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Long-term trend of ozone pollution in China during 2014-2020: distinct seasonal and spatial characteristics

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Abstract: In the past decade, ozone (O_3) pollution has become a severe environmental problem in major cities in China. Here, based on available observational records, we investigated the long-term trend of ozone pollution in China during 2014–2020. Ozone concentrations were higher in urban areas than in non-urban areas. During these seven years, the highest ozone concentrations primarily occurred in summer in northern China, and in autumn or spring in southern China. Although ozone precursors, including nitrogen oxides (NO_X) and carbon monoxide (CO), continuously decreased throughout the seven years, four ozone metrics that were used to characterize ozone exposure levels increased from 2014 to 2017 and reached a plateau after 2017. The long-term trend of ozone concentrations differed across seasons; especially from 2019 to 2020 when ozone concentrations decreased in summer and increased in winter. To analyze the causes of this observed trend, a photochemical box model was used to investigate the change in ozone sensitivity regime in two representative cities – Beijing and Shanghai. Our model simulations suggest that the summertime ozone sensitivity regime in urban areas of China has changed from a VOC-limited regime to a transition regime during 2014–2020; by 2020, the urban photochemistry is in a transition regime in summer but in a VOC-limited regime in winter. This study helps to understand the distinct trends of ozone in China and provides insights into efficient future ozone control strategies in different regions and seasons.

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1 Introduction

Tropospheric ozone (O_3) is an air pollutant that is detrimental to human health, vegetation and ecosystem productivity (Ainsworth et al., 2012;Mills et al., 2018;Monks et al., 2015;Fiore et al., 2009). The inhalation of ozone impairs the functioning of the human respiratory and cardiovascular systems through its reaction with the lining of the lung and other surfaces in the respiratory tract (Jindal, 2007). Ozone is also an important greenhouse gas that leads to positive radiative forcing (Stocker et al., 2014). A comprehensive characterization of the spatial (latitude, longitude and altitude) and temporal distribution of tropospheric ozone is critical to our understanding of these issues. Here we summarize this distribution over China from the available observational records to the extent possible.

China has undergone rapid economic development, leading to higher demand for energy, and greater usage of fossil fuels during the past several decades. As a result, high anthropogenic emissions to the atmosphere have produced severe ozone pollution in urban areas of China, where daily maximum 8-hour average (MDA8) ozone concentrations often exceed the standard of 80 ppb (Li et al., 2014;Li et al., 2019a;Zhang et al., 2014;Lu et al., 2018). In contrast to the generally decreasing ozone levels in the United States and Europe, available surface ozone observations have widely shown significant upward trends in China since 1990 in rural areas (Xu et al., 2008;Wang et al., 2009;Ma et al., 2016;Sun et al., 2016;Xu et al., 2016;Xu et al., 2018), urban areas (Li et al., 2019a;Wang et al., 2020;Zhang et al., 2014;Lu et al., 2018), and over regional scales (Verstraeten et al., 2015;Xu and Lin, 2011). China has become a global hot spot of ground-level ozone pollution. The present annual mortality attributed to long-term ozone exposure in China is estimated to be ~50,000 to 316,000 deaths (Liu et al., 2018;Malley et al., 2017).

Global model simulations suggest that the average lifetime of ozone in the troposphere is about 22 days (Stevenson et al., 2006;Young et al., 2013). In the free troposphere at northern midlatitudes, where prevailing westerly winds dominate, the

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net ozone lifetime is considerably longer, and is greater than the transport time around the Earth (Trickl et al., 2011). Consequently, the increase in ozone in China not only influenced domestic public health, but also influenced downwind countries (Brown-Steiner and Hess, 2011;Lin et al., 2012) and thus increased global background ozone concentrations. Several studies indicate that rising Asian emissions influenced baseline ozone concentrations in America and Europe through the hemispheric transmission of ozone and its precursors (Cooper et al., 2010;Verstraeten et al., 2015). The baseline ozone concentrations at northern midlatitudes increased at an average rate of ~ 0.60 ppb year^{-1} from 1980 to 2000 (Parrish et al., 2020). Such an increase in baseline ozone concentrations makes it more difficult to further reduce ozone in America and Europe. Therefore, a detailed characterization of ozone pollution in China will aid understanding of the variation in baseline ozone and guide the reduction of ozone throughout northern midlatitudes.

Despite several studies of the ozone trends in specific rural or urban sites in China (Ma et al., 2016;Gao et al., 2017;Zhang et al., 2014;Lu et al., 2018), detailed characterization of spatial distribution and temporal trend of ozone in different seasons across China remains scarce. In addition, it is not yet well understood how changes in precursor emissions influence the trend of ozone in China. Chinese government launched the Air Pollution Prevention and Control Action Plan in 2013–2017 and the Clean Air Action plan in 2018–2020 to reduce anthropogenic emissions (Cheng et al., 2019). Since 2013, the surface monitoring network has been greatly expanded, and detailed hourly data across China became available from China Ministry of Ecology and Environment (https://quotsoft.net/air/). The goal of this study is to elucidate the spatial distribution, seasonal variation and temporal trends of ozone in China by using these surface ozone observations to characterize multiple ozone metrics relevant to human health. Quantifying the detailed spatial distribution and temporal trend of ozone across China will provide a better understanding of the response of ozone to emission reductions, and inform the development of control measures to effectively mitigate ozone in the future.

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2 Method

2.1 Measurements

Hourly surface O_3, nitrogen dioxide (NO_2) and carbon monoxide (CO) concentrations during 2014–2020 were obtained from the public website of the China Ministry of Ecology and Environment (MEE) (https://quotsoft.net/air/). The surface monitoring network covered 940 stations in summer 2014, and extended to 1669 stations by 2020 (~330 cities). The O_3 concentrations at 750 sites with continuous observations over these 7 years were analyzed. These measurements document the air quality in Chinese cities and have been analyzed in recent studies (Li et al., 2019a;Lu et al., 2018). Total solar radiation data in 2013 were acquired from the meteorological data set of fundamental meteorological elements of China national weather station (V3.0), but data after 2013 are unavailable.

Urban and non-urban sites are distinguished by population density. Population density data were acquired from Gridded Population of the World (GPW), v4; https://sedac.ciesin.columbia.edu/data/set/gpw-v4-population-density-rev11. This is a dataset of the world population gridded at ~5 km resolution. According to the China Statistical Yearbook in 2018 (China Statistics Press, 2018), China's urban population density was 2500 people/km^2. In the following analysis, urban sites correspond to population density ≥2500 people/km^2, and non-urban sites correspond to population density <2500 people/km^2.

To reflect the breadth of different health-related indicators used globally, four metrics—AVGMDA8, 4MDA8, NDGT70, 3MMDA1—are used here to characterize ozone pollution levels. The definitions of these metrics are given in **Table 1**. AVGMDA8 represents the mean value of daily maximum 8 h average (MDA8) ozone concentrations, and 4MDA8 represents the annual 4th highest (MDA8) ozone concentrations; 3MMDA1 represents the annual maximum of the 3-month running mean of the daily maximum 1-hour (MDA1) ozone concentrations. Since these metrics are determined from different parts of the distribution of ozone concentrations, their

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spatial distribution and temporal variation may differ. We derived all metrics from the hourly measurements that were filtered by data quality control procedures following the Tropospheric Ozone Assessment Report (TOAR) data completeness requirements and procedures. The calculations of AVGMDA8, 4MDA8, NDGT70 are based on MDA8 ozone concentrations. MDA8 ozone concentration is the maximum value of 8-hour running averages calculated from 0 h to 23 h local time. Note that if the data availability at a certain site is less than 60% (i.e., less than 5 hours for 8-hour averages or 15 hours for one day), the MDA8 value was considered missing. For the calculation of these metrics (AVGMDA8, 4MDA8, NDGT70), if less than 60% of MDA8 values are available (i.e., less than 220 MDA8values for a year or 55 MDA8 values for a season), the annual mean or seasonal mean values of the metrics at a certain site were considered missing. The calculation of 3MMDA1 was based on the daily maximum 1-hour (MDA1) ozone value. Similar to the MDA8 ozone value, if less than 60% of data are available (i.e. less than 15 hours for one day), the MDA1 value at a certain site was considered missing. For the calculation of 3MMDA1, if less than 60% of MDA1 values are available (i.e. less than 55 MDA1 values for three months and 220 MDA1 values for a year), the 3MMDA1 value at a certain site was considered missing.

Beijing and Shanghai are the two largest cities in China and have undergone severe ozone pollution in the past decade (Wang et al., 2020;Xu et al., 2019). Measurement data in Beijing and Shanghai were analyzed to show the variation characteristic of ozone sensitivity regimes. The data of O_3, NO_2 and CO were acquired from the public website of the China Ministry of Ecology and Environment (https://quotsoft.net/air/). Volatile organic compounds (VOCs), and meteorological factors (including temperature, relative humidity, photolysis frequencies and air pressure) were measured during 2014–2020 at two representative urban sites in Beijing and Shanghai: Peking University and Shanghai Academy of Environmental Sciences. Temporal trends and composition of VOCs at the two sites were considered to be representative across Beijing and Shanghai (Wang et al., 2010;Xu et al., 2011;Zhang et al., 2012). VOCs were measured using a commercial GC-FID/PID system (Syntech Spectra GC955 series 600/800 analyzer).

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2.2 Zero-dimension photochemical box model

A zero-dimension photochemical box model was used to simulate the sensitivity of ozone production and loss to its precursor concentrations. Compared with regional 3-D models, the box model has the advantage that it can be constrained by comprehensive measurements to eliminate the uncertainty from emission inventories. The box model includes MCM v.3.3.1 as the chemical mechanism. Hourly averages of CO, NO_2, NO, O_3, SO_2, VOCs (56 species), formaldehyde, acetaldehyde, photolysis frequencies, temperature, air pressure, and relative humidity were used as model constraints. HONO was not measured, and thus was calculated according to the concentration of NO_2 and the observed ratio of HONO to NO_2 in Beijing (Hendrick et al., 2014). The model simulations were performed in a time-dependent mode with spin-up of two days. For physical removal processes, a 24-h lifetime was assumed for all simulated species, which approximately simulates the effects of dilution and surface deposition. This modeling approach has been used previously (Wang et al., 2019;Wang et al., 2020).

RO_2, HO_2 and OH radicals were simulated by the box model to calculate the net ozone production rate ($P(O_3)$) and ozone loss rate ($L(O_3)$)as shown in Equation (1) and (2) as derived by Mihelcic et al. (2003).

$$P(O_3) = k_{HO_2+NO}[HO_2][NO] + \sum_i (k^i_{RO_2+NO}[RO^i_2][NO]) - k_{OH+NO_2}[OH][NO_2] - L(O_3) \quad (1)$$

$$L(O_3) = (\theta j(O^1D) + k_{OH+O_3}[OH] + k_{HO_2+O_3}[HO_2] + \sum_j (k^j_{alkene+O_3}[alkene^j])[O_3] \quad (2)$$

where θ is the fraction of O^1D from ozone photolysis that reacts with water vapour, and i and j represent the number of species of RO_2 and alkenes, respectively.

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3 Results and Discussion

3.1 The spatial distribution and seasonal variation of ozone pollution

Figure 1 presents the spatial distributions of the mean values of four ozone metrics (AVGMDA8, 4MDA8, NDGT70 and 3MMDA1) at non-urban and urban sites in China during 2014–2020. The spatial distribution was similar between urban and non-urban sites for all four metrics; for example, warm-season AVGMDA8 O_3 concentrations at 74% of urban sites and 67% of non-urban sites exceed the air quality standard Grade 1 limit of 50 ppb. Hot spots of ozone pollution mainly occurred in the more economically developed areas in northern, eastern and central China. At both urban and non-urban sites, the highest regional average ozone concentrations occur in the North China Plain with the average warm-season AVGMDA8 O_3 concentration of 66 ppb, significantly higher than the corresponding national average value of 54 ppb. Although the solar radiation in the North China Plain is not the strongest across China (Jiang et al., 2019), North China Plain has a large density of urban and industrial activities. Previous studies denote that the North China Plain has the highest NO_X and VOCs emissions over China (Liu et al., 2016;Li et al., 2019b). This clearly indicates that ozone pollution is closely related to anthropogenic activities.

The month in which the 3MMDA1 ozone concentration occurred is defined as the middle month in the 3 months of 3MMDA1, which can indicate the season when maximum ozone pollution occurred. As shown in **Fig. 2**, the month in which the 3MMDA1 ozone concentration occurred shows a significant spatial variation. In most years, the 3MMDA1 ozone concentration in northern China (north of the Yangtze River) occurred mainly in summer (June, July and August), whereas in southern China (south of the Yangtze River), it occurred in autumn (September, October and November) or spring (March, April, May). In northern China, sunlight intensity is highest in summer and photochemical production from anthropogenic and biogenic precursors maximizes. In southern China, the southwest monsoon prevails in summer leading to an inflow of marine air with low ozone concentrations and reduced photochemical ozone production

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due to more cloudy and rainy weather (Yin et al., 2019); thus in this region the highest ozone usually appears in autumn when sunlight intensity maximizes.

It is notable that the 3MMDA1 ozone concentration mainly occurred in spring in both Heilongjiang and Yunnan provinces, which are located in northeast and southwest China, respectively. This is consistent with a previous study reporting that Yunnan province and northeast China had peak O_3 in spring 2014–2017 (Yin et al., 2019). A springtime maximum was also found for the column O_3 in Yunnan retrieved from satellite data (Xiao and Jiang, 2013). The occurrence of maximum ozone concentrations in spring has been attributed to several factors, including 1) the peak occurrence of stratospheric intrusions, 2) photochemistry of precursors built up during winter, and 3) biomass-burning either as forest fires or for land clearance (Monks et al., 2015). Heilongjiang province is located in the northernmost part of China (43°26′N–53°33′N) with relatively low temperature and light intensity, and thus its photochemical production of ozone is weak all year round. We surmise that the springtime maxima of ozone in this province is due to the first two causes: the stratospheric intrusion of ozone in spring (Stohl et al., 2003), and ozone production in spring from accumulated precursors that were emitted from coal burning for heating during the wintertime. Yunnan province is located in a plateau area with average altitude of 2000 m; the elevated terrain of this province is more likely to be influenced by the descending free tropospheric air masses with high ozone concentrations from the stratospheric origin (Stohl et al., 2003;Cooper et al., 2012). Additionally, higher sunlight intensity in spring at this lower latitude province is also conducive to photochemical production of ozone.

We also compared the seasonal variations in MDA1 ozone concentrations in three typical Chinese city clusters, Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) (**Fig.3**). In each city cluster there is a distinct seasonal ozone pattern: a sharp unimodal distribution with a summer maximum in BTH, a broad distribution with a spring maximum in YRD, and a less distinct, unimodal distribution with an autumn maximum in PRD. Meteorological factors determine the different ozone distribution patterns; most importantly PRD and YRD received more precipitation in summer than BTH, and that PRD was especially affected by the inflow

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of marine air during the southwest monsoon. Furthermore, in PRD typhoons led to less cloud cover, and thus more solar radiation in autumn, which accelerated O_3 production (Qu et al., 2021). As shown in **Fig. 3**, the seasonal variations in ozone in the three city clusters are overall consistent with those of solar radiation in representative cities of the three city clusters (Beijing in BTH, Shanghai in YRD and Guangzhou in PRD). This result indicates that the local photochemistry driven by solar radiation plays a crucial role in ozone seasonal variations.

3.2 Temporal trend of ozone pollution

Ozone trends measured on regional scales, in urban areas, and at rural sites in previous studies are summarized in **Table 2**. For direct comparison of these results reported in different units, we have included estimated trends in units of % yr^{-1} for all studies. Xu and Lin (2011) and Verstraeten et al. (2015) have reported that tropospheric ozone concentrations increased in summer during 1979–2005 in the North China Plain and 2005–2010 in Eastern China at a rate of 1.1% and 3.0% yr^{-1}, respectively, based on satellite measurements. Urban ozone concentrations increased significantly in Beijing, Shanghai, Hongkong, Sichuan Basin and other cities during the past one to two decades at rates of 2.0 to 6.7% yr^{-1} (Gao et al., 2017;Cheng et al., 2016;Wang et al., 2020;Chen et al., 2021;Li et al., 2020;Lu et al., 2018). A significant increase in ozone (+1.6% yr^{-1}) was detected at Shangdianzi, a rural site in the North China Plain (Ma et al., 2016). A moderate increase was detected at the global background site (Waliguan site) in western China (+0.44% yr^{-1}) (Xu et al., 2018;Xu et al., 2020). No significant trend was detected at either the eastern coastal Changdao site (Wang et al., 2020) or the Longfengshan site on the northeastern edge of China (Xu et al., 2020). The only significant decrease was reported at the Akedala site on the northwestern edge of China (-3.3% yr^{-1}) (Xu et al., 2020). In general, these studies show that ozone concentrations in China have risen in the past three to four decades.

Figure 4 summarizes variations of four ozone metrics (warm season AVGMDA8, 4MDA8, 3MMDA1 and NDGT70) during 2014–2020 for Chinese urban and non-urban

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sites. **Figure A1** presents the spatial distribution of warm season AVGMDA8 ozone concentrations at urban and non-urban sites for each year during 2014–2020. These four metrics at urban sites were overall higher than at non-urban sites by 3.5%, 6.6%, 4.2% and 16%, respectively (**Fig. 4**). These results in China differ from those in Europe and North America, where the mean levels of these metrics at non-urban sites are similar to those at urban sites (Fleming et al., 2018), although there are differences in the approaches to classifying sites as urban or non-urban. The possible reason for the difference is that transported background ozone dominates over local and regional photochemical production in Europe and North America, while local photochemistry dominates ozone levels in China. From 2014 to 2020, the trends of ozone were generally similar between urban and non-urban sites. The four metrics all increased from 2014 to 2017, but remained relatively stable after 2017 despite being significantly higher in 2019. The elevated ozone level in 2019 was related to higher temperatures in the warm season (Li et al., 2020). Overall, the rapid increase in ozone concentrations in China has either slowed or ended (depending upon metric) after 2017. In **Fig. 4**, the variations in the four metrics are fitted by quadratic functions. The quadratic polynomial coefficients are all negative and statistically significant for the four metrics, which is strong evidence that the increasing trend has slowed.

Because the trends of ozone are generally similar between urban and non-urban sites (**Fig. 4**), the nationwide (including both urban and non-urban) AVGMDA8 was used to analyze ozone trends for different seasons. **Figure 5** shows variations in seasonal and annual AVGMDA8 during 2014–2020. For the national average, AVGMDA8 was highest in summer, followed by spring, autumn, and winter. This metric increased in all four seasons from 2014 to 2017, with the fastest increase in spring (3.1 ppb yr^{-1}, r^2=0.94), followed by winter (2.9 ppb yr^{-1}, r^2=0.91), summer (2.0 ppb yr^{-1}, r^2=0.90) and autumn (1.2 ppb yr^{-1}, r^2=0.81). The annual average increased at a rate of 2.0 ppb yr^{-1} (r^2=0.95) from 2014 to 2017. The more rapid increase of ozone concentration in spring than in summer resulted in a decrease in the gap between the two seasons. This is consistent with a recent study reporting that ozone pollution in the North China Plain extended to the spring season (Li et al., 2021). After 2017,

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AVGMDA8 remained relatively stable in summer and spring, but still increased significantly in autumn and winter. Compared with 2019, the seasonal average MDA8 ozone concentration decreased by 5.5 ppb in summer 2020, but increased by 5.1 ppb in winter 2020. **Figure 6** illustrates the spatial patterns of the summer and winter changes in seasonal average MDA8 O_3 from 2019 to 2020. In summer ozone decreased significantly in most regions of China, with greater decreases in central China and the North China Plain. In winter, ozone increased significantly throughout China. The cause of these changes will be discussed in Section 3.3.

The trends of the ozone precursors, NO_2 and CO, were investigated based on the observational data. As shown in **Fig. 7**, both NO_2 and CO decreased significantly from 2014 to 2020 for both annual and seasonal averages. Notably, NO_2 decreased faster after 2017 than before 2017. Both the MEIC inventory and OMI NO_2 data show a decrease during 2013-2019 (Shah et al., 2020), which is consistent with our result. The emission inventory suggests that VOC emissions were stable during 2013-2019 in China (Li et al., 2019b;Zheng et al., 2021). In 2020 VOCs, CO and NO_X emissions decreased significantly in winter but only slightly in summer, compared to 2019 (Zheng et al., 2021), which is consistent with the changes of measured NO_2 and CO (**Fig. 7**).

3.3 The impact of photochemistry on ozone temporal trend

Ozone concentrations are influenced by photochemical processes that depend on precursor concentrations and meteorological conditions. Changes in ozone precursor emissions, particularly VOC and NO_X, are the primary factors driving ozone trends in China. The relationship between O_3 and its precursor concentrations is generally nonlinear—a decrease in precursor concentrations does not necessarily result in a corresponding decrease in O_3 concentration. Differing responses of ozone production to VOC and NOx emission changes allow three ozone sensitivity regimes to be distinguished: VOC-limited, NOx-limited and transition regimes (Kleinman, 1994;Kleinman et al., 1997). In this section, based on comprehensive measurements in Beijing, the impact of photochemical regimes on the temporal trend of ozone in urban

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areas of China was discussed.

As discussed in Section 3.2, in summer when ozone pollution is most severe, ozone increased from 2014 to 2017, but remained relatively stable after 2017 (**Fig. 5**). To explore the impact of photochemical regimes on the temporal trend of ozone in summer, the zero-dimensional photochemical box model constrained by long-term measurements in Beijing and Shanghai was used to examine the variation in the sensitivity of ozone to precursor emissions. In this study, we focused on the effects of photochemistry on ozone sensitivity to precursors; transport effects were not considered. The ozone sensitivity regime was diagnosed by testing the response of $P(O_3)$ as calculated from Equation (1) to the changes of VOCs and NO_X concentrations (**Fig. 8**). The box model simulations suggest that in Beijing VOC reduction would significantly decrease ozone during all seven years, while NO_X reduction would significantly increase ozone during 2014–2017 but only slightly increase ozone in 2018 and slightly decrease ozone during 2019–2020. The 2014–2018 results are consistent with the VOC-limited regime in which a reduction in VOCs is effective in mitigating ozone production, while a reduction in NO_X increases ozone production. The 2019–2020 results are consistent with the transition regime in which reductions of either VOCs or NO_X can decrease ozone production. These results indicated that the summertime photochemical environment in Beijing shifted from a VOC-limited regime to a transition regime. The Shanghai simulations show similar behavior in terms of the shift in the photochemical regime.

Previous 3-D model studies have reported results similar to our box model simulation; urban areas in China were in the VOC-limited regime in the summer of 2013-2017, but in the transition regime after 2017 (Shao et al., 2021;Kang et al., 2021;Li et al., 2019a). Tang et al. (2021) showed that ozone production in Beijing was transitioning from VOC-sensitive to NO_X-sensitive over the 2013-2018 period. The sharp decrease in NO_X combined with a smaller change in VOCs in Shanghai has led to a shift in the O_3 production from a VOC-limited regime to a NO_X-limited regime over the past decade (Xu et al., 2019). In addition to model studies, satellite-observed $HCHO/NO_2$ ratios also indicate that there is a shift in the North China Plain, YRD and

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PRD from the VOC-limited regime to the transitional regime, which is associated with a rapid drop in anthropogenic NO_X emissions from 2016 to 2019 (Wang et al., 2021). These studies agree that ozone sensitivity in summer in urban areas of China has gradually changed from a VOC-limited regime to a transition or NO_X-limited regime due to faster decreases in NO_X emissions than in VOC emissions over the past decade. Therefore, we surmise that the rapid increase of summertime ozone during 2013-2017 is due to the decrease in NO_X under VOC-limited conditions, and that the slowing of the summertime ozone increase after 2017 is due to decreased NO_X emissions and relatively stable VOC emissions under the transition regime conditions. This finding lends more confidence to the effective reduction in summertime ozone through continued reductions in VOC and NO_X emissions.

Another issue is that compared to 2019, MDA8 ozone concentrations decreased in summer but increased in winter in 2020 (**Fig. 5 and 6**) despite the decrease of both NO_X and CO concentrations (**Fig. 7**). Based on measurements in Beijing in 2019, the observation-based box model was used to examine the sensitivity of ozone to precursors in summer and winter. As shown in **Fig. 9**, in the summer of 2019, Beijing was in the transition regime, when reductions in VOCs and NO_x both decreased the integrated ozone production rate. In winter it was in the VOC-limited regime, when reduction in VOCs decreased, but reduction in NO_x increased, the ozone production rate. This result demonstrates that summer and winter had different ozone sensitivity characteristics in 2019. Based on WRF-Chem model simulations, Kang et al. (2021) also reported that ozone sensitivity entered the transition or NO_X-limited regime in summer 2020, but was still in the VOC-limited regime in winter 2020. In addition, the WRF-Chem model results by Le et al. (2020) indicate that the chemical regime was VOC-limited during the COVID-19 pandemic lockdown in winter 2020 in China and the decrease in NO_X led to significant ozone increases. These studies are consistent with our simulation results in Beijing. The difference in ozone sensitivity regimes between winter and summer is likely to be a crucial cause of opposite ozone changes between winter and summer in 2020. Although ozone production rates and concentrations are much smaller in winter, ozone can influence particulate matter (PM) formation through increasing the atmospheric oxidizing capacity in this season (Le et al., 2020). Therefore, different ozone sensitivity regimes between winter and summer should be fully considered to effectively mitigate both ozone and PM in the two seasons.

4 Conclusions

During the past decade, China has devoted substantial resources to improving the environment. These efforts reduced atmospheric particulate matter loading, but ambient ozone levels increased (Shao et al., 2021). We present a detailed characterization of the spatial distribution and temporal trend of ozone over China based on nationwide hourly ozone observations, and find that:

(1) Maximum ozone concentrations primarily occur in summer in northern China, but in autumn or spring in southern China. Meteorological factors, especially solar radiation and the southwestern monsoon, play key roles in the regional contrast of the seasonal variations.

(2) Four ozone metrics (AVGMDA8, 4MDA8, NDGT70, 3MMDA1) increased from 2014 to 2017, and remained relatively stable after 2017. These metrics were generally higher at urban sites than at non-urban sites. The trend of ozone concentrations differed across seasons; especially from 2019 to 2020 when ozone concentrations decreased in summer and increased in winter.

(3) Simulations by an observationally constrained box model and previous 3-D model simulations agree that the ozone sensitivity in summer in urban areas of China gradually changed from the VOC-limited regime to a transition regime. This increases our confidence in the reduction of both VOC and NO_X emissions as an effective approach to further reducing summertime ozone. Box model simulations also indicate that the urban photochemistry is still in the VOC-limited regime in winter in 2020.

Our study provides an improved understanding of the past and future response of ozone to emission reductions in China, and can inform control measures for effective future reductions of ozone.

Data availability

The observational data and model code used in this study are available from corresponding authors upon request (h.su@mpic.de).

Author contributions

HS and WW designed the research. WW and HS prepared the manuscript with contributions from other authors. WW performed data analysis with contributions from DP, SW, RN, FB and YC. HW, XL, SY collected data.

Competing interests

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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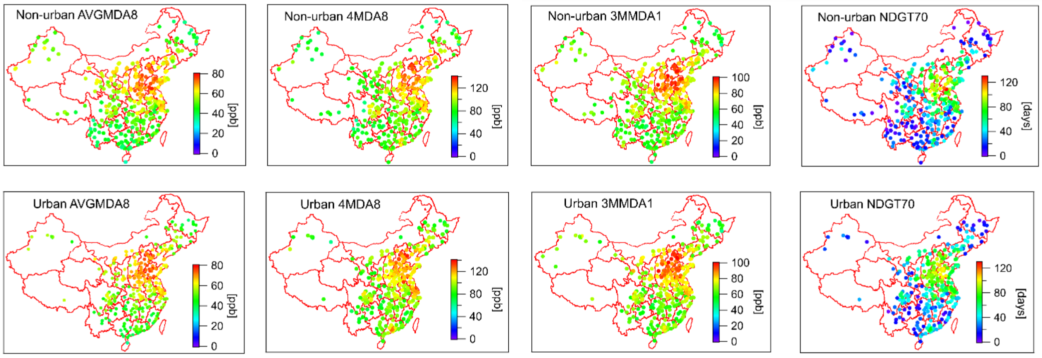

Figure 1. Spatial distribution of four ozone metrics (AVGMDA8, 4MDA8, 3MMDA1, NDGT70) at urban and non-urban sites averaged over 2014–2020. AVGMDA8 is the mean MDA8 O_3 in the warm season (April to September); other metrics are annual values.

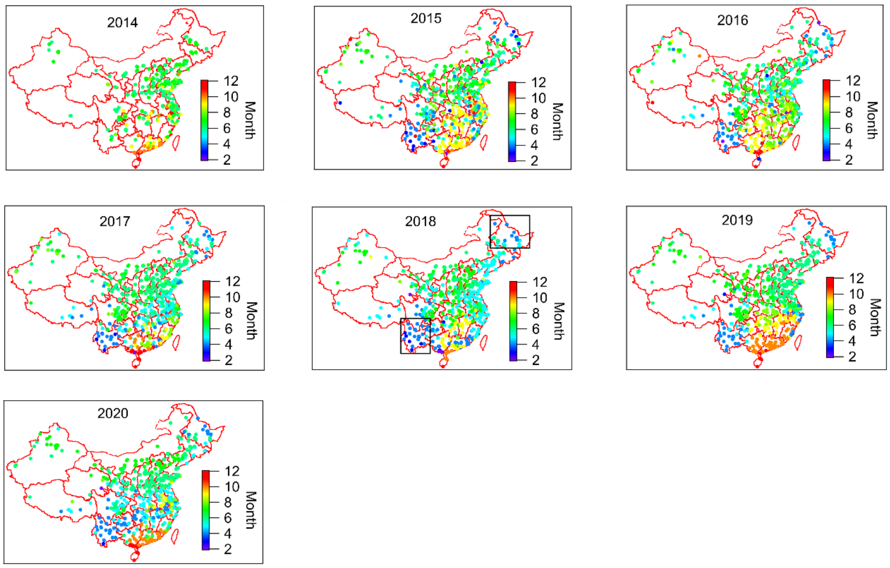

Figure 2. Spatial distribution of the month in which 3MMDA1 ozone concentration occurred during 2014-2020. Rectangles included in the 2018 map in the northeast and southwest China represents the Heilongjiang and Yunnan provinces, respectively.

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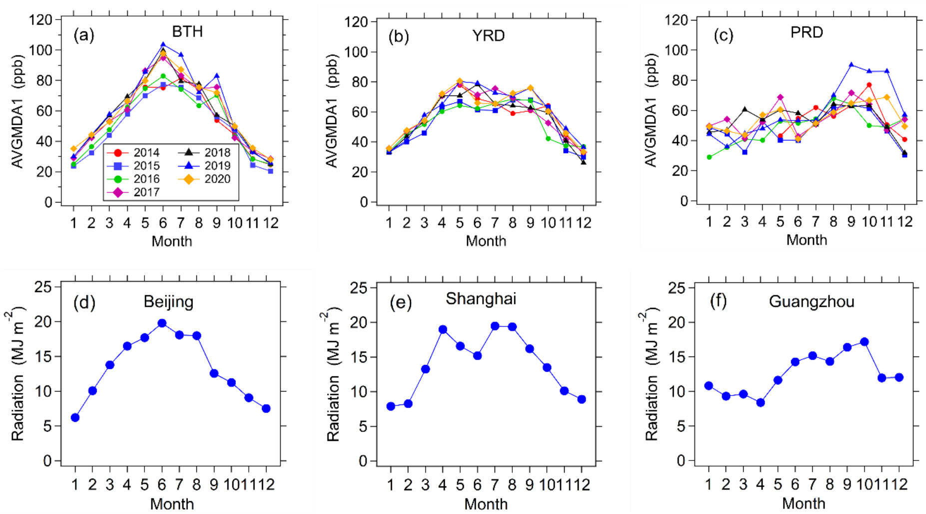

Figure 3. (a), (b) (c): Seasonal variations in monthly mean MDA1 ozone concentrations over all sites in BTH, YRD and PRD during 2014-2020. (d), (e), (f): Seasonal variations in monthly mean solar radiation in representative cities of the three city clusters (Beijing in BTH, Shanghai in YRD and Guangzhou in PRD) in 2013.

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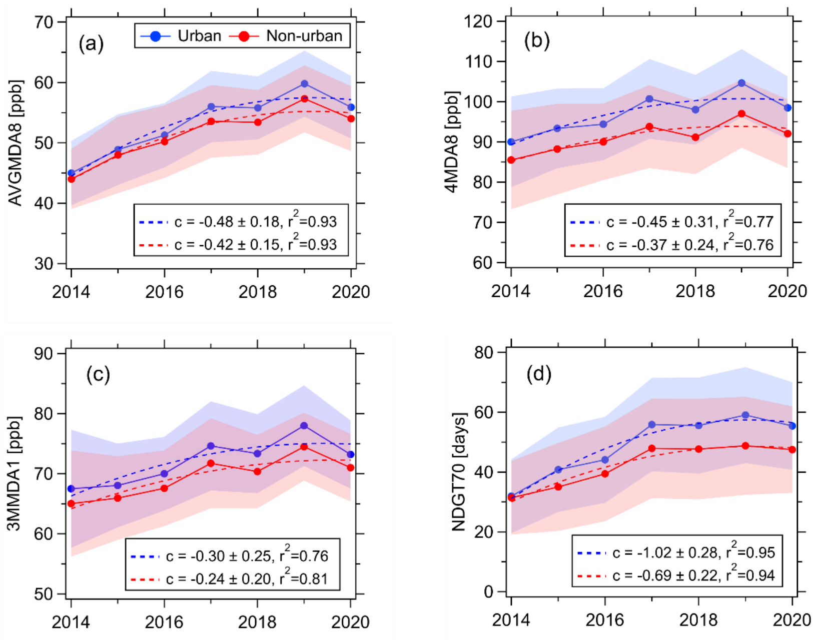

Figure 4. Variations in four ozone metrics (AVGMDA8, 4MDA8, 3MMDA1 and NDGT70) at urban and non-urban sites during 2014-2020. AVGMDA8 is the mean MDA8 O_3 in the warm season (April to September), and the other metrics are annual values. Shaded areas represent the range of mean values ± the 50% standard deviation for each metric. The dashed lines are fitted by the polynomial function ($y=a+bx+cx^2$). The quadratic polynomial coefficient c (± one standard deviation) and the determination coefficient r^2 are given.

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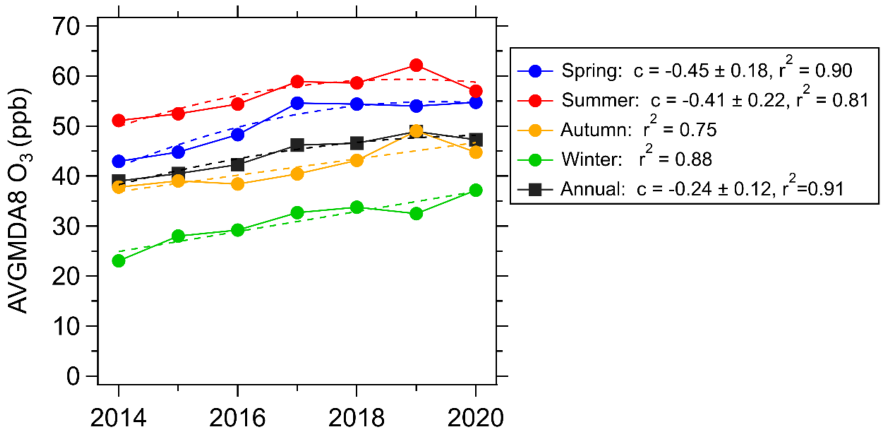

Figure 5. Variations in seasonal and annual AVGMDA8 O_3 levels during 2014–2020. The trends for spring, summer and annual averages are fitted by the polynomial function ($y=a+bx+cx^2$) and the trends for autumn and winter are fitted by the linear function ($y=a+bx$). The quadratic polynomial coefficient c (± one standard deviation) and the determination coefficient r^2 are given.

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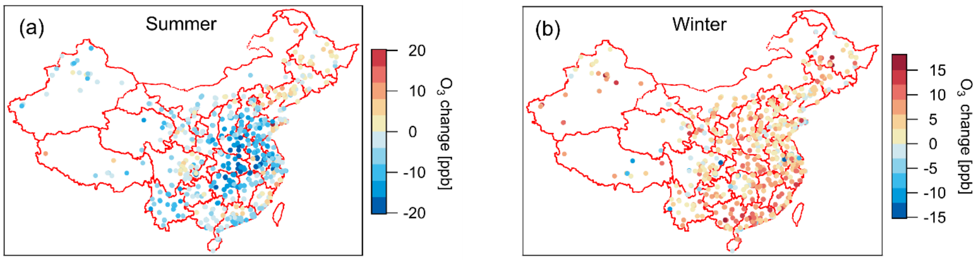

Figure 6. The change in seasonal averages of MDA8 O_3 from 2019 to 2020 in summer (a) and winter (b), China.

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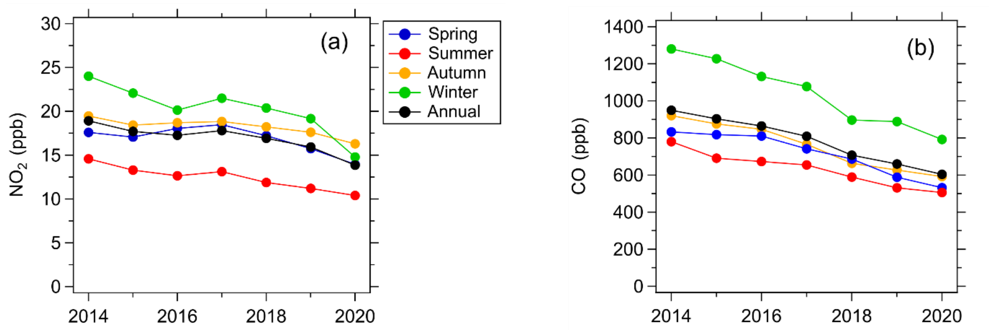

Figure 7. Variations in seasonal and annual average concentrations of NO_2 and CO measured during 2014–2020 in China.

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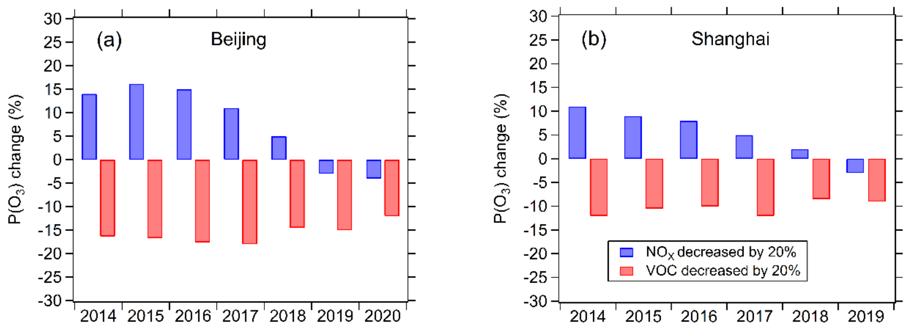

Figure 8. Sensitivity of summertime mean daytime ozone production rate [$P(O_3)$] to VOCs and NO_X simulated by the photochemical box model during 2014–2020 in Beijing (a) and Shanghai (b). VOCs and NO_X are decreased by 20% to test the fractional change of $P(O_3)$.

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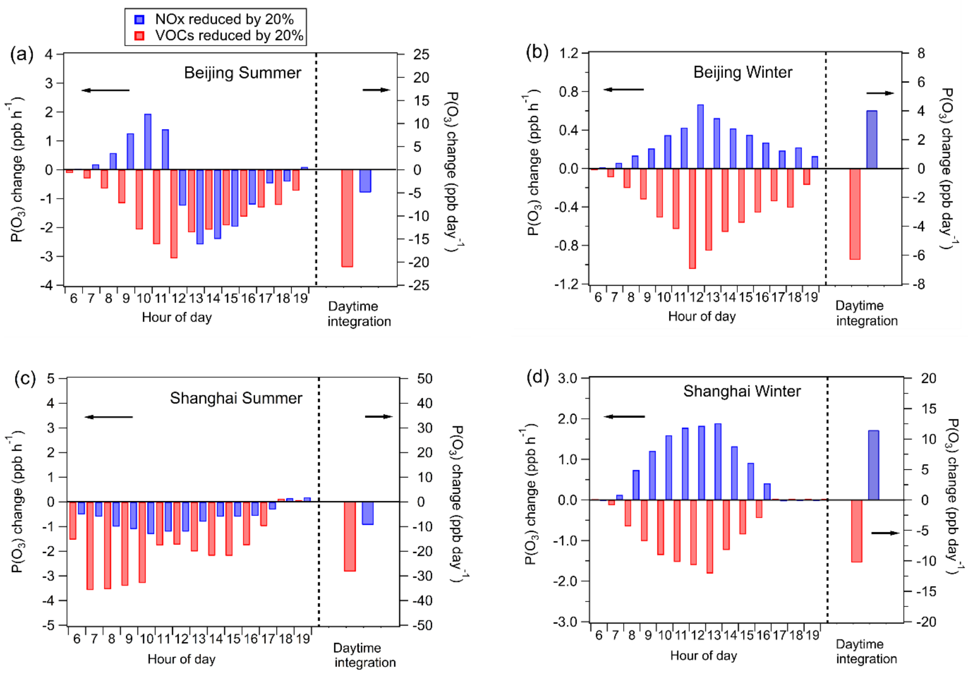

Figure 9. Sensitivity of ozone production rate [P(O_3)] to 20% reductions in VOCs and NO_X emissions for summer and winter 2019 in Beijing and Shanghai.

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Table 1. Description of ozone metrics used in this study.

Metric	Definition
MDA8 (ppb)	daily maximum 8 h average, AVGMDA8 represents mean MDA8 O_3 in the focused period.
MDA1 (ppb)	daily maximum 1 h average; AVGMDA1 represents mean MDA1 in the focused period
4MDA8 (ppb)	The annual 4th highest MDA8 O_3.
NDGT70 (days)	The annual total number of days with MDA8 O_3 >70 ppb.
3MMDA1	The annual maximum of the 3-month running mean of the daily maximum 1-hour ozone value. This metric has been used to quantify mortality attributable to long-term ozone exposure. The month in which the 3MMDA1 ozone concentration occurred is the middle month in the 3 months of 3MMDA1.

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Table 2. The reported trends of ozone concentration in China.

Spatial scale	Region	Period	Metrics	Ozone trend	References
Regional scale	Eastern China	2005–2010	average ozone column	+ 0.23 DU (+1.1%) yr^{-1}	Verstraeten et al. (2015)
	North China Plain	1979–2005	average tropospheric ozone residual	+1.1 DU (~+3%) $decade^{-1}$	Xu and Lin (2011)
Urban areas	Beijing	2006–2016	MDA8	+2.6 ppb (+3.3%) yr^{-1}	Wang et al. (2020)
	Beijing	1995–2005	daytime average	+1.0 ppb (+2.0%) yr^{-1}	Ding et al. (2008)
	Beijing	2001–2006	daily average	+1.1 ppb (+4.1%) yr^{-1}	Tang et al. (2009)
	Beijing	2002–2010	average ozone column	+1.6 DU (+3.1 %) yr^{-1}	Wang et al. (2012)
	Beijing	2004–2015	MDA8	+1.14 ppb (+2.9%) yr^{-1}	Cheng et al. (2016)
	Shanghai	2006–2016	daily average	+1.1 ppb (+6.7%) yr^{-1}	Gao et al. (2017)
	Hong Kong	1994–2007	daytime average	+0.58 ppb (+2.0%) yr^{-1}	Wang et al. (2009)
	Pearl River Delta	2006–2019	95^{th} percentile	+0.71 ppb (+1.3%) yr^{-1}	Li et al. (2022)
	Sichuan Province	2013–2020	MDA8	+2.0 ppb (+4.8%) yr^{-1}	Chen et al. (2021)
	Chinese urban sites	2013–2019	MDA8	+2.4 ppb (+5%) yr^{-1}	Lu et al. (2020)
Rural sites	Shangdianzi	2004–2016	daytime average	+0.67 ppb (+1.6%) yr^{-1}	Ma et al. (2016); Xu et al. (2020)
	Waliguan	1994–2016	daytime average	+0.21 ppb (+0.44%) yr^{-1}	Xu et al. (2020)
	Akedala	2009–2016	daytime average	-1.3 ppb (-3.3%) yr^{-1}	Xu et al. (2020)
	Longfengshan	2005–2016	daytime average	No trend	Xu et al. (2020)
	Lin'an	2005–2016	daytime average	No trend	Xu et al. (2020)
	Xianggelila	2007–2016	daytime average	No trend	Xu et al. (2020)
	Changdao	2013–2019	MDA8	No trend	Wang et al. (2020)

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Appendix

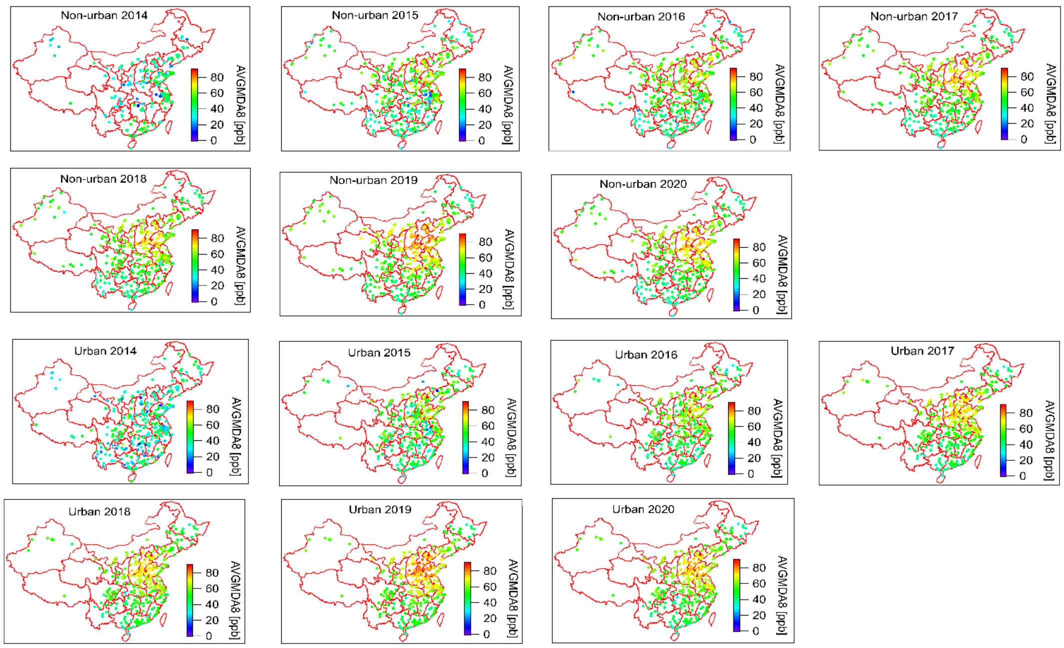

Figure A1. Spatial distribution of warm-season AVGMDA8 ozone concentrations in urban and non-urban areas during 2014-2020.