

TOPICAL REVIEW • **OPEN ACCESS**

## Ground-level ozone pollution in China: a synthesis of recent findings on influencing factors and impacts

To cite this article: Tao Wang *et al* 2022 *Environ. Res. Lett.* **17** 063003

View the [article online](#) for updates and enhancements.

You may also like

- [Changes in extreme rainfall over mainland China induced by landfalling tropical cyclones](#)  
Jun Su, Guoyu Ren, Yingxian Zhang *et al.*
- [Analysis of variation in reference evapotranspiration and its driving factors in mainland China from 1960 to 2016](#)  
Dong Wu, Shibo Fang, Xingyuan Tong *et al.*
- [Divergent trends in irrigation-water withdrawal and consumption over mainland China](#)  
Ling Zhang, Donghai Zheng, Kun Zhang *et al.*

ENVIRONMENTAL RESEARCH  
LETTERS

## TOPICAL REVIEW

## OPEN ACCESS

## RECEIVED

30 December 2021

## REVISED

21 April 2022

## ACCEPTED FOR PUBLICATION

25 April 2022

## PUBLISHED

24 May 2022

Original content from this work may be used under the terms of the [Creative Commons Attribution 4.0 licence](#).

Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.



## Ground-level ozone pollution in China: a synthesis of recent findings on influencing factors and impacts

Tao Wang<sup>1,\*</sup> , Likun Xue<sup>2</sup>, Zhaozhong Feng<sup>3</sup>, Jianing Dai<sup>1,4</sup>, Yingnan Zhang<sup>2</sup> and Yue Tan<sup>1</sup><sup>1</sup> Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong 999077, People's Republic of China<sup>2</sup> Environment Research Institute, Shandong University, Qingdao 266237, People's Republic of China<sup>3</sup> School of Applied Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, People's Republic of China<sup>4</sup> Max Planck Institute for Meteorology, Hamburg, Germany

\* Author to whom any correspondence should be addressed.

E-mail: [tao.wang@polyu.edu.hk](mailto:tao.wang@polyu.edu.hk)**Keywords:** ozone, O<sub>3</sub>, China, ozone trend, ozone impact on crops and forests, ozone formation and transport, ozone controlSupplementary material for this article is available [online](#)

## Abstract

Ozone (O<sub>3</sub>) in the troposphere is an air pollutant and a greenhouse gas. In mainland China, after the Air Pollution Prevention and Action Plan was implemented in 2013—and despite substantial decreases in the concentrations of other air pollutants—ambient O<sub>3</sub> concentrations paradoxically increased in many urban areas. The worsening urban O<sub>3</sub> pollution has fuelled numerous studies in recent years, which have enriched knowledge about O<sub>3</sub>-related processes and their impacts. In this article, we synthesise the key findings of over 500 articles on O<sub>3</sub> over mainland China that were published in the past six years in English-language journals. We focus on recent changes in O<sub>3</sub> concentrations, their meteorological and chemical drivers, complex O<sub>3</sub> responses to the drastic decrease in human activities during coronavirus disease 2019 lockdowns, several emerging chemical processes, impacts on crops and trees, and the latest government interventions.

## 1. Introduction

While ozone (O<sub>3</sub>) in the stratosphere is beneficial for life on Earth by filtering out Sun's harmful ultra-violet radiation, at ground level, O<sub>3</sub> adversely affects human health and vegetation. O<sub>3</sub> also regulates the oxidative capacity of the troposphere, and is a powerful greenhouse gas that contributes to climate warming. Elevated O<sub>3</sub> concentrations in the lower part of the atmosphere remain a persistent environmental problem in many of the world's urban and industrialised regions [1–3]. In mainland China (henceforth 'China'), the rigorous control of emissions following the implementation of the Air Pollution Prevention and Action Plan (2013–2017) has led to remarkable reductions in air pollution caused by sulphur dioxide, particulate matter and nitrogen oxides [4]. However, ground-level O<sub>3</sub> concentrations in major population centres have been increasing in recent years [5, 6],

which has led to a surge in the numbers of studies on O<sub>3</sub> in the past six years. From this body of work, new findings on the factors influencing O<sub>3</sub> production and distribution and on the O<sub>3</sub> impacts have emerged. On the policy side, mitigating O<sub>3</sub> pollution is now on the agenda of the national and local governments planning the next phase of air-pollution controls. Therefore, the time is ripe for a synthesis of recent findings concerning the key chemical and physical influences on ground-level O<sub>3</sub> concentrations, which will help provide scientific support for the development of evidence-based O<sub>3</sub> control strategies.

Several reviews on the O<sub>3</sub> situation in China have been conducted in the past five years. The first comprehensive review [3] covered the fundamental chemical and meteorological processes involved in O<sub>3</sub> production and transport, the history of O<sub>3</sub> monitoring and research in China, the O<sub>3</sub> concentrations in major city-clusters, the relationships between O<sub>3</sub>

concentrations and their precursors, early findings on the roles of nitrous acid (HONO) and nitryl chloride (ClNO<sub>2</sub>) in O<sub>3</sub> production, and the impacts of O<sub>3</sub> on crops, forests and human health. Subsequently, five additional topical reviews were published [7–11]. Lu *et al* [7] reviewed findings on the O<sub>3</sub> formation regimes and their temporal (inter-annual, seasonal and diurnal) changes for period up to 2016. Lu *et al* [8] focused on the impacts of meteorology and climate on natural emissions of O<sub>3</sub> precursors, chemistry and deposition, and transport patterns. Fu *et al* [9] reviewed impacts of emission, multiscale meteorology, land use change, aerosol effect on O<sub>3</sub>, and the influence of tropospheric O<sub>3</sub> change on radiative forcing and temperature. Liu *et al* [10] reviewed the methodological aspects of O<sub>3</sub> and precursor apportionment, including back trajectories, observation-based chemical models, receptor models for volatile organic compounds (VOCs), emission-based chemical transport model, and their respective results. Xu [11] provided a short review of the impacts of O<sub>3</sub> on crop yield and economic loss, tree risk exposure to O<sub>3</sub>, and cardio-vascular mortality with O<sub>3</sub> exposure dose. The O<sub>3</sub> spatial distribution and changes across China from 2013 up to 2019 were presented in three of the above reviews [9–11], and the decadal trends of O<sub>3</sub> at limited number of sites were reviewed in two of them [9, 11].

The present review focuses on the multiple novel insights that were gained in the recent six years, but were not covered by the prior reviews. They include (1) the recent situation of O<sub>3</sub>-formation regimes amid decreasing NO<sub>x</sub> and increasing VOC emissions, (2) the recent results on the impact of HONO and ClNO<sub>2</sub> on O<sub>3</sub> production and the new findings on the impact of other reactive halogen (Cl<sub>2</sub> and BrCl) in polluted regions, (3) the winter photochemistry of O<sub>3</sub> (an understudied topic), (4) the complex responses of O<sub>3</sub> to reduced emissions under recent control measures and during China's coronavirus disease 2019 (COVID-19) lockdown, (5) the impact of O<sub>3</sub> on the quality of grain crops, tree productivity and water use efficiency. Additionally, we update the results on surface O<sub>3</sub> changes across China to the year of 2021, which reveals possible signs of O<sub>3</sub> levelling off or decrease in some urban areas, and (6) introduce recent government efforts to control O<sub>3</sub> in summer. We also give a graphic summary of multi-scale transport patterns.

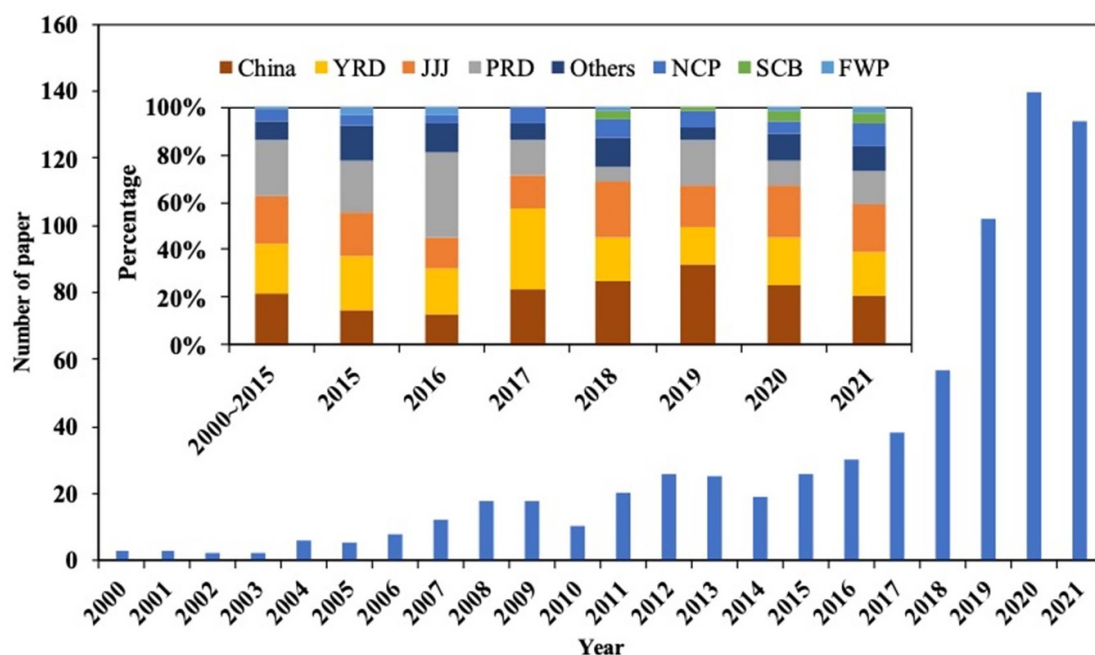
## 2. Literature search and results

We adopted an evidence-based approach to systematically search three research databases (Web of Science, Scopus and Google Scholar) for English-language, peer-reviewed studies that were published (online) between 1 January 2000 and 30 June 2021, using three

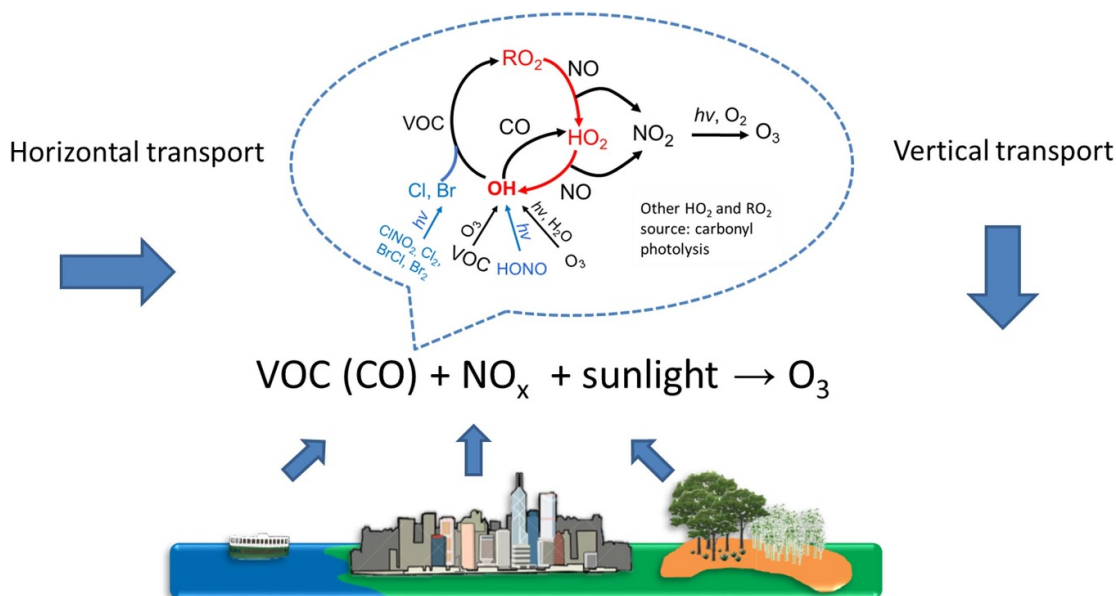
search methods. Here, we included the papers published before 2015 in the search with the aim to show the rapid development of ozone research in China. The first search method used keywords that combined 'ozone' or 'O<sub>3</sub>' with the name of a specific region, namely 'China', 'Chinese', 'Pearl River Delta' (or 'PRD'), 'Yangtze River Delta' (or 'YRD'), 'North China Plain' (or 'NCP'), 'Beijing–Tianjin–Hebei' (or 'BTH'), 'Jing-Jin-Ji' (or 'JJJ'), 'Sichuan Basin' (or 'SCB'), Tibetan Plateau (or 'TP'), 'Beijing', 'Shanghai', 'Guangzhou' or 'Hong Kong'. This returned 656 articles that had been published since 2000. The second search method targeted publications from active O<sub>3</sub>-research groups across 21 universities (or research institutes) that were known to the authors (table S1 (available online at [stacks.iop.org/ERL/17/063003/mmedia](https://stacks.iop.org/ERL/17/063003/mmedia))). The search yielded 54 additional papers that had not been found in the previous search. At this stage, we also identified four additional papers that cited Wang *et al* [3]. The third search method used the keywords 'O<sub>3</sub>' and 'COVID-19', and produced 37 papers. A total of 751 relevant papers were identified (see appendix for the complete list), 571 of which were published between 1 January 2015 and 30 June 2021.

Figure 1 shows the number of O<sub>3</sub>-related papers published from 2000 to the end of the June 2021, which highlights the rapid growth of O<sub>3</sub> research in China since 2015. Ozone studies in China have focused on the three most urban and industrial conglomerates: the PRD (including Hong Kong) in the south, the YRD in the east, and the Jing-Jin-Ji (Beijing–Tianjin–Hebei) region in the NCP. In recent years, an increasing proportion of publications have focused on the northern parts of China (figure 1, insert). This trend reflects the fact that earlier O<sub>3</sub> studies had mainly been conducted in southern parts of the country, whereas research (and mitigation) in the north had been focused on the more pressing problems of sulphur and particulate pollution [3]. Increasing recognition of the serious O<sub>3</sub> pollution in Beijing [12, 13] has motivated more studies on O<sub>3</sub> in the NCP. There have also been more publications on O<sub>3</sub> processes and impacts across the whole of China since 2013, when data on ambient concentrations of O<sub>3</sub> and other regulated pollutants were made available by the China National Environmental Monitoring Center network.

The topics of investigation in O<sub>3</sub> studies in China have changed over the years. Due to limited research funding, earlier studies were largely exploratory in nature. Subsequent rapid increases in research facilities and the research community, and a large increase in research funding, has led to recent in-depth and rigorous studies on specific processes contributing to O<sub>3</sub> concentrations and the impacts of O<sub>3</sub> on vegetation and human health.



**Figure 1.** Number of journal papers published during 1 January 2000–30 June 2021 and their covered regions (insert). JJJ: Jing-Jin-Ji, i.e. Beijing-Tianjin-Hebei; YRD: Yangtze River Delta; NCP: North China Plain (excluding JJJ); PRD: Pearl River Delta; SCB: Sichuan Basin; FWP: Fenwei Plain (refer to figure 6 for their geographic locations); China: papers focusing on the whole country.



**Figure 2.** Illustration depicting the sources of ground-level ozone ( $O_3$ ) and radicals that initiate oxidation of VOC and CO in polluted regions. The ‘conventional’ radicals are labelled in red, and understudied halogen radicals and HONO (an OH source) are shown in blue. The  $O_3$  loss pathways, radical termination steps and their depositions are not shown.

### 3. Brief review of the factors affecting ground-level $O_3$ concentrations

The basic chemical and physical factors affecting the formation and distribution of ground-level  $O_3$  have been well established, and reader is referred to previous review articles [3, 14–22]. Here, we only give a brief summary. Ground-level  $O_3$  is produced via

the chemical reactions of the precursors  $NO_x$  (nitric oxide (NO) and nitrogen dioxide ( $NO_2$ )), VOCs, and CO (carbon monoxide) under sunlight (figure 2). In urban and industrial regions,  $NO_x$  is mainly emitted from the combustion of fossil fuels, such as in vehicles and industrial settings, whereas VOCs are released from more diverse sources such as vehicle exhaust, evaporating fuels and solvents, consumer products,

and trees. In polluted regions, the photochemical production of  $O_3$  begins with OH radical (or an analogue, such as Cl atom) reacting with VOC (and CO) to form organic peroxy radical ( $RO_2$ ) and hydroperoxyl radical ( $HO_2$ ), which subsequently oxidise NO to  $NO_2$ . Then, photolysis of  $NO_2$  produces O atoms, which react with  $O_2$  to form  $O_3$  (figure 2, also see Wang *et al* [3]). Therefore, the sources and sinks of OH radical profoundly affect the rate at which  $O_3$  is formed. It has been well established that  $O_3$  has a non-linear relationship with its precursors; that is,  $NO_x$  can lead to either decreases or increases in the concentration of  $O_3$ , depending on the relative ratio of  $NO_x$  to VOC. In general, the production of  $O_3$  in urban areas with high  $NO_x$ /VOC ratios is VOC-limited, such that a reduction in  $NO_x$  emissions will tend to increase  $O_3$  concentrations due to the decreased titration of  $O_3$  and radicals by  $NO_x$ . In contrast, in rural areas where  $NO_x$ /VOCs ratios are often low, decreasing  $NO_x$  emissions will decrease  $O_3$  concentrations. Aerosols can also influence  $O_3$  concentrations by altering solar irradiance, and via the chemical reactions that occur on aerosol surfaces. In addition to *in-situ* photochemistry,  $O_3$  is transported from higher altitudes of the atmosphere to ground level, and from one region to another. Meteorological conditions can also strongly affect  $O_3$  production and distribution by changing patterns of air transport, the wet and dry depositions of gases and aerosols, and the rates of chemical reactions and natural emissions.

## 4. $O_3$ spatiotemporal distributions and formation regimes

### 4.1. Decadal change

Long-term (>10 years) changes in  $O_3$  concentrations over China have been summarized in recent reviews [9, 11], and factors contributing to the  $O_3$  changes were reviewed by Fu *et al* [9]. The reader is referred to these reviews for the detail results on the decadal change in  $O_3$ . Here, we give a brief summary. In mainland China,  $O_3$  measurements lasting longer than 15 years were limited, and were mainly conducted at several regional background sites [9, 11]. Additionally,  $O_3$  decadal changes have been reported in major cities such as Beijing and Shanghai and in the PRD region [9, 11]. The results from these relatively long measurements indicate increasing surface  $O_3$  in or near the developed regions of China. The increasing rate was typically lower at rural sites than at urban cores in a given region (e.g. JJJ), and the surface  $O_3$  concentrations at several rural sites (Lin'an, Longfengshan, Gucheng and Hok Tsui) appeared to have stopped increasing or even decreased during the recent decade [23, 24]. The decreases in  $O_3$  concentrations in the rural areas may be a consequence of decreased  $NO_x$  emissions since 2011. Figure 3 shows a marked decrease in  $NO_x$  emissions after 2011, but continued increase in VOC emissions, which is in

contrast to the decrease in both  $NO_x$  and VOC emissions in the U.S. and Europe (figure 3).

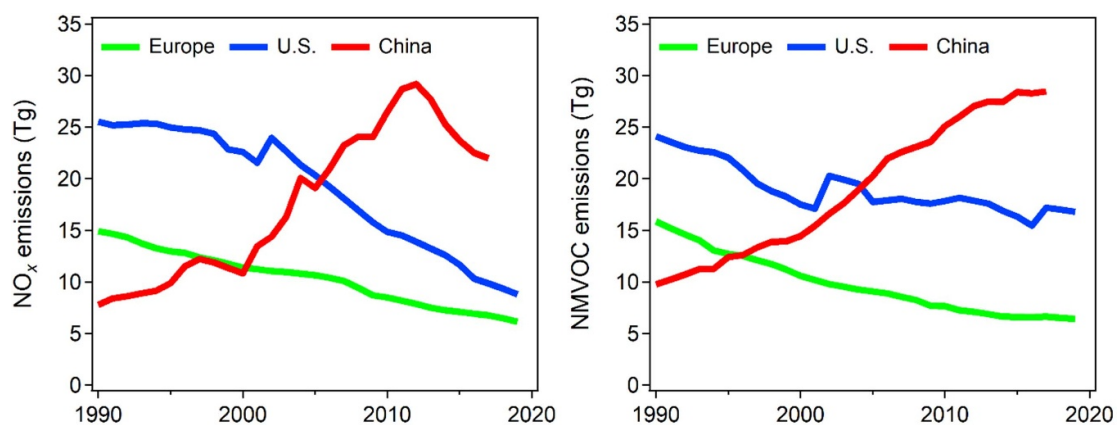
### 4.2. Urban $O_3$ since 2013

After public release of nation-wide data from the China National Environmental Monitoring Center network since 2013, a much clearer picture has emerged on the  $O_3$  changes in urban areas across China. Numerous recent studies have analysed the data to show rapidly increasing urban  $O_3$  concentrations during years up to 2019 [9, 11]. In the present review, we extend the prior analysis of the trend result to the year of 2021. Figure 4 shows the rates of change in maximum daily 8-hour average (MDA8)  $O_3$  mixing ratios during 2013–2021 in 162 cities in which  $O_3$  concentrations have been monitored since 2013 (figure 4(a)), the spatial distribution of the average MDA8  $O_3$  mixing ratios at 1497 sites in 2021 (figure 4(b)), and the average MDA8 mixing ratios in seven representative cities in regions that experience frequent  $O_3$  pollution (figure 4(c)) in warm seasons (May–October). Figure 5 depicts the respective results for cool seasons (November–April). The seven cities include Beijing (in Jing-Jin-Ji, northern part of NCP), Jinan (in southern part of NCP), Xi'an (in FWP), Shanghai (in YRD), Wuhan (in city clusters of central China), Chengdu (in SCB) and Guangzhou (in PRD). Several insights can be obtained from these plots. First, the rates of increase in MDA8  $O_3$  mixing ratios have slowed down in the recent two years, as indicated by the smaller positive rates for 2013–2021 than those for 2013–2018 in many cities. This may be in part a consequence of the efforts to control  $O_3$  precursors emissions that were implemented in summers of 2020 and 2021 (see section 10). Second, the rates of increases were larger in the cool seasons compared to those in the warm seasons (figures 4(a) and 5(a)). Third, the  $O_3$  increase rates were generally larger in the central-eastern regions of China (i.e. parts of Hebei, Shanxi, Henan and Shandong province), compared to the northeastern and southern regions. Consistent with the previous results, the 2021 data show that  $O_3$  pollution was more severe in urbanized and industrialized regions such as NCP, FWP, YRD, and PRD. The findings of the effects of meteorology and emission changes on the urban  $O_3$  changes during 2013–2017 are summarized in section 7.

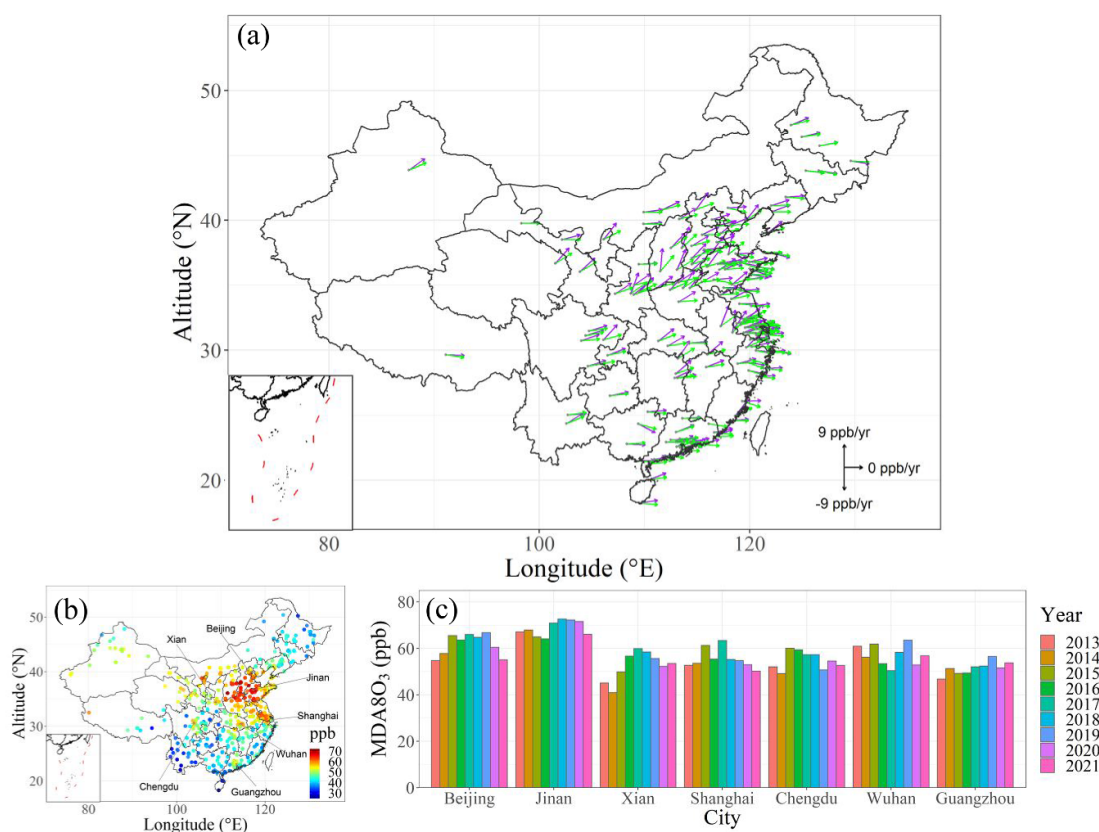
### 4.3. $O_3$ formation regime

Previous findings on  $O_3$  formation regimes up to the year of 2016 have been reviewed by Wang *et al* [3] and Lu *et al* [7], which indicated that in most urban areas, chemical production of  $O_3$  was controlled by VOCs. Observation-based results from the past 5 years suggest that this situation has largely remained despite decrease in  $NO_x$  emissions and increase in VOC emissions, e.g. in urban areas of Shanghai [28], Nanjing [29], and Wuhan [30]. The recent studies have expanded to less-developed cities





**Figure 3.** Trends in the annual anthropogenic emissions of nitrogen oxides (NO<sub>x</sub>) (1990–2017) and non-methane VOCs (NMVOCs) (1990–2017) in China, and a comparison with the trends in the U.S. and Europe. [Unit: Teragram (Tg)]. The NO<sub>x</sub> emissions are counted as the mass of NO<sub>x</sub> (NO + NO<sub>2</sub>) in China and the U.S., and as the mass equivalent to NO<sub>2</sub> in Europe. NMVOCs are the total mass of non-methane hydrocarbons and oxygenated VOCs. The emission data of NO<sub>x</sub> and NMVOC for China were obtained from Li *et al* [25], Zheng *et al* [26] and the China statistical yearbook [27]. The anthropogenic emission data of NO<sub>x</sub> and NMVOC for the U.S. and Europe were obtained from USEPA ([www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data](http://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data)) and EEA ([www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-4](http://www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-4)), respectively.



**Figure 4.** (a) The rates of change in maximum daily 8-hour average (MDA8) O<sub>3</sub> mixing ratios during 2013–2018 (purple arrows) and during 2013–2021 (green arrows) in 162 cities of Mainland China, (b) the spatial distribution of average MDA8 O<sub>3</sub> mixing ratios at 1497 sites in 2021, and (c) the average MDA8 mixing ratios in seven cities. The results are for warm seasons (May–October). The O<sub>3</sub> data were obtained from <https://air.cnemc.cn:18007/>.

such as Dongying (in NCP) [31], Xuzhou, Yancheng, and Nantong (in YRD) [32], Shantou (in Guangdong province) [33] and Weinan (in FWP) [34]. For most of the newly-studied cities, urban areas were found in a VOC-limited regime, with two exceptions in Weinan

and Nantong, both of which were in a mixed-limited regime [32, 34]. However, transition from a VOC-limited [35] to a mixed-limited O<sub>3</sub> formation regime [28] was indicated in some suburban areas of the YRD, mainly owing to decreases in NO<sub>x</sub> emissions in

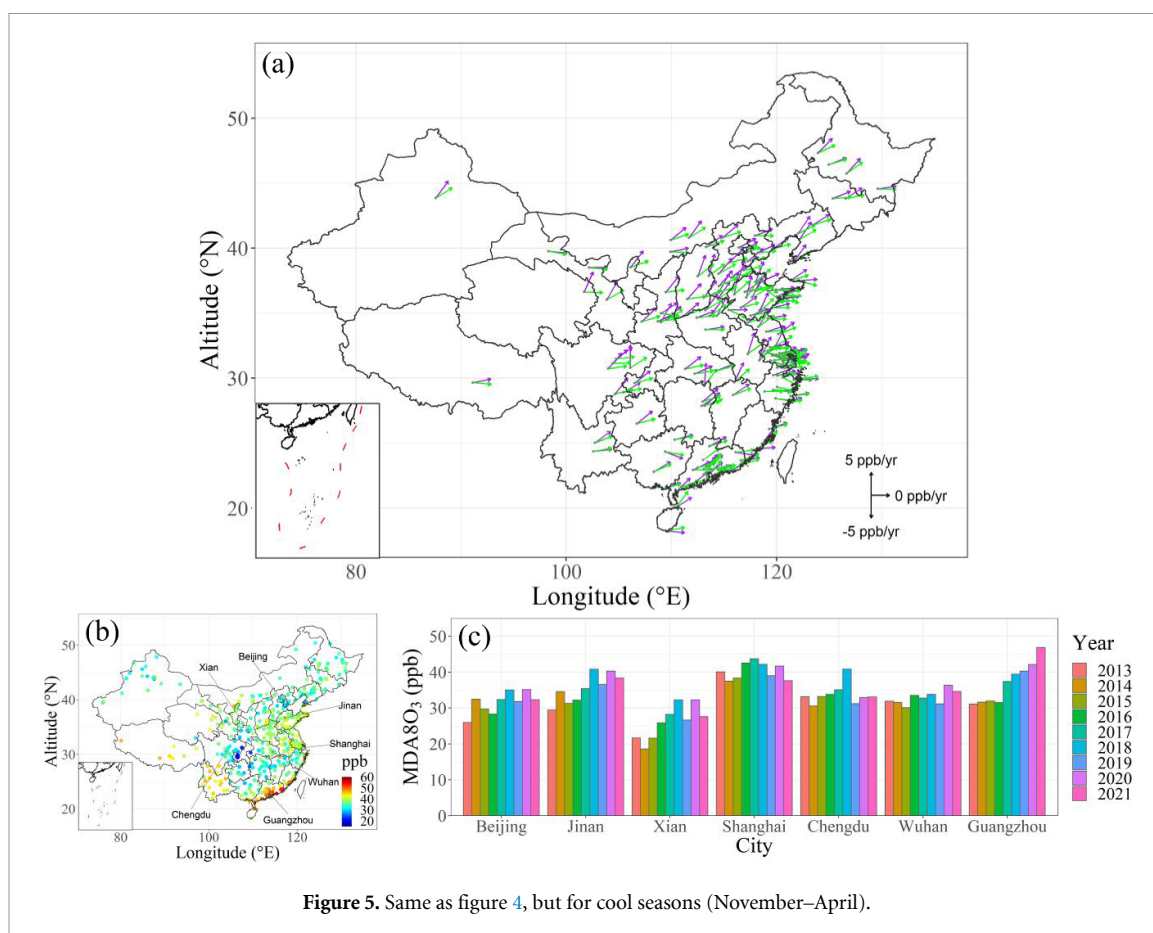


Figure 5. Same as figure 4, but for cool seasons (November–April).

the recent years. As to altitude dependence, an analysis of airborne measurements in the NCP showed that O<sub>3</sub> formation became more sensitive to NO<sub>x</sub> emissions with increasing heights [36].

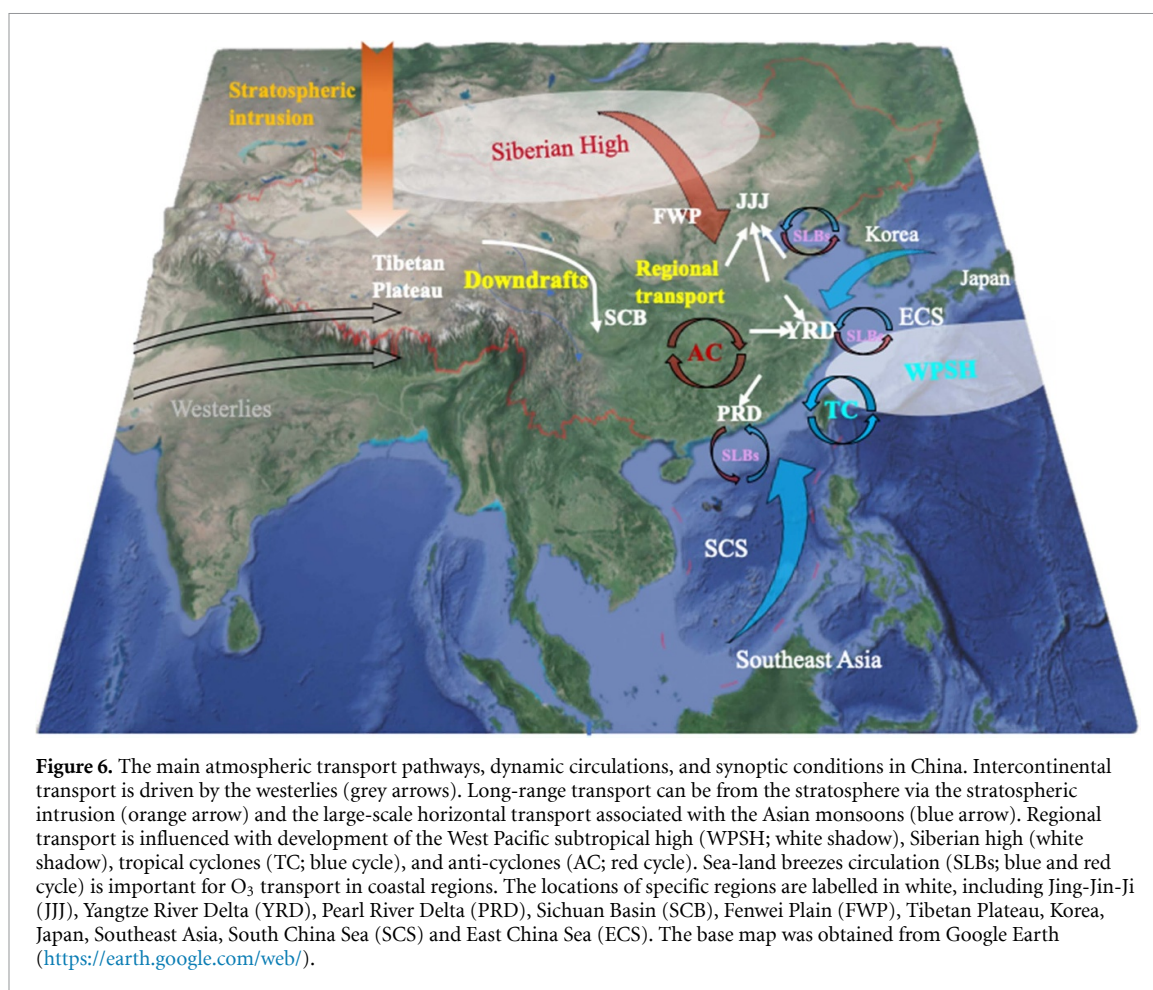
Recent analysis of *in-situ* measurements indicated that alkenes and aromatics were the major reactive VOC groups for O<sub>3</sub> production in urban areas. For example, alkenes were the dominant VOCs in the NCP [37], FWP [34], and SCB [38], while aromatics were more important in the YRD [28, 39] and PRD [40]. The importance of aromatics to O<sub>3</sub> formation typically increased along with decreasing latitudes. These results are generally similar to those obtained from the early studies [3, 7], indicating insignificant changes in VOC species that controlled O<sub>3</sub> formation in these regions. Biogenic VOCs play an important role in some occasions [39].

Recent emission-based model studies [41] and analysis of satellite data [42] have suggested some changes in the O<sub>3</sub> formation regime over eastern China, with the areas of mixed-limited regime becoming larger due to NO<sub>x</sub> emission reductions. Quantitatively, Wang *et al* [41] found that the areas in mixed-limited regime in JJJ, YRD, and PRD expanded by 17.1%, 20.8%, and 20.3%, respectively, from 2012 to 2016 according to the simulations with a Weather Research Forecast–Community Multiscale Air Quality (WRF-CMAQ). The expansion mainly occurred in suburban and rural areas, whereas urban areas in

Beijing, Tianjin, Shanghai, Nanjing, and Guangzhou remained in a VOC-limited regime. The results from emission-based models are generally consistent with the findings from observation-based analysis, which indicate that the reduction in NO<sub>x</sub> emissions in the past decade has affected the O<sub>3</sub> formation regime, but is insufficient to change the typical urban areas from a VOC-limited to a NO<sub>x</sub>-limited regime.

## 5. Roles of multi-scale atmospheric transport

O<sub>3</sub> is a secondary pollutant with a lifetime that ranges between several days and several weeks [16], and thus can be transported from one place to another. In this review, we consider three scales at which the atmospheric transport of O<sub>3</sub> occurs: the intercontinental-, long-range- and regional scales. Intercontinental transport of O<sub>3</sub> occurs between, for instance, Europe and Asia. Long-range transport of O<sub>3</sub> occurs from other Asian countries to China and from the stratosphere to ground level. We separate regional transport of O<sub>3</sub> into super-regional (i.e. inter-regional) and intra-regional scales, which represent transport between different regions and different areas within the same region, respectively. Previous reviews have summarized the findings of the impact of multiscale meteorological conditions on O<sub>3</sub> accumulation and transport up to the year of 2018 [8, 9] and the source



apportionment of  $O_3$  and its precursors up to the year of 2019 [10]. The present review synthesizes the main transport patterns, synoptic conditions, and dynamic features in a graphic form (figure 6) and update the results published in the past two years. We also review the findings on intercontinental transport, which was not covered in the prior reviews.

### 5.1. Intercontinental and long-range transport

Several studies have investigated the impact of intercontinental transport on  $O_3$  in China [43–45]. Their results showed that the strength and location of the westerlies strongly influenced the transport pathway and seasonal variation of the amount of  $O_3$  transported to China. Using Goddard Earth Observing System (GEOS)-Chem model and measurement data from 31 locations, Ni *et al* [45] quantified the contributions of other continents to  $O_3$  in China in a spring season. Their simulations showed that Europe contributes 2.1–3.0 ppb to surface  $O_3$  over China's northern border and that North America contributes 0.9–2.7 ppb of surface  $O_3$  over most parts of China. These studies also indicated that intercontinental contributions are generally larger in the middle and upper troposphere than at surface.

It has been known that the transport of  $O_3$  between China and other Asian countries can be

influenced by the Asian monsoons [8, 9]. Long-range transport of increased emissions from Southeast Asia, under the influence of the Asian Summer Monsoons, has been suggested to contribute to increased  $O_3$  concentrations in remote or rural sites in western and southern China [23, 46, 47]. Anthropogenic emissions in Japan and Korea have also been shown in model studies to contribute to the  $O_3$  concentrations over China's eastern coasts. For example, GEOS-chem simulations indicated a contribution of 0.6–2.1 ppb in a spring season [45]. A recent analysis of a week-long  $O_3$  episode during autumn in eastern China showed that under the influence of eastward movement of the Mongolian high-pressure system, transport of  $O_3$  produced from emissions in Japan and Korea could contribute up to 30 ppb of  $O_3$  (or ~45%) at the peak, according to WRF-CAMQ simulations [48].

Stratosphere-troposphere exchange has been known to affect tropospheric  $O_3$  [8]. Recent studies have provided additional evidence to show that the stratosphere intrusion (SI) can be a source of  $O_3$  in the lower troposphere. Analysis of ozonesonde data showed that the elevated concentrations of surface  $O_3$  in Lhasa on the Tibetan Plateau were associated with the SI [49] and that the SI also affected the ground-level  $O_3$  in the PRD (Yangjiang and Hong Kong)



during some periods of a year [47, 50]. Model studies have further demonstrated the considerable role that the SI events played in O<sub>3</sub> pollution over eastern and southern China [49, 51–53].

## 5.2. Regional transport

The tropical cyclones (low-pressure systems) over the western pacific, continental anti-cyclones (high-pressure system) and the West Pacific Subtropical High are the typical synoptic conditions leading to the formation and transport of O<sub>3</sub> pollution in China [9]. Recent studies have provided additional result on their impact on O<sub>3</sub> pollution in central (Wuhan), eastern (YRD) and southern (PRD) China [54–62]. Smaller-scale circulations, such as sea–land breezes (SLBs) and mountain–valley breezes (MVBs), can strongly affect O<sub>3</sub> concentrations in the coastal and mountainous areas under weak synoptic forcing [3]. Several recent studies presented more cases to demonstrate the impact of the SLBs on O<sub>3</sub> concentrations in coastal areas of the Bohai Gulf [63], the East China Sea [64] and the South China Sea [60, 65]. MVBs have also been shown to strongly influence the diurnal variation in O<sub>3</sub> concentrations at Mt Waliguan, Mt Huang and Mt Tai Mo Shan in the western, central and southern China, respectively [66–69].

Recent model studies have quantified inter-regional transport [70–73]. The surface O<sub>3</sub> concentrations in the JJJ region were strongly influenced by transport from the province of Shandong, Henan, Jiangsu, and Anhui, with a collective contribution up to 36% of the O<sub>3</sub> concentrations in JJJ [74]. Similarly, emissions from non-YRD regions have been found to be the dominant contributor to O<sub>3</sub> pollution in the YRD, with the peak contribution of 63% [71, 75, 76]. In the PRD region, intercity transport played an important role, with Guangzhou and Foshan being the major sources of emissions [72, 77]. In SCB, downward transport of upper stratospheric air from the Tibetan Plateau was found to be a key contributor to ground-level O<sub>3</sub> pollution over the city clusters of the basin [73]. These studies highlight the importance of cross-boundary transport in developing O<sub>3</sub> mitigation strategies for a specific city/region.

## 6. Emerging chemical processes important to O<sub>3</sub> production in polluted regions

### 6.1. HONO and photolabile halogens

As discussed in section 1, any factors affecting the sources and sinks of OH or its analogues also affect the photochemical production of O<sub>3</sub>. In the past decade, the photolysis of HONO has gained increasing recognition as an important source of OH, not only in the morning but also throughout the day [3, 15, 78]. The results of limited studies that were conducted up to year of 2016 were given in a previous review [3].

In the past 5 years, the number of HONO studies in China has increased rapidly, allowing assessment of the HONO impact on OH and O<sub>3</sub> in more and diverse areas. The available studies thus far have shown very high daytime concentrations of HONO (on order of a few ppbs) in many urban and rural areas of China, in both summer and winter [79–88]. Calculations with observed HONO and O<sub>3</sub> concentrations show that the photolysis of HONO dominated the OH primary sources in the early morning and contributed 40%–50% at noon during photochemical episodes in southern China [89]. Similarly, the HONO photolysis dominated the OH primary sources at an agricultural site in the NCP, and was nearly five times of the production rates from O<sub>3</sub> photolysis after fertilization [87]. Chemistry-transport models (CTMs) incorporating known HONO sources have attempted to quantify the HONO contribution to O<sub>3</sub> concentrations [90–93]. Simulations with a Weather Research Forecast-Chemistry (WRF-Chem) model showed that HONO enhanced the O<sub>3</sub> peak concentrations by 4%–10% in the HK-PRD region of southern China during a summer O<sub>3</sub> episode [90]; another study using a WRF-CAMQ model showed 34% increase in regional O<sub>3</sub> concentrations in a heavy winter pollution episode in the same region [91]. In other regions of China, WRF-Chem simulations suggested monthly O<sub>3</sub> increase of 5%–44% in Beijing-Tianjin-Hebei region in August [92]. Another WRF-Chem study indicated 6%–13% increase in surface O<sub>3</sub> concentrations in the developed regions of eastern China in July [93]. Direct comparison of these model results is difficult due to the differences in the treatment of the HONO sources and other aspects of model configurations in these studies. Nonetheless, the results of these studies have demonstrated important impact of HONO as a source of OH on O<sub>3</sub> production in polluted regions.

The high HONO concentrations (>10 ppb) reported in China were mainly attributed to the heterogeneous reactions of NO<sub>2</sub> on ground and aerosol surfaces, and to the photolysis of nitrate (NO<sub>3</sub><sup>−</sup>) aerosols [91]. However, the kinetic parameters in the real atmospheric conditions remain uncertain (i.e. the NO<sub>2</sub> uptake coefficient, HONO production yields and NO<sub>3</sub><sup>−</sup> photolysis rates). Additionally, recent field observations have shown that agricultural fertilisation can increase the emissions of HONO from soil [87, 94, 95]. Most state-of-the-art air quality models cannot properly simulate this HONO source due to a lack of information on the key factors determining HONO emissions from fertilised soils, such as fertiliser types, and biotic and abiotic factors. A very recent study developed a parametrisation linking soil HONO emissions to three commonly used fertilisers in China, and the temperature and water-holding capacity of soil samples, and incorporation of this parameterization in a WRF-CMAQ model improved

simulations of HONO and increased regionally averaged  $O_3$  concentrations by 8% in the agriculture-intensive NCP [96].

Other radicals that can produce  $O_3$  are chlorine (Cl) or bromine (Br) atoms. Ambient  $ClNO_2$  is a potential source of Cl radical (and a reservoir for  $NO_2$ ). The early findings on the impact of  $ClNO_2$  observed in 2013–2014 in China using a box model were given in a previous review [3], which showed that Cl produced by the photolysis of  $ClNO_2$  increased daytime  $O_3$  production by up to 41% in top of the PBL of southern China during winter [97], and by up to 13% at a polluted rural site (Wangdu) in NCP [98]. Since then,  $ClNO_2$  has been measured in more sites/seasons across China [99] and more model studies have been conducted. Recent measurements during winter at several sites in northern China showed that  $ClNO_2$  concentrations can increase during daytime, leading to greater daytime  $O_3$  production [99]. A WRF-Chem model incorporating laboratory-determined heterogeneous production of  $ClNO_2$  was used to show that there was a 5%–6% increase in  $O_3$  concentrations within the PBL of NCP and the YRD region [93] in the summer of 2014. The same model has been used to evaluate the effect of the  $ClNO_2$  and HONO chemistry on the designation of the  $O_3$  formation regimes, and the result showed that the ‘new’ nitrogen chemistry changed the  $O_3$  sensitivity regime for nearly 40% of the simulated area with human influence in China, mainly from VOC-sensitive or  $NO_x$ -sensitive regimes to mixed-sensitive regime [100]. However, current parametrisations of  $ClNO_2$  production (including the uptake coefficient of nitrogen pentoxide ( $N_2O_5$ ) and the yields of  $ClNO_2$ ) are subject to large uncertainties and need further improvement [101, 102].

Molecular chlorine ( $Cl_2$ ) is another potentially important photolytic source of Cl.  $Cl_2$  mixing ratios of up to 400 ppt were observed at a polluted rural site in Wangdu in northern China during the summer of 2014, and corresponding box model calculations suggested that  $Cl_2$  and  $ClNO_2$  increased the  $O_3$  production rate by 19% [103]. A recent study during the coal-burning period of winter documented surprisingly high concentrations of bromine chloride (BrCl) and  $Cl_2$  at a site in Wangdu, and calculations from an observation-constrained model indicated that Cl and Br atoms collectively increased the total  $O_x$  ( $O_3 + NO_2$ ) mixing ratios at this site by 50% [104]. Another very recent study reported unprecedented levels of  $Cl_2$  (up to 1 ppb) at a polluted coastal site of Hong Kong, and such high concentrations of  $Cl_2$  increased  $O_x$  daytime production by 16% [105]. However, existing CTMs cannot account for observed daytime  $Cl_2$  or BrCl concentrations because they lack appropriate mechanistic and kinetic information. Nonetheless, several CTM studies have attempted to examine the impact of previously known chlorine sources on the atmospheric

oxidation capacity and  $O_3$  concentrations and suggested potential importance of halogen chemistry in the lower troposphere [106–109].

## 6.2. Active winter photochemistry in northern China

It is typically assumed that the photochemical processes that produce  $O_3$  are slow during winter in places like the NCP, owing to the substantially lower temperatures and levels of solar radiation during winter than in the warm seasons. However, several field studies in Beijing and its surrounding areas have indicated that photochemical processes could be rapid during polluted days in winter, which has been attributed to the presence of very high concentrations of  $HO_x$  precursors (i.e. HONO and oxygenated VOCs) and halogen atom precursors (BrCl and  $Cl_2$ ), and to the rapid recycling of OH due to high NO concentrations [104, 110–113]. Chemical models have suggested that rapid photochemical process was likely to occur throughout the NCP region and its surrounding areas [113, 114]. In these studies, the noontime rates of  $O_3$  production were 15–100 ppb  $h^{-1}$  under high  $NO_x$  conditions, which is comparable to those during the summer. However, the concentration of  $O_3$  was suppressed due to the simultaneous and rapid loss of  $O_3$  by NO titration, the subsequent loss of  $NO_2$  via gas-phase reactions with OH and the heterogeneous loss of  $N_2O_5$ , which led to the rapid production of nitric acid and secondary aerosols [112, 114]. Thus, decreasing emissions VOCs would slow the winter photochemical processes that generate  $O_3$  and thereby help to alleviate the severity of the winter haze covering Beijing and a large part of the NCP.

## 7. Factors causing increased urban $O_3$ concentrations since 2013

### 7.1. Emissions change in 2013–2017

In 2013, the Chinese government implemented the Air Pollution Prevention and Control Action Plan to mitigate the severe haze in many parts of the country, and consequently, anthropogenic emissions of sulphur dioxide ( $SO_2$ ),  $NO_x$ , CO and 2.5  $\mu m$  or smaller particulate matter ( $PM_{2.5}$ ) in China were reduced by 59%, 21%, 23%, and 33% from 2013 to 2017, respectively [4]. However, the emissions of VOCs and ammonia increased slightly in the same period [26]. Data from national environmental monitoring stations has shown corresponding decreases in the ambient concentrations of  $SO_2$ ,  $NO_x$  and PM, but increases in ground-level  $O_3$  concentrations in numerous urban areas [1, 6, 115, 116] (see figures 4 and 5).

### 7.2. The effect of meteorology change

CTMs have been used to quantify the roles of meteorological variations and emission changes in

the  $O_3$  increase during 2013–2017. To determine the contribution from the meteorology, the model mainly compares the  $O_3$  concentrations from simulations with a fixed emission but with the meteorology data of different years. For example, Wang *et al* [117] applied a WRF-CMAQ to evaluate the impact of meteorological variations on summer  $O_3$  concentrations over China from 2013 to 2015. Lu *et al* [118] used a GEOS-Chem model to explore the meteorological factors contributing to changes in  $O_3$  concentrations over China in 2016 and 2017. Liu and Wang [116] performed comprehensive simulations (across multiple years, and for combined and individual meteorological factors) to investigate the effect of meteorological factors on summer trends in  $O_3$  concentrations across China from 2013 to 2017, which showed that the meteorological impacts on  $O_3$  concentrations tended to vary regionally. This study showed that changes in meteorological conditions since 2013 have made relatively little contribution to the increased  $O_3$  concentrations in Beijing compared to the effects of emissions, but have made relatively larger contributions (either positive or negative) than emissions to the increased  $O_3$  concentrations in Shanghai and Guangzhou for some years from 2014 to 2017. Meteorological changes, including those affecting in the long-range transport of  $O_3$  to China from places outside China, were the predominant cause of the increase in  $O_3$  concentrations in the western Qinghai–Tibet Plateau during 2013–2017 [116]. The above findings from various studies highlight the important roles of meteorological factors in determining inter-annual variations in summer  $O_3$  concentrations, and the necessity of considering meteorological effects when evaluating the efficacy of emission control measures.

### 7.3. The effect of emission change

Several CTMs studies have attempted to evaluate the contribution of emission change to the recent increase in  $O_3$  concentrations in China, with varied approaches. For instance, using the CMAQ model, Wang *et al* [117] concluded that variation in MDA8  $O_3$  concentrations was mainly attributed to emission changes in 2013–2015. They obtained this result by subtracting the simulated change in MDA8  $O_3$  concentrations due to meteorology from the total observed change. Their results also showed that the increase in  $O_3$  concentrations from 2014 to 2015 relative to 2013 was mainly due to changes in emissions. However, inter-annual emissions were not explicitly accounted for in their simulations of the effects of emission changes on  $O_3$  concentrations. In other work, Li *et al* [119] used the GEOS-Chem model to simulate the effects of changes in ambient  $PM_{2.5}$  concentrations and anthropogenic emissions of  $O_3$  precursors ( $NO_x$  and VOCs) on MDA8  $O_3$  concentrations in 2013 and 2017. Their results indicated that the large decrease in  $PM_{2.5}$  concentrations ( $\sim 40\%$ )

reduced the uptake of  $HO_2$  on aerosol surfaces, leading to an increase in  $O_3$  concentrations in the NCP. Wang *et al* [41] used the WRF-CMAQ model to simulate the effect of a reduction in  $NO_x$  emissions from 2012 to 2016. Their results suggested that a decrease in  $NO_x$  emissions was the main reason for the increase in ground-level  $O_3$  concentrations in urban areas in eastern China during this period. Liu and Wang [115] used a WRF-CMAQ model, which was modified to include key heterogeneous reactions, to evaluate the impact of changes in the emissions of multiple and individual anthropogenic pollutants during 2013 and 2017 on the ground-level  $O_3$  concentrations in China. Their results showed that decreased emissions of  $NO_x$ , primary PM and  $SO_2$ , and increased emissions of VOCs, were the main cause of the increased urban  $O_3$  concentrations in China. However, the nature of these effects varied between regions. For example, in Beijing (in the north) and Chengdu (in the central west), large reductions in primary PM and  $SO_2$  emissions were the dominant drivers of the increase in  $O_3$  concentrations than the reduction of  $NO_x$  emissions. In contrast, in Shanghai (in the central east) and Guangzhou (in the south), the decrease in  $NO_x$  emission and increase in VOC emission were the dominant drivers of increased  $O_3$  concentrations [115]. The same study also highlighted the importance of the uptake of  $HO_2$  and  $O_3$  by PM in driving changes in  $O_3$  concentrations.

A key policy-relevant conclusion that can be drawn from the above studies is that while the nationwide control measures from 2013 to 2017 have successfully reduced emissions of primary pollutants, they have also led to increased  $O_3$  concentrations in urban areas of China, due to the non-linear dependence of  $O_3$  on  $NO_x$  and aerosol feedbacks. Therefore, the control of VOC emissions should be included in future  $O_3$  control strategies. A positive outcome of these control measures for  $O_3$  is that  $O_3$  concentrations in rural areas have been reduced according to model simulations [116] and observations at several non-urban sites [24], which can be attributed to  $NO_x$ -limited  $O_3$  formation regime that exists in rural areas [23, 116].

## 8. Effects of China's COVID-19 lockdown on ground-level $O_3$ concentrations

The outbreak of coronavirus disease 2019 (COVID-19) pandemic in 2019, and attempts to control it, have severely impacted human activities worldwide. China was the first country that imposed nationwide measures (from 23 January to 13 February 2020) that aimed to prevent the spread of severe acute respiratory syndrome coronavirus 2, which causes COVID-19. These various restrictions drastically reduced emissions of air pollutants [120, 121], which offered an unprecedented opportunity to

investigating the effects of large and nationwide decreases in emissions on O<sub>3</sub> concentrations. A number of studies have analysed data on ground-level O<sub>3</sub> concentrations based on data from China National Environmental Monitoring Center network, using different methodologies. Some directly compared ambient O<sub>3</sub> concentrations during the COVID-19 lockdown period to those during pre-lockdown periods (in the same year or in previous years), whereas others have combined statistical transport models and CTMs to separate the effects of emission reductions and meteorological factors on O<sub>3</sub> concentrations. The emissions of air pollutants during the COVID-19 lockdown period have been estimated based on activity data or estimates (that were available at the time) for the transportation and power-generation industries and the residential sector [120–122]. The reductions in emissions were found to vary between regions, and were estimated to be ~45%–54% for NO<sub>x</sub> and ~22%–43% for VOCs in northern, central and southern China, with larger decreases in VOCs in the south [122].

Most studies of changes in O<sub>3</sub> concentrations during the COVID-19 lockdown have focused on the increases in ground-level O<sub>3</sub> concentrations that occurred in many of the urban areas in northern and central China during their lockdown period amid large decreases in the concentrations of primary pollutants such as NO<sub>2</sub> and SO<sub>2</sub> in the same areas [123–125]. A few studies have also investigated the decrease in O<sub>3</sub> concentrations in southern provinces such as Guangdong and Guangxi [122, 126]. Studies using models and data on the estimated emission reductions during the COVID-19 lockdown period have found that meteorological factors had strong (or stronger than emissions) effects on O<sub>3</sub> concentrations during this period [122, 125]. These findings re-affirm the critical role of meteorological factors in driving short-term air quality, as proposed in many previous studies on air-quality changes subsequent to short-term reductions in emissions [122]. The model simulations have attributed increases or decreases in O<sub>3</sub> concentrations to the non-linear relationship between O<sub>3</sub> concentrations and those of its precursors, which led to an increase in O<sub>3</sub> concentrations in most cities due to O<sub>3</sub> titration by NO<sub>x</sub> under NO<sub>x</sub>-saturated or VOC-limited conditions, and a decrease in O<sub>3</sub> concentrations in the south due to the lower NO<sub>x</sub> to VOC ratios there. Model-calculated indicators of the O<sub>3</sub>-formation regime (i.e. the ratio of the production rate of H<sub>2</sub>O<sub>2</sub> to that of HNO<sub>3</sub>) suggest that most of the populated and industrialised parts of the NCP and YRD, and the core of the PRD remained a typical VOC-limited O<sub>3</sub>-formation regime during the COVID-19 lockdown, but other large parts of these regions transitioned to a mixed regime. The reduction in VOC emissions in northern China during the COVID-19 lockdown were insufficient to counteract the increase in O<sub>3</sub> concentrations

due to decreased NO<sub>x</sub> titration [122]. Moreover, the increased O<sub>3</sub> concentrations in many northern cities could have enhanced the oxidative capacity of the atmosphere and consequently the production of secondary aerosols in the winter haze in these cities, such as Beijing [121].

## 9. Impacts of O<sub>3</sub> on crops and trees

There was limited research on crop responses to O<sub>3</sub> in China in the earlier years, but a rapid increase in studies on rice and wheat and those involving experimental and model investigations after 2008. A recent review highlighted the magnitudes of crop yield loss and tree biomass reduction across China or some regions of China due to rising O<sub>3</sub> concentration [11]. Currently, there has been a general consensus that O<sub>3</sub> pollution in China has substantially decreased crop yields [11, 127–130]. A very recent assessment, which is based on the latest O<sub>3</sub> dose-yield response relationship obtained from the Asian field experiments and hourly O<sub>3</sub> concentrations in 2018–2020 at 1400 air quality stations in China, suggests that the ambient O<sub>3</sub> concentration in China has caused the national yield loss of wheat, hybrid rice, inbred rice and maize by 32.8%, 29.8%, 12.2% and 8.6%, respectively [131]. These new estimates of crop yield loss were a bit higher than the previous studies, in which O<sub>3</sub> dose response relationship was based on the manipulation experiment from one or two sites [127–130]. While numerous studies have focused on crop yield [11], little is known about how O<sub>3</sub> pollution affects grain quality in China. The few available studies on grain quality have largely focused on wheat or rice. O<sub>3</sub> pollution has been shown to affect the starch content and composition of grain [132, 133], which influences grain processing, appearance and cooking quality. A study on Chinese hybrid rice cultivars found that decreased grain cooking quality and increased proportions of chalky and cracked kernels are associated with high O<sub>3</sub> concentrations [134]. In addition, elevated O<sub>3</sub> concentrations have been found to lead to decreases in the total grain content of nutrients per plant, but have also been found to increase grain concentrations of protein and mineral nutrients (e.g. potassium, calcium and manganese) [135, 136]. This increase in grain nutrient concentrations is related to advanced grain filling or the ‘concentration effects’ of yield loss under elevated O<sub>3</sub> concentrations [136, 137]. These findings suggest that although O<sub>3</sub> pollution can improve grain nutrient quality in some cases, it generally leads to a deterioration of grain quality.

Ground-level O<sub>3</sub> pollution has also been shown to affect the health of Chinese forests. It has been suggested that over 98% of forested areas in China are threatened by O<sub>3</sub> pollution [138]. However, only a few studies have estimated the impacts of O<sub>3</sub> on Chinese forest productivity. Northern temperate



forests in China are exposed to relatively higher O<sub>3</sub> concentrations than sub-tropical forests [139]. However, as tropical and subtropical evergreen forests have longer growing seasons (and are thus exposed to O<sub>3</sub> for longer durations) but are more resistant to O<sub>3</sub> than temperate deciduous forests, O<sub>3</sub> pollution was found to reduce tree biomass by similar amounts in these two forest types (13% vs. 11%) [138]. In addition, broadleaved forests have generally been shown to be more vulnerable to elevated O<sub>3</sub> concentrations than needle-leaved forests [140], probably due to the lower O<sub>3</sub> uptake per unit leaf mass by thicker or denser leaves [141].

The ratio of photosynthetic carbon uptake to water vapour loss, or water-use efficiency (WUE), is a key forest function that is commonly used to describe the coupling of the carbon and water cycles in terrestrial ecosystems. Evidence from studies on individual plants suggests that elevated O<sub>3</sub> concentrations can influence photosynthetic CO<sub>2</sub> assimilation and stomatal conductance, thereby ultimately altering WUE [142, 143].

## 10. Government regulations and efforts to control O<sub>3</sub>

To improve regional air quality, the Chinese central government has enacted a series of long-term regulations including the Air Pollution Prevention and Control Action Plan (2013–2017) and the Blue Sky Protection Campaign (2018–2020) ([www.mee.gov.cn](http://www.mee.gov.cn), accessed 1 December 2021). These regulations have set specific targets for reducing emissions and/or ambient PM<sub>2.5</sub> concentrations, or the number of heavy pollution days. The regulations in 2018 mandated a 15% reduction in the emissions of NO<sub>x</sub> and SO<sub>2</sub> and a 10% reduction in the emissions of VOC in 2020, relative the emissions in 2015. The implementation of these regulations in various economic sectors has led to significant decreases in the concentrations of PMs (including PM<sub>2.5</sub> and PM<sub>10</sub>) and other routinely monitored pollutants such as NO<sub>x</sub>, CO, and SO<sub>2</sub> [26, 115, 144], but have not been effective in reducing O<sub>3</sub> pollution. This is in part because previous control measures mainly aimed to alleviate the notorious haze or PM<sub>2.5</sub> pollution in China (especially in northern China during the winter), but assigned a lower priority to reductions in O<sub>3</sub> concentrations. Nonetheless, during some important large-scale events held in China, such as the 2016 Group of Twenty (G20) Summit in Hangzhou, specific measures to reduce NO<sub>x</sub> and VOCs emissions were implemented to reduce O<sub>3</sub> pollution, and the responses of ambient O<sub>3</sub> concentrations to these reductions varied—decreased in some areas but increased others [122, 145].

In its 14th Five-Year Plan for 2021–2025, China has identified VOC emission management as a major target, in parallel with reductions in NO<sub>x</sub>

emissions ([www.mee.gov.cn](http://www.mee.gov.cn), accessed 1 December 2021). Specifically, as part of the O<sub>3</sub> Pollution Prevention and Control Action Campaign, which was launched by the Ministry of Ecology and Environment (MEE) to mitigate increasing summertime O<sub>3</sub> pollution, VOC emission reduction measures were implemented in many cities between June and September in 2020 and 2021. Inspection teams led by officials from MEE, Provincial and City's Bureau of Ecology and Environment and assisted by scientists, visited major VOC-emitting industries (such as petrochemical, chemical, solvent, packaging and fuel storage industries), and helped them to find ways to reduce their VOC emissions. The results from ambient monitoring suggest that these measures were effective, and that O<sub>3</sub> concentrations decreased in the summer of 2020 and 2021 in major cities, compared to previous years (figure 4). This was especially true for Jing-Jin-Ji and its surrounding regions. It is expected that such top-down enforcement will continue in the coming years. It is important that these somewhat ad hoc approaches are developed into a more systematic and clearly formulated long-term effort.

## 11. Summary and recommendations

In this work, we have synthesised the findings of studies on the O<sub>3</sub> situation in China that have been published in the past six years. The main findings are summarised below.

- Many studies have used the data released by the China National Environmental Monitoring Center network (since in 2013) to investigate the temporal variations (i.e. inter-annual, seasonal and episodic) and spatial distribution of O<sub>3</sub> in China. O<sub>3</sub> concentrations in many urban and sub-urban areas have increased since 2013, but appear to have stabilised in the most recent two years. The highest O<sub>3</sub> concentrations occur in the vast central-eastern plain region (which has megacities such as Beijing in the north and Shanghai in the south, and major industries and intensive agricultural activities), the Pearl River Delta (in the southern coast), the Fenwei plain (in central China) and in several other isolated cities. Limited long-term observations (>10 years) at several rural sites suggest that there have not been significant increases in O<sub>3</sub> concentrations in rural areas in the past decade.
- Despite the continuous reductions in NO<sub>x</sub> emissions, observation- and emission-based studies indicate that major urban areas of China remain under VOC-limited O<sub>3</sub> formation regimes, although some sites can be under VOC–NO<sub>x</sub> co-limited or NO<sub>x</sub>-limited O<sub>3</sub>-formation regimes during certain periods. These results highlight the necessity of joint NO<sub>x</sub>–VOC controls to alleviate O<sub>3</sub> pollution.

- A number of studies have assessed the atmospheric transport of O<sub>3</sub> and its precursors in different regions of China. These studies have highlighted the important roles of various scales of transport, including inter-regional transport between eastern China and the PRD, between the YRD and the Jing–Jin–Ji region, and intraregional (or inter-city) transport within these populated and industrialised regions. These results re-affirm the need for cooperation between regional governments to reduce O<sub>3</sub> pollution.
- The drivers of the increase in urban O<sub>3</sub> concentrations from 2013 to 2019 have been investigated in numerous studies. Their findings suggest that meteorological factors play an important role in modulating inter-annual variations in O<sub>3</sub> concentrations, and that reductions in the emissions of NO<sub>x</sub>, SO<sub>2</sub> and PM have complex effects on ground-level O<sub>3</sub> concentrations. The studies have also suggested that control of VOC emission could have prevented the increase in O<sub>3</sub> concentrations. Similarly, studies on the response of O<sub>3</sub> concentrations to the COVID-19 lockdown have underscored the complex relationships between O<sub>3</sub> and primary pollutants, and the critical effect of meteorological factors on short-term changes in O<sub>3</sub> concentrations.
- There have been several advances on the chemical processes important to O<sub>3</sub> production in the past few years. These include improved understanding of the roles of HONO and ClNO<sub>2</sub> and discovery of the important roles of other reactive halogen compounds (Cl<sub>2</sub> and BrCl), the aerosol uptake of radicals and reactive gases, and the activity of photochemical processes in winter. These findings have refined knowledge on classic O<sub>3</sub> formation mechanisms in high NO<sub>x</sub> and high PM environments.
- The past five years have also seen the publication of many studies on the impacts of O<sub>3</sub> on vegetation. Their findings provide strong scientific evidence that confirms the negative impacts of O<sub>3</sub> on the crop yields and grain quality. The available studies have also shown that deciduous species are at more risk to O<sub>3</sub> than evergreen species, and increased O<sub>3</sub> concentrations can affect water use of trees.
- In terms of policy, although previous control regulations and measures focused on PM<sub>2.5</sub>, those in the past two years have paid increasing attention to O<sub>3</sub>. The enforced control of VOC emissions in the two most recent summers has had the desired effect on ambient O<sub>3</sub> concentrations, which appear to show signs of decreasing in many O<sub>3</sub>-affected cities.

We would like to provide the following recommendations for future research and policy development.

- Data on long-term ambient VOC compositions in different regions of China is needed. While

China National Environmental Monitoring Center network has provided unprecedented data on the concentrations of O<sub>3</sub>, NO<sub>2</sub> and other regulated pollutants since 2013, there remains a lack of data on the concentrations and composition of ambient VOCs. Despite the reported establishment of dozens of VOC measurement sites across the country, their data have not been made available. This impedes efforts to assess the roles of VOCs in O<sub>3</sub> formation and to validate VOC emission inventories.

- More research is needed on the role of newly discovered chemical processes in O<sub>3</sub> formation, given the rapid changes that will occur in chemical environments in future years (i.e. the expected large decrease in the emissions of NO<sub>x</sub> and VOCs, and the continuing reductions in the emissions of PM<sub>2.5</sub>). Air quality models should incorporate these processes to enable more accurate predictions.
- More research is needed to quantify the complex relationship between O<sub>3</sub> and PM<sub>2.5</sub> in different regions. This will help support the development of a co-control strategy for O<sub>3</sub> and PM<sub>2.5</sub>. Research for future O<sub>3</sub>-control policies should consider the expected transformative changes in energy production and transportation industries in response to the national commitment to peak carbon emission in 2030 and carbon neutrality by 2060.
- More O<sub>3</sub> monitors need to be installed in the countryside and at forest reserves to facilitate scientific assessments on the ecological impacts of O<sub>3</sub>, and the contribution of BVOC emissions to O<sub>3</sub> generation.
- More research is needed to develop measures to reduce the negative effects of O<sub>3</sub> on plants. Such measures could include protectants, management of water use and the breeding of O<sub>3</sub>-tolerant cultivars or species.

## Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

## Acknowledgments

T W, J D, and Y T are supported by the National Natural Science Foundation (NSFC, Project No. 91844301) and the Hong Kong Research Grants Council (Project No. T24-504/17-N). L X and Y Z are supported by the NSFC (Project No. 41922051) and the Shandong Provincial Science Foundation for Distinguished Young Scholars (ZR2019JQ09). Z F is supported by the NSFC (Project No. 42130714).

## Author contributions

T W coordinated the review. Section 1: T W; section 2: J D, Y Z, and T W; section 3: T W; section 4: Y Z, Y T, L X, and T W; section 5: J D and T W; sections 6–8: T W; section 9: Z F; section 10: L X, Y Z, and T W; section 11: T W, Z F, and L X. All authors commented on the full manuscript.

## ORCID iD

Tao Wang  <https://orcid.org/0000-0002-4765-9377>

## References

- [1] Lu X *et al* 2018 Severe surface ozone pollution in China: a global perspective *Environ. Sci. Technol. Lett.* **5** 487–94
- [2] Lefohn A S *et al* 2018 Tropospheric ozone assessment report: global ozone metrics for climate change, human health, and crop/ecosystem research *Elementa* **6** 27
- [3] Wang T, Xue L K, Brimblecombe P, Lam Y F, Li L and Zhang L 2017 Ozone pollution in China: a review of concentrations, meteorological influences, chemical precursors, and effects *Sci. Total Environ.* **575** 1582–96
- [4] Zhang Q *et al* 2019 Drivers of improved PM<sub>2.5</sub> air quality in China from 2013 to 2017 *Proc. Natl Acad. Sci. USA* **116** 24463–9
- [5] Lu X, Zhang L, Wang X L, Gao M, Li K, Zhang Y Z, Yue X and Zhang Y H 2020 Rapid increases in warm-season surface ozone and resulting health impact in China since 2013 *Environ. Sci. Technol. Lett.* **7** 240–7
- [6] Wang Y *et al* 2020 Contrasting trends of PM<sub>2.5</sub> and surface-ozone concentrations in China from 2013 to 2017 *Natl Sci. Rev.* **7** 1331–9
- [7] Lu H, Lyu X, Cheng H, Ling Z and Guo H 2019 Overview on the spatial-temporal characteristics of the ozone formation regime in China *Environ. Sci. Process. Impacts* **21** 916–29
- [8] Lu X, Zhang L and Shen L 2019 Meteorology and climate influences on tropospheric ozone: a review of natural sources, chemistry, and transport patterns *Curr. Pollut. Rep.* **5** 238–60
- [9] Fu Y, Liao H and Yang Y 2019 Interannual and decadal changes in tropospheric ozone in china and the associated chemistry-climate interactions: a review *Adv. Atmos. Sci.* **36** 975–93
- [10] Liu H, Zhang M and Han X 2020 A review of surface ozone source apportionment in China *Atmos. Ocean. Sci. Lett.* **13** 470–84
- [11] Xu X 2021 Recent advances in studies of ozone pollution and impacts in China: a short review *Curr. Opin. Environ. Sci. Health* **19** 100225
- [12] Wang T, Ding A, Gao J and Wu W S 2006 Strong ozone production in urban plumes from Beijing, China *Geophys. Res. Lett.* **33** 21
- [13] Shao M, Tang X Y, Zhang Y H and Li W J 2006 City clusters in China: air and surface water pollution *Front. Ecol. Environ.* **4** 353–61
- [14] Monks P S *et al* 2015 Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer *Atmos. Chem. Phys.* **15** 8889–973
- [15] Archibald A T *et al* 2020 Tropospheric ozone assessment report *Elementa* **8** 1
- [16] National Research Council 1992 *Rethinking the Ozone Problem in Urban and Regional Air Pollution* (Washington, DC: National Academies Press)
- [17] NARSTO 2000 *An Assessment of Tropospheric Ozone Pollution: A North American Perspective* (Pasco, WA: North American Research Strategy for Tropospheric Ozone)
- [18] Hidy G 2000 Ozone process insights from field experiments—part I: overview *Atmos. Environ.* **34** 2001–22
- [19] Jenkin M E and Clemitshaw K C 2000 Ozone and other secondary photochemical pollutants: chemical processes governing their formation in the planetary boundary layer *Atmos. Environ.* **34** 2499–527
- [20] Kleinman L I 2000 Ozone process insights from field experiments—part II: observation-based analysis for ozone production *Atmos. Environ.* **34** 2023–33
- [21] Solomon P, Cowling E, Hidy G and Furiness C 2000 Comparison of scientific findings from major ozone field studies in North America and Europe *Atmos. Environ.* **34** 1885–920
- [22] Jacob D J 2000 Heterogeneous chemistry and tropospheric ozone *Atmos. Environ.* **34** 2131–59
- [23] Wang T, Dai J, Lam K S, Nan Poon C and Brasseur G P 2019 Twenty-five years of lower tropospheric ozone observations in tropical East Asia: the influence of emissions and weather patterns *Geophys. Res. Lett.* **46** 11463–70
- [24] Xu X B *et al* 2020 Long-term changes of regional ozone in China: implications for human health and ecosystem impacts *Elementa* **8** 13
- [25] Li M *et al* 2019 Persistent growth of anthropogenic non-methane volatile organic compound (NMVOC) emissions in China during 1990–2017: drivers, speciation and ozone formation potential *Atmos. Chem. Phys.* **19** 8897–913
- [26] Zheng B *et al* 2018 Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions *Atmos. Chem. Phys.* **18** 14095–111
- [27] National Bureau of Statistics 1991–2019 *China Statistical Yearbooks* (Beijing: Statistical Press)
- [28] Lin H, Wang M, Duan Y, Fu Q, Ji W, Cui H, Jin D, Lin Y and Hu K 2020 O<sub>3</sub> sensitivity and contributions of different NMHC sources in O<sub>3</sub> formation at urban and suburban sites in Shanghai *Atmosphere* **11** 295
- [29] Wang M, Chen W, Zhang L, Qin W, Zhang Y, Zhang X and Xie X 2020 Ozone pollution characteristics and sensitivity analysis using an observation-based model in Nanjing, Yangtze River Delta Region of China *J. Environ. Sci.* **93** 13–22
- [30] Zhu J, Cheng H, Peng J, Zeng P, Wang Z, Lyu X and Guo H 2020 O<sub>3</sub> photochemistry on O<sub>3</sub> episode days and non-O<sub>3</sub> episode days in Wuhan, Central China *Atmos. Environ.* **223** 117236
- [31] Chen T *et al* 2020 Volatile organic compounds and ozone air pollution in an oil production region in northern China *Atmos. Chem. Phys.* **20** 7069–86
- [32] Huang W, Zhao Q, Liu Q, Chen F, He Z, Guo H and Ling Z 2020 Assessment of atmospheric photochemical reactivity in the Yangtze River Delta using a photochemical box model *Atmos. Res.* **245** 105088
- [33] Shen H *et al* 2021 Significance of carbonyl compounds to photochemical ozone formation in a coastal city (Shantou) in eastern China *Sci. Total Environ.* **764** 144031
- [34] Hui L, Ma T, Gao J, Wang Z, Xue L, Liu H and Liu J 2021 Characteristics and sources of volatile organic compounds during high ozone episodes: a case study at a site in the eastern Guanzhong Plain, China *Chemosphere* **265** 129072
- [35] Xue L K *et al* 2014 Ground-level ozone in four Chinese cities: precursors, regional transport and heterogeneous processes *Atmos. Chem. Phys.* **14** 13175–88
- [36] Benish S E *et al* 2020 Measurement report: aircraft observations of ozone, nitrogen oxides, and volatile organic compounds over Hebei Province, China *Atmos. Chem. Phys.* **20** 14523–45
- [37] Wang Z, Wang H, Zhang L, Guo J, Li Z, Wu K, Zhu G, Hou D, Su H and Sun Z 2021 Characteristics of volatile organic compounds (VOCs) based on multisite

- observations in Hebei province in the warm season in 2019 *Atmos. Environ.* **256** 118435
- [38] Tan Z, Lu K, Jiang M, Su R, Dong H, Zeng L, Xie S, Tan Q and Zhang Y 2018 Exploring ozone pollution in Chengdu, southwestern China: a case study from radical chemistry to O<sub>3</sub>-VOC-NO<sub>x</sub> sensitivity *Sci. Total Environ.* **636** 775–86
- [39] Zhao Y, Chen L, Li K, Han L, Zhang X, Wu X, Gao X, Azzi M and Cen K 2020 Atmospheric ozone chemistry and control strategies in Hangzhou, China: application of a 0-D box model *Atmos. Res.* **246** 105109
- [40] Yu D *et al* 2020 An explicit study of local ozone budget and NO<sub>x</sub>-VOCs sensitivity in Shenzhen China *Atmos. Environ.* **224** 117304
- [41] Wang N, Lyu X P, Deng X J, Huang X, Jiang F and Ding A J 2019 Aggravating O<sub>3</sub> pollution due to NO<sub>x</sub> emission control in eastern China *Sci. Total Environ.* **677** 732–44
- [42] Cheng N *et al* 2019 Ground ozone variations at an urban and a rural station in Beijing from 2006 to 2017: trend, meteorological influences and formation regimes *J. Clean. Prod.* **235** 11–20
- [43] Zhu Y, Liu J, Wang T, Zhuang B, Han H, Wang H, Chang Y and Ding K 2017 The impacts of meteorology on the seasonal and interannual variabilities of ozone transport from North America to East Asia *J. Geophys. Res. Atmos.* **122** 10,612–10,36
- [44] Han H, Liu J, Yuan H, Zhuang B, Zhu Y, Wu Y, Yan Y and Ding A 2018 Characteristics of intercontinental transport of tropospheric ozone from Africa to Asia *Atmos. Chem. Phys.* **18** 4251–76
- [45] Ni R, Lin J, Yan Y and Lin W 2018 Foreign and domestic contributions to springtime ozone over China *Atmos. Chem. Phys.* **18** 11447–69
- [46] Xu W, Xu X, Lin M, Lin W, Tarasick D, Tang J, Ma J and Zheng X 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 *Atmos. Chem. Phys.* **18** 773–98
- [47] Liao Z H, Ling Z H, Gao M, Sun J R, Zhao W, Ma P K, Quan J N and Fan S J 2021 Tropospheric ozone variability over Hong Kong based on recent 20 years (2000–2019) ozonesonde observation *J. Geophys. Res. Atmos.* **126** 3
- [48] Zheng Y, Jiang F, Feng S, Cai Z, Shen Y, Ying C, Wang X and Liu Q 2021 Long-range transport of ozone across the eastern China seas: a case study in coastal cities in southeastern China *Sci. Total Environ.* **768** 144520
- [49] Jiang Y, Zhao T, Liu J, Xu X, Tan C, Cheng X, Bi X, Gan J, You J and Zhao S 2015 Why does surface ozone peak before a typhoon landing in southeast China? *Atmos. Chem. Phys.* **15** 13331–8
- [50] Zhang W *et al* 2021 Characteristics of the vertical distribution of tropospheric ozone in late autumn at Yangjiang station in Pearl River Delta (PRD), China. Part I: observed event *Atmos. Environ.* **244** 117898
- [51] Luo J, Liang W, Xu P, Xue H, Zhang M, Shang L and Tian H 2019 Seasonal features and a case study of tropopause folds over the Tibetan Plateau *Adv. Meteorol.* **2019** 1–12
- [52] Wang Y, Wang H and Wang W 2020 A stratospheric intrusion-influenced ozone pollution episode associated with an intense horizontal-trough event *Atmosphere* **11** 164
- [53] Zhao K, Hu C, Yuan Z, Xu D, Zhang S, Luo H, Wang J and Jiang R 2021 A modeling study of the impact of stratospheric intrusion on ozone enhancement in the lower troposphere over the Hong Kong regions, China *Atmos. Res.* **247** 105158
- [54] Jiang Z, Li J, Lu X, Gong C, Zhang L and Liao H 2021 Impact of western Pacific subtropical high on ozone pollution over eastern China *Atmos. Chem. Phys.* **21** 2601–13
- [55] Zhao Z and Wang Y 2017 Influence of the West Pacific subtropical high on surface ozone daily variability in summertime over eastern China *Atmos. Environ.* **170** 197–204
- [56] Yin Z, Cao B and Wang H 2019 Dominant patterns of summer ozone pollution in eastern China and associated atmospheric circulations *Atmos. Chem. Phys.* **19** 13933–43
- [57] Mao J, Wang L, Lu C, Liu J, Li M, Tang G, Ji D, Zhang N and Wang Y 2020 Meteorological mechanism for a large-scale persistent severe ozone pollution event over eastern China in 2017 *J. Environ. Sci.* **92** 187–99
- [58] Dong Y, Li J, Guo J, Jiang Z, Chu Y, Chang L, Yang Y and Liao H 2020 The impact of synoptic patterns on summertime ozone pollution in the North China Plain *Sci. Total Environ.* **735** 139559
- [59] Shu L, Xie M, Wang T, Gao D, Chen P, Han Y, Li S, Zhuang B and Li M 2016 Integrated studies of a regional ozone pollution synthetically affected by subtropical high and typhoon system in the Yangtze River Delta region, China *Atmos. Chem. Phys.* **16** 15801–19
- [60] Wang H, Lyu G H, Wang Y, Zou S, Ling Z, Xinming W, Jiang F and Zeren Y 2018 Ozone pollution around a coastal region of south China sea: interaction between marine and continental air *Atmos. Chem. Phys.* **18** 6
- [61] Zeng P, Lyu X P, Guo H, Cheng H R, Jiang F, Pan W Z, Wang Z W, Liang S W and Hu Y Q 2018 Causes of ozone pollution in summer in Wuhan, Central China *Environ. Pollut.* **241** 852–61
- [62] Xu Z, Huang X, Nie W, Chi X, Xu Z, Zheng L, Sun P and Ding A 2017 Influence of synoptic condition and holiday effects on VOCs and ozone production in the Yangtze River Delta region, China *Atmos. Environ.* **168** 112–24
- [63] Bei N, Zhao L, Jiarui W, Xia L and Tian F 2018 Impacts of sea-land and mountain-valley circulations on the air pollution in Beijing-Tianjin-Hebei (BTH): a case study *Environ. Pollut.* **234** 429–38
- [64] Xu J, Huang X, Wang N, Li Y and Ding A 2020 Understanding ozone pollution in the Yangtze River Delta of eastern China from the perspective of diurnal cycles *Sci. Total Environ.* **752** 141928
- [65] Wang Y, Guo H, Lyu X, Zhang L, Zeren Y, Zou S and Ling Z 2019 Photochemical evolution of continental air masses and their influence on ozone formation over the South China Sea *Sci. Total Environ.* **673** 424–34
- [66] Wang N, Guo H, Jiang F, Ling Z and Wang T 2015 Simulation of ozone formation at different elevations in mountainous area of Hong Kong using WRF-CMAQ model *Sci. Total Environ.* **505** 939–51
- [67] Zhang L, Jin L, Zhao T, Yin Y, Zhu B, Shan Y, Guo X, Tan C, Gao J and Wang H 2015 Diurnal variation of surface ozone in mountainous areas: case study of Mt. Huang, East China *Sci. Total Environ.* **538** 583–90
- [68] Xu W Y *et al* 2015 Long-term trends of surface ozone and its influencing factors at the Mt. Waliguan GAW station, China—part 1: overall trends and characteristics *Atmos. Chem. Phys.* **16** 30987–1024
- [69] Gao J, Zhu B, Xiao H, Kang H and Miao Q 2017 Diurnal variations and source apportionment of ozone at the summit of Mount Huang, a rural site in Eastern China *Environ. Pollut.* **222** 513–22
- [70] Han X, Zhu L, Wang S, Meng X, Zhang M and Hu J 2018 Modeling study of impacts on surface ozone of regional transport and emissions reductions over North China Plain in summer 2015 *Atmos. Chem. Phys.* **18** 12207–21
- [71] Shu L, Wang T, Xie M, Li M, Zhao M, Zhang M and Zhao X 2019 Episode study of fine particle and ozone during the CAPUM-YRD over Yangtze River Delta of China: characteristics and source attribution *Atmos. Environ.* **203** 87–101
- [72] Yang L F, Luo H H, Yuan Z B, Zheng J Y, Huang Z J, Li C, Lin X H, Louie P K K, Chen D H and Bian Y H 2019 Quantitative impacts of meteorology and precursor emission changes on the long-term trend of ambient ozone over the Pearl River Delta, China, and implications for ozone control strategy *Atmos. Chem. Phys.* **19** 12901–16
- [73] Zhao S, Yu Y, Qin D, Yin D, Dong L and He J 2019 Analyses of regional pollution and transportation of PM<sub>2.5</sub> and



- ozone in the city clusters of Sichuan Basin, China *Atmos. Pollut. Res.* **10** 374–85
- [74] Gong C, Liao H, Zhang L, Yue X, Dang R and Yang Y 2020 Persistent ozone pollution episodes in North China exacerbated by regional transport *Environ. Pollut.* **265** 115056
- [75] Li L, An J, Shi Y, Zhou M, Yan R, Huang C, Wang H, Lou S, Wang Q and Lu Q 2016 Source apportionment of surface ozone in the Yangtze River Delta, China in the summer of 2013 *Atmos. Environ.* **144** 194–207
- [76] Li L, An J, Huang L, Yan R, Huang C and Yarwood G 2019 Ozone source apportionment over the Yangtze River Delta region, China: investigation of regional transport, sectoral contributions and seasonal differences *Atmos. Environ.* **202** 269–80
- [77] Yang W, Chen H, Wang W, Wu J, Li J, Wang Z, Zheng J and Chen D 2019 Modeling study of ozone source apportionment over the Pearl River Delta in 2015 *Environ. Pollut.* **253** 393–402
- [78] Kleffmann J 2005 Daytime formation of nitrous acid: a major source of OH radicals in a forest *Geophys. Res. Lett.* **32** 5
- [79] Li X, Brauers T, Häsel R, Bohn B, Fuchs H, Hofzumahaus A, Holland F, Lou S, Lu K and Rohrer F 2012 Exploring the atmospheric chemistry of nitrous acid (HONO) at a rural site in Southern China *Atmos. Chem. Phys.* **12** 1497–513
- [80] Bernard F, Cazaunau M, Grosselin B, Zhou B, Zheng J, Liang P, Zhang Y, Ye X, Daële V and Mu Y 2016 Measurements of nitrous acid (HONO) in urban area of Shanghai, China *Environ. Sci. Pollut. Res.* **23** 5818–29
- [81] Xu Z, Wang T, Wu J, Xue L, Chan J, Zha Q, Zhou S, Louie P K and Luk C W 2015 Nitrous acid (HONO) in a polluted subtropical atmosphere: seasonal variability, direct vehicle emissions and heterogeneous production at ground surface *Atmos. Environ.* **106** 100–9
- [82] Liu Y et al 2019 A comprehensive model test of the HONO sources constrained to field measurements at rural North China Plain *Environ. Sci. Technol.* **53** 3517–25
- [83] Wang J, Zhang X, Guo J, Wang Z and Zhang M 2017 Observation of nitrous acid (HONO) in Beijing, China: seasonal variation, nocturnal formation and daytime budget *Sci. Total Environ.* **587** 350–9
- [84] Li D, Xue L, Wen L, Wang X, Chen T, Mellouki A, Chen J and Wang W 2018 Characteristics and sources of nitrous acid in an urban atmosphere of northern China: results from 1-yr continuous observations *Atmos. Environ.* **182** 296–306
- [85] Gu R, Zheng P, Chen T, Dong C, Liu Y, Liu Y, Luo Y, Han G, Wang X and Zhou X 2020 Atmospheric nitrous acid (HONO) at a rural coastal site in North China: seasonal variations and effects of biomass burning *Atmos. Environ.* **229** 117429
- [86] Gu R et al 2022 Investigating the sources of atmospheric nitrous acid (HONO) in the megacity of Beijing, China *Sci. Total Environ.* **812** 152270
- [87] Xue C et al 2021 Evidence for strong HONO emission from fertilized agricultural fields and its remarkable impact on regional O<sub>3</sub> pollution in the summer North China Plain *ACS Earth Space Chem.* **5** 340–7
- [88] Liu Y, Nie W, Xu Z, Wang T, Wang R, Li Y, Wang L, Chi X and Ding A 2019 Semi-quantitative understanding of source contribution to nitrous acid (HONO) based on 1 year of continuous observation at the SORPES station in eastern China *Atmos. Chem. Phys.* **19** 13289–308
- [89] Xue L et al 2016 Oxidative capacity and radical chemistry in the polluted atmosphere of Hong Kong and Pearl River Delta region: analysis of a severe photochemical smog episode *Atmos. Chem. Phys.* **16** 9891–903
- [90] Zhang L, Wang T, Zhang Q, Zheng J Y, Xu Z and Lv M Y 2016 Potential sources of nitrous acid (HONO) and their impacts on ozone: a WRF-chem study in a polluted subtropical region *J. Geophys. Res. Atmos.* **121** 3645–62
- [91] Fu X et al 2019 The significant contribution of HONO to secondary pollutants during a severe winter pollution event in southern China *Atmos. Chem. Phys.* **19** 1–14
- [92] Zhang J, An J, Qu Y, Liu X and Chen Y 2019 Impacts of potential HONO sources on the concentrations of oxidants and secondary organic aerosols in the Beijing-Tianjin-Hebei region of China *Sci. Total Environ.* **647** 836–52
- [93] Zhang L, Li Q Y, Wang T, Ahmadov R, Zhang Q, Li M and Lv M Y 2017 Combined impacts of nitrous acid and nitryl chloride on lower-tropospheric ozone: new module development in WRF-chem and application to China *Atmos. Chem. Phys.* **17** 9733–50
- [94] Tang K et al 2019 A dual dynamic chamber system based on IBBCEAS for measuring fluxes of nitrous acid in agricultural fields in the North China Plain *Atmos. Environ.* **196** 10–19
- [95] Xue C, Ye C, Zhang Y, Ma Z, Liu P, Zhang C, Zhao X, Liu J and Mu Y 2019 Development and application of a twin open-top chambers method to measure soil HONO emission in the North China Plain *Sci. Total Environ.* **659** 621–31
- [96] Wang Y, Fu X, Wu D, Wang M, Lu K, Mu Y, Liu Z, Zhang Y and Wang T 2021 Agricultural fertilization aggravates air pollution by stimulating soil nitrous acid emissions at high soil moisture *Environ. Sci. Technol.* **55** 14556–66
- [97] Wang T et al 2016 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** 2476–89
- [98] Tham Y J et al 2016 Significant concentrations of nitryl chloride sustained in the morning: investigations of the causes and impacts on ozone production in a polluted region of northern China *Atmos. Chem. Phys.* **16** 14959–77
- [99] Xia M et al 2021 Winter ClNO<sub>2</sub> formation in the region of fresh anthropogenic emissions: seasonal variability and insights into daytime peaks in northern China *Atmos. Chem. Phys.* **21** 15985–6000
- [100] Li Q, Zhang L, Wang T, Wang Z, Fu X and Zhang Q 2018 'New' reactive nitrogen chemistry reshapes the relationship of ozone to its precursors *Environ. Sci. Technol.* **52** 2810–8
- [101] Yu C et al 2020 Heterogeneous N<sub>2</sub>O<sub>5</sub> reactions on atmospheric aerosols at four Chinese sites: improving model representation of uptake parameters *Atmos. Chem. Phys.* **20** 4367–78
- [102] Wang Z, Wang W, Tham Y J, Li Q, Wang H, Wen L, Wang X and Wang T 2017 Fast heterogeneous N<sub>2</sub>O<sub>5</sub> uptake and ClNO<sub>2</sub> production in power plant and industrial plumes observed in the nocturnal residual layer over the North China Plain *Atmos. Chem. Phys.* **17** 12361–78
- [103] Liu X et al 2017 High levels of daytime molecular chlorine and nitryl chloride at a rural site on the North China Plain *Environ. Sci. Technol.* **51** 9588–95
- [104] Peng X et al 2021 An unexpected large continental source of reactive bromine and chlorine with significant impact on wintertime air quality *Natl Sci. Rev.* **8** nwaa304
- [105] Peng X, Wang T, Wang W, Ravishankara A, George C, Xia M, Cai M, Li Q, Salvador C M and Lau C 2022 Photodissociation of particulate nitrate as a source of daytime tropospheric Cl<sub>2</sub> *Nat. Commun.* **13** 1–10
- [106] Li Q et al 2020 Potential effect of halogens on atmospheric oxidation and air quality in China *J. Geophys. Res. Atmos.* **125** 9
- [107] Qiu X H et al 2019 Significant impact of heterogeneous reactions of reactive chlorine species on summertime atmospheric ozone and free-radical formation in north China *Sci. Total Environ.* **693** 133580
- [108] Wang X, Jacob D J, Fu X, Wang T, Breton M L, Hallquist M, Liu Z, McDuffie E E and Liao H 2020 Effects of anthropogenic chlorine on PM<sub>2.5</sub> and ozone air quality in China *Environ. Sci. Technol.* **54** 9908–16

- [109] Zhang Y *et al* 2021 Impacts of chlorine emissions on secondary pollutants in China *Atmos. Environ.* **246** 118177
- [110] Tan Z *et al* 2018 Wintertime photochemistry in Beijing: observations of RO<sub>x</sub> radical concentrations in the North China Plain during the BEST-ONE campaign *Atmos. Chem. Phys.* **18** 12391–411
- [111] Whalley L K *et al* 2021 Evaluating the sensitivity of radical chemistry and ozone formation to ambient VOCs and NO<sub>x</sub> in Beijing *Atmos. Chem. Phys.* **21** 2125–47
- [112] Lu K *et al* 2019 Fast photochemistry in wintertime haze: consequences for pollution mitigation strategies *Environ. Sci. Technol.* **53** 10676–84
- [113] Li K *et al* 2021 Ozone pollution in the North China Plain spreading into the late-winter haze season *Proc. Natl Acad. Sci. USA* **118** 10
- [114] Fu X, Wang T, Gao J, Wang P, Liu Y, Wang S, Zhao B and Xue L 2020 Persistent heavy winter nitrate pollution driven by increased photochemical oxidants in Northern China *Environ. Sci. Technol.* **54** 3881–9
- [115] Liu Y M and Wang T 2020 Worsening urban ozone pollution in China from 2013 to 2017—part 2: the effects of emission changes and implications for multi-pollutant control *Atmos. Chem. Phys.* **20** 6323–37
- [116] Liu Y M and Wang T 2020 Worsening urban ozone pollution in China from 2013 to 2017—part 1: the complex and varying roles of meteorology *Atmos. Chem. Phys.* **20** 6305–21
- [117] Wang P F, Guo H, Hu J L, Kota S H, Ying Q and Zhang H 2019 Responses of PM<sub>2.5</sub> and O<sub>3</sub> concentrations to changes of meteorology and emissions in China *Sci. Total Environ.* **662** 297–306
- [118] Lu X, Zhang L, Chen Y F, Zhou M, Zheng B, Li K, Liu Y M, Lin J T, Fu T M and Zhang Q 2019 Exploring 2016–2017 surface ozone pollution over China: source contributions and meteorological influences *Atmos. Chem. Phys.* **19** 8339–61
- [119] Li K, Jacob D J, Liao H, Shen L, Zhang Q and Bates K H 2019 Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China *Proc. Natl Acad. Sci. USA* **116** 422–7
- [120] Doumbia T, Granier C, Elguindi N, Bouarar I, Darras S, Brasseur G, Gaubert B, Liu Y, Shi X and Stavrakou T 2021 Changes in global air pollutant emissions during the COVID-19 pandemic: a dataset for atmospheric modeling *Earth Syst. Sci. Data* **13** 4191–206
- [121] Huang X *et al* 2021 Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China *Natl Sci. Rev.* **8** nwaa137
- [122] Liu Y, Wang T, Stavrakou T, Elguindi N, Doumbia T, Granier C, Bouarar I, Gaubert B and Brasseur G P 2021 Diverse response of surface ozone to COVID-19 lockdown in China *Sci. Total Environ.* **789** 147739
- [123] Shi X and Brasseur G P 2020 The response in air quality to the reduction of Chinese economic activities during the COVID-19 outbreak *Geophys. Res. Lett.* **47** e2020GL088070
- [124] Wang P, Chen K, Zhu S, Wang P and Zhang H 2020 Severe air pollution events not avoided by reduced anthropogenic activities during COVID-19 outbreak *Resour. Conserv. Recycl.* **158** 104814
- [125] Zhao Y B, Zhang K, Xu X T, Shen H Z, Zhu X, Zhang Y X, Hu Y T and Shen G F 2020 Substantial changes in nitrogen dioxide and ozone after excluding meteorological impacts during the COVID-19 outbreak in Mainland China *Environ. Sci. Technol. Lett.* **7** 402–8
- [126] Fu S, Guo M, Fan L, Deng Q, Han D, Wei Y, Luo J, Qin G and Cheng J 2021 Ozone pollution mitigation in Guangxi (south China) driven by meteorology and anthropogenic emissions during the COVID-19 lockdown *Environ. Pollut.* **272** 115927
- [127] Lin Y, Jiang F, Zhao J, Zhu G, He X, Ma X, Li S, Sabel C E and Wang H 2018 Impacts of O<sub>3</sub> on premature mortality and crop yield loss across China *Atmos. Environ.* **194** 41–47
- [128] Zhao H, Zheng Y, Zhang Y and Li T 2020 Evaluating the effects of surface O<sub>3</sub> on three main food crops across China during 2015–2018 *Environ. Pollut.* **258** 113794
- [129] Zhang T *et al* 2021 Modeling the joint impacts of ozone and aerosols on crop yields in China: an air pollution policy scenario analysis *Atmos. Environ.* **247** 118216
- [130] Feng Z, Kobayashi K, Li P, Xu Y, Tang H, Guo A, Paoletti E and Calatayud V 2019 Impacts of current ozone pollution on wheat yield in China as estimated with observed ozone, meteorology and day of flowering *Atmos. Environ.* **217** 116945
- [131] Feng Z *et al* 2022 Ozone pollution threatens the production of major staple crops in East Asia *Nat. Food* **3** 47–56
- [132] Broberg M C, Feng Z, Xin Y and Pleijel H 2015 Ozone effects on wheat grain quality—a summary *Environ. Pollut.* **197** 203–13
- [133] Broberg M C, Xu Y, Feng Z and Pleijel H 2021 Harvest index and remobilization of 13 elements during wheat grain filling: experiences from ozone experiments in China and Sweden *Field Crops Res.* **271** 108259
- [134] Shen S, Zhang D, Yang K, Wang Y, Zhu J, Yang L and Wang Y 2016 Effect of elevated surface layer ozone concentration on grain quality of two rice cultivars—a face study *Chin. J. Eco-Agric.* **24** 1231–8
- [135] Wang Y, Yang L, Han Y, Zhu J, Kobayashi K, Tang H and Wang Y 2012 The impact of elevated tropospheric ozone on grain quality of hybrid rice: a free-air gas concentration enrichment (FACE) experiment *Field Crops Res.* **129** 81–89
- [136] Zheng F, Wang X, Zhang W, Hou P, Lu F, Du K and Sun Z 2013 Effects of elevated O<sub>3</sub> exposure on nutrient elements and quality of winter wheat and rice grain in Yangtze River Delta, China *Environ. Pollut.* **179** 19–26
- [137] Feng Z, Kazuhiko K and Elizabeth A 2008 Impact of elevated ozone concentration on growth, physiology, and yield of wheat (*Triticum aestivum* L.): a meta-analysis *Glob. Change Biol.* **14** 2696–708
- [138] Feng Z, De Marco A, Anav A, Gualtieri M, Sicard P, Tian H, Fornasier F, Tao F, Guo A and Paoletti E 2019 Economic losses due to ozone impacts on human health, forest productivity and crop yield across China *Environ. Int.* **131** 104966
- [139] Li P, De Marco A, Feng Z, Anav A, Zhou D and Paoletti E 2018 Nationwide ground-level ozone measurements in China suggest serious risks to forests *Environ. Pollut.* **237** 803–13
- [140] Li P, Feng Z, Catalayud V, Yuan X, Xu Y and Paoletti E 2017 A meta-analysis on growth, physiological, and biochemical responses of woody species to ground-level ozone highlights the role of plant functional types *Plant. Cell Environ.* **40** 2369–80
- [141] Feng Z, Buker P, Pleijel H, Emberson L, Karlsson P E and Uddling J 2018 A unifying explanation for variation in ozone sensitivity among woody plants *Glob. Change Biol.* **24** 78–84
- [142] Hoshika Y, Fares S, Pellegrini E, Conte A and Paoletti E 2020 Water use strategy affects avoidance of ozone stress by stomatal closure in Mediterranean trees—a modelling analysis *Plant. Cell Environ.* **43** 611–23
- [143] Li P, Feng Z, Shang B and Uddling J 2021 Combining carbon and oxygen isotopic signatures to identify ozone-induced declines in tree water-use efficiency *Tree Physiol.* **41** 2234–44
- [144] Zeng Y, Cao Y, Qiao X, Seyler B C and Tang Y 2019 Air pollution reduction in China: recent success but great challenge for the future *Sci. Total Environ.* **663** 329–37
- [145] Wang Y and Liao H 2020 Effect of emission control measures on ozone concentrations in Hangzhou during G20 meeting in 2016 *Chemosphere* **261** 127729