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

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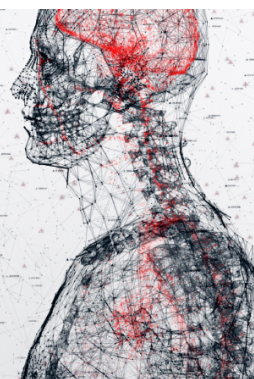
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Ground-level ozone pollution in China: a synthesis of recent findings on influencing factors and impacts

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E-mail: tao.wang@polyu.edu.hk**Keywords:** ozone, O₃, China, ozone trend, ozone impact on crops and forests, ozone formation and transport, ozone controlSupplementary material for this article is available [online](#)**Abstract**

Ozone (O₃) 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 O₃ concentrations paradoxically increased in many urban areas. The worsening urban O₃ pollution has fuelled numerous studies in recent years, which have enriched knowledge about O₃-related processes and their impacts. In this article, we synthesise the key findings of over 500 articles on O₃ over mainland China that were published in the past six years in English-language journals. We focus on recent changes in O₃ concentrations, their meteorological and chemical drivers, complex O₃ 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 (O₃) in the stratosphere is beneficial for life on Earth by filtering out Sun's harmful ultra-violet radiation, at ground level, O₃ adversely affects human health and vegetation. O₃ also regulates the oxidative capacity of the troposphere, and is a powerful greenhouse gas that contributes to climate warming. Elevated O₃ 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 O₃ 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 O₃ in the past six years. From this body of work, new findings on the factors influencing O₃ production and distribution and on the O₃ impacts have emerged. On the policy side, mitigating O₃ 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 O₃ concentrations, which will help provide scientific support for the development of evidence-based O₃ control strategies.

Several reviews on the O₃ 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 O₃ production and transport, the history of O₃ monitoring and research in China, the O₃ concentrations in major city-clusters, the relationships between O₃

concentrations and their precursors, early findings on the roles of nitrous acid (HONO) and nitryl chloride (ClNO₂) in O₃ production, and the impacts of O₃ on crops, forests and human health. Subsequently, five additional topical reviews were published [7–11]. Lu *et al* [7] reviewed findings on the O₃ 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 O₃ precursors, chemistry and deposition, and transport patterns. Fu *et al* [9] reviewed impacts of emission, multiscale meteorology, land use change, aerosol effect on O₃, and the influence of tropospheric O₃ change on radiative forcing and temperature. Liu *et al* [10] reviewed the methodological aspects of O₃ 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 O₃ on crop yield and economic loss, tree risk exposure to O₃, and cardio-vascular mortality with O₃ exposure dose. The O₃ 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 O₃ 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 O₃-formation regimes amid decreasing NO_x and increasing VOC emissions, (2) the recent results on the impact of HONO and ClNO₂ on O₃ production and the new findings on the impact of other reactive halogen (Cl₂ and BrCl) in polluted regions, (3) the winter photochemistry of O₃ (an understudied topic), (4) the complex responses of O₃ to reduced emissions under recent control measures and during China's coronavirus disease 2019 (COVID-19) lockdown, (5) the impact of O₃ on the quality of grain crops, tree productivity and water use efficiency. Additionally, we update the results on surface O₃ changes across China to the year of 2021, which reveals possible signs of O₃ levelling off or decrease in some urban areas, and (6) introduce recent government efforts to control O₃ in summer. We also give a graphic summary of multi-scale transport patterns.

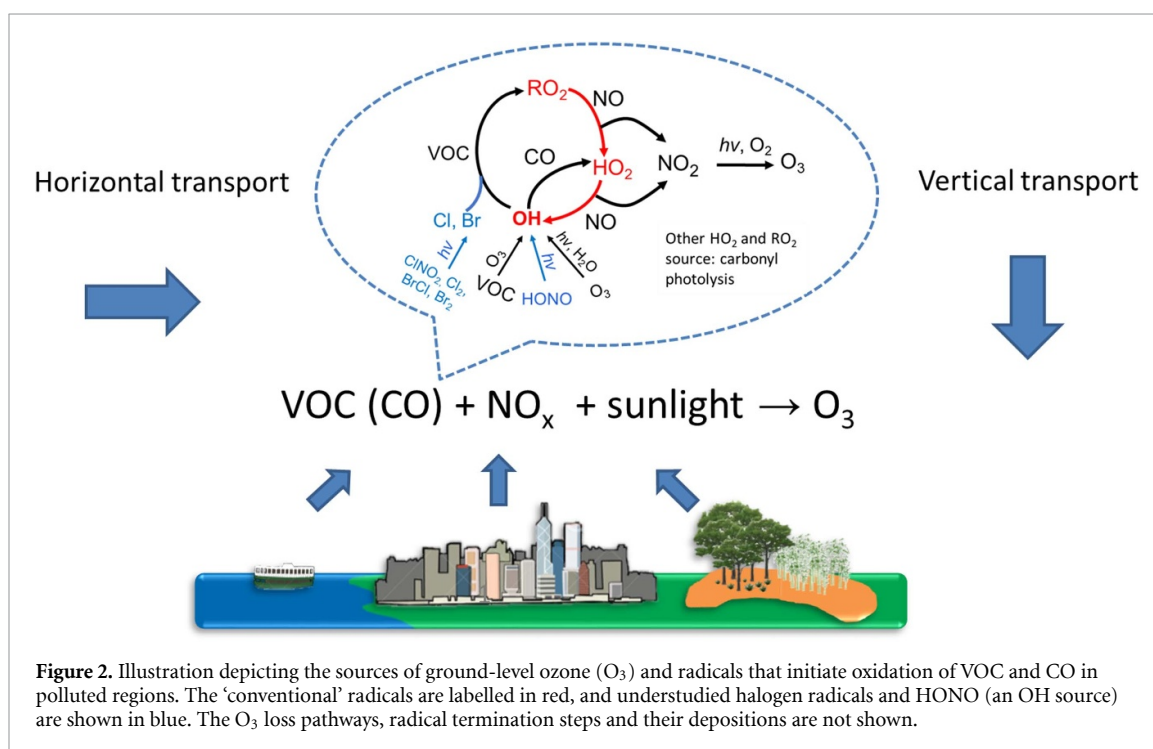
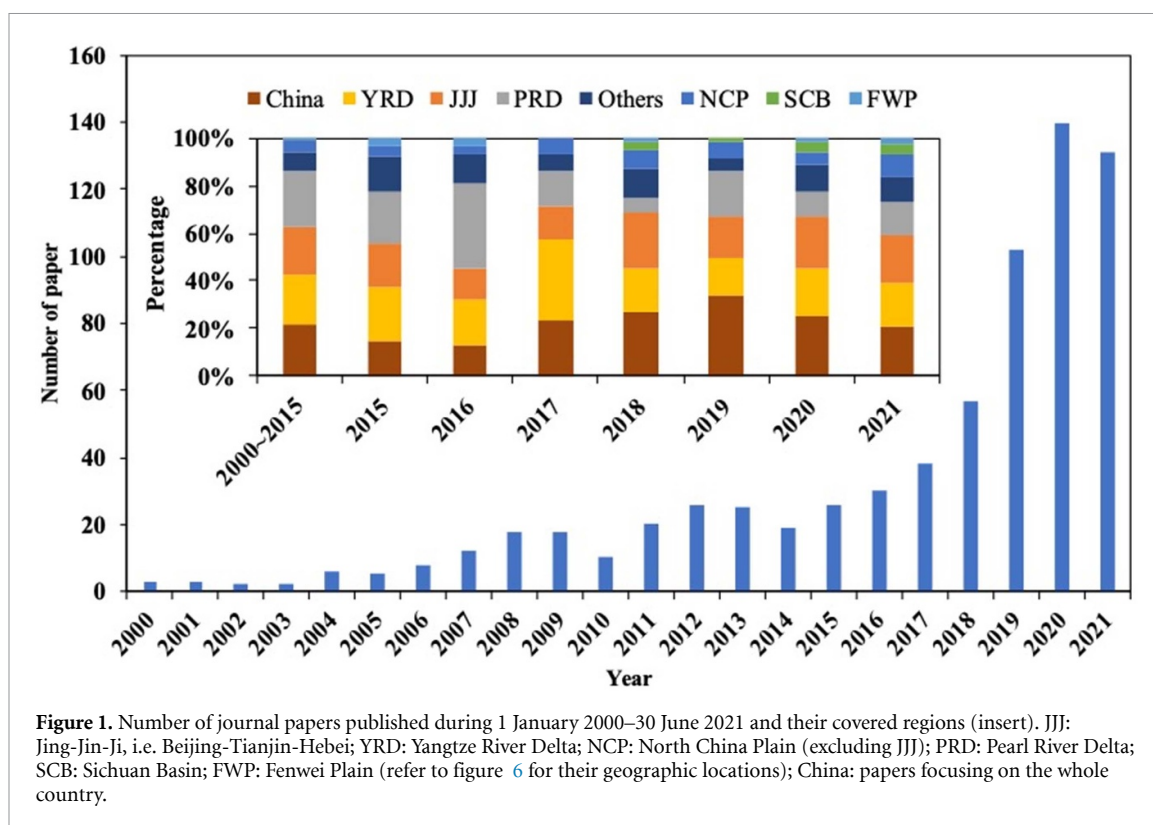
2. Literature search and results

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 'O₃' 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 O₃-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)). 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 'O₃' 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 O₃-related papers published from 2000 to the end of the June 2021, which highlights the rapid growth of O₃ 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 O₃ 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 O₃ pollution in Beijing [12, 13] has motivated more studies on O₃ in the NCP. There have also been more publications on O₃ processes and impacts across the whole of China since 2013, when data on ambient concentrations of O₃ and other regulated pollutants were made available by the China National Environmental Monitoring Center network.

The topics of investigation in O₃ 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 O₃ concentrations and the impacts of O₃ on vegetation and human health.



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

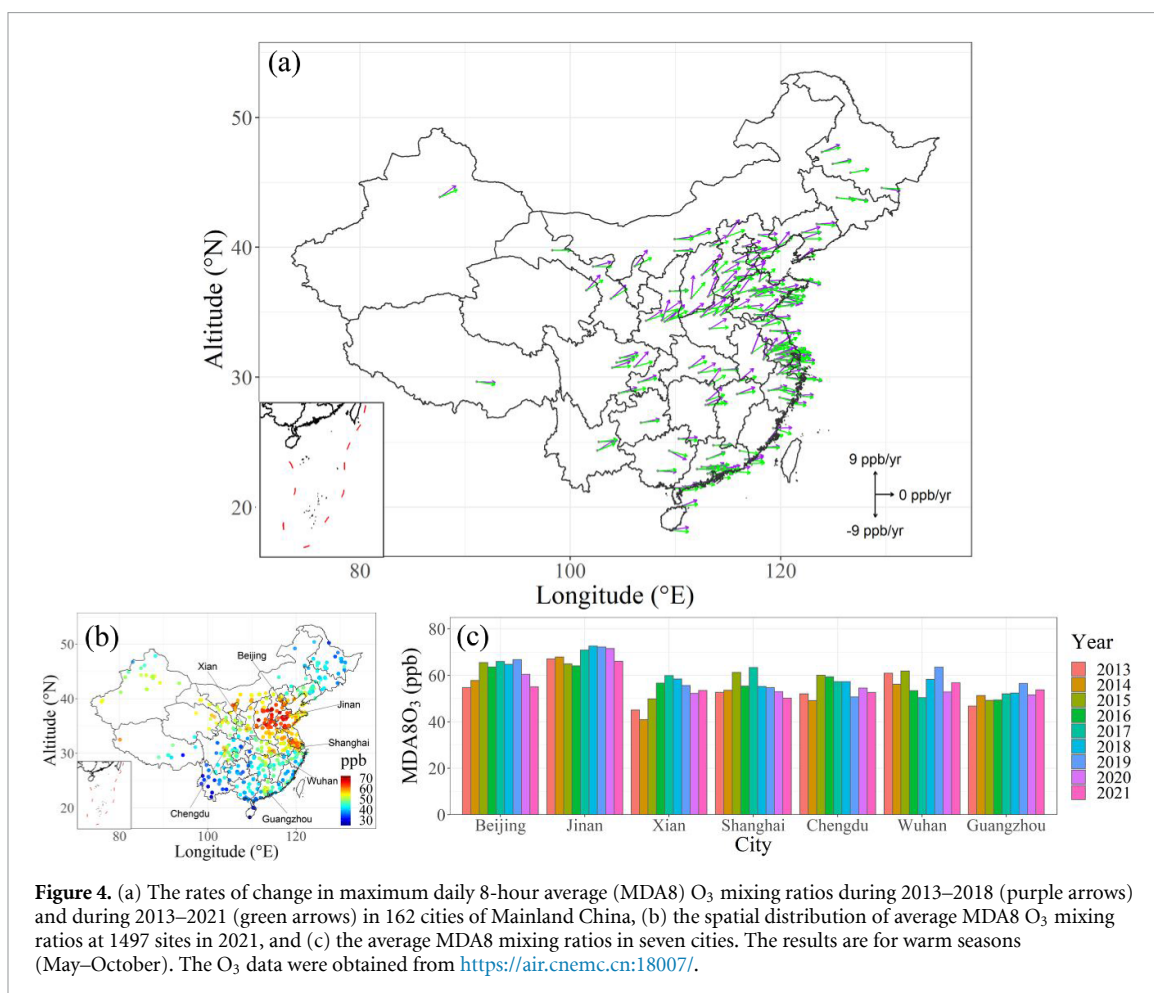
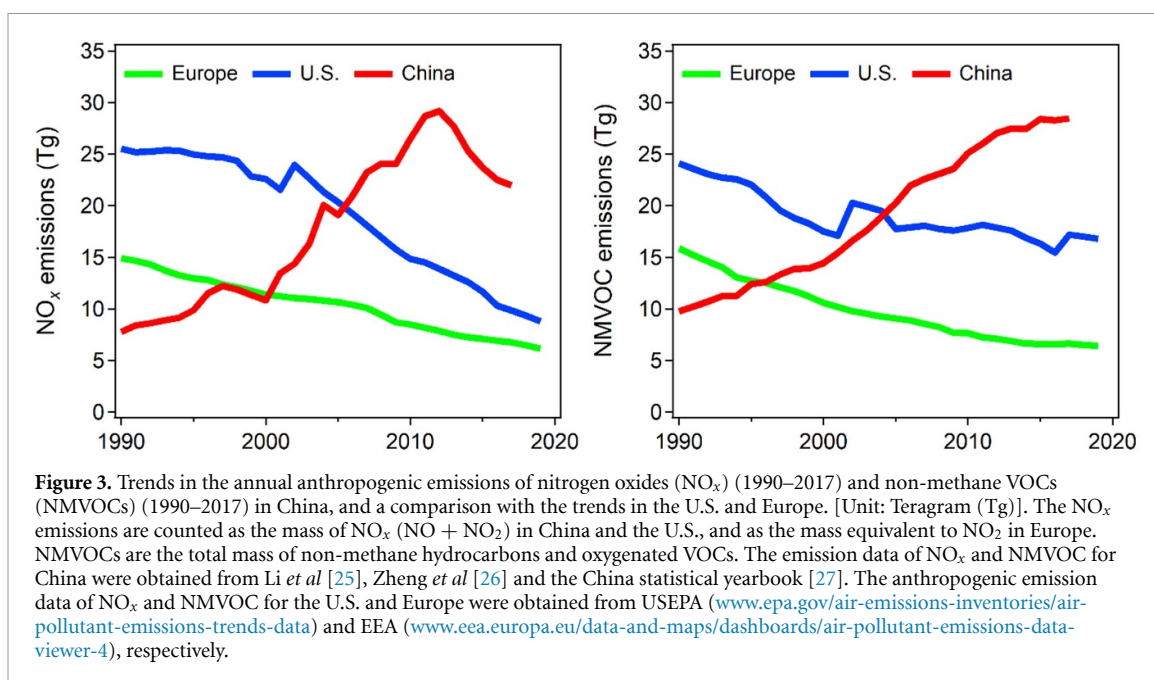
contrast to the decrease in both NO_x and VOC emissions in the U.S. and Europe (figure 3).

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



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_3 formation regime [28] was indicated in some suburban areas of the YRD, mainly owing to decreases in NO_x emissions in

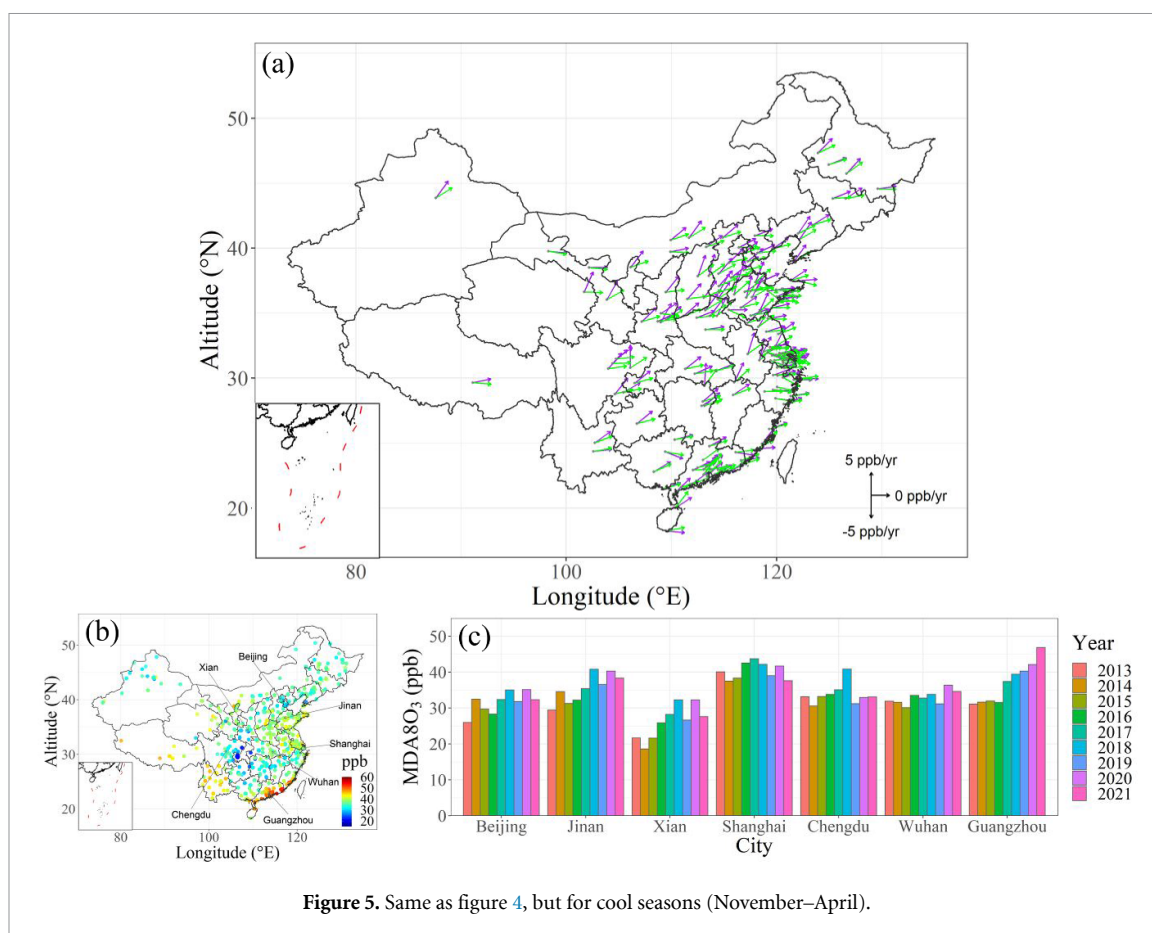


Figure 5. Same as figure 4, but for cool seasons (November–April).

the recent years. As to altitude dependence, an analysis of airborne measurements in the NCP showed that O_3 formation became more sensitive to NO_x emissions with increasing heights [36].

Recent analysis of *in-situ* measurements indicated that alkenes and aromatics were the major reactive VOC groups for O_3 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_3 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_3 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_3 formation regime over eastern China, with the areas of mixed-limited regime becoming larger due to NO_x 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_x emissions in the past decade has affected the O_3 formation regime, but is insufficient to change the typical urban areas from a VOC-limited to a NO_x -limited regime.

5. Roles of multi-scale atmospheric transport

O_3 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_3 occurs: the intercontinental-, long-range- and regional scales. Intercontinental transport of O_3 occurs between, for instance, Europe and Asia. Long-range transport of O_3 occurs from other Asian countries to China and from the stratosphere to ground level. We separate regional transport of O_3 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_3 accumulation and transport up to the year of 2018 [8, 9] and the source

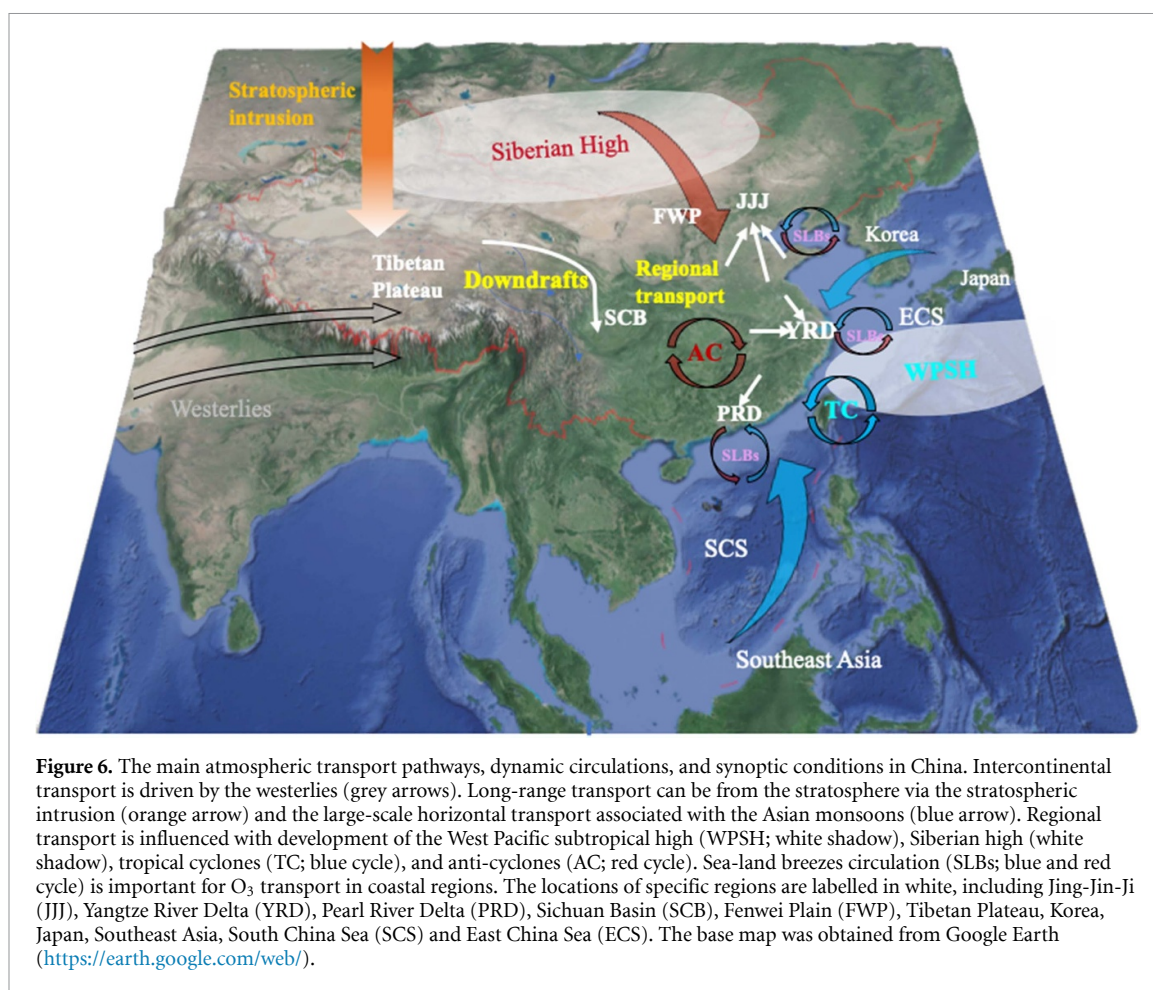


Figure 6. The main atmospheric transport pathways, dynamic circulations, and synoptic conditions in China. Intercontinental transport is driven by the westerlies (grey arrows). Long-range transport can be from the stratosphere via the stratospheric intrusion (orange arrow) and the large-scale horizontal transport associated with the Asian monsoons (blue arrow). Regional transport is influenced with development of the West Pacific subtropical high (WPSH; white shadow), Siberian high (white shadow), tropical cyclones (TC; blue cycle), and anti-cyclones (AC; red cycle). Sea-land breezes circulation (SLBs; blue and red cycle) is important for O_3 transport in coastal regions. The locations of specific regions are labelled in white, including Jing-Jin-Ji (JJJ), Yangtze River Delta (YRD), Pearl River Delta (PRD), Sichuan Basin (SCB), Fenwei Plain (FWP), Tibetan Plateau, Korea, Japan, Southeast Asia, South China Sea (SCS) and East China Sea (ECS). The base map was obtained from Google Earth (<https://earth.google.com/web/>).

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 O₃ 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 O₃ pollution in China [9]. Recent studies have provided additional result on their impact on O₃ 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 O₃ 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 O₃ 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 O₃ 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 O₃ 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 O₃ concentrations in JJJ [74]. Similarly, emissions from non-YRD regions have been found to be the dominant contributor to O₃ 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 O₃ pollution over the city clusters of the basin [73]. These studies highlight the importance of cross-boundary transport in developing O₃ mitigation strategies for a specific city/region.

6. Emerging chemical processes important to O₃ 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 O₃. 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 O₃ 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 O₃ 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 O₃ photolysis after fertilization [87]. Chemistry-transport models (CTMs) incorporating known HONO sources have attempted to quantify the HONO contribution to O₃ concentrations [90–93]. Simulations with a Weather Research Forecast-Chemistry (WRF-Chem) model showed that HONO enhanced the O₃ peak concentrations by 4%–10% in the HK-PRD region of southern China during a summer O₃ episode [90]; another study using a WRF-CAMQ model showed 34% increase in regional O₃ concentrations in a heavy winter pollution episode in the same region [91]. In other regions of China, WRF-Chem simulations suggested monthly O₃ increase of 5%–44% in Beijing-Tianjin-Hebei region in August [92]. Another WRF-Chem study indicated 6%–13% increase in surface O₃ 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 O₃ production in polluted regions.

The high HONO concentrations (>10 ppb) reported in China were mainly attributed to the heterogeneous reactions of NO₂ on ground and aerosol surfaces, and to the photolysis of nitrate (NO₃[−]) aerosols [91]. However, the kinetic parameters in the real atmospheric conditions remain uncertain (i.e. the NO₂ uptake coefficient, HONO production yields and NO₃[−] 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 O₃ concentrations by 8% in the agriculture-intensive NCP [96].

Other radicals that can produce O₃ are chlorine (Cl) or bromine (Br) atoms. Ambient ClNO₂ is a potential source of Cl radical (and a reservoir for NO₂). The early findings on the impact of ClNO₂ 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 ClNO₂ increased daytime O₃ 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, ClNO₂ 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 ClNO₂ concentrations can increase during daytime, leading to greater daytime O₃ production [99]. A WRF-Chem model incorporating laboratory-determined heterogeneous production of ClNO₂ was used to show that there was a 5%–6% increase in O₃ 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 ClNO₂ and HONO chemistry on the designation of the O₃ formation regimes, and the result showed that the ‘new’ nitrogen chemistry changed the O₃ 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 ClNO₂ production (including the uptake coefficient of nitrogen pentoxide (N₂O₅) and the yields of ClNO₂) are subject to large uncertainties and need further improvement [101, 102].

Molecular chlorine (Cl₂) is another potentially important photolytic source of Cl. Cl₂ 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 Cl₂ and ClNO₂ increased the O₃ production rate by 19% [103]. A recent study during the coal-burning period of winter documented surprisingly high concentrations of bromine chloride (BrCl) and Cl₂ at a site in Wangdu, and calculations from an observation-constrained model indicated that Cl and Br atoms collectively increased the total O_x (O₃ + NO₂) mixing ratios at this site by 50% [104]. Another very recent study reported unprecedented levels of Cl₂ (up to 1 ppb) at a polluted coastal site of Hong Kong, and such high concentrations of Cl₂ increased O_x daytime production by 16% [105]. However, existing CTMs cannot account for observed daytime Cl₂ 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 O₃ 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 O₃ 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 Cl₂), 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 O₃ production were 15–100 ppb h⁻¹ under high NO_x conditions, which is comparable to those during the summer. However, the concentration of O₃ was suppressed due to the simultaneous and rapid loss of O₃ by NO titration, the subsequent loss of NO₂ via gas-phase reactions with OH and the heterogeneous loss of N₂O₅, 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 O₃ 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 O₃ 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 (SO₂), NO_x, CO and 2.5 μm 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 SO₂, NO_x and PM, but increases in ground-level O₃ 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 O₃ increase during 2013–2017. To determine the contribution from the meteorology, the model mainly compares the O₃ 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 O₃ 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 O₃ 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 O₃ concentrations across China from 2013 to 2017, which showed that the meteorological impacts on O₃ concentrations tended to vary regionally. This study showed that changes in meteorological conditions since 2013 have made relatively little contribution to the increased O₃ concentrations in Beijing compared to the effects of emissions, but have made relatively larger contributions (either positive or negative) than emissions to the increased O₃ concentrations in Shanghai and Guangzhou for some years from 2014 to 2017. Meteorological changes, including those affecting in the long-range transport of O₃ to China from places outside China, were the predominant cause of the increase in O₃ 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 O₃ 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 O₃ concentrations in China, with varied approaches. For instance, using the CMAQ model, Wang *et al* [117] concluded that variation in MDA8 O₃ concentrations was mainly attributed to emission changes in 2013–2015. They obtained this result by subtracting the simulated change in MDA8 O₃ concentrations due to meteorology from the total observed change. Their results also showed that the increase in O₃ 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 O₃ 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 O₃ precursors (NO_x and VOCs) on MDA8 O₃ concentrations in 2013 and 2017. Their results indicated that the large decrease in PM_{2.5} concentrations (~40%)

reduced the uptake of HO₂ on aerosol surfaces, leading to an increase in O₃ 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 O₃ 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 O₃ concentrations in China. Their results showed that decreased emissions of NO_x, primary PM and SO₂, and increased emissions of VOCs, were the main cause of the increased urban O₃ 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 SO₂ emissions were the dominant drivers of the increase in O₃ 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 O₃ concentrations [115]. The same study also highlighted the importance of the uptake of HO₂ and O₃ by PM in driving changes in O₃ 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 O₃ concentrations in urban areas of China, due to the non-linear dependence of O₃ on NO_x and aerosol feedbacks. Therefore, the control of VOC emissions should be included in future O₃ control strategies. A positive outcome of these control measures for O₃ is that O₃ 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 O₃ formation regime that exists in rural areas [23, 116].

8. Effects of China's COVID-19 lockdown on ground-level O₃ concentrations

The outbreak of coronavirus disease 2019 (COVID-19) pandemic in 2020, 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 O₃ concentrations. A number of studies have analysed data on ground-level O₃ concentrations based on data from China National Environmental Monitoring Center network, using different methodologies. Some directly compared ambient O₃ 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 O₃ 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_x 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 O₃ concentrations during the COVID-19 lockdown have focused on the increases in ground-level O₃ 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 NO₂ and SO₂ in the same areas [123–125]. A few studies have also investigated the decrease in O₃ 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 O₃ 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 O₃ concentrations to the non-linear relationship between O₃ concentrations and those of its precursors, which led to an increase in O₃ concentrations in most cities due to O₃ titration by NO_x under NO_x-saturated or VOC-limited conditions, and a decrease in O₃ concentrations in the south due to the lower NO_x to VOC ratios there. Model-calculated indicators of the O₃-formation regime (i.e. the ratio of the production rate of H₂O₂ to that of HNO₃) 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 O₃-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 O₃ concentrations

due to decreased NO_x titration [122]. Moreover, the increased O₃ 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 O₃ on crops and trees

There was limited research on crop responses to O₃ 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 O₃ concentration [11]. Currently, there has been a general consensus that O₃ pollution in China has substantially decreased crop yields [11, 127–130]. A very recent assessment, which is based on the latest O₃ dose-yield response relationship obtained from the Asian field experiments and hourly O₃ concentrations in 2018–2020 at 1400 air quality stations in China, suggests that the ambient O₃ 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 O₃ 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 O₃ pollution affects grain quality in China. The few available studies on grain quality have largely focused on wheat or rice. O₃ 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 O₃ concentrations [134]. In addition, elevated O₃ 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 O₃ concentrations [136, 137]. These findings suggest that although O₃ pollution can improve grain nutrient quality in some cases, it generally leads to a deterioration of grain quality.

Ground-level O₃ 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 O₃ pollution [138]. However, only a few studies have estimated the impacts of O₃ on Chinese forest productivity. Northern temperate

forests in China are exposed to relatively higher O₃ concentrations than sub-tropical forests [139]. However, as tropical and subtropical evergreen forests have longer growing seasons (and are thus exposed to O₃ for longer durations) but are more resistant to O₃ than temperate deciduous forests, O₃ 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 O₃ concentrations than needle-leaved forests [140], probably due to the lower O₃ 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 O₃ concentrations can influence photosynthetic CO₂ assimilation and stomatal conductance, thereby ultimately altering WUE [142, 143].

10. Government regulations and efforts to control O₃

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, accessed 1 December 2021). These regulations have set specific targets for reducing emissions and/or ambient PM_{2.5} concentrations, or the number of heavy pollution days. The regulations in 2018 mandated a 15% reduction in the emissions of NO_x and SO₂ 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_{2.5} and PM₁₀) and other routinely monitored pollutants such as NO_x, CO, and SO₂ [26, 115, 144], but have not been effective in reducing O₃ pollution. This is in part because previous control measures mainly aimed to alleviate the notorious haze or PM_{2.5} pollution in China (especially in northern China during the winter), but assigned a lower priority to reductions in O₃ 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_x and VOCs emissions were implemented to reduce O₃ pollution, and the responses of ambient O₃ 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_x

emissions (www.mee.gov.cn, accessed 1 December 2021). Specifically, as part of the O₃ Pollution Prevention and Control Action Campaign, which was launched by the Ministry of Ecology and Environment (MEE) to mitigate increasing summertime O₃ 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 O₃ 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 O₃ 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 O₃ in China. O₃ 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 O₃ 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 O₃ concentrations in rural areas in the past decade.
- Despite the continuous reductions in NO_x emissions, observation- and emission-based studies indicate that major urban areas of China remain under VOC-limited O₃ formation regimes, although some sites can be under VOC–NO_x co-limited or NO_x-limited O₃-formation regimes during certain periods. These results highlight the necessity of joint NO_x–VOC controls to alleviate O₃ pollution.

- A number of studies have assessed the atmospheric transport of O₃ 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 O₃ pollution.
 - The drivers of the increase in urban O₃ 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 O₃ concentrations, and that reductions in the emissions of NO_x, SO₂ and PM have complex effects on ground-level O₃ concentrations. The studies have also suggested that control of VOC emission could have prevented the increase in O₃ concentrations. Similarly, studies on the response of O₃ concentrations to the COVID-19 lockdown have underscored the complex relationships between O₃ and primary pollutants, and the critical effect of meteorological factors on short-term changes in O₃ concentrations.
 - There have been several advances on the chemical processes important to O₃ production in the past few years. These include improved understanding of the roles of HONO and ClNO₂ and discovery of the important roles of other reactive halogen compounds (Cl₂ 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 O₃ formation mechanisms in high NO_x and high PM environments.
 - The past five years have also seen the publication of many studies on the impacts of O₃ on vegetation. Their findings provide strong scientific evidence that confirms the negative impacts of O₃ on the crop yields and grain quality. The available studies have also shown that deciduous species are at more risk to O₃ than evergreen species, and increased O₃ concentrations can affect water use of trees.
 - In terms of policy, although previous control regulations and measures focused on PM_{2.5}, those in the past two years have paid increasing attention to O₃. The enforced control of VOC emissions in the two most recent summers has had the desired effect on ambient O₃ concentrations, which appear to show signs of decreasing in many O₃-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 O₃, NO₂ 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 O₃ formation and to validate VOC emission inventories.

- More research is needed on the role of newly discovered chemical processes in O₃ 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_x and VOCs, and the continuing reductions in the emissions of PM_{2.5}). Air quality models should incorporate these processes to enable more accurate predictions.
- More research is needed to quantify the complex relationship between O₃ and PM_{2.5} in different regions. This will help support the development of a co-control strategy for O₃ and PM_{2.5}. Research for future O₃-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 O₃ monitors need to be installed in the countryside and at forest reserves to facilitate scientific assessments on the ecological impacts of O₃, and the contribution of BVOC emissions to O₃ generation.
- More research is needed to develop measures to reduce the negative effects of O₃ on plants. Such measures could include protectants, management of water use and the breeding of O₃-tolerant cultivars or species.

Data availability statement

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

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Author contributions

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