb Sounder), OMI-SOA (Smithsonian Astrophysical Observatory), OMI-RAL (Rutherford Appleton Laboratory), and SCIAMACHY (Scanning Imaging Spectrometer for Atmospheric Chartography) (Gaudel et al., 2018). Note that the SCIAMACHY time series ended in 2012 and cannot be continued because the satellite became inoperable and later crashed. Xu and Lin (2011c) investigated the decadal trend of tropospheric O_3 over China during 1979–2005 using the NASA satellite product of tropospheric ozone residue (TOR), a surrogate of tropospheric column O_3, and found a decreasing trend of TOR over the PRD and the Sichuan Basin and a significant increasing trend of TOR over the NCP for all seasons except winter. Based on Tropospheric Emission Spectrometer (TES) measurements of tropospheric O_3 and NO_2, Verstraeten et al. (2015) reported that tropospheric O_3 concentrations over China increased by approximately 7% between 2005 and 2010, as a result of the 21% increase in Chinese emissions and the enhanced downward transport of stratospheric O_3. Hayashida et al. (2015) analyzed O_3 in the lower troposphere over central and eastern China on the basis of OMI O_3 profiles over 2005–13 and pointed out that O_3 enhancement was the largest in June among all the months. However, using the lower-tropospheric columns derived from the IASI satellite instrument, Dufour et al. (2018) found an overall trend of −0.77% yr^{-1} over the NCP for 2008–16, and large-scale

dynamical processes such as QBO (quasi-biennial oscillation) and ENSO (El Niño–Southern Oscillation) explain more than half of the observed trend, with a possible reduction in stratosphere-to-troposphere transport being the main contributor. It is noted that the negative trend derived in the lower troposphere from IASI is inconsistent with the positive trend derived from the surface measurements over the NCP (Ma et al., 2016; Sun et al., 2016). A possible explanation for this inconsistency is that the lower troposphere and surface O$_3$ respond differently to the recent reductions in NO$_x$. In addition, the uncertainties in satellite retrievals of tropospheric O$_3$ trends should be addressed in future studies.

3. Mechanisms for the variations of tropospheric O$_3$ in China

As mentioned in the introduction, tropospheric O$_3$ is a secondary air pollutant formed from a combination of nitrogen oxides (NO$_x$ = NO$_2$ + NO), VOCs, CO and CH$_4$ in the presence of sunlight. Several studies and reviews are available in the literature that describe the O$_3$ photochemistry reactions (e.g. Monks, 2005; Monks et al., 2015). For better understanding of the variations in tropospheric O$_3$ and its precursors, readers are referred to those papers for a comprehensive description of the chemical mechanisms of O$_3$.

3.1. *Impacts of changes in anthropogenic emissions*

The most important O$_3$ precursors in the atmosphere include NO$_x$, CO and VOCs, which emit from both anthropogenic and biogenic sources. For example, the majority of NO$_x$ emitted into the atmosphere is from anthropogenic fossil fuel combustion (i.e., industry, power, residential, and transportation), with the remaining emissions from biomass burn-

ing, soil, and lightning. Non-methane VOCs have large anthropogenic sources, but vegetation also represents a significant source depending on the region.

Many researchers have put tremendous efforts into developing emissions inventories covering China (Streets et al., 2003, 2006; Ohara et al., 2007; Zhang et al., 2009; Kurokawa et al., 2013; Li et al., 2017b). Table 2 summarizes several Asian emissions inventories for China that are widely used for atmospheric chemistry–climate research. Previous studies have reported the characteristics and the trends of anthropogenic emissions in China (Zhang et al., 2012a; Zhao et al., 2013; Wang et al., 2014; Wu et al., 2016; Zheng et al., 2018). For instance, Kurokawa et al. (2013) showed that emissions of NO$_x$, CO and non-methane VOCs over China increased by 89%, 42% and 71%, respectively, from 2000 to 2008. Wang et al. (2014) also found that the emissions of NO$_x$ and non-methane VOCs in China increased by 25% and 15%, respectively, during 2005–10, due to inadequate control strategies. Wu et al. (2016) reported that China's anthropogenic VOC emissions increased by 7.38% (from 22.45 Tg in 2008 to 29.85 Tg in 2012). Facing the big challenge of improving air quality, emission mitigation measures were implemented gradually in China during the 11th Five-Year Plan (2006–10) and the 12th Five-Year Plan (2011–15). Xia et al. (2016) estimated the emissions of SO$_2$, NO$_x$ and CO for 2000–14 in China to evaluate the effectiveness of national air pollution control policies and found that the estimated NO$_x$ emissions increased from 11.7 Tg in 2000 to 29.8 Tg in 2011, and then decreased to 27.7 Tg in 2014. Recently, Zheng et al. (2018) showed that, during 2013–17, anthropogenic NO$_x$ emissions decreased by 21% (from 27.7 Tg in 2013 to 22.0 Tg in 2017), while non-methane VOCs emissions did not show a significant change (from 28.1 Tg in 2013 to 28.6 Tg in 2017), indicating the absence of effective control measures for non-

Table 2. Summary of regional anthropogenic emissions inventories covering China.

Inventory name	Description	Time periods	Reference
TRACE-P inventory	Asian emissions inventory under the support of the Transport and Chemical Evolution over the Pacific Campaign	2000	Streets et al. (2003, 2006)
INTEX-B inventory	Emissions inventory for Asia as an update and improvement of the TRACE-P inventory under NASA's Intercontinental Chemical Transport Experiment Phase B mission	2006	Zhang et al. (2009)
REAS	Regional Emissions Inventory in Asia, which includes SO$_2$, NO$_x$, CO, non-methane VOCs, PM$_{10}$, PM$_{2.5}$, BC, OC, NH$_3$, CH$_4$, N$_2$O, and CO$_2$	Historical periods (1980–2003) Projection (2010–20) 2000–08 (REAS 2.1)	Ohara et al. (2007) Kurokawa et al. (2013)
MIX	Anthropogenic emissions inventory under the Model Inter-Comparison Study for Asia project in phase III and the Task Force on Hemispheric Transport of Air Pollution	2008 and 2010	Li et al. (2017b)
MEIC	Multi-resolution Emissions Inventory for China, developed by Tsinghua University under an open access model framework that provides model-ready emissions data over China	1990–2015	Hong et al. (2017) Zheng et al. (2018) http://www.meicmodel.org/

methane VOCs in current policies.

Based on existing emissions inventories, the air quality degradation associated with elevated O_3 levels in China has been demonstrated to be primarily driven by increasing anthropogenic emissions of O_3 precursors. Some modeling studies have highlighted the importance of historical changes in anthropogenic emissions. Lou et al. (2015) found that the interannual variations in anthropogenic emissions in China over 2004–12 led to changes in summertime O_3 by 2–2.5 ppbv in the Sichuan Basin and South China. Fu and Tai (2015) showed that multidecadal changes in anthropogenic emissions between 1985 and 2005 enhanced summertime O_3 by up to 25 ppbv in eastern China. Some modeling studies have also examined the possible impact of future changes in anthropogenic emissions on O_3 concentrations. Liu et al. (2013b) indicated that the changes in anthropogenic emissions between 2005–07 and 2055–57 following the A1B scenario of the IPCC Special Report on Emission Scenarios would result in an average change of surface-layer O_3 by 12.8 ppbv in October over southern China. Wang et al. (2013b) also showed that surface O_3 would increase by 3.4 ± 4.8 ppbv in East China, due to the 2000–50 changes in anthropogenic emissions under the IPCC A1B scenario. By using the future changes in emissions under the IPCC Representative Concentration Pathways (RCP2.6, RCP4.5, RCP6.0 and RCP8.5), Zhu and Liao (2016) simulated that, relative to the year 2000, the maximum increases in the annual mean surface O_3 concentrations of 6–12 ppbv would occur over southern China in 2020–30 under RCP8.5 and in 2040–50 under RCP6.0. Meanwhile, extremely high numbers of O_3 exceedance days would be found over most developed regions of China (i.e., the NCP, YRD and PRD) in 2050 under RCP6.0 and in 2030 under RCP8.5.

To improve O_3 air quality, some studies have evaluated the effects of possible emission control regulations on O_3 concentrations at the regional scale. Li et al. (2013) showed that NO_x regulation in the PRD region, especially in regions where O_3 formation was found to be predominantly VOC-limited, could increase the mean O_3 level but decrease the peak O_3 concentration. Liu et al. (2013a) found that the simulated O_3 non-attainment rate (days in a year that exceed the air quality standard according to the grade II NAAQS for protection of residential areas) in Guangzhou would increase from 7.1% in 2010 to 12.9% in 2025 under a $PM_{2.5}$-based emission control scenario, in which the emissions of NO_x and VOCs in 2025 would be reduced by 51.9% and 41.3%, respectively, compared to 2010. Ou et al. (2016) explored the feasibility of O_3 control measures in the PRD by using a set of control scenarios in a numerical model and concluded that VOC-focused controls would be efficient for reductions of peak 1-h O_3 concentration and for long-term O_3 regulation (achieving the O_3 air quality standard). Lu et al. (2018) showed that observed surface O_3 concentrations increased continuously in China over 2013–17 when NO_x emissions were reduced, suggesting that O_3 chemical production in eastern China was VOC-limited and decreases in NO_x emissions or increases in VOCs emissions would en-

hance O_3 pollution. Recently, Li et al. (2019) also investigated the trend in summertime O_3 in China over 2013–17, and attributed the increasing O_3 trends of 1–3 ppbv yr^{-1} in eastern China to the approximate 40% decrease in $PM_{2.5}$ concentrations. The reductions in $PM_{2.5}$ slow down the aerosol sink of hydroperoxy (HO_2) radicals and thus produce more O_3. These findings indicate that O_3 control (e.g., VOCs control) must be taken into account when designing $PM_{2.5}$ control strategies and VOCs/NO_x reduction ratios must be further investigated.

3.2. *Impacts of multiscale meteorological conditions and climate change on O_3 concentrations*

Other than anthropogenic emissions, changes in meteorological variables, including temperature, precipitation, humidity, winds, clouds and stagnation events, are expected to affect O_3 concentrations via changing physical and chemical processes within the atmosphere (i.e., photochemical production and loss, ventilation, dilution, and removal processes), altering natural emissions of O_3 precursors and disturbing the stratosphere–troposphere exchange (Fiore et al., 2015).

3.2.1. *Impacts of changes in meteorological conditions on O_3 concentrations*

A number of studies have examined local meteorological conditions during severe O_3 pollution episodes in China based on observations. They showed that high temperature, intense solar radiation, low relative humidity, low wind velocity and low planetary boundary layer height are conducive to the photochemical production and accumulation of O_3 and its precursors (Xu et al., 2011b; Zhao et al., 2016). Changes in wind direction and wind speed can also perturb regional O_3 pollution transport and thereby affect the O_3 air quality in downwind locations (Ma et al., 2011). Some regional studies have demonstrated that synoptic-scale circulation/weather patterns such as tropical cyclones in the western Pacific, continental anticyclones (high pressure systems), cold fronts, and the intensity of the western Pacific subtropical high (WPSH) are critical meteorological conditions aggravating the occurrence and maintenance of surface O_3 pollution in different regions (Ding et al., 2013; Zhang et al., 2013; Liao et al., 2017; Zhao and Wang, 2017). For instance, O_3 episodes with hourly concentrations exceeding 120 ppbv during the period of 1999–2011 in Hong Kong were mainly due to regional transport under the influence of tropical cyclones and photochemical reactions upon the prevalence of the anticyclonic circulation (Zhang et al., 2013). The co-occurrence of high O_3 and $PM_{2.5}$ episodes in the YRD region is also related to anticyclones, since they may cause favorable local meteorological conditions for O_3 production and pollution accumulation, such as sunny weather and low wind speeds (Ding et al., 2013). The variability of the WPSH intensity has been identified as a meteorological driver affecting surface O_3 variations in eastern China. Zhao and Wang (2017) found that a stronger WPSH can enhance the southwesterly transport of moisture into South China, which is not conducive to O_3 formation (i.e., higher RH, more cloudiness and precipitation, less UV

radiation, and lower temperatures), leading to a decrease in O$_3$ over South China. Meanwhile, a strong WPSH creates dry and sunny conditions over North China and therefore leads to an increase in O$_3$ there.

Several modeling studies have examined high O$_3$ pollution events in China. Most global and regional chemical transport models (CTMs) (e.g., GEOS-Chem, WRF-Chem, CMAQ, RegCM-CHEM) are able to capture the magnitude and distribution of O$_3$ concentrations, as well as the occurrence of O$_3$ pollution episodes, although model biases still exist in reproducing the highest or lowest values due to the uncertainties related to meteorological fields, emissions and model parameterizations (Hu et al., 2016; Li et al., 2017a). By using the regional chemistry–climate model RegCM-CHEM4, Pu et al. (2017) simulated the impact of a heat wave on a high O$_3$ event in the YRD region that occurred in the summer of 2013 and found that chemical reactions associated with the high temperature played the most important role, which resulted in a 12 ppbv O$_3$ enhancement compared to a non-heat wave period. Modeling analysis also indicated that the enhanced emission of biogenic isoprene under dry and warm conditions is another significant contributor to the extreme high O$_3$ pollution events (Geng et al., 2011; Wang et al., 2012). Meanwhile, both anthropogenic and biogenic O$_3$ precursors can be transported far from the sources, which can result in high O$_3$ concentrations in the downwind locations (Wang et al., 2012; Xue et al., 2014). Hu et al. (2018) explored the underlying mechanism of a severe O$_3$ episode over the PRD region using the WRF-Chem model and indicated that the regional O$_3$ transport contributed considerably to the rapid increase in the surface O$_3$ level.

Previous studies have reported that variations in meteorological fields lead to large interannual variations of O$_3$ concentration in East Asia. Jeong and Park (2013) examined the variability on O$_3$ concentration for the period of 1985–2006 using the global chemical transport model GEOS-Chem and showed that changes in cloud cover and temperature were important for O$_3$ variability. Fu and Liao (2012) found that the interannual variations in meteorological parameters changed the summertime O$_3$ concentrations over 2001–06 by 3–5 ppbv. Lou et al. (2015) indicated that, in North China during 2004–12 (Fig. 3), the interannual variation in simulated surface O$_3$ concentrations induced by variations in meteorology were larger than those caused by changes in anthropogenic emissions, and the variations in winds were found to have the largest impact on the interannual variation of surface-layer O$_3$ among all meteorological parameters, followed by temperature and specific humidity. On the basis of these studies, the interannual variations in surface O$_3$ concentrations driven by meteorological parameters alone are in the range of 0.5–5.0 ppbv in China.

3.2.2. *Impacts of the East Asian monsoon on O$_3$ concentrations*

The East Asian monsoon, which is the most important pattern of general circulation that drives meteorology over East Asia, has been demonstrated to influence the seasonal and interannual variation of O$_3$ in China (Tang et al., 2013; Zhou et al., 2013; Yang et al., 2014a; Li et al., 2018a). Tang et al. (2013) explored five years (2007–11) of surface O$_3$ measurements at an agricultural site in the northern YRD region and found a bimodal pattern in the seasonal variation of O$_3$ with peaks in June and October, which was mostly due to variations in marine air mass incursions driven by the Asian summer monsoon and open crop residue burning across the regions. By analyzing 11 years of ozonesonde data in Hong Kong with the aid of Lagrangian dispersion modeling, Zhou et al. (2013) indicated that the interannual variability in the boundary layer O$_3$ was mainly associated with the intensity of the East Asian monsoon, with high O$_3$ mixing ratios being linked to northeasterly circulation anomalies. Yang et al. (2014a) quantified the impact of the EASM on summertime surface O$_3$ concentrations over China for 1986–2006 and reported positive correlation between summertime O$_3$ concentrations and the EASM strength. The largest difference in O$_3$ concentration between the strongest and weakest EASM years exceeded 6% in northeastern and southwestern China, which was mostly attributed to the differences in the transboundary transport of O$_3$ between strongest and weakest EASM years. Li et al. (2018a) analyzed the impact of EASM intensity on O$_3$ concentrations using the RegCM4-Chem model and illustrated that the chemistry was the uppermost process controlling summertime O$_3$ over eastern China during monsoon seasons, through changing winds in the lower troposphere, cloud cover, and downward shortwave radiation.

3.2.3. *Impacts of climate change on future O$_3$ concentrations*

By coupling a general circulation model (GCM) and CTMs, some studies have examined the future changes in surface O$_3$ over China under different climate scenarios and the potential drivers for the changes. These model results have generally showed that a warmer climate would lead to increased O$_3$ levels overpopulated areas as a result of enhanced biogenic hydrocarbon emissions and accelerated decomposition of peroxyacetyl nitrate (PAN) to form NO$_x$ at higher temperature (Jacob and Winner, 2009; Doherty et al., 2013). However, tropospheric O$_3$ levels in remote regions were simulated to decrease at higher temperature, because increased water vapor accompanied by the higher temperature would increase O$_3$ destruction and therefore cause a shorter O$_3$ lifetime (Fiore et al., 2012). Through sensitivity simulations, Lee et al. (2015) compared the contributions of climate change and emission changes to the changes in summertime MDA8 O$_3$ concentrations over 2000–50 under the A2 scenario and found that the regional climate change would lead to a decrease in O$_3$ by 2.2 ppbv in China, owing to an increase in precipitation and cloud fraction in the 2050s relative to the year 2000. Liu et al. (2013b) reported that surface O$_3$ concentrations in the afternoon in autumn could be changed by ±5 ppbv in southern China over 2005–55 owing to the changes in both meteorology and biogenic emissions under the IPCC A1B scenario. Wang et al. (2013b) also reported that sum-
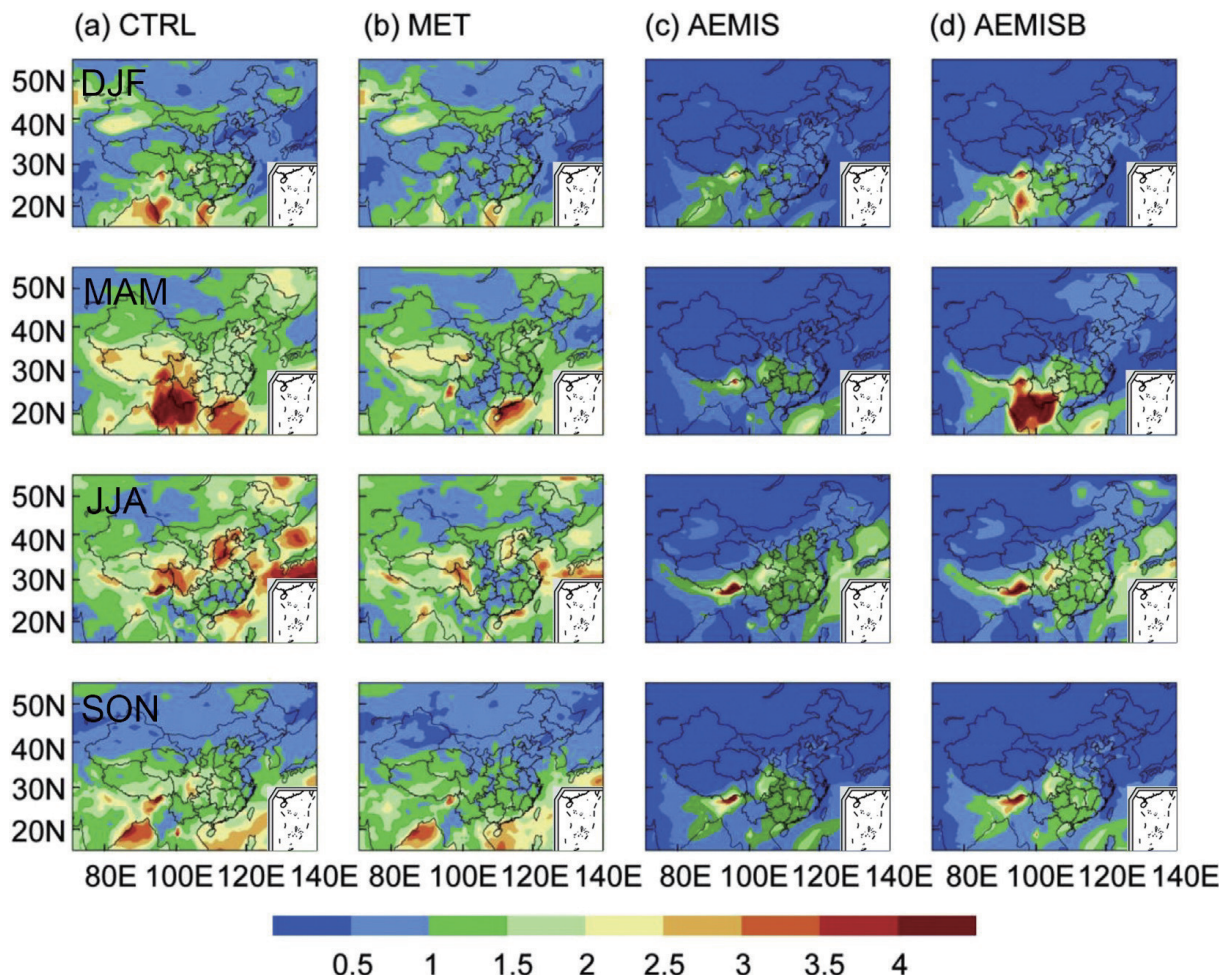
Fig. 3. Mean absolute deviation (units: ppbv) of seasonal mean surface O_3 concentrations obtained from simulations of (a) CTRL, (b) MET, (c) AEMIS, and (d) AEMISB for the period 2014–12 over China. CTRL represents the interannual variation (IAVs) of O_3 driven by both meteorological fields and emissions (including anthropogenic, natural, and biomass emissions). MET represents the IAVs of O_3 driven by meteorological fields alone. AEMIS represents the IAVs of O_3 driven by anthropogenic emissions alone. AEMISB represents the IAVs of O_3 driven by biomass burning emissions change alone. [Reprinted from Lou et al. (2015)].

mertime O_3 would increase largely over eastern China, with local increases of up to 10 ppbv due to the climate change alone over 2000–50 under the A1B scenario, representing a significant "climate change penalty". In contrast, the surface O_3 over western China was simulated to decrease with the 2050 climate, with the largest reduction of 5 ppbv, implying a "climate change benefit". The aforementioned studies have indicated that changes in BVOCs under future climate tend to modify O_3 concentrations, but, currently, the effect of BVOCs on O_3 formation under the future climate scenarios remains uncertain. Some studies have found that atmospheric CO_2, O_3 and soil moisture affect BVOCs emissions (Peñuelas and Staudt, 2010), although the biochemical mechanisms have not been fully understood (Guenther et al., 2012). For example, Tai et al. (2013) predicted that climate change over 2000–50 could enhance summertime O_3 by up to 6 ppbv in eastern China, while upon an inclusion of the inhibition effect of CO_2 on isoprene, the projected O_3 changes

due to climate change were substantially modified.

In summary, studies so far have shown that O_3 concentrations are sensitive to meteorological conditions on synoptic, interannual, and decadal time scales, which have important implications for O_3 air quality planning. Scientifically, more studies are needed for systematic understanding of the meteorological conditions for the occurrence of severe O_3 pollution (duration, intensity etc.) at regional scales. For the development of O_3 air quality control strategies for different time scales, if meteorological conditions in the regions are not favorable for O_3 air quality, extra efforts are needed to reduce emissions.

3.3. *Impacts of land use and land cover changes on O_3 concentrations*

Land use and land cover (LULC) change often occurs in response to both human influences (e.g., urbanization, agriculture development, deforestation or afforestation practices,

and agricultural waste burning) and climate influences (e.g., CO_2 fertilization, climate-driven biome shifts and wildfires). LULC change has impacts on climate by perturbing surface albedo, the water cycle, and concentrations of atmospheric chemical components. For instance, the terrestrial biosphere is a significant emission source of isoprene, a dominant biogenic hydrocarbon species with a global annual emission comparable to anthropogenic emissions. The emission of isoprene depends on the type and density of vegetation and other environmental conditions (Guenther et al., 2012). In polluted regions with high NO_x concentrations, the oxidation of isoprene contributes to O_3 production. In remote regions with low NO_x, isoprene may reduce O_3 by ozonolysis or by sequestering NO_x as isoprene nitrate. In addition, dry deposition onto the surface, and specifically the uptake by vegetation, represents a dominant sink within the boundary layer for trace gases and particles.

A growing number of high-resolution modeling studies have investigated the impacts of regional-scale LULC change on O_3 using satellite-derived and land survey data. By using the WRF-Chem model and dynamical vegetation datasets derived from MODIS and AVHRR measurements, Li et al. (2015) found that a twofold increase in BVOC emissions by LULC in the PRD from the early 1990s to 2006 increased surface O_3 concentrations in the downwind region by 0.9–4.6 ppbv, whereas the large increase in BVOC emissions during 2006–12 tended to reduce the peak surface O_3 concentrations by 1.6–2.5 ppbv in rural regions. Liao et al. (2015) simulated, by using the WRF-Chem model, that urbanization between 1992 and 2004 caused an increase of O_3 by up to 6.8 ppbv (2.3 ppbv) during daytime in July (January) in the YRD region. Li et al. (2016b) reported that urbanization-induced changes in atmospheric circulation could lead to decreases in daytime O_3 concentration by about 1.3 ppbv. The recent work by Tao et al. (2018) reported that LULC change over the BTH region could lead to an increase in summertime O_3 of 0.4–4.3 ppbv during daytime in Beijing but a decrease in O_3 by up to 3.4 ppbv at noon in Tianjin.

Historical and future changes in LULC have also been shown to have important ramifications for tropospheric O_3 in China. Fu and Liao (2014) found that the seasonal mean surface O_3 concentrations changed by −4 to +6 ppbv in China between the late 1980s and mid-2000s owing to changes in both climate and land cover. Further, Fu and Tai (2015) showed that LULC change alone between the periods of 1981–85 and 2007–11 led to decreases in summertime O_3 by up to 4 ppbv in China, and the sensitivity of surface O_3 to land cover change is more dependent on dry deposition than on isoprene emission in most of the study area (Fig. 4).

3.4. *Impacts of aerosols on O_3 concentrations*

The presence of aerosol in the atmosphere has a large influence on photochemical reactions. The formation and growth of secondary aerosols, i.e., sulfate and nitrate aerosol, are related to the abundance of the precursor gases (SO_2, NO_x) and atmospheric oxidants such as OH radical, hydrogen peroxide (H_2O_2), and O_3 (Seinfeld and Pandis, 2006; Unger

et al., 2006). Thereby, the changes in concentrations of OH, H_2O_2, and NO_x resulting from the formation of aerosols are expected to influence concentrations of tropospheric O_3. Additionally, the heterogeneous reactions of chemicals such as dinitrogen pentoxide (N_2O_5), NO_3, NO_2, and HO_2 on aerosol surfaces (sulfate, nitrate, ammonium, organic carbon, sea salt, and mineral dust) have considerable impacts on the O_3 concentration (Liao and Seinfeld, 2005). Moreover, aerosol can influence the climate (temperature, atmospheric ventilation, cloud formation precipitation etc.) by direct, indirect and semi-direct radiative effects, which can further influence O_3 concentration.

Some existing modeling studies have endeavored to identify the influence of aerosols on O_3 concentration in China by heterogeneous reactions and by altering photolysis rates (Li et al., 2011; Xu et al., 2012; Lou et al., 2014). Li et al. (2011) found that the summertime O_3 in the boundary layer was reduced by 5% in highly polluted regions over central and eastern China, driven by the 53% reduction in daytime average $O_3 \rightarrow O\,(^1D)$ photolysis frequency in the lower troposphere. Xu et al. (2012) revealed that the absorption of NO_2 by aerosols could increase O_3 at noon by 4–20 ppbv in VOC-limited urban areas but decrease O_3 by 4–10 ppbv in NO_x-limited suburban areas during a high O_3 episode that occurred in Beijing during June 26–27 2000, suggesting that the impact of heterogeneous reactions on O_3 depends on the local VOC/NO_x ratio. They also showed that the reduced photolysis rate by aerosol radiative effects led to a large decrease in O_3 concentration of up to 73 ppbv in Beijing. Lou et al. (2014) reported that the impact of aerosols on O_3 by heterogeneous reactions was simulated to be much larger than that of aerosols on O_3 by altering the photolysis rate in China. Feng et al. (2016) investigated a heavy air pollution episode with high concentrations of O_3 and $PM_{2.5}$, and found that high aerosol concentrations in Xi'an and the surrounding areas significantly reduced the photolysis frequencies and hence reduced O_3 concentrations by about 25 ppb on average. A recent study by Qu et al. (2018) suggested that the relative roles of heterogeneous reactions and the changes in photolysis rates by aerosols depend on the concentration of aerosols. In their simulations for Nanjing, heterogeneous reactions dominated O_3 reduction when aerosol levels were low ($PM_{2.5} < 50$ μg m^{-3}), whereas changes in photolysis by aerosols led to a greater O_3 reduction when aerosol concentrations were high ($PM_{2.5} > 50$ μg m^{-3}).

Few studies have quantified the impacts of aerosol formation on O_3 concentrations in China. For example, Yang et al. (2014b) showed that the chemical reactions associated with sulfate formation were simulated to increase O_3 concentrations over the NCP and Sichuan Basin of China, with maximum increases of ~2.0 ppbv in the NCP and ~3.0 ppbv in the Sichuan Basin in spring. The effect of nitrate formation on O_3 concentration is opposite, leading to a decrease in O_3 concentration by up to 1.0 ppbv in summer in eastern China.

Changes in meteorology induced by radiative effects of aerosols can also influence O_3 concentrations. By using a coupled meteorology–air quality model, Xing et al. (2015)
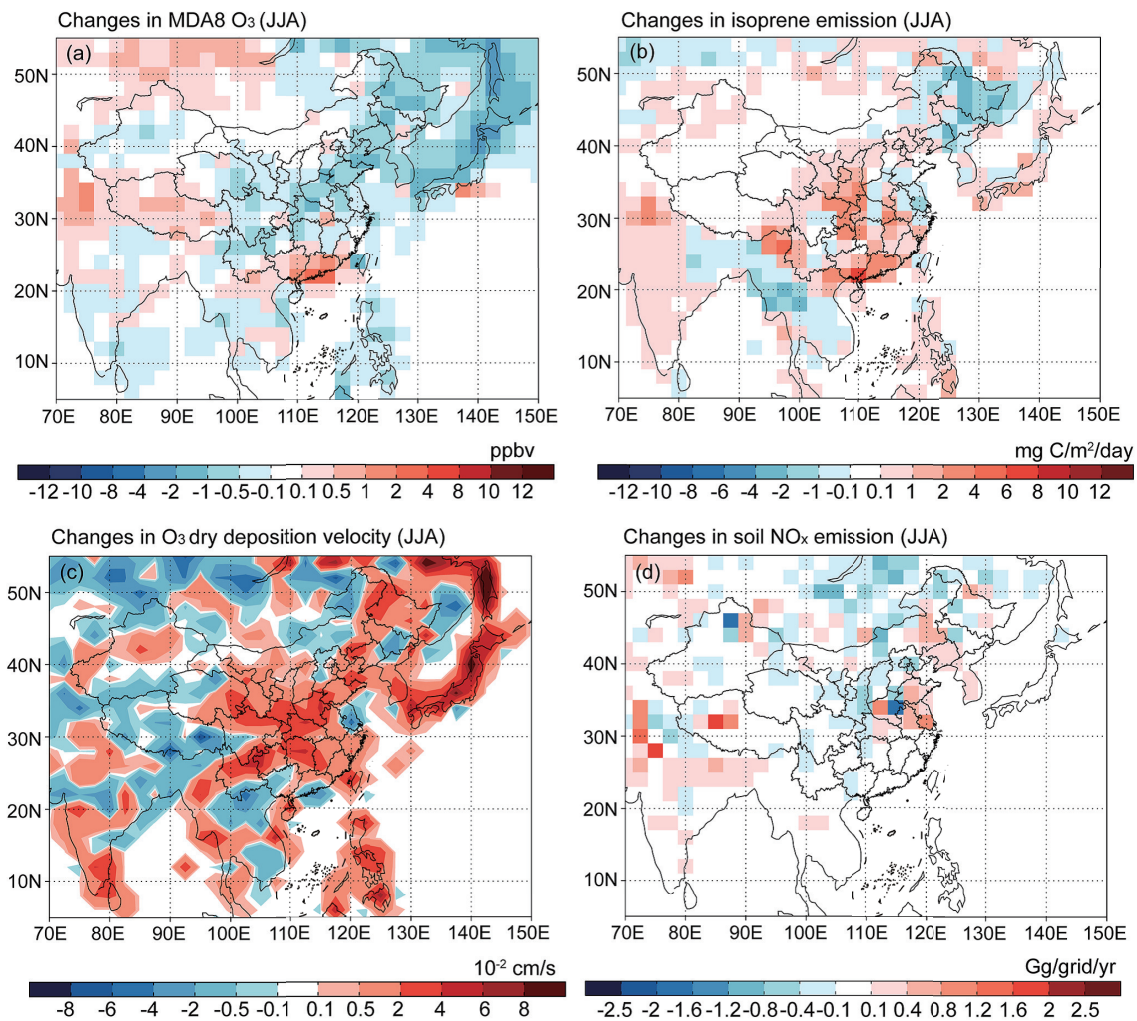
Fig. 4. Changes in summertime (a) surface maximum daily 8-h average O_3 concentration (MDA8 O_3), (b) isoprene emissions, (c) O_3 dry deposition velocity, and (d) soil NO_x emissions, driven by changes in land cover and land use alone over 1980–2010. [Reprinted from Fu and Tai (2015)].

found that the inclusion of the aerosol direct radiative effect led to an increase in surface O_3 by 2%–3% in eastern China, as averaged over 1990–2010, which mainly resulted from the aerosol-induced lower planetary boundary layer height and the increased precursor concentrations. Wang et al. (2016a) simulated the direct effect of aerosols on O_3 air quality in March 2005, and found that the surface daily maximum 1-h O_3(MD1O_3) was reduced by up to 12% in eastern China as a result of less radiation reaching the surface. Xing et al. (2017) found that the aerosol direct effect reduced the surface MD1O_3 in eastern China by up to 11 ppbv in January but increased it in most areas of China by up to 2 ppbv in July due to the aerosol-induced changes in atmospheric stability, suggesting that the reductions in aerosols will have the potential risk of increasing O_3 in winter but the benefit of reducing the maximum O_3 in summer.

The studies cited above underscore the considerable effects of aerosols on O_3 concentrations in China. However, the mechanism of the heterogeneous reactions and some co-

efficients in related processes remain uncertain. Subsequent experiments, including in-situ measurements and model simulations, should be carried out in future research.

4. Influence of tropospheric O_3 on climate

Tropospheric O_3 is distinguished from other well-mixed greenhouse gases (i.e., CO_2 and CH_4) by having significant spatial and temporal heterogeneity due to its relatively short lifetime of a few days to several weeks. Changes in O_3 concentrations perturb the radiative energy budget of the earth–atmosphere system, which may further alter the climate. Furthermore, O_3 can affect climate indirectly via modification of the surface fluxes of energy and water as well as the land carbon sink. Many field experiments have demonstrated the damaging effect of surface O_3 on vegetation productivity. The phytotoxicity of tropospheric O_3 has been shown to reduce the ability of plants to absorb CO_2, then to decrease photosynthesis, plant growth and biomass accumulation by

entering leaves through the stomata, generating other reactive oxygen species and causing oxidative stress (Ainsworth et al., 2012; Feng et al., 2015). In this review, we describe recent estimates of radiative forcing (RF) by tropospheric O_3, as well as the RF and regional climate resulted from the changes in tropospheric O_3 over China.

4.1. *RF of tropospheric O_3*

Due to the significant increases in anthropogenic emissions since pre-industrial times, the increases in tropospheric O_3 have amplified its greenhouse effect, and thus contributes to the RF. Relying on a limited number of studies for the period of 1998–2011 with regard to the RF of short-lived components (Skeie et al., 2011; Søvde et al., 2011; Shindell et al., 2013), the IPCC Fifth Assessment Report (AR5) suggested that the global mean RF due to tropospheric O_3 was 0.4 (0.2–0.6) W m^{-2} over 1750–2011 (Myhre et al., 2013). The recent estimate by Myhre et al. (2017) showed that the global mean RF due to the changes in tropospheric O_3 and aerosol over 1990–2015 was 0.17 ± 0.08 W m^{-2}, in which approximately one-third was due to tropospheric O_3 (0.06 W m^{-2}), and this value was larger than that reported in IPCC AR5 for the same time period. Understanding the contribution that individual countries have made to global RF is of particular importance for implementation of the agreement on "common but differentiated responsibilities" reached by the United Nations Framework Convention on Climate Change. However, few studies have examined the RF induced by tropospheric O_3 over recent decades in China. Table 3 summarizes the simulated annual mean tropospheric O_3 RF over China. Early estimates of annual mean RF of tropospheric O_3 over the domain of China were in the range of 0.45–0.87 W m^{-2} for the present day (year 2000) relative to pre-industrial times (Berntsen et al., 2000; Wang et al., 2005; Chang et al., 2009; Chang and Liao, 2009), indicating the large positive contribution of tropospheric O_3 over China to global RF. Li et al. (2016a) quantified the relative and absolute contributions of historical anthropogenic emissions of well-mixed greenhouse gases, O_3 precursors and aerosols in China to global RF over 1750–2010 and found that China contributed approximately $12\% \pm 2\%$ (0.48 ± 0.09 W m^{-2}) of the global positive RF from well-mixed greenhouse gases, tropospheric O_3, and black carbon (BC). The RF induced by China through emissions of precursors of O_3 was as much as +0.2 W m^{-2} over eastern China. Zhu and Liao (2016) estimated an averaged RF of tropospheric O_3 of +0.58 W m^{-2} over eastern China for the period of 1850–2000, with high RF values of 0.6–0.8 W m^{-2} found over central and eastern China (i.e., the Sichuan Basin and the provinces of Hunan, Hubei and Jiangxi) and low RF values of 0.2–0.4 W m^{-2} over the Tibetan Plateau.

Zhu and Liao (2016) projected future changes (2000–50) in the annual mean RF of tropospheric O_3 under the four RCP scenarios (RCP2.6, RCP4.5, RCP6.0 and RCP8.5) and reported that the four RCP scenarios provide a wide range of possible future changes in O_3 RF, from −0.11 W m^{-2} under RCP2.6 to +0.14 W m^{-2} under RCP8.5, in 2050 relative to 2000, which offer options for mitigating warming over eastern China from 2000 to 2050 (Fig. 5).

4.2. *Climate effects of tropospheric O_3*

The climate effects of tropospheric O_3 have drawn widespread attention, but only a small number of studies have examined the impacts of tropospheric O_3 on climate in China. Xie et al. (2016), by using an aerosol–climate coupled model, BCC_AGCM2.0.1_CUACE/Aero, found that the changes in tropospheric O_3 over 1850–2013 increased the surface temperature by 0.2°C–0.8°C, whereas they reduced precipitation in most of China. Zhang et al. (2018) reported that the changes in short-lived climatic pollutants (SLCPs), including CH_4, tropospheric O_3 and BC, under the three RCP scenarios, would lead to different climate responses for the period 2010–50, especially in regional climate. For example, by 2050, changes in the SLCPs would increase surface temperature in north China under RCP8.5 but reduce surface temperature by up to 0.6 K (0.8 K) under RCP4.5 (RCP2.6) in the same region. However, they examined only the climate responses to changes in all of CH_4, tropospheric O_3 and BC, and did not distinguish the role of tropospheric O_3. Recently, Li et al. (2018b) examined the radiative effect of tropospheric O_3 on monsoon circulation and the possible feedback mechanism by using the regional climate model RegCM4, and reported that changes in tropospheric O_3 led to an increase in summertime temperature by 0.06 K and an increase in precipitation by 0.22 mm d^{-1} over eastern China.

5.   Summary and recommendations

This review is focused on the recent advances in existing literature on tropospheric O_3 in China from 2011 to 2018, including the observed O_3 abundance and its variations in the troposphere, the factors that determine O_3 formation and distribution, and the influences of tropospheric O_3 on regional

---

Table 3. Summary of the simulated annual mean tropospheric O_3 radiative forcing over China.

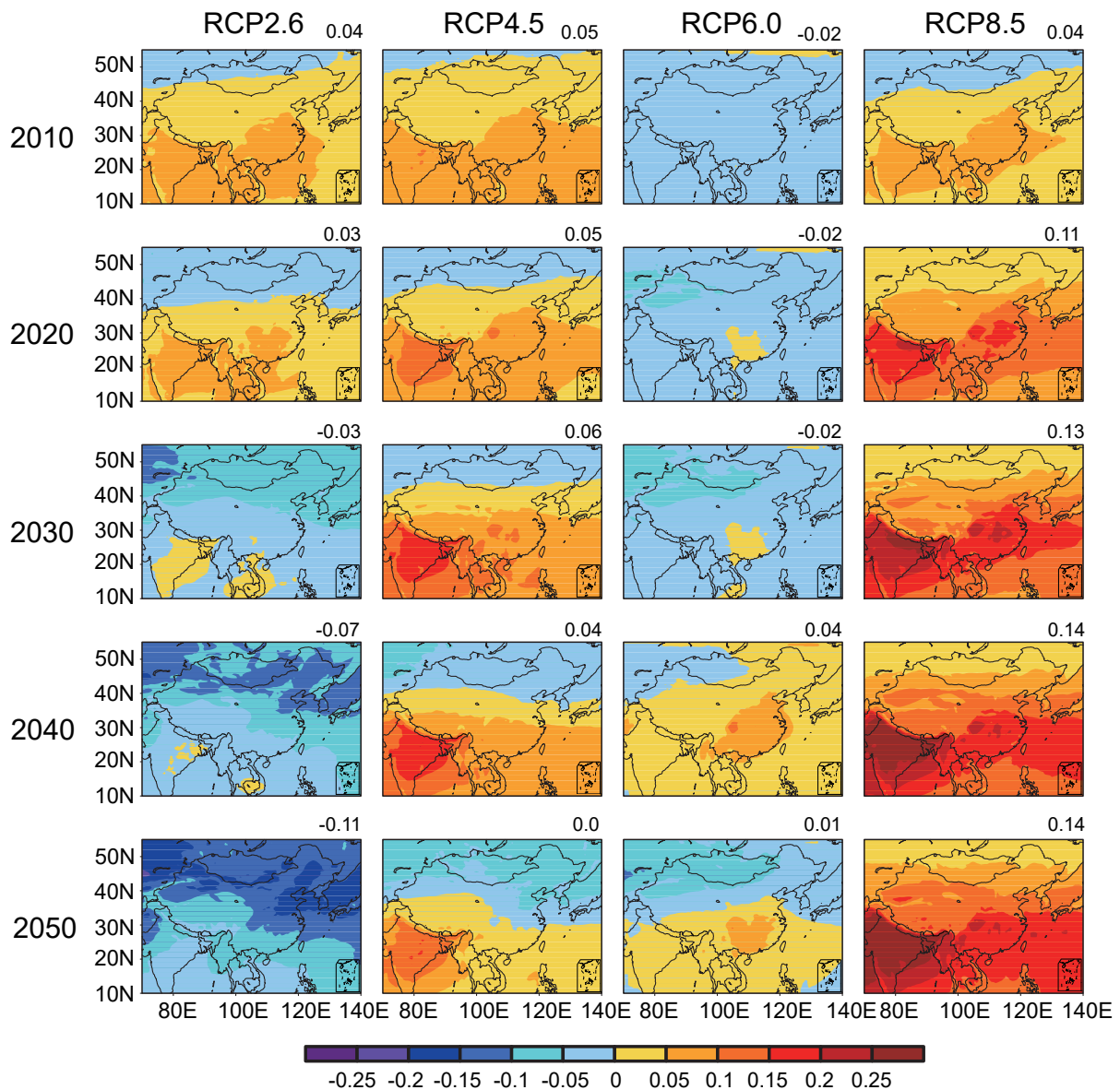| Region	Model	Time period	Radiative forcing (W m^{-2})	Reference
China and adjacent area (6°–52°N, 85°–130°E)	RegCM2	1860–2000	0.65	Wang et al. (2005)
Eastern China (20°–50°N, 100°–130°E)	GISS GCM II	1950–2000	0.87	Chang and Liao (2009)
Eastern China (18°–45°N, 95°–125°E)	GISS GCM II	1850–2000	0.53	Chang and Liao (2009)
China	GEOS-Chem	1850–2000	0.48	Zhu and Liao (2016)
China	RegCM4	1850–2010	0.68	Li et al. (2018b)

Fig. 5. Projected future changes in annual mean tropospheric O_3 radiative forcing (units: W m^{-2}) at the tropopause over 2010–50 for every decade (relative to the year 2000) under the RCP scenarios. The annual mean O_3 radiative forcing averaged over eastern China (18°–45°N, 95°–125°E) is shown in the top-right corner of each panel. [Reprinted from Zhu and Liao (2016)].

climate in China. We offer the following conclusions and recommendations:

(1) The available observational studies have shown an increasing trend of tropospheric O_3 in China and more frequent extreme pollution episodes. In the past five years, severe O_3 pollution events were detected in city clusters of China (e.g. the North China Plain, the Yangtze River Delta, and the Pearl River Delta). Previous studies have shown that the O_3 levels increased at a rate of 1.0–3.0 ppbv yr^{-1} at several sites in eastern China over the past two decades, which was higher than the overall rate of increase (0.45 ppbv yr^{-1}) in other places of East Asia (Japan and Korea). However, knowledge of the interannual variation and decadal trend of O_3 concentration on a nationwide scale is still limited. With the development of the National Air Quality Monitoring Network and the regular

monitoring of surface O_3 around China, more studies should be focused on the spatial and temporal variations in the whole of China to better understand O_3 air quality in China.

(2) Measurements and modeling studies have highlighted the important role of anthropogenic emissions in the short-term and long-term variations of surface O_3 concentrations in China. However, estimates of the changes in O_3 concentrations caused by the changes in anthropogenic emissions in China are still subject to large uncertainties. The major uncertainty arises from anthropogenic emissions inventories. In this regard, it is strongly suggested that consistent and up-to-date emissions inventories should be made available for improving model performance and for assessing the effectiveness of emission regulations on current and short-term trends of O_3 air quality. Additionally, in terms of regional emission

controls, future work should be focused on a more systematic analysis of the relationships between O$_3$ and its precursors, especially the spatial variability of O$_3$ sensitivity to changes in O$_3$ precursors, which may help to determine the regional-scale O$_3$ chemical formation regimes in China.

(3) Modeling studies in China have suggested that, on the interannual time scale, the variations in meteorological conditions may have larger influences on surface O$_3$ concentrations than the changes in anthropogenic emissions. The East Asian monsoon, as a critical climate pattern in Asia, has been found to have large influences on the seasonal and interannual variability of O$_3$ in China. Therefore, if meteorological conditions are not favorable for O$_3$ air quality, short-term air quality control may need extra effort to reduce emissions. A systematic understanding of the relationships among large-scale circulation, synoptic-scale circulation patterns, and tropospheric O$_3$ concentrations in China should be addressed in future studies. Likewise, on the decadal scale, climate change may at least partly offset or aggravate the effect of changes in emissions regionally, representing a "climate change benefit" or "climate change penalty" on O$_3$ air quality. Therefore, the effects of climate change should receive particular attention to achieve given air quality goals. Some studies have suggested that the ambient CO$_2$ levels could affect biogenic isoprene emissions and may partly offset the effects of climate warming, which is a source of uncertainty for O$_3$ air quality in the coming century. In this regard, it is recommended that more research be conducted on the effects of CO$_2$-isoprene inhibition, as well as soil moisture, on isoprene emissions for a comprehensive assessment of the impact of climate on O$_3$ air quality.

(4) The variation characteristics of severe O$_3$ pollution, and the possible factors affecting the occurrence of high O$_3$ episodes are discussed. However, most studies were focused on analyzing a small number of high O$_3$ pollution days or O$_3$ air quality in a single city or on a local scale. There is a lack of systematic understanding of the characteristics (e.g., duration and intensity) of severe O$_3$ pollution in China. The mechanisms of medium-term and long-term variations in severe O$_3$ pollution episodes are still unclear. Further studies are needed to examine how atmospheric circulation and weather types affect the formation and development of high O$_3$ events.

(5) LULC driven by climate change and human activities may substantially impact O$_3$ air quality in China, although this effect is less considerable than the effects of changes in climate and anthropogenic emissions. Future studies should be focused on a more systematic analysis of the spatial variability of O$_3$ sensitivity to vegetation changes in China. Anthropogenic land use changes, such as urbanization, crop expansion, biofuel development, afforestation, and some agricultural practices, may further complicate the effects of land cover changes, and the implications for O$_3$ need to be considered in future studies, which may benefit both long-term land use policies and air quality regulations.

(6) Research on the effects of aerosols on O$_3$ concentrations in China is limited, and the mechanism of the heterogeneous reactions, the combined effect of particles on both photolysis rates and heterogeneous reactions, and some coefficients in related processes have not been thoroughly elucidated to date. Subsequent experiments, including in-situ measurements and model simulations, should be addressed in future research. In particular, in recent years, the Chinese government has implemented stringent control actions for reducing aerosol concentrations. Whether such extensive aerosol controls will aggravate O$_3$ pollution remains an unclear issue, and the impact of aerosols on O$_3$ pollution needs further quantification and assessment for air pollution cleaning strategies. Such studies will benefit both scientific cognition and policy-making for fighting against the complex air pollution in China.

(7) Large changes in regional emissions of O$_3$ precursors, aerosol precursors, and aerosols have occurred over China, and the net effect of these emission changes on the Earth's radiative balance is less clear. Therefore, an improved understanding of the impacts of O$_3$ on climate over China is very necessary to evaluate the climate impacts of recent and future air quality regulations and the potential to mitigate near-term warming by selective emission reductions of SLCPs.

REFERENCES

Ainsworth, E. A., C. R. Yendrek, S. Sitch, W. J. Collins, and L. D. Emberson, 2012: The effects of tropospheric ozone on net primary productivity and implications for climate change. *Annual Review of Plant Biology*, **63**, 637–661, https://doi.org/10.1146/annurev-arplant-042110-103829.

An, J. L., J. N. Zou, J. X. Wang, X. Lin, and B. Zhu, 2015: Differences in ozone photochemical characteristics between the megacity Nanjing and its suburban surroundings, Yangtze River Delta, China. *Environmental Science and Pollution Research*, **22**, 19 607–19 617, https://doi.org/10.1007/s11356-015-5177-0.

Berntsen, T. K., G. Myhre, F. Stordal, and I. S. A. Isaksen, 2000: Time evolution of tropospheric ozone and its radiative forcing. *J. Geophys. Res.: Atmos.*, **105**, 8915–8930, https://doi.org/10.1029/1999JD901139.

Chang, K.-L., I. Petropavlovskikh, O. R. Cooper, M. G. Schultz, and T. Wang, 2017: Regional trend analysis of surface ozone observations from monitoring networks in eastern North America, Europe and East Asia. *Elementa Science of the Anthropocene*, **5**, 50, http://doi.org/10.1525/elementa.243.

Chang, W. Y., and H. Liao, 2009: Anthropogenic direct radiative forcing of tropospheric ozone and aerosols from 1850 to 2000 estimated with IPCC AR5 emissions inventories. *Atmospheric and Oceanic Science Letters*, **2**, 201–207, https://doi.org/10.1080/16742834.2009.11446804.

Chang, W. Y., H. Liao, and H. J. Wang, 2009: Climate responses to direct radiative forcing of anthropogenic aerosols, tropospheric ozone, and long-lived greenhouse gases in eastern China over 1951-2000. *Adv. Atmos. Sci.*, **26**, 748–762, https://

doi.org/10.1007/s00376-009-9032-4.

Chen, W., L. Yan, and H. M. Zhao, 2015: Seasonal variations of atmospheric pollution and air quality in Beijing. *Atmosphere*, **6**, 1753–1770, https://doi.org/10.3390/atmos6111753.

Cooper, O. R., and Coauthors, 2014: Global distribution and trends of tropospheric ozone: An observation-based review. *Elementa Science of the Anthropocene*, **2**, 000029, http://doi.org/10.12952/journal.elementa.000029.

Ding, A. J., T. Wang, V. Thouret, J. P. Cammas, and P. Nédélec, 2008: Tropospheric ozone climatology over Beijing: Analysis of aircraft data from the MOZAIC program. *Atmospheric Chemistry and Physics*, **8**, 1–13, https://doi.org/10.5194/acp-8-1-2008.

Ding, A. J., and Coauthors, 2013: Ozone and fine particle in the western Yangtze River Delta: An overview of 1 yr data at the SORPES station. *Atmospheric Chemistry and Physics*, **13**, 5813–5830, https://doi.org/10.5194/acp-13-5813-2013.

Doherty, R. M., and Coauthors, 2013: Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study. *J. Geophys. Res.: Atmos.*, **118**, 3744–3763, https://doi.org/10.1002/jgrd.50266.

Dufour, G., and Coauthors, 2018: Lower tropospheric ozone over the North China Plain: Variability and trends revealed by IASI satellite observations for 2008-2016. *Atmospheric Chemistry and Physics*, **18**, 16 439–16 459, https://doi.org/10.5194/acp-18-16439-2018.

Feng, T., and Coauthors, 2016: Summertime ozone formation in Xi'an and surrounding areas, China. *Atmospheric Chemistry and Physics*, **16**, 4323–4342, https://doi.org/10.5194/acp-16-4323-2016.

Feng, Z. Z., E. Z. Hu, X. K. Wang, L. J. Jiang, and X. J. Liu, 2015: Ground-level O$_3$ pollution and its impacts on food crops in China: A review. *Environmental Pollution*, **199**, 42–48, https://doi.org/10.1016/j.envpol.2015.01.016.

Fiore, A. M., and Coauthors, 2012: Global air quality and climate. *Chemical Society Reviews*, **41**, 6663–6683, https://doi.org/10.1039/c2cs35095e.

Fiore, A. M., V. Naik, and E. M. Leibensperger, 2015: Air quality and climate connections. *Journal of the Air & Waste Management Association*, **65**, 645–685, https://doi.org/10.1080/10962247.2015.1040526.

Fu, Y., and H. Liao, 2012: Simulation of the interannual variations of biogenic emissions of volatile organic compounds in China: Impacts on tropospheric ozone and secondary organic aerosol. *Atmos. Environ.*, **59**, 170–185, https://doi.org/10.1016/j.atmosenv.2012.05.053.

Fu, Y., and H. Liao, 2014: Impacts of land use and land cover changes on biogenic emissions of volatile organic compounds in China from the late 1980s to the mid-2000s: implications for tropospheric ozone and secondary organic aerosol. *Tellus B: Chemical and Physical Meteorology*, **66**, 24987.

Fu, Y., and A. P. K. Tai, 2015: Impact of climate and land cover changes on tropospheric ozone air quality and public health in East Asia between 1980 and 2010. *Atmospheric Chemistry and Physics*, **15**, 10 093–10 106, https://doi.org/10.5194/acp-15-10093-2015.

Gaudel, A., and Coauthors, 2018: Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation. *Elementa Science of the Anthropocene*, **6**, 39, http://doi.org/10.1525/elementa.291.

Geng, F., X. Tie, A. Guenther, G. Li, J. Cao, and P. Harley, 2011:

Effect of isoprene emissions from major forests on ozone formation in the city of Shanghai, China. *Atmospheric Chemistry and Physics*, **11**, 10 449-10 459.

Guenther, A. B., X. Jiang, C. L. Heald, T. Sakulyanontvittaya, T. Duhl, L. K. Emmons, and X. Wang, 2012: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): An extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development*, **5**, 1471–1492, https://doi.org/10.5194/gmd-5-1471-2012.

Hayashida, S., X. Liu, A. Ono, K. Yang, and K. Chance, 2015: Observation of ozone enhancement in the lower troposphere over East Asia from a space-borne ultraviolet spectrometer. *Atmospheric Chemistry and Physics*, **15**, 9865–9881, https://doi.org/10.5194/acp-15-9865-2015.

Hong, C. P., Q. Zhang, K. B. He, D. B. Guan, M. Li, F. Liu, and B. Zheng, 2017: Variations of China's emission estimates: Response to uncertainties in energy statistics. *Atmospheric Chemistry and Physics*, **17**, 1227–1239, https://doi.org/10.5194/acp-17-1227-2017.

Hu, J. L., J. J. Chen, Q. Ying, and H. L. Zhang, 2016: One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system. *Atmospheric Chemistry and Physics*, **16**, 10 333–10 350, https://doi.org/10.5194/acp-16-10333-2016.

Hu, J., and Coauthors, 2018: An important mechanism of regional O$_3$ transport for summer smog over the Yangtze River Delta in Eastern China. *Atmospheric Chemistry and Physics*, **18**, 16 239–16 251, https://doi.org/10.5194/acp-18-16239-2018.

Huang, J., and Coauthors, 2015: Origin of springtime ozone enhancements in the lower troposphere over Beijing: In situ measurements and model analysis. *Atmospheric Chemistry and Physics*, **15**, 5161–5179, https://doi.org/10.5194/acp-15-5161-2015.

Jacob, D. J., 2000: Heterogeneous chemistry and tropospheric ozone. *Atmos. Environ.*, **34**, 2131–2159, https://doi.org/10.1016/S1352-2310(99)00462-8.

Jacob, D. J., and D. A. Winner, 2009: Effect of climate change on air quality. *Atmos. Environ.*, **43**, 51–63, https://doi.org/10.1016/j.atmosenv.2008.09.051.

Jeong, J. I., and R. J. Park, 2013: Effects of the meteorological variability on regional air quality in East Asia. *Atmos. Environ.*, **69**, 46–55, https://doi.org/10.1016/j.atmosenv.2012.11.061.

Kurokawa, J., T. Ohara, T. Morikawa, S. Hanayama, G. Janssens-Maenhout, T. Fukui, K. Kawashima, and H. Akimoto, 2013: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2. *Atmospheric Chemistry and Physics*, **13**, 11 019–11 058, https://doi.org/10.5194/acp-13-11019-2013.

Lee, J. B., and Coauthors, 2015: Projections of summertime ozone concentration over East Asia under multiple IPCC SRES emission scenarios. *Atmos. Environ.*, **106**, 335–346, https://doi.org/10.1016/j.atmosenv.2015.02.019.

Li, B. G., and Coauthors, 2016a: The contribution of China's emissions to global climate forcing. *Nature*, **531**, 357–361, https://doi.org/10.1038/nature17165.

Li, D., and J. C. Bian, 2015: Observation of a summer tropopause fold by ozonesonde at Changchun, China: Comparison with reanalysis and model simulation. *Adv. Atmos. Sci.*, **32**, 1354–1364, https://doi.org/10.1007/s00376-015-5022-x.

Li, G. H., and Coauthors, 2017a: Widespread and persistent

ozone pollution in eastern China during the non-winter season of 2015: Observations and source attributions. *Atmospheric Chemistry and Physics*, **17**, 2759–2774, https://doi.org/10.5194/acp-17-2759-2017.

Li, J., and Coauthors, 2011: Impacts of aerosols on summertime tropospheric photolysis frequencies and photochemistry over Central Eastern China. *Atmos. Environ.*, **45**, 1817–1829, https://doi.org/10.1016/j.atmosenv.2011.01.016.

Li, J. F., and Coauthors, 2014: Fast increasing of surface ozone concentrations in Pearl River Delta characterized by a regional air quality monitoring network during 2006-2011. *Journal of Environmental Sciences*, **26**, 23–36, https://doi.org/10.1016/S1001-0742(13)60377-0.

Li, K., D. J. Jacob, H. Liao, L. Shen, Q. Zhang, and K. H. Bates, 2019: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China. *Proceedings of the National Academy of Sciences of the United States of America*, **116**, 422–427, https://doi.org/10.1073/pnas.1812168116.

Li, M. M., Y. Song, M. X. Liu, H. Yao, X. Huang, X. S. Wang, and Y. H. Zhang, 2015: Impacts of decadal variations in natural emissions due to land-cover changes on ozone production in southern China. *Tellus B: Chemical and Physical Meteorology*, **67**, 27676, https://doi.org/10.3402/tellusb.v67.27676.

Li, M. M., Y. Song, Z. C. Mao, M. X. Liu, and X. Huang, 2016b: Impacts of thermal circulations induced by urbanization on ozone formation in the Pearl River Delta region, China. *Atmos. Environ.*, **127**, 382–392, https://doi.org/10.1016/j.atmosenv.2015.10.075.

Li, M., and Coauthors, 2017b: MIX: A mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. *Atmospheric Chemistry and Physics*, **17**, 935–963, https://doi.org/10.5194/acp-17-935-2017.

Li, S., and Coauthors, 2018a: Impact of East Asian summer monsoon on surface ozone pattern in China. *J. Geophys. Res.: Atmos.*, **123**, 1401–1411, https://doi.org/10.1002/2017JD027190.

Li, S., T. J. Wang, P. Zanis, D. Melas, and B. L. Zhuang, 2018b: Impact of tropospheric ozone on summer climate in China. *J. Meteor. Res.*, **32**, 279–287, https://doi.org/10.1007/s13351-018-7094-x.

Li, Y., A. K. H. Lau, J. C. H. Fung, J. Y. Zheng, and S. Liu, 2013: Importance of NO$_x$ control for peak ozone reduction in the Pearl River Delta region. *J. Geophys. Res.: Atmos.*, **118**, 9428–9443, https://doi.org/10.1002/jgrd.50659.

Liao, H., and J. H. Seinfeld, 2005: Global impacts of gas-phase chemistry-aerosol interactions on direct radiative forcing by anthropogenic aerosols and ozone. *J. Geophys. Res.: Atmos.*, **110**, https://doi.org/10.1029/2005JD005907.

Liao, J. B., and Coauthors, 2015: WRF/Chem modeling of the impacts of urban expansion on regional climate and air pollutants in Yangtze River Delta, China. *Atmos. Environ.*, **106**, 204–214, https://doi.org/10.1016/j.atmosenv.2015.01.059.

Liao, Z. H., M. Gao, J. R. Sun, and S. J. Fan, 2017: The impact of synoptic circulation on air quality and pollution-related human health in the Yangtze River Delta region. *Science of the Total Environment*, **607–608**, 838–846, https://doi.org/10.1016/j.scitotenv.2017.07.031.

Lim, S. S., and Coauthors, 2012: A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: A systematic analysis for the Global Burden of Disease Study 2010. *The Lancet*, **380**, 2224–2260.

Lin, M. Y., A. M. Fiore, L. W. Horowitz, A. O. Langford, S. J. Oltmans, D. Tarasick, and H. E. Rieder, 2015: Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions. *Nature Communications*, **6**, 7105, https://doi.org/10.1038/ncomms8105.

Liu, H., X. M. Wang, J. M. Pang, and K. B. He, 2013a: Feasibility and difficulties of China's new air quality standard compliance: PRD case of PM$_{2.5}$ and ozone from 2010 to 2025. *Atmospheric Chemistry and Physics*, **13**, 12 013–12 027, https://doi.org/10.5194/acp-13-12013-2013.

Liu, J., D. Wu, S. J. Fan, Z. H. Liao, and T. Deng, 2017: Impacts of precursors and meteorological factors on ozone pollution in Pearl River Delta. *China Environmental Science*, **37**, 813–820, https://doi.org/10.3969/j.issn.1000-6923.2017.03.002. (in Chinese with English abstract)

Liu, Q., K. S. Lam, F. Jiang, T. J. Wang, M. Xie, B. L. Zhuang, and X. Y. Jiang, 2013b: A numerical study of the impact of climate and emission changes on surface ozone over South China in autumn time in 2000–2050. *Atmos. Environ.*, **76**, 227–237, https://doi.org/10.1016/j.atmosenv.2013.01.030.

Lou, S. J., H. Liao, and B. Zhu, 2014: Impacts of aerosols on surface-layer ozone concentrations in China through heterogeneous reactions and changes in photolysis rates. *Atmos. Environ.*, **85**, 123–138, https://doi.org/10.1016/j.atmosenv.2013.12.004.

Lou, S. J., H. Liao, Y. Yang, and Q. Mu, 2015: Simulation of the interannual variations of tropospheric ozone over China: Roles of variations in meteorological parameters and anthropogenic emissions. *Atmos. Environ.*, **122**, 839–851, https://doi.org/10.1016/j.atmosenv.2015.08.081.

Lu, K. D., and Coauthors, 2012: Observation and modelling of OH and HO$_2$ concentrations in the Pearl River Delta 2006: A missing OH source in a VOC rich atmosphere. *Atmospheric Chemistry and Physics*, **12**, 1541–1569, https://doi.org/10.5194/acp-12-1541-2012.

Lu, X., and Coauthors, 2018: Severe surface ozone pollution in China: A global perspective. *Environmental Science & Technology Letters*, **5**, 487–494, https://doi.org/10.1021/acs.estlett.8b00366.

Ma, J. Z., X. B. Xu, C. S. Zhao, and P. Yan, 2012: A review of atmospheric chemistry research in China: Photochemical smog, haze pollution, and gas-aerosol interactions. *Adv. Atmos. Sci.*, **29**, 1006–1026, https://doi.org/10.1007/s00376-012-1188-7.

Ma, Z. Q., X. L. Zhang, J. Xu, X. J. Zhao, and W. Meng, 2011: Characteristics of ozone vertical profile observed in the boundary layer around Beijing in autumn. *Journal of Environmental Sciences*, **23**, 1316–1324, https://doi.org/10.1016/S1001-0742(10)60557-8.

Ma, Z. Q., J. Xu, W. J. Quan, Z. Y. Zhang, W. L. Lin, and X. B. Xu, 2016: Significant increase of surface ozone at a rural site, north of eastern China. *Atmospheric Chemistry and Physics*, **16**, 3969–3977, https://doi.org/10.5194/acp-16-3969-2016.

Monks, P. S., 2005: Gas-phase radical chemistry in the troposphere. *Chemical Society Reviews*, **34**, 376–395, https://doi.org/10.1039/b307982c.

Monks, P. S., and Coauthors, 2015: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. *Atmospheric Chemistry and Physics*, **15**, 8889–8973, https://doi.org/10.5194/acp-15-8889-2015.

Myhre, G., and Coauthors, 2013: Anthropogenic and natural radiative forcing. *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, T. F. Stocker et al., Eds., Cambridge University Press.

Myhre, G., and Coauthors, 2017: Multi-model simulations of aerosol and ozone radiative forcing due to anthropogenic emission changes during the period 1990–2015. *Atmospheric Chemistry and Physics*, **17**, 2709–2720, https://doi.org/10.5194/acp-17-2709-2017.

Newell, R. E., and M. J. Evans, 2000: Seasonal changes in pollutant transport to the North Pacific: The relative importance of Asian and European sources. *Geophys. Res. Lett.*, **27**, 2509–2512, https://doi.org/10.1029/2000GL011501.

Ohara, T., H. Akimoto, J. Kurokawa, N. Horii, K. Yamaji, X. Yan, and T. Hayasaka, 2007: An Asian emission inventory of anthropogenic emission sources for the period 1980-2020. *Atmospheric Chemistry and Physics*, **7**, 4419–4444, https://doi.org/10.5194/acp-7-4419-2007.

Ou, J. M., and Coauthors, 2016: Ambient ozone control in a photochemically active region: Short-term despiking or long-term attainment? *Environmental Science & Technology*, **50**, 5720–5728, https://doi.org/10.1021/acs.est.6b00345.

Peñuelas, J., and M. Staudt, 2010: BVOCs and global change. *Trends in Plant Science*, **15**, 133–144, https://doi.org/10.1016/j.tplants.2009.12.005.

Pu, X., T. J. Wang, X. Huang, D. Melas, P. Zanis, D. K. Papanastasiou, and A. Poupkou, 2017: Enhanced surface ozone during the heat wave of 2013 in Yangtze River Delta region, China. *Science of the Total Environment*, **603–604**, 807–816, https://doi.org/10.1016/j.scitotenv.2017.03.056.

Qu, Y. W., and Coauthors, 2018: Influence of atmospheric particulate matter on ozone in Nanjing, China: Observational study and mechanistic analysis. *Adv. Atmos. Sci.*, **35**, 1381–1395, https://doi.org/10.1007/s00376-018-8027-4.

Schnell, J. L., and M. J. Prather, 2017: Co-occurrence of extremes in surface ozone, particulate matter, and temperature over eastern North America. *Proceedings of the National Academy of Sciences of the United States of America*, **114**, 2854–2589, https://doi.org/10.1073/pnas.1614453114.

Schultz, M. G., and Coauthors, 2017: Tropospheric ozone assessment report: Database and metrics data of global surface ozone observations. *Elementa Science of the Anthropocene*, **5**, 58, http://doi.org/10.1525/elementa.244.

Seinfeld, J. H., and S. N. Pandis, 2006: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. 2nd ed. John Wiley and Sons.

Shen, L., L. J. Mickley, and A. P. K. Tai, 2015: Influence of synoptic patterns on surface ozone variability over the eastern United States from 1980 to 2012. *Atmospheric Chemistry and Physics*, **15**, 10 925–10 938, https://doi.org/10.5194/acp-15-10925-2015.

Shi, C. Z., and Coauthors, 2015: A study of aerosol optical properties during ozone pollution episodes in 2013 over Shanghai, China. *Atmospheric Research*, **153**, 235–249, https://doi.org/10.1016/j.atmosres.2014.09.002.

Shindell, D., and Coauthors, 2013: Attribution of historical ozone forcing to anthropogenic emissions. *Nature Climate Change*, **3**, 567–570, https://doi.org/10.1038/nclimate1835.

Skeie, R. B., T. K. Berntsen, G. Myhre, K. Tanaka, M. M. Kvalevåg, and C. R. Hoyle, 2011: Anthropogenic radiative forcing time series from pre-industrial times until 2010. *Atmospheric Chemistry and Physics*, **11**, 11 827–11 857, https://doi.org/10.5194/acp-11-11827-2011.

Sofen, E. D., D. Bowdalo, and M. J. Evans, 2016: How to most effectively expand the global surface ozone observing network. *Atmospheric Chemistry and Physics*, **16**, 1445–1457, https://doi.org/10.5194/acp-16-1445-2016.

Søvde, O. A., C. R. Hoyle, G. Myhre, and I. S. A. Isaksen, 2011: The HNO_3 forming branch of the $HO_2 + NO$ reaction: Pre-industrial-to-present trends in atmospheric species and radiative forcings. *Atmospheric Chemistry and Physics*, **11**, 8929–8943, https://doi.org/10.5194/acp-11-8929-2011.

Stohl, A., and Coauthors, 2003: Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO. *J. Geophys. Res.: Atmos.*, **108**, 8516, https://doi.org/10.1029/2002JD002490.

Streets, D. G., and Coauthors, 2003: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res.: Atmos.*, **108**, 8809, https://doi.org/10.1029/2002JD003093.

Streets, D. G., Q. Zhang, L. T. Wang, K. B. He, J. M. Hao, Y. Wu, Y. H. Tang, and G. R. Carmichael, 2006: Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *J. Geophys. Res.: Atmos.* **111**, D14306, https://doi.org/10.1029/2006JD007118.

Sun, L., and Coauthors, 2016: Significant increase of summertime ozone at Mount Tai in Central Eastern China. *Atmospheric Chemistry and Physics*, **16**, 10 637–10 650, https://doi.org/10.5194/acp-16-10637-2016.

Tai, A. P. K., L. J. Mickley, C. L. Heald, and S. L. Wu, 2013: Effect of CO_2 inhibition on biogenic isoprene emission: Implications for air quality under 2000 to 2050 changes in climate, vegetation, and land use. *Geophys. Res. Lett.*, **40**, 3479–3483, https://doi.org/10.1002/grl.50650.

Tang, G., Y. Wang, X. Li, D. Ji, S. Hsu, and X. Gao, 2012: Spatial-temporal variations in surface ozone in Northern China as observed during 2009–2010 and possible implications for future air quality control strategies. *Atmospheric Chemistry and Physics*, **12**, 2757–2776, https://doi.org/10.5194/acp-12-2757-2012.

Tang, H. Y., G. Liu, J. G. Zhu, Y. Han, and K. Kobayashi, 2013: Seasonal variations in surface ozone as influenced by Asian summer monsoon and biomass burning in agricultural fields of the northern Yangtze River Delta. *Atmospheric Research*, **122**, 67–76, https://doi.org/10.1016/j.atmosres.2012.10.030.

Tao, H. R., J. Xing, H. S. Zhou, X. Chang, G. J. Li, L. Chen, and J. H. Li, 2018: Impacts of land use and land cover change on regional meteorology and air quality over the Beijing-Tianjin-Hebei region, China. *Atmos. Environ.*, **189**, 9–21, https://doi.org/10.1016/j.atmosenv.2018.06.033.

Unger, N., D. T. Shindell, D. M. Koch, and D. G. Streets, 2006: Cross influences of ozone and sulfate precursor emissions changes on air quality and climate. *Proceedings of the National Academy of Sciences of the United States of America*, **103**, 4377–4380, https://doi.org/10.1073/pnas.0508769103.

Verstraeten, W. W., J. L. Neu, J. E. Williams, K. W. Bowman, J. R. Worden, and K. F. Boersma, 2015: Rapid increases in tropospheric ozone production and export from China. *Nature Geoscience*, **8**, 690–695, https://doi.org/10.1038/ngeo2493.

Wang, J., D. J. Allen, K. E. Pickering, Z. Q. Li, and H. He, 2016a: Impact of aerosol direct effect on East Asian air quality during

the EAST-AIRE campaign. *J. Geophys. Res.: Atmos.*, **121**, 6534–6554, https://doi.org/10.1002/2016JD025108.

Wang, S. X., and Coauthors, 2014: Emission trends and mitigation options for air pollutants in East Asia. *Atmospheric Chemistry and Physics*, **14**, 6571–6603, https://doi.org/10.5194/acp-14-6571-2014.

Wang, T., X. L. Wei, A. J. Ding, C. N. Poon, K. S. Lam, Y. S. Li, L. Y. Chan, and M. Anson, 2009: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994-2007. *Atmospheric Chemistry and Physics*, **9**, 6217–6227, https://doi.org/10.5194/acp-9-6217-2009.

Wang, T., and Coauthors, 2016b: Observations of nitryl chloride and modeling its source and effect on ozone in the planetary boundary layer of southern China. *J. Geophys. Res.: Atmos.*, **121**, 2476–2489, https://doi.org/10.1002/2015JD024556.

Wang, T., L. K. Xue, P. Brimblecombe, Y. F. Lam, L. Li, and L. Zhang, 2017a: Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects. *Science of the Total Environment*, **575**, 1582–1596, https://doi.org/10.1016/j.scitotenv.2016.10.081.

Wang, W. G., J. Wu, H. N. Liu, S. C. Guo, X. M. Chen, and Y. Luo, 2005: Researches on the influence of pollution emission on tropospheric ozone variation and radiation over China and its adjacent area. *Chinese Journal of Atmospheric Sciences*, **29**, 734–746, https://doi.org/10.3878/j.issn.1006-9895.2005.05.07. (in Chinese with English abstract)

Wang, W. N., and Coauthors, 2017b: Assessing spatial and temporal patterns of observed ground-level ozone in China. *Scientific Reports*, **7**, 3651, https://doi.org/10.1038/s41598-017-03929-w.

Wang, X., Z. X. Shen, J. J. Cao, L. M. Zhang, L. Liu, J. J. Li, S. X. Liu, and Y. F. Sun, 2012: Characteristics of surface ozone at an urban site of Xi'an in Northwest China. *Journal of Environmental Monitoring*, **14**, 116–126, https://doi.org/10.1039/C1EM10541H.

Wang, Y., and Coauthors, 2017c: Long-term O₃–precursor relationships in Hong Kong: Field observation and model simulation. *Atmospheric Chemistry and Physics*, **17**, 10 919–10 935, https://doi.org/10.5194/acp-17-10919-2017.

Wang, Y. H., B. Hu, G. Q. Tang, D. S. Ji, H. X. Zhang, J. H. Bai, X. K. Wang, and Y. S. Wang, 2013a: Characteristics of ozone and its precursors in Northern China: A comparative study of three sites. *Atmospheric Research*, **132–133**, 450–459, https://doi.org/10.1016/j.atmosres.2013.04.005.

Wang, Y. X., L. L. Shen, S. L. Wu, L. Mickley, J. W. He, and J. M. Hao, 2013b: Sensitivity of surface ozone over China to 2000-2050 global changes of climate and emissions. *Atmos. Environ.*, **75**, 374–382, https://doi.org/10.1016/j.atmosenv.2013.04.045.

Wang, Z., and Coauthors, 2015: Ground-level ozone in urban Beijing over a 1-year period: Temporal variations and relationship to atmospheric oxidation. *Atmospheric Research*, **164-165**, 110-117.

Wei, W., Z. F. Lv, S. Y. Cheng, L. L. Wang, D. S. Ji, Y. Zhou, L. H. Han, and L. T. Wang, 2015: Characterizing ozone pollution in a petrochemical industrial area in Beijing, China: A case study using a chemical reaction model. *Environmental Monitoring and Assessment*, **187**, 377, https://doi.org/10.1007/s10661-015-4620-5.

Wu, R. R., Y. Bo, J. Li, L. Y. Li, Y. Q. Li, and S. D. Xie, 2016: Method to establish the emission inventory of anthropogenic volatile organic compounds in China and its applica-

tion in the period 2008-2012. *Atmos. Environ.*, **127**, 244–254, https://doi.org/10.1016/j.atmosenv.2015.12.015.

Xia, Y. M., Y. Zhao, and C. P. Nielsen, 2016: Benefits of China's efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014. *Atmos. Environ.*, **136**, 43–53, https://doi.org/10.1016/j.atmosenv.2016.04.013.

Xie, B., H. Zhang, Z. L. Wang, S. Y. Zhao, and Q. Fu, 2016: A modeling study of effective radiative forcing and climate response due to tropospheric ozone. *Adv. Atmos. Sci.*, **33**, 819–828, https://doi.org/10.1007/s00376-016-5193-0.

Xing, J., S. X. Wang, C. Jang, Y. Zhu, and J. M. Hao, 2011: Nonlinear response of ozone to precursor emission changes in China: A modeling study using response surface methodology. *Atmospheric Chemistry and Physics*, **11**, 5027–5044, https://doi.org/10.5194/acp-11-5027-2011.

Xing, J., R. Mathur, J. Pleim, C. Hogrefe, C. M. Gan, D. C. Wong, C. Wei, and J. D. Wang, 2015: Air pollution and climate response to aerosol direct radiative effects: A modeling study of decadal trends across the northern hemisphere. *J. Geophys. Res.: Atmos.*, **120**, 12 221–12 236, https://doi.org/10.1002/2015JD023933.

Xing, J., and Coauthors, 2017: Impacts of aerosol direct effects on tropospheric ozone through changes in atmospheric dynamics and photolysis rates. *Atmospheric Chemistry and Physics*, **17**, 9869–9883, https://doi.org/10.5194/acp-17-9869-2017.

Xu, J., and Coauthors, 2011a: Measurements of ozone and its precursors in Beijing during summertime: Impact of urban plumes on ozone pollution in downwind rural areas. *Atmospheric Chemistry and Physics*, **11**, 12 241–12 252, https://doi.org/10.5194/acp-11-12241-2011.

Xu, J., Y. H. Zhang, S. Q. Zheng, and Y. J. He, 2012: Aerosol effects on ozone concentrations in Beijing: A model sensitivity study. *Journal of Environmental Sciences*, **24**, 645–656, https://doi.org/10.1016/S1001-0742(11)60811-5.

Xu, W. Y., and Coauthors, 2011b: Characteristics of pollutants and their correlation to meteorological conditions at a suburban site in the North China Plain. *Atmospheric Chemistry and Physics*, **11**, 4353–4369, https://doi.org/10.5194/acp-11-4353-2011.

Xu, W. Y., W. L. Lin, X. B. Xu, J. Tang, J. Q. Huang, H. Wu, and X. C. Zhang, 2016: Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China — Part 1: Overall trends and characteristics. *Atmospheric Chemistry and Physics*, **16**, 6191–6205, https://doi.org/10.5194/acp-16-6191-2016.

Xu, W. Y., X. B. Xu, M. Y. Lin, W. L. Lin, D. Tarasick, J. Tang, J. Z. Ma, and X. D. Zheng, 2018: Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China — Part 2: The roles of anthropogenic emissions and climate variability. *Atmospheric Chemistry and Physics*, **18**, 773–798, https://doi.org/10.5194/acp-18-773-2018.

Xu, X. B., and W. L. Lin, 2011c: Trends of Tropospheric Ozone over China Based on Satellite Data (1979–2005). *Advances in Climate Change Research*, **2**(1), 43–48, https://doi.org/10.3724/SP.J.1248.2011.00043.

Xue, L. K., T. Wang, P. K. K. Louie, C. W. Y. Luk, D. R. Blake, and Z. Xu, 2014: Increasing external effects negate local efforts to control ozone air pollution: A case study of Hong Kong and implications for other Chinese Cities. *Environmental Science & Technology*, **48**, 10 769–10 775, https://doi.org/10.1021/

es503278g.

Yan, Y. Y., J. T. Lin, and C. L. He, 2018: Ozone trends over the United States at different times of day. *Atmospheric Chemistry and Physics*, **18**, 1185–1202, https://doi.org/10.5194/acp-18-1185-2018.

Yang, Y., H. Liao, and J. Li, 2014a: Impacts of the East Asian summer monsoon on interannual variations of summertime surface-layer ozone concentrations over China. *Atmospheric Chemistry and Physics*, **14**, 6867–6879, https://doi.org/10.5194/acp-14-6867-2014.

Yang, Y., H. Liao, and S. J. Lou, 2014b: Simulated impacts of sulfate and nitrate aerosol formation on surface-layer ozone concentrations in China. *Atmospheric and Oceanic Science Letters*, **7**, 441–446, https://doi.org/10.3878/j.issn.1674-2834.14.0033.

Zhang, H., B. Xie, and Z. Wang, 2018: Effective radiative forcing and climate response to short-lived climate pollutants under different scenarios. *Earth's Future*, **6**, 857–866, https://doi.org/10.1029/2018EF000832.

Zhang, Q., and Coauthors, 2009: Asian emissions in 2006 for the NASA INTEX-B mission. *Atmospheric Chemistry and Physics*, **9**, 5131–5153, https://doi.org/10.5194/acp-9-5131-2009.

Zhang, Q., K. B. He, and H. Huo, 2012a: Cleaning China's air. *Nature*, **484**, 161–162, https://doi.org/10.1038/484161a.

Zhang, Q., and Coauthors, 2014: Variations of ground-level O_3 and its precursors in Beijing in summertime between 2005 and 2011. *Atmospheric Chemistry and Physics*, **14**, 6089–6101, https://doi.org/10.5194/acp-14-6089-2014.

Zhang, X. Y., Y. Q. Wang, T. Niu, X. C. Zhang, S. L. Gong, Y. M. Zhang, and J. Y. Sun, 2012b: Atmospheric aerosol compositions in China: Spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols. *Atmospheric Chemistry and Physics*, **12**, 779–799, https://doi.org/10.5194/acp-12-779-2012.

Zhang, Y., H. T. Mao, A. J. Ding, D. R. Zhou, and C. B. Fu, 2013: Impact of synoptic weather patterns on spatio-temporal variation in surface O_3 levels in Hong Kong during 1999–2011. *Atmos. Environ.*, **73**, 41–50, https://doi.org/10.1016/j.atmosenv.2013.02.047.

Zhang, Y. Q., O. R. Cooper, A. Gaudel, A. M. Thompson, P. Nédélec, S.-Y. Ogino, and J. J. West, 2016: Tropospheric ozone change from 1980 to 2010 dominated by equatorward redistribution of emissions. *Nature Geoscience*, **9**, 875–879, https://doi.org/10.1038/ngeo2827.

Zhao, B., and Coauthors, 2013: NO_x emissions in China: Historical trends and future perspectives. *Atmospheric Chemistry and Physics*, **13**, 9869–9897, https://doi.org/10.5194/acp-13-9869-2013.

Zhao, W., S. J. Fan, H. Guo, B. Gao, J. R. Sun, and L. G. Chen, 2016: Assessing the impact of local meteorological variables on surface ozone in Hong Kong during 2000-2015 using quantile and multiple line regression models. *Atmos. Environ.*, **144**, 182–193, https://doi.org/10.1016/j.atmosenv.2016.08.077.

Zhao, Z. J., and Y. X. Wang, 2017: Influence of the West Pacific subtropical high on surface ozone daily variability in summertime over eastern China. *Atmos. Environ.*, **170**, 197–204, https://doi.org/10.1016/j.atmosenv.2017.09.024.

Zheng, B., and Coauthors, 2018: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. *Atmospheric Chemistry and Physics*, **18**, 14 095–14 111, https://doi.org/10.5194/acp-18-14095-2018.

Zheng, J. Y., L. J. Zhong, T. Wang, P. K. K. Louie, and Z. C. Li, 2010: Ground-level ozone in the Pearl River Delta region: Analysis of data from a recently established regional air quality monitoring network. *Atmos. Environ.*, **44**, 814–823, https://doi.org/10.1016/j.atmosenv.2009.11.032.

Zhou, D. R., and Coauthors, 2013: Impacts of the East Asian monsoon on lower tropospheric ozone over coastal South China. *Environmental Research Letters*, **8**, 044011, https://doi.org/10.1088/1748-9326/8/4/044011.

Zhu, J., and H. Liao, 2016: Future ozone air quality and radiative forcing over China owing to future changes in emissions under the Representative Concentration Pathways (RCPs). *J. Geophys. Res.: Atmos.*, **121**, 1978–2001, https://doi.org/10.1002/2015JD023926.

Zou, Y., and Coauthors, 2015: Characteristics of 1 year of observational data of VOCs, NO_x and O_3 at a suburban site in Guangzhou, China. *Atmospheric Chemistry and Physics*, **15**, 6625-6636.