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TOPICAL REVIEW

Ambient fine particulate matter and ozone pollution in China: synergy in anthropogenic emissions and atmospheric processes

Yueqi Jiang\textsuperscript{1,2}, Shuxiao Wang\textsuperscript{2,3,4,5,}, Jia Xing\textsuperscript{6,}, Bin Zhao\textsuperscript{1,2}, Shengyue Li\textsuperscript{7,}, Xing Chang\textsuperscript{8,}, Shuping Zhang\textsuperscript{1,2} and Zhaoxin Dong\textsuperscript{1,2}

1 State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, People's Republic of China
2 State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing 100084, People's Republic of China
* Author to whom any correspondence should be addressed.

E-mail: shxwang@mail.tsinghua.edu.cn

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Abstract

Since 2013, China has taken a series of actions to relieve serious PM\textsubscript{2.5} pollution. As a result, the annual PM\textsubscript{2.5} concentration decreased by more than 50% from 2013 to 2021. However, ozone pollution has become more pronounced, especially in the North China Plain. Here, we review the impacts of anthropogenic emissions, meteorology, and atmospheric processes on ambient PM\textsubscript{2.5} loading and components and O\textsubscript{3} pollution in China. The reported influence of interannual meteorological changes on PM\textsubscript{2.5} and O\textsubscript{3} pollution during 2013–2019 ranged from 10%–20% and 20%–40%, respectively. During the same period, the anthropogenic emissions of NO\textsubscript{x}, SO\textsubscript{2}, primary PM\textsubscript{2.5}, NMVOC and NH\textsubscript{3} are estimated to decrease by 38%, 51%, 35%, 11% and 17%, respectively. Such emission reduction is the main cause for the decrease in PM\textsubscript{2.5} concentration across China. However, the imbalanced reductions in various precursors also result in the variation in nitrate gas-particle partitioning and hence an increase in the nitrate fraction in PM\textsubscript{2.5}. The increase of ozone concentration and the enhancement of atmospheric oxidation capacity can also have substantial impact on the secondary components of PM\textsubscript{2.5}, which partly explained the growth of organic aerosols during haze events and the COVID-19 shutdown period. The uneven reduction in NO\textsubscript{x} and NMVOC is suggested to be the most important reason for the rapid O\textsubscript{3} increase after 2013. In addition, the decrease in PM\textsubscript{2.5} may also have affected O\textsubscript{3} formation via radiation effects and heterogeneous reactions. Moreover, climate change is expected to influence both anthropogenic emissions and atmospheric processes. However, the extent and pathways of the PM\textsubscript{2.5}–O\textsubscript{3} interplay and how it will be impacted by the changing emission and atmospheric conditions making the synergetic control of PM\textsubscript{2.5} and O\textsubscript{3} difficult. Further research on the interaction of PM\textsubscript{2.5} and O\textsubscript{3} is needed to provide basis for a scientifically-grounded and effective co-control strategy.

1. Introduction

Through efforts in recent years, China has made significant advances in alleviating fine particulate matter (PM\textsubscript{2.5}) pollution (Wang \textit{et al} 2017a, Zheng \textit{et al} 2018, Zhou \textit{et al} 2021). According to observational data, the PM\textsubscript{2.5} concentration decreased by more than 50% from 2013 to 2021 (Bulletin of China Ecological Environment 2013 and Bulletin of China Ecological Environment 2021, available on [www.mee.gov.cn/hjzl/stxjzk/zghjzkgb/, in Chinese]). However, ozone concentration has shown an increasing trend. According to Lu \textit{et al} (2020), the proportion of sites with mean daily maximum 8-h average (MDA8) O\textsubscript{3} concentration during the warm season (April–September) over 60 ppb increased from 11.1% in 2013 to 49.2% in 2019. The synergistic control of PM\textsubscript{2.5} and ozone pollution in China has become a major challenge and subject of high research interest. In the past few years, the number of articles on this
topic has steadily increased. The number of related articles published in 2021 (569 articles) is more than five times that in 2013 (104 articles) according to the search results on the Web of Science with theme as ['PM$_{2.5}$' or 'particle' or 'aerosol', 'O$_3$' or 'ozone', and 'China'] (as of 10th March 2022). Some studies have focused on the trends and characteristics of PM$_{2.5}$ and O$_3$ pollution (Chu et al. 2020, Kuerban et al. 2020, Wei et al. 2021a). More has attempted to explain the change in terms of variations in atmospheric processes and anthropogenic emissions. Numerical model analysis found that the contribution of interannual meteorological changes was 10%–20% and 20%–40% for PM$_{2.5}$ and O$_3$, respectively (Ding et al. 2019b, Chen et al. 2020b, Li et al. 2020a). The impacts of anthropogenic emissions changes are greater but more complex (Zhang et al. 2019b, Wang et al. 2020b, 2021a). The changes in precursor emissions not only changed the PM$_{2.5}$ and O$_3$ concentrations, but also affected the chemical composition of PM$_{2.5}$ as well as the interactions between PM$_{2.5}$ and O$_3$. Previous reviews have mostly focused on PM$_{2.5}$ or O$_3$ (Wang et al. 2017b, An et al. 2019, Lu et al. 2019a, Chen et al. 2020e), but neglected the synergistic effects between the two pollutants. It is necessary and imperative to summarize the current understanding of how the synergy in anthropogenic emissions and atmospheric processes affects PM$_{2.5}$ and O$_3$ pollution, and provide ground for scientifically-informed air pollution control policymaking.

In this article, we first briefly review the spatial and temporal characteristics of PM$_{2.5}$ and O$_3$ pollution and the correlation between PM$_{2.5}$ and O$_3$ concentrations in China based on literature review and observation data analysis. Next, we review the synergistic effects between PM$_{2.5}$ and O$_3$ as influenced by three different factors: (a) the impact of anthropogenic emissions; (b) the interaction between PM$_{2.5}$ and O$_3$ through atmospheric processes; (c) the impact of climate change.

2. Methods

2.1. Data source and processing

This review is mainly based on two sets of data:

(a) The surface observation data of PM$_{2.5}$ and O$_3$ in China from 2013 to 2021

We obtained the surface observation data from the China National Environmental Monitoring Center network (https://air.cnemc.cn:18007/). Data processing of PM$_{2.5}$ and O$_3$ concentration was conducted according to National Ambient Air Quality Standards (GB3095-2012) (available on www.mee.gov.cn/ywgz/fgbz/bzwb/dqhyjhb/dqhyjlzbz/201203/W020120410330232398521.pdf, in Chinese) and China's Technical Regulation for Ambient Air Quality Assessment (tentative) (HJ 633-2013) (available on www.mee.gov.cn/ywgz/fgbz/bzwb/jcfhbx/201309/W020131105548549111863.pdf, in Chinese). It should be noted here the standard state data were used before 2019, and the actual state data were used starting in 2019 for the assessment of ambient air quality. The O$_3$ concentration depended significantly on this modification. We multiplied the O$_3$ observation data for 2019, 2020, and 2021 by 1.09 for consistent conversion (1.09 was calculated according to difference between the Bulletin of China Ecological Environment 2019 and Bulletin of China Ecological Environment 2018, available on www.mee.gov.cn/hjzl/sthjzg/zghjzkgb/, in Chinese).

(b) The anthropogenic air pollutants emissions inventory of China

We obtained anthropogenic air pollutants emissions data from five databases, including ABaCAS-EI (Zhao et al. 2013, Wang et al. 2014, Cai et al. 2017, Zheng et al. 2019; www.abacas-dss.com/abacas/), Community Emissions Data System (CEDS, O’Rourke et al. 2021), Emissions Database for Global Atmospheric Research (EDGARv5.0, https://edgar.jrc.ec.europa.eu/overview.php?v=50_AP), Multi-resolution Emission Inventory for China (MEIC, Zheng et al. 2018, 2021; http://meicmodel.org/?page_id=560), and Regional Emission inventory in Asia (REASv3.1, Kurokawa and Ohara 2020). ABaCAS-EI and MEIC also provided the emissions data of each province in China.

2.2. Literature search and selection strategy

The literature cited in this study was obtained by searching in the Web of Science (www.webofscience.com) database. The latest search date is 10 June 2022. The search terms are included in table S1 of the supplementary materials. We need to uncover suitable literature from many research domains because just a few papers have provided a thorough grasp of the synergy between PM$_{2.5}$ and O$_3$. When choosing the literature, we followed the following rules: (a) literature which receives a lot of citations or is extensively discussed; (b) literature which focuses on China or typical locations; and (c) literature that proposes novel phenomena and arguments for their explanations. Additionally, in order to track the most recent advancements in adjacent fields of research, we primarily focused on literature released after 2017 (152 references). Also a number of classic works of literature released before 2017 are included (18 references).

3. PM$_{2.5}$ and O$_3$ pollution across China

3.1. Spatial and temporal distribution of PM$_{2.5}$ and O$_3$

Based on air quality observation data in China, we analyzed spatial and temporal distribution of PM$_{2.5}$
Figure 1. Annual evaluation values of PM$_{2.5}$ (average PM$_{2.5}$ concentration in one year) and O$_3$ (the 90th percentile of daily maximum 8-hour average O$_3$ in one year) in China and five key regions from 2013 to 2021. Here, observation data of some cities in FW and SCB are absent in 2013 and 2014.

and O$_3$ from 2013 to 2021. Annual evaluation values of PM$_{2.5}$ (average PM$_{2.5}$ concentration in one year) and O$_3$ (the 90th percentile of daily maximum 8-hour average O$_3$ in one year) in China and five key regions are shown in figure 1 (annual evaluation values of PM$_{2.5}$ and O$_3$ of all sites are shown in figure S1 in supplementary materials).

Generally, the annual PM$_{2.5}$ concentration in China continuously decreased from 72 µg·m$^{-3}$ to 30 µg·m$^{-3}$ from 2013 to 2021. The annual PM$_{2.5}$ concentrations of the Jing-Jin-Ji (JJJ), the Yangtze River Delta (YRD) and the Pearl River Delta (PRD) regions decreased by 63 µg·m$^{-3}$ (59%), 36 µg·m$^{-3}$ (54%), and 26 µg·m$^{-3}$ (55%), respectively. The annual PM$_{2.5}$ concentrations in Beijing, Shanghai and Guangzhou all met the national standard level 2 in 2021, according to the National Air Quality Standards (GB3095-2012). The spatial pattern of PM$_{2.5}$ is remained similar from 2013 to 2021, with higher PM$_{2.5}$ concentrations in eastern and northern China and lower PM$_{2.5}$ concentrations in western and southern China (Wang et al 2017a, Xu et al 2020). The North China Plain (NCP) is typically a polluted region in China. Seasonal variation of PM$_{2.5}$ has been described in many previous studies (Zhao et al 2016, Li et al 2017c, Chen et al 2018, Zhao et al 2018, Ye et al 2018b, Ma et al 2019b). As shown in figure 2(a), PM$_{2.5}$ is higher in winter and lower in summer. During 2013–2021, the averaged PM$_{2.5}$ concentration in winter was 2–3 times that in summer in China. In spring and autumn, the PM$_{2.5}$ concentration has different variation characteristics for different regions. In JJJ, the Fenwei Plain (FW) and PRD, PM$_{2.5}$ in autumn is typically higher than that in spring, while the trends in the YRD and Sichuan Basin (SCB) are the opposite, as shown in figure S2 in the supplementary materials.

For O$_3$, as shown in figure 1, the 90th MDA8 O$_3$ in China, JJJ, YRD, PRD, and FW were the highest in 2019. The highest 90th MDA8 O$_3$ in SCB appeared in 2020. Compared with 2013, the 90th MDA8 O$_3$ in 2019 in JJJ, YRD, and PRD increased by 59 µg·m$^{-3}$ (38%), 35 µg·m$^{-3}$ (24%), and 37 µg·m$^{-3}$ (24%), respectively. In terms of spatial distribution, eastern China is more polluted than western China, which is similar to the spatial distribution of PM$_{2.5}$. Due to higher volatile organic compounds (VOCs) emissions and stronger radiation, O$_3$ is higher in the warm season (Zhang et al 2015, Cheng et al 2017, Wang et al 2018, Yang et al 2020). Figure 2(b) shows the monthly averaged MDA8 O$_3$ concentrations of JJJ, YRD, PRD, FW and SCB in 2021. For JJJ and FW, the peak of monthly averaged MDA8 O$_3$ appears in June. For SCB, O$_3$ is highest in July or August. For YRD and FW, the peak of monthly averaged MDA8 O$_3$ appears in June. For SCB, O$_3$ is highest in July or August. For PRD, O$_3$ is the highest in June or September. PRD has a distinct pattern, with the highest O$_3$ in September or October. Thus, the warm season of China is usually considered to be April to September or April to October.
Figure 2. Monthly (a) averaged PM$_{2.5}$ and (b) MDA8 O$_3$ concentrations in JJJ, YRD, PRD, FW and SCB in 2021.

Table 1. Summary of studies on the Pearson correlation coefficient between PM$_{2.5}$ and O$_3$ over China.

| Region  | Data                                      | Period                               | Pearson correlation coefficient | References       |
|---------|-------------------------------------------|--------------------------------------|---------------------------------|------------------|
| China   | Hourly and daily PM$_{2.5}$ and O$_3$ concentration | 1 March 2013–28 February 2017         | Lower in the northwestern China; higher in the southeastern China in the winter; Positive in the summer Lower at around 10:00; Higher at around 16:00 | Chen et al (2019a) |
| China   | Daily PM$_{2.5}$ and MDA8 O$_3$            | 2013–2018                            | Generally positive across NCP, YRD, PRD and SCB | Li et al (2019c)  |
| China   | Daily PM$_{2.5}$ and MDA8 O$_3$            | 2016                                 | Strong negative in Northern China during January; Strong positive in Southern China during July | Zhu et al (2019)  |
| China   | Hourly PM$_{2.5}$ and O$_3$ concentration  | 2013–2018                            | Negative in the North; Positive in the South $-0.13, -0.21, -0.14, and 0.13$ for China, JJJ, YRD and PRD, respectively | Chu et al (2020)  |
| NCP     | Monthly PM$_{2.5}$ and O$_3$ concentration | 2016–early 2019                      | $-0.6389$                       | Tian et al (2020) |
| YRD     | Daily PM$_{2.5}$ and MDA8 O$_3$            | April–October in 2013–2019           | 0.2–0.6                         | Dai et al (2021)  |
| YRD     | Daily PM$_{2.5}$ and MDA8 O$_3$ (Shanghai, Nanjing) | January, April, July and October in 2017 | Independent when AOC$^a$ is low ($r^2 = 0.01$); highly correlated when AOC is high ($r^2 = 0.4–0.7$) | Qin et al (2022)  |
| PRD     | Monthly PM$_{2.5}$ and O$_3$ concentration | 2006–2019                            | $-0.02$ on average; $-0.21, 0.38, -0.16$ and $-0.01$ for spring, summer, autumn and winter, respectively | Hu et al (2021)   |
| Lanzhou | Daily PM$_{2.5}$ and O$_3$ concentration  | Spring-summer period in 2014–2017     | $-0.335$                        | Filonchyk et al (2018) |

$^a$ AOC: atmospheric oxidizing capacity.

3.2. Correlation between PM$_{2.5}$ and O$_3$ pollution

Many studies have analyzed the correlation between PM$_{2.5}$ and O$_3$ by calculating the Pearson correlation coefficients based on monitoring data, as shown in table 1. From the spatial distribution, as pointed out in many studies, PM$_{2.5}$ and O$_3$ tend to be negatively correlated in North China and positively correlated in South China. In addition, Chen et al (2019a) found that PM$_{2.5}$ and O$_3$ in coastal areas tend to be positively correlated but negatively correlated in inland areas. In this study, we also analyzed the correlation between the daily PM$_{2.5}$ and MDA8 O$_3$ concentrations in JJJ, YRD, PRD, FW and SCB in 2021, as shown in figures S3 and S4 (remove seasonal cycle of PM$_{2.5}$ and O$_3$). It
can be seen that the correlation between PM$_{2.5}$ and O$_3$ is more positive in PRD than in other regions, which is consistent with the conclusion above. From the temporal characterization, there is an obvious seasonal variation in that PM$_{2.5}$ and O$_3$ tend to be more positively correlated in the summer but tend to be more negatively correlated in the winter, as shown in both previous studies and our results. The daily variation shows that the peak value of the Pearson correlation coefficient appears at approximately 16:00 (Chen et al 2019a).

The secondary generation of PM$_{2.5}$ and O$_3$ is the key to explain the spatial-temporal variation of PM$_{2.5}$ and O$_3$ correlations. Qin et al (2022) associated atmospheric oxidizing capacity (AOC) with the correlation between PM$_{2.5}$ and O$_3$ in YRD. They found that the daily PM$_{2.5}$ and O$_3$ were highly correlated at moderate to high AOC levels. The promoted formation of secondary pollutants by AOC is the main reason (Liu et al 2020, Zhu et al 2020, Chen et al 2020c, Wang et al 2022a). We analyzed the correlation between daily O$_3$ (O$_3$ + NO$_x$) and PM$_{2.5}$ (figure S5). The result showed that O$_3$ and PM$_{2.5}$ is positively correlated in almost all key regions and seasons in 2021, indicating promoted production of secondary components of PM$_{2.5}$ by AOC. Chu et al (2020) suggested that the correlation between PM$_{2.5}$ and O$_3$ would become positive with the decrease of PM$_{2.5}$ in China due to the influence of PM$_{2.5}$ on solar radiation and heterogeneous reactions of HO$_2$. In summer or south China, where PM$_{2.5}$ concentration is relatively low, PM$_{2.5}$ has little impact on solar irradiation and HO$_2$ radicals, thus PM$_{2.5}$ and O$_3$ tend to have a positive correlation with secondary generation from NO$_x$ and VOCs emissions. Chen et al (2019a) suggested similar causes for regional differences. Besides, they reported the climate and meteorological conditions would cause the regional differences by influencing the formation and removal of PM$_{2.5}$ and O$_3$. Lei et al (2022) revealed that the Spearman correlation coefficients between PM$_{2.5}$ and O$_3$ in key regions (JJJ, YRD, PRD, SCB and FW) were much higher in 2020 than in 2019. They emphasized the urgent demand for the co-control of PM$_{2.5}$ and O$_3$ as the primary emissions decreased steadily. We therefore discuss the synergy of PM$_{2.5}$ and O$_3$ as influenced by three factors: (a) the impact of emissions; (b) interactions between the two pollutants; (c) future challenges under climate change in the following sections.

4. Changes of anthropogenic emissions and their influence on PM$_{2.5}$ and O$_3$

4.1. Anthropogenic emissions change in China

A large number of studies have been conducted to analyze the emission changes in China in recent years. Researchers have provided detailed reviews of both pollutants separated and sectors separated emissions (Wang et al 2014, Li et al 2017b, Li et al 2021a); thus, we briefly discuss the emissions change to provide support for later sections. Figure 3 shows the emission trends of SO$_2$, primary PM$_{2.5}$, NO$_x$, non-methane volatile organic compounds (NMVOC), and NH$_3$ from different studies and databases (Zhao et al 2013, Wang et al 2014, Crippa et al 2018, Zheng et al 2018, 2019, Kurokawa and Ohara 2020, O’Rourke et al 2021, Zheng et al 2021). The trends of SO$_2$ emissions and PM$_{2.5}$ emissions were similar. From 2000 to 2005, SO$_2$ emissions and PM$_{2.5}$ emissions increased by 32%–70% (EDGARv5.0, REASv3.1, CEDS) and 15%–21% (EDGARv5.0, REASv3.1), respectively, and then decreased from 2006 to 2010. Wide application of flue-gas desulfurization devices and end-of-pipe dust collectors in power plants is reported as one of the most effective measures (Lu et al 2010, Li et al 2017b). Small increases of SO$_2$ and PM$_{2.5}$ emissions appeared from 2011 to 2013. After 2013, SO$_2$ and PM$_{2.5}$ emissions decreased rapidly as a result of the implementation of the ‘Air Pollution Prevention and Control Action Plan’ (Cai et al 2017, Zheng et al 2019). Pollutant control strategies in industry, including strengthening industrial emission standards and removing pollutant emissions, were recognized as the main drivers of this reduction (Zheng et al 2018, Zhang et al 2019b). NO$_x$ emissions increased by 80%–128% (EDGARv5.0, REASv3.1, CEDS) from 2000 to 2013. From 2013 to 2017, rapid implementation of an effective NO$_x$ emissions control strategy (e.g. application of selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) in power plants and industry) caused a 20%–37% (AbA CAS-EI, MEIC, CEDS) decrease of NO$_x$ emissions in China. However, the emissions of NMVOC increased slowly and NH$_3$ emissions were stable from 2000 to 2017, due to the lack of effective control policies.

Herein, we also analyzed the emissions variation in key regions in AbA CAS-EI, as shown in figure 4. Compared to 2007, the trends of SO$_2$ and primary PM$_{2.5}$ were consistent in different regions, decreasing by 73%–88% and 57%–73% in 2019, respectively. However, the emissions of NO$_x$ were highest in 2013 in all 5 regions. Compared to 2013, the emissions of NO$_x$ in JJJ, YRD, PRD, FW and SCB decreased by 42%, 41%, 27%, 52%, and 33%, respectively, in 2019. In contrast, the emissions of NMVOC varied less from 2013 to 2019, by −33%, −6%, −5%, −10% and +5% in JJJ, YRD, PRD, FW, and SCB, respectively. The emission of NH$_3$ was more stable from 2007 to 2019.

4.2. Anthropogenic emissions driven PM$_{2.5}$ change

As listed in table 2, the changes of anthropogenic emissions is the major driver of PM$_{2.5}$ change in recent years in China. Ding et al (2019b) revealed that the variation of anthropogenic emissions contributed 70.8%, 95.6% and 99.7% to the total decrease of annual PM$_{2.5}$ concentration in JJJ, Guangdong and Sichuan-Chongqing region from 2013 to 2017. Control measures on industry were evaluated as the
largest source of PM$_{2.5}$ decline across China, which was consistent with the anthropogenic emissions change (Zheng et al 2019, Cheng et al 2019a, Zhang et al 2019b).

In addition to the PM$_{2.5}$ concentrations, the chemical compositions and source apportionment of PM$_{2.5}$ changed significantly due to the changes of anthropogenic emissions. Dong et al (2020) and Cao et al (2022) stated that the local contribution to PM$_{2.5}$ of Beijing decreased largely for strict pollutant control measures from 2013 to 2017, reflecting the uneven emissions reduction in different cities. Chen et al (2019b) reported that compared with the pollution episode in March 2013, the relative contribution of coal combustion decreased by 29%, while the relative contribution of vehicle exhaust increased by 35% in the pollution episode in March 2018 in Beijing. They suggested that different emissions control measures on emission sources were the main cause. Li et al (2021c) also found an increased contribution of vehicles and a decreased contribution of coal combustion to PM$_{2.5}$ in Heze (a city in the central NCP) from 2017 to 2019.

In addition, more studies have focused on the chemical composition of PM$_{2.5}$. Sulfate decreased markedly due to the large reduction of SO$_2$ emissions in the past decade. For JJ, YRD, PRD and SCB, the sulfate decreased by 51.1%, 40.8%, 37.5% and 36.2%, respectively, as reported by Cao et al (2021). However, nitrate showed different trends. Geng et al (2019) claimed that the proportion of nitrate in PM$_{2.5}$ increased by 5%, 7%, 4% and 3% in eastern China,
J JJ, YRD and PRD from 2013 to 2017, respectively, even if NOx emissions had been reduced. Explosive growth of nitrate during haze events was also observed (Kong et al 2018, Li et al 2018, 2019a, Zhou et al 2019, Xu et al 2019a, Fu et al 2020, Sun et al 2020, Zhang et al 2020). A simple explanation for the phenomenon was that a large reduction of SO2 left more NH3 available to form nitrate. However, some studies reported that the North China Plain was in an ammonia-rich atmosphere (Xu et al 2019b, Bhattarai et al 2020), contradicting the above explanation. The second explanation was that photochemical oxidants increased due to the reduction of NOx emissions and constant VOCs emissions in NCP, so that most regions were VOC-limited in winter (Xing et al 2019, Lu et al 2019a, Fu et al 2020, Leung et al 2020), increasing the conversion efficiency of NOx to HNO3. Fu et al (2020) reported ~30% increase of O3 and OH and 38.7% increase of conversion efficiency of NOx to HNO3 from 2010 to 2017 in NCP. Leung et al (2020) showed that the above two explanations both accounted for high NO3− concentrations in eastern and central China. Another explanation was proposed by Zhai et al (2021). A large reduction of SO2 emissions, smaller reduction of NOx emissions (compared to SO2 emissions), and stable NH3 emissions were conducive for the transformation of gas-phase nitrate to particle-phase nitrate. By simulation with the Goddard Earth Observing System (GEOS)-Chem model, they found that the deposition of the total nitrate (gas + particle) decreased as the particulate fraction of total nitrate approached unity. As a result, nitrate sometimes showed a weak response to NOx emissions reduction and an increase in haze events.

4.3. Anthropogenic emissions drive O3 change

Unlike PM2.5 which is influenced by a multitude of precursor emissions, the O3 concentration is primarily affected by NOx and VOCs emissions. However, the anthropogenic emissions driven O3 change varied in different studies, as shown in table 2. Ding et al (2019a) showed that changes of anthropogenic emissions decreased O3 in China and key regions from 2013 to 2017, while Liu and Wang (2020b) found that anthropogenic emissions increased O3 in urban areas and decreased O3 in rural areas. Lin et al (2021) projected that anthropogenic emissions changes caused O3 to rise in eastern and central China, but meteorological conditions played the opposite role in the area from 2013 to 2020. Studies also showed that both radiative effect of particles and aerosol chemistry have significant effects on O3 pollution (Li et al 2019b, Wang et al 2020a), which will be discussed in the next section. There are many factors that may cause different results, such as model selections, mechanism settings, amount and distribution of emissions. One of the most important factors is the definition of the temporal and spatial scope. Due to distinctive regional and temporal characteristics of O3, same change of anthropogenic emissions may cause different variation of O3 in different regions and times.

Herein, we mainly focused on the influence of anthropogenic emissions on the O3 formation regime, which is closely associated with the control strategy, and has been considered in many studies as listed in table 3. As NOx plays a dual role in O3 formation, the O3 formation regime is classified as VOC-limited regime (VOC-sensitive regime), transition regime and NOx-limited regime (NOx-sensitive regime) (Sillman 1995, Wang et al 2019b). Lu et al (2019a) reviewed the spatial-temporal characteristics of the O3 formation regime across China. O3 formation during high O3 seasons in most urban areas was in the VOC-limited regime due to large amounts of anthropogenic emissions, while rural areas were in the NOx-limited regime (Xing et al 2019, ...
Table 2. Summary of studies on drivers of PM$_{2.5}$ and O$_3$ variation.

| Period      | Pollutants | Impact of anthropogenic emissions | Pollutants changes (if provided) | Impact of meteorological conditions | References             |
|-------------|------------|-----------------------------------|----------------------------------|------------------------------------|-------------------------|
| 2013–2017   | PM$_{2.5}$ | 70.8% of PM$_{2.5}$ decrease in JJJ | 95.6% of PM$_{2.5}$ decrease in Guangdong | 99.7% of PM$_{2.5}$ decrease in Sichuan-Chongqing | Ding et al (2019b) |
|             |            | −46 µg·m$^{-3}$·a$^{-1}$ across China | 95.6% of PM$_{2.5}$ decrease in Guangdong | 99.7% of PM$_{2.5}$ decrease in Sichuan-Chongqing | Ding et al (2019b) |
|             |            | −8.0 ± 1.1 µg·m$^{-3}$·a$^{-1}$ in JJJ | −6.3 ± 0.9 µg·m$^{-3}$·a$^{-1}$ in YRD | −2.2 ± 0.5 µg·m$^{-3}$·a$^{-1}$ in PRD | Zhai et al (2019)     |
|             |            | −4.9 ± 0.9 µg·m$^{-3}$·a$^{-1}$ in SCB | −5.0 ± 1.9 µg·m$^{-3}$·a$^{-1}$ in FW |                                    |                         |
| 2013–2017   | PM$_{2.5}$ | −18.1 µg·m$^{-3}$ in China        | −28.7 µg·m$^{-3}$ in JJJ         | −18.6 µg·m$^{-3}$ in YRD          | Zhang et al (2019b)   |
|             |            | −8.9 µg·m$^{-3}$ in PRD            |                                    |                                    |                         |
| 2013–2017   | PM$_{2.5}$ | 87.9% of PM$_{2.5}$ decrease in Beijing (local emissions reduction contributed 65.4%) |                                    |                                    | Cheng et al (2019a)   |
| 2014–2017   | PM$_{2.5}$ | More than 70% of PM$_{2.5}$ decrease in JJJ |                                    |                                    | Dong et al (2020)     |
| 2013–2015   | PM$_{2.5}$ | −23.9 µg·m$^{-3}$ in 2014 in 31 capital cities in China | −43.5 µg·m$^{-3}$ in 2015 in 31 capital cities in China | +11.5 µg·m$^{-3}$ in 2014 in 31 capital cities in China | Wang et al (2019c)   |
| O$_3$ in summer |            | +16.7 ppb in 2014 in 31 capital cities in China | +18.3 ppb in 2015 in 31 capital cities in China |                                    |                         |
| 2014–2018   | PM$_{2.5}$ | −23.5 µg·m$^{-3}$ in NCP         | −18.0 µg·m$^{-3}$ in YRD         | −18.4 µg·m$^{-3}$ in FW           | Chen et al (2020b)    |
|             |            | −2.6 µg·m$^{-3}$ in YRD           | −3.5 µg·m$^{-3}$ in FW            |                                    |                         |
| O$_3$       |            | +13.6 µg·m$^{-3}$ in NCP         | +11.2 µg·m$^{-3}$ in YRD         | +22.1 µg·m$^{-3}$ in FW            |                         |
|             |            | +1.8 µg·m$^{-3}$ in NCP           | +1.3 µg·m$^{-3}$ in YRD           |                                    |                         |
|             |            | +3.8 µg·m$^{-3}$ in FW            |                                    |                                    |                         |

(Continued.)
| Period | Pollutants | Impact of anthropogenic emissions | Impact of meteorological conditions | References |
|--------|------------|----------------------------------|------------------------------------|------------|
| 2013–2017 | O₃ in warm season | +2.1 ± 1.0 ppb in North China | +0.2 ± 1.1 ppb in North China | Ding et al (2019a) |
| | | +2.2 ± 0.8 ppb in North-east China | +0.3 ± 0.8 ppb in North-east China | |
| | | +2.6 ± 1.2 ppb in East China | +2.0 ± 1.7 ppb in East China | |
| | | +2.7 ± 0.7 ppb in Central China | +0.5 ± 1.5 ppb in South China | |
| | | +1.5 ± 1.0 ppb in South China | | |
| | | +0.9 ± 1.3 ppb in South-west China | | |
| | | +1.1 ± 0.9 ppb in North-west China | | |
| | | +0.2 ± 1.1 ppb in North China | | |
| | | +0.3 ± 0.8 ppb in North-east China | | |
| | | +2.0 ± 1.7 ppb in East China | | |
| | | +2.6 ± 1.2 ppb in East China | | |
| | | +1.5 ± 1.0 ppb in South China | | |
| | | +0.9 ± 1.3 ppb in South-west China | | |
| | | +1.1 ± 0.9 ppb in North-west China | | |
| 2013–2017 | O₃ in summer | +1–3 ppbv·a⁻¹ in megacity clusters of eastern China (Rapid decrease PM₂.₅ was the major factor by slowing down the uptake of HO₂ radicals) | | Li et al (2019b) |
| 2003–2015 | O₃ in July | +3.5 ± 1.4 ppbv over the central part of central eastern China | +5.8 ± 3.9 ppbv over the central part of central eastern China | Sun et al (2019) |
| | | +5.6 ± 1.8 ppbv over the eastern part of central eastern China | −0.8 ± 3.5 ppbv over the eastern part of central eastern China | |
| 2005–2016 | O₃ | Large decrease of PM₂.₅ in Beijing was the main driver of increased O₃ production. | | Wang et al (2020a) |
| 2013–2019 | O₃ in summer | +1.2 ppbv·a⁻¹ in China | +0.7 ppbv·a⁻¹ in China | Li et al (2020a) |
| | | +1.9 ppbv·a⁻¹ in NCP | +1.4 ppbv·a⁻¹ in NCP | |
| | | +0.9 ppbv·a⁻¹ in YRD | +0.7 ppbv·a⁻¹ in YRD | |
| | | +0.3 ppbv·a⁻¹ in PRD | +0.8 ppbv·a⁻¹ in PRD | |
| | | +1.0 ppbv·a⁻¹ in SCB | −0.2 ppbv·a⁻¹ in SCB | |
| 2013–2017 | O₃ in summer | Increase ozone (~2 ppbv) in urban areas | −12.7 ppbv to +15.3 ppbv over China in 2014–2017 relative to 2013 | Liu and Wang (2020a), Liu and Wang (2020b) |
| | | Decrease ozone (~1 ppbv) in rural areas | Little impact in Beijing and Chengdu | |
| | | +8 ppbv in Beijing and Shanghai in 2017 | Comparable with the impact of anthropogenic emissions in Shanghai | |
| | | +3 ppbv in Guangzhou and Chengdu in 2017 | | |
| | | (estimated according to figure 4 in the Liu and Wang (2020b)) | | |
| 2012–2017 | O₃ in summer | +39% in NCP | +49% in NCP | Dang et al (2021) |
| | | +13% in YRD | +84% in YRD | |
| Period          | Pollutants | Impact of anthropogenic emissions | Impact of meteorological conditions | References                        |
|----------------|------------|-----------------------------------|-------------------------------------|-----------------------------------|
| 2015–2019      | $O_3$      | $+68\%$ in JJJ                    | $+32\%$ in JJJ                      | Mousavinezhad et al (2021)        |
| 2016–2018      | 90th $O_3$-h in May (compared to 2017) | $+0.6 \mu g \cdot m^{-3}$ in 2016 in Shandong Province | Major factor in YRD and PRD (compared to 2017) | Li et al (2021b)                  |
| 2013–2020      | $O_3$ in summer | Increase $O_3$ in eastern and central China | Comparable to or greater than the impact of anthropogenic emissions in eastern, central and southern China | Lin et al (2021)                  |
Table 3. Summary of studies on O\textsubscript{3} formation regime change in China.

| Region                        | Period                  | Method                                      | Major result                                                                                     | References                  |
|-------------------------------|-------------------------|---------------------------------------------|--------------------------------------------------------------------------------------------------|-----------------------------|
| Beijing                       | 2006–2017               | Statistic method based on observation data  | The ozone formation regime tended to be VOC-NO\textsubscript{x}-limited regime due to NO\textsubscript{x}-oriented emissions reduction. | Cheng et al (2019b)         |
| Eastern China                 | 2012–2016               | WRF-CMAQ simulation                        | O\textsubscript{3} formation has changed from VOCs sensitive regime to the mix sensitive (transition) regime due to NO\textsubscript{x} reductions. | Wang et al (2019a)         |
| Guangdong-Hong Kong-Macao     | 2012–2016               | Analysis based on OMI remote sensing data   | Ozone formation sensitivity tended to be NO\textsubscript{x}-limited due to 33.1\% decrease of NO\textsubscript{x} emission and 35.2\% increase of VOCs emission. | Chen et al (2020d)         |
| China                         | 2005–2019               | Polynomial simulation; convergent cross-mapping (CCM) based on observation data | The ozone formation regime has changed to the transitional and NO\textsubscript{x}-limited regime all over China according to the reverse trends of HCHO and NO\textsubscript{2}. | Li et al (2021e)           |
| China                         | 2016–2019, COVID-19 lock down | Observation data and Chemistry Land-surface Atmosphere Soil Slab model | As rapid reduction of NO\textsubscript{x} emissions, some cities changed from VOC-limited regime to transitional regime. | Wang et al (2021c)         |
| JJJ                           | 2014–2018               | Brute-force method based on WRF-Chem simulation | VOC-sensitive regimes had changed from 8.4\%–44.2\% to 5.6\%–33.3\% with the rapid reduction of NO\textsubscript{x} emissions. | Wei et al (2021b)          |
| Shanghai                      | 2008–2019               | Statistic method based on observation data  | Ozone formation regime transformed from VOC-limited to NO\textsubscript{x}-limited with the NO\textsubscript{x} emissions reduction. | Yang et al (2021b)         |

Yu et al (2020). In the VOC-limited regime, a reduction of NO\textsubscript{x} emissions may lead to an increase of ozone. Thus, a rapid decrease of NO\textsubscript{x} was identified as one of the reasons for ozone increases in urban areas in recent years and during COVID-19 (Sciard et al 2020, Xing et al 2020a, Chen et al 2020c, Huang et al 2021). With the change of anthropogenic NO\textsubscript{x} and VOCs emissions, the formation mechanism of ozone changes dynamically, especially after 2013. In JJJ, the proportion of VOC-limited regime areas varied from 8.4\%–44.2\% to 5.6\%–33.3\% from 2014 to 2018 (Wei et al 2021b). Urban areas changed from VOC-limited regime to transitional regime, and rural areas were always in transitional regime or NO\textsubscript{x}-limited regime (Cheng et al 2019b, Li et al 2021e). In YRD, the transitional regime accounted for more than half, and rural areas tend to be NO\textsubscript{x}-limited (Wang et al 2019a). In PRD, more than half of urban areas belonged to the transitional regime, and most rural areas were NO\textsubscript{x}-limited (Chen et al 2020d).

4.4. Coordinated control strategy of PM\textsubscript{2.5} and O\textsubscript{3}
In terms of synergistic effects between PM\textsubscript{2.5} and ozone, joint control becomes possible. Based on empirical kinetic modeling approach (EKMA) curve, a reduction of VOCs emissions was proven to conduct under the current emission situation (Wang et al 2019a, Huang et al 2021). Studies conducted by Xiang et al (2020) and Ding et al (2021) revealed that earlier VOCs emission reductions were more cost-effective in reducing both PM\textsubscript{2.5} and O\textsubscript{3} in JJJ and surrounding regions. For further improvement of air quality in China, reduction of NO\textsubscript{x} emission is still essential (Xing et al 2019, Li et al 2019c, Ding et al 2021). Based on the response surface model, Dong et al (2022) suggested that the emissions of NO\textsubscript{x}, SO\textsubscript{2}, NH\textsubscript{3}, VOCs and primary PM\textsubscript{2.5} should be reduced by 50\%, 26\%, 28\%, 28\% and 55\%, respectively, to achieve the National Ambient Air Quality Standard (NAAQS) in YRD region. Huang et al (2020) suggested that emissions of NO\textsubscript{x}, VOCs and primary PM\textsubscript{2.5} should be reduced by 22\%, 12\% and 30\%, respectively, to achieve NAAQS in PRD region.

Furthermore, many studies focused on control policies aimed at both carbon and air pollution reduction (Xing et al 2020b, Yang et al 2021a). Note here that there is a special issue for China referred to as achieving carbon neutrality by 2060. Several studies proposed a strategy to achieve carbon neutrality and evaluated the influence on air quality. Shi et al (2021) projected SO\textsubscript{2}, NO\textsubscript{x}, primary PM\textsubscript{2.5} and NMVOCs in China to reduce by 93\%, 93\%, 90\% and
61% in 2060, respectively, compared to 2019, decreasing more than 70% PM$_{2.5}$ and 25% 90th MDA8 O$_3$. They expected PM$_{2.5}$ concentrations and O$_3$ concentrations in more than half of 337 cities to meet the World Health Organization (WHO) guidance (2006) in 2060. Cheng et al (2021) and Zhang et al (2021a) also highlighted that emission mitigations to achieve China’s carbon neutrality goal could improve PM$_{2.5}$ exposure to below 10 µg·m$^{-3}$ for the majority of China’s population.

5. Interaction of PM$_{2.5}$ and O$_3$ through atmospheric processes

Both secondary pollutants, PM$_{2.5}$ and O$_3$, have an impact on one another through various atmospheric processes. In this section, we discuss three dominant pathways: atmospheric oxidation capacity, heterogeneous reactions, and aerosol radiative effects, as shown in figure 5.

5.1. Atmospheric oxidation capacity

AOC refers to the ability of atmospheric chemical processes to oxidize primary pollutants. Oxidants (HO$_x$, NO$_3$…) concentrations and reaction rates are always used to characterize AOC. AOC plays an important role in the formation of both PM$_{2.5}$ and O$_3$. As one of the important atmospheric oxidants, O$_3$ concentration is often positively correlated with AOC (Zhu et al 2020). In clear air, O$_3$ is generated by NO$_2$ photolysis, and consumed in the oxidation of NO, without the accumulation of O$_3$ in the process. However, when OH oxidizes VOCs and generates HO$_2$ or RO$_2$ radicals, they replace O$_3$ to oxidize NO, leading to an increase of the O$_3$ concentration. Another product of VOCs oxidized by OH is oxygenated compounds, leading to the formation of secondary organic aerosol (SOA). At the same time, the increase of atmospheric oxidants also accelerates the formation of secondary inorganic aerosol (SNA). SOA and SNA are reported to account for more than 50% of the PM$_{2.5}$ concentration in most cities in China (Yin et al 2020). Wang et al (2021b) analyzed simulation and observation data of oxidants in 2013 and 2020. The results showed that the level of AOC did not decrease significantly (Chen et al 2020c), and even increased slightly in NCP and PRD, causing an increase of secondary pollutants. The importance of AOC was again evident in studies on COVID-19. Zhu et al (2021b) revealed that enhanced AOC during COVID-19 contributed 40% and 53% of O$_3$ in NCP and YRD, respectively. Wang et al (2021d) reported that the changes of AOC and O$_3$ concentrations in YRD were consistent during the COVID-19 lockdown. Huang et al (2021) found that an increase of AOC due to large decreases of NO$_x$ emissions promoted the formation of secondary particles. Moreover, Chen et al (2020a) revealed that the stratosphere-to-troposphere (STE) exchange of O$_3$ would increase PM$_{2.5}$ by 2.289 µg·m$^{-3}$ in winter and 2.034 µg·m$^{-3}$ in spring by enhancing AOC.

Several studies analyzed the nonlinear relationship between AOC or oxidants and precursors, and then proposed a control strategy for NO$_x$ and VOCs to avoid the increase of secondary pollutants. On the basis of O$_3$, OH and NO$_3$ isopleths, Kang et al (2021) found that NO$_x$ emission reduction was beneficial for O$_3$ and PM$_{2.5}$ in summertime. Wang et al (2021b) found that NO$_x$ emission reduction would decrease the AOC in summer, but increase the winter AOC in NCP and YRD. Both studies also proposed that the emission reduction of VOCs were beneficial for the further control of O$_3$ and PM$_{2.5}$. Furthermore, the effects of some other species on AOC have also been found. Ge et al (2019) showed that NH$_3$ could enhance AOC by accelerating the formation of gas-phase nitrous acid (HONO). Li et al (2021d) demonstrated that halogens increased PM$_{2.5}$ by 21% in northern China, and the enhancement of AOC was the main reason (Li et al 2020b).

5.2. Heterogeneous reactions

Heterogeneous chemistry refers to reactions on the surface of aerosols, including reactions and uptake of gases. Thus, it can lead to changes in both the size and components of particles (Farmer et al 2015). For example, Tian et al (2021) reported that heterogeneous reactions (including the uptake of SO$_2$, NO$_2$, HNO$_3$ and H$_2$SO$_4$ on CaCO$_3$ and mineral oxides) contributed 20%–30% to sulfate formation over northern China during a dust pollution event. Wang et al (2017c) showed that heterogeneous reactions promote both fine mode and coarse mode nitrate and sulfate to be internally mixed with dust particles.

Heterogeneous reactions related to sulfate formation have been a concern since the end of the last century. Early studies revealed that SO$_2$ could be oxidized by oxidants (H$_2$O$_2$, OH, O$_3$…) and NO$_2$ on mineral dust (Zhang and Carmichael 1999, Ye et al 2018a), and by dissolved oxidants in the aqueous phase (Cheng et al 2016, Li et al 2017a). Recently,
Gen et al (2019) proposed NO₂ and OH radicals produced from the photolysis of particle nitrate could oxidize aqueous-phase SO₂, which was a less explored pathway for sulfate formation. Song et al (2019) proposed that heterogeneous reactions of hydroxymethanesulfonate (HMS) might be important and could account for up to 1/3 of unexplained sulfate formation in an air quality model. In addition, many studies have focused on heterogeneous reactions of HONO. HONO, as an important source of OH radicals, is highly related to the secondary components of particles. Cui and Wang (2021) found that in polluted regions (JJJ, YRD, and PRD), the HONO concentration in winter was highest. However, the HONO concentration was typically underestimated in chemical transport models (CTM), particularly during haze events (Xue et al 2020). By considering heterogeneous reactions of HONO, the performance of HONO, OH, O₃, and secondary components in particles was improved significantly. Zhang et al (2019a) found that heterogeneous reactions (NO₂ and HONO) contributed approximately 85% of the daytime HONO concentration and improved the SOA concentration by more than 40% in Beijing. Fu et al (2019) reported that heterogeneous reactions of HONO on the surfaces (2NO₂ + H₂O → HONO + HNO₃) were the major source (72%) of HONO concentration during a severe pollution in PRD. Compared to the simulation without any HONO sources, the daytime averaged O₃ and mean concentration of TNO₃ (HNO₃ and fine particle NO₃⁻) increased 70% and 67%, respectively. Zhang et al (2021b) revealed that heterogeneous reactions on ground contributed nearly 70% of total HONO production. In Beijing, heterogeneous reactions of HONO increased the OH concentration and SOA concentration by 98% and 50%, respectively. Heterogeneous reactions were proven to effectively improve the simulation of HONO and SOA, but there still exists a gap between simulations and observations, and more studies are needed.

In addition, heterogeneous reactions were proven to influence ozone concentration by impacting the production and loss of nitrogen oxides (NO₂, NO₃, and N₂O₅), hydrogen radicals (HO₂, OH, and H), ozone and halogen radicals (Jacob 2000), as shown in table 4. Li et al (2019b) projected that a 40% decrease of PM₂.₅ was the most important driver of the O₃ increase in the North China Plain from 2013 to 2017, by slowing the uptake of HO₂. However, another study by Qu et al (2018) proved that effect of heterogeneous reactions of H₂O₂ (OH, HO₂, H₂O₂) was less than that of heterogeneous reactions of nitrogen oxides and ozone. Based on field

| Reactions | Uptake coefficients | References |
|----------|---------------------|------------|
| HO₂(g) → 0.5H₂O₂(g) | 0.2 (range 0.1–1) | Jacob (2000) |
| NO₂(g) → 0.5HONO(g) + 0.5HNO₃(g) | 10⁻⁴ (range 10⁻⁶–10⁻³) | |
| NO₃(g) → HNO₂(g) | 10⁻² (range 2 × 10⁻⁴–10⁻²) | |
| N₂O₅(g) → 2HNO₃(g) | 0.1 (range 0.01–1) | |
| OH(g) → OH(ads) | Dust: 0.1 (range 0.01–0.2) | Deng et al (2010) |
| HO₂(g) → HO₂(ads) | Dust: 0.1 (range 0.01–0.2) | Sea salt: 0.04 |
| H₂O₂(g) → H₂O₂(ads) | Dust: 10⁻⁴ (range 10⁻³–2 × 10⁻⁴) | BC: 0.1 |
| O₃(g) → O₃(ads) → O₂ | Dust: 10⁻⁴ (range 10⁻⁴–10⁻³) | Sea salt: 5 × 10⁻⁵ |
| NO₂(g) → NO₂(ads) → NO₃⁻ | Dust: 4.4 × 10⁻⁵ (range 10⁻³–10⁻⁴) | BC: 4.5 × 10⁻⁵ |
| NO₃(g) → NO₃(ads) → NO₅⁻ | Dust: 0.1 (range 0.01–0.2) | Sea salt: 0.046 |
| HNO₃(g) → HNO₃(ads) → NO₅⁻ | Dust: 0.01 (range 0.001–0.1) | BC: 0.001 |

Table 4. Heterogeneous reactions related to ozone concentration.
campaign measurements of radicals and trace gases, Tan et al (2020) conducted a sensitivity test of HO₂ uptake by using different uptake coefficients. The results showed that the performance of all radicals (OH, HO₂, and RO₂) was poor when using an uptake coefficient value of 0.2 (same as Li et al 2019b). When using an uptake coefficient of 0.08, the difference between the simulation and observation was much smaller, and the influence of aerosol uptake on HO₂ concentration was only 17%. Tan et al (2019) also found that the RO₂ and HO₂ budgets of the PRD closed without aerosol characterization, indicating that heterogeneous reactions of HO₂ were not important. In summary, the impact of heterogeneous reactions on ozone is subjected to much debate. Both comprehensive field measurement and modeling studies are suggested.

5.3. Aerosol radiative effects

Aerosol was proven to have direct effects on radiative forcing by absorbing or scattering radiation and indirect effects by participating in cloud formation (Bond et al 2013, Hong et al 2017, Liu and Matsui 2021). Gao et al (2022) reported anthropogenic aerosols decreased shortwave radiative force at the bottom of the atmosphere by 0.45–140.00 W m⁻² in Asia. Thus, the radiative effect can influence ozone formation via its impact on thermal and photolysis reactions. Previous studies revealed that aerosol direct effects lead to a lower photolysis rate and less ozone. (Xing et al 2017, Mukherjee et al 2020, Qu et al 2021, Zhao et al 2021). Mukherjee et al (2020) reported that the direct radiative effects of black carbon reduced more than 30% ozone concentrations by changing the photolysis rate in South Asia during a heavy dust and biomass burning event. Besides, aerosol direct effects can change meteorological factors, such as the temperature, wind speed, humidity, planetary boundary layer height, by influencing the energy balance in atmosphere (Li et al 2017d, Wu et al 2020, Wang et al 2022b). Li et al (2022) estimated that aerosol radiative effect decreased 2 m air temperature and planetary boundary layer height by 0.4 °C–1.8 °C and 30.9–183.6 m, respectively, during severe winter haze events in the North China Plain. Impact of aerosol radiative effects on meteorological factors is proven to influence O₃ concentration. Xing et al (2017) found that changes of atmospheric dynamics caused by aerosol radiative effect decreased the daily maximum 1 h O₃ in January by up to 39 µg m⁻³ but increased the peak ozone in July by up to 4 µg m⁻³ in China. Moreover, Qu et al (2021) found that aerosol direct effects also influenced ozone concentration through the vertical transmission of water vapor. They projected that the annual ozone concentration decreased by 14.9%, 8.7%, 4.3% and 7.1% in JJJ, YRD, PRD and SCB, respectively, due to the combined impact of aerosol direct effects.

Regarding the impact of aerosol radiative effects on ozone, some studies have pointed out that rapid aerosol reduction in China may tend to enhance ozone production. According to Zhao et al (2021), from 2005 to 2019, the photolysis rate of nitrogen dioxide in Beijing increased by 5 × 10⁻³ s⁻¹ per year, while the annual aerosol effect decreased in this period. Wang et al (2020a) got the similar conclusion that the decrease of particles enhanced solar radiation and thus led to an increase in the photolysis rate of nitrogen dioxide of 3.6 ± 0.8% per year. However, whether the aerosol radiative effects play an important role in ozone variation is contentious. Ma et al (2021) showed that aerosol radiative effects contributed 23% of the summertime ozone change in the NCP from 2013 to 2019. Zhu et al (2021a) proved aerosol radiative effects contributed to a 7.8% ozone rise over the NCP during COVID-19. However, Li et al (2019b), Liu and Wang (2020b), and Li et al (2021) all showed that the influence of aerosol radiative effects on ozone was weak, generally less than 1 ppb MDA8 O₃ from 2013 to 2017 across China. The varied mechanism of aerosol radiative effects in different methods and models was an important reason for the difference between studies, which should be set more carefully and accurately.

6. Influence of climate change on PM_{2.5} and O₃

As one of the important components of the earth’s ecosystem, human activities have been proven to significantly influence climate. According to AR6 by the IPCC, the total anthropogenic driving effective radiative forcing (ERF) increased by 2.72 (very likely (5%–95%) range is 1.96–3.48) W m⁻² from 1750 to 2019. Among the contributing forcing agents, ozone and aerosols contributed 0.47 (0.24–0.71) W m⁻² and −1.1 (−1.7 to −0.4) W m⁻² to ERF, respectively. Climate change could in turn affect the production, transport, and deposition of PM_{2.5} and O₃ (Neu et al 2014, von Schneidemesser et al 2015, Xu et al 2017, Fu and Tian 2019, Lu et al 2019b, Cao and Yin 2020, Chen et al 2020e). The impact of a future warming climate without anthropogenic emissions change on air quality is defined as the ‘climate change penalty’ (Fiore et al 2012). Previous studies have shown that increasing regional stagnation enhances PM_{2.5} and O₃ concentrations (Zhang et al 2018, Liu et al 2021a, Liu et al 2021b). For ozone, increasing contributions from biogenic CH₃ and soil NOₓ emissions were expected (Xie et al 2017, Ma et al 2019a). Moreover, STE was likely to increase in the future. Meul et al (2018) reported that the global mean annual influx of STE would increase by 53% from 2000 to 2100 under the Representative Concentration Pathway 8.5 (RCP8.5) scenario. For PM_{2.5}, the influence of meteorological variables (e.g. mixing
depth and precipitation) cannot be neglected (Jacob and Winner 2009, Fiore et al 2013) (in contrast to O$_3$). The impacts of climate-sensitive processes and meteorological variables on PM$_{2.5}$ and O$_3$ were discussed in detail by Jacob and Winner (2009), Fiore et al (2012) and Fu and Tian (2019).

Many studies forecasted PM$_{2.5}$ and O$_3$ concentrations under climate change scenarios (Special Report on Emission Scenarios (SRES), RCPs, and Shared Socioeconomic Pathways (SSPs)) proposed by the Intergovernmental Panel on Climate Change (IPCC, O’Neill et al 2013, 2017). Turnock et al (2020) estimated that PM$_{2.5}$ and O$_3$ would decrease across all regions globally by up to 12 $\mu$g m$^{-3}$ and 15 ppb in the late 20th century under SSP126 scenario, but would increase in most regions by up to 8 $\mu$g m$^{-3}$ and 12 ppb under the SSP370 scenario. Hong et al (2019) found that compared with 2006–2010, climate change (air pollutant emissions were fixed) increased the mean annual PM$_{2.5}$ concentration and ozone season (April to September) averages of MDA1 O$_3$ by up to 9 $\mu$g m$^{-3}$ and 2–8 ppb across eastern China in 2046–2050 under the RCP4.5 scenario. However, studies on climate change always exhibit large uncertainties, due to dependence on model and scenario assumptions (Rao et al 2017, Tokarska et al 2020). Even if anthropogenic pollutant emissions are expected to decrease in China to in the context of climate change (Xing et al 2020b), climate change would result in additional challenges for air quality management.

7. Conclusion and outlook

In this study, we summarize the spatiotemporal distribution and correlation of PM$_{2.5}$ and O$_3$, and review the synergistic effects between PM$_{2.5}$ and O$_3$ as influenced by three factors: (a) anthropogenic precursor emissions; (b) the interaction between PM$_{2.5}$ and O$_3$ through atmospheric processes; (c) the impact of climate change.

According to surface observation data, PM$_{2.5}$ concentration in China has decreased by more than 50% since 2013, however the O$_3$ concentration, particularly in urban areas, has increased. According to literature review, the main factor influencing changes in PM$_{2.5}$ and O$_3$ is the evolution of anthropogenic emissions. Sources and components of PM$_{2.5}$ also have shown great changes. We mainly focus on the increase of secondary components and nitrate in NCP. For O$_3$, change of anthropogenic emissions have an impact on O$_3$ formation regime, which tend to be more sensitive to NO$_x$. The studies on co-control of PM$_{2.5}$ and O$_3$ generally point to the importance of VOC emissions reduction in the future. Furthermore, we review three important atmospheric processes related to interactions between PM$_{2.5}$ and O$_3$. Numerous studies suggest that increased atmospheric oxidation capacity is crucial for the growth of secondary components as well as O$_3$. Many studies have suggested the significance of heterogeneous reactions and the radiative effect when considering how PM$_{2.5}$ affects ozone. Finally, we review how climate change affects China’s PM$_{2.5}$ and O$_3$ levels. The co-control of PM$_{2.5}$ and O$_3$ is challenging due to the high level of uncertainty in this field.

Overall, the following gaps exist and need to be explored in the future:

(a) Despite of the rapid increase of air pollutants monitoring sites in China, there still lack the observation data of PM$_{2.5}$ components, key oxidants and VOC species. We expect future studies based on monitoring to provide basis for mechanistic understanding.

(b) Though the heterogeneous reactions and aerosol radiative feedback are significant, relevant mechanism and parameterization scheme are missing or unclear in three-dimensional air quality model. We recommend the development of models based on solid experiments.

(c) Influence of climate change on PM$_{2.5}$ and O$_3$ is hard to be clarified by limited observation data and field measurements, which makes research on pertinent mechanisms more challenging. Considering the synergistic effect of PM$_{2.5}$ and O$_3$ through atmospheric processes, we look forward to the progress of research on how these atmospheric processes change and influence PM$_{2.5}$ and O$_3$ in the future. We also advise the research focusing on China with various climate models and scenarios.

(d) As the common precursors of PM$_{2.5}$ and O$_3$, a coordinated control strategy of NO$_x$ and VOCs is crucial, but with lots of limitations in past research. Based on advanced models, we propose that a control strategy should take into account different meteorological conditions, regional emission characteristics, and the feasibility of implementing actual control measures.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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