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Severe Surface Ozone Pollution in China: A Global Perspective

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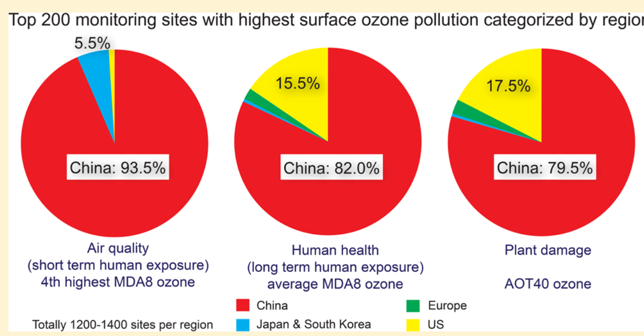
Supporting Information

ABSTRACT: The nationwide extent of surface ozone pollution in China and its comparison to the global ozone distribution have not been recognized because of the scarcity of Chinese monitoring sites before 2012. Here we address this issue by using the latest 5 year (2013−2017) surface ozone measurements from the Chinese monitoring network, combined with the recent Tropospheric Ozone Assessment Report (TOAR) database for other industrialized regions such as Japan, South Korea, Europe, and the United States (JKEU). We use various human health and vegetation exposure metrics. We find that although the median ozone values are comparable between Chinese and JKEU cities, the magnitude and frequency of high-ozone events are much larger in China. The national warm-season (April−September) fourth highest daily maximum 8 h average (4MDA8) ozone level (86.0 ppb) and the number of days with MDA8 values of >70 ppb (NDGT70, 29.7 days) in China are 6.3−30% (range of regional mean differences) and 93−575% higher, respectively, than the JKEU regional averages. Health exposure metrics such as warm-season mean MDA8 and annual SOMO35 (sum of ozone means over 35 ppb) are 6.3−16 and 25−95% higher in China, respectively. We also find an increase in the surface ozone level over China in 2016 and 2017 relative to 2013 and 2014. Our results show that on the regional scale the exposure of humans and vegetation to ozone is greater in China than in other developed regions of the world with comprehensive ozone monitoring.



1. INTRODUCTION

Surface ozone is an air pollutant that is detrimental to human health and vegetation growth.[1] There is evidence of risks of respiratory and cardiovascular mortality due to short-term (acute) exposure to high ambient ozone, and recent studies also reveal long-term (chronic) exposure effects even at low ozone levels.[2−4] Ozone near the surface is mainly generated by photochemical oxidation of carbon monoxide (CO) and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x = NO + NO_2) and sunlight. Severe ozone pollution in many European and U.S. urban areas has now been extensively alleviated because of stringent emission control measures since the 1990s.[5−8] In the meantime, South and East Asia have experienced rapid urbanization and industrialization, causing significant increases in anthropogenic ozone precursor emissions[9,10] and potentially shifting the worldwide air pollution hot spots to these populous regions.[11]

Eastern China experiences severe fine particulate matter ($PM_{2.5}$) pollution in winter,[12] and this issue has been the main focus of the government's air pollution control strategy over the past 5 years.[13] More recently, summertime ozone pollution has become an emerging concern in China. The past summer of 2017 saw particularly high surface ozone levels in a large number of Chinese cities, with the 90th percentile of the daily

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Table 1. Description of Ozone Metrics Used in This Study[a]

metric	definition	aggregation period
median (ppb)	50th percentile of hourly concentrations	April–September
Perc98 (ppb)	98th percentile of hourly concentrations	April–September
DTAvg (ppb)	daytime average ozone is the average of hourly ozone concentrations for the 12 h period from 08:00 to 19:59 local time	April–September
MDA1 (ppb)	daily maximum 1 h average; AVGMDA1 represents mean MDA1 in the aggregation period	annual and April–September
MDA8 (ppb)	daily maximum 8 h average; AVGMDA8 represents mean MDA8 in the aggregation period	annual and April–September
4MDA8 (ppb)	4th highest MDA8	April–September
SOMO35 (ppb day)	sum of positive differences between MDA8 and a cutoff concentration of 35 ppb	annual
NDGT70 (day)	total number of days with MDA8 values of >70 ppb	April–September
AOT40 (ppb h)	cumulative hourly ozone concentrations of >40 ppb; daily 12 h AOT40 is calculated using hourly values for the 12 h period from 08:00 to 19:59 local time, and warm-season total AOT40 is presented	April–September
W126 (ppb h)	daily W126 is calculated using the formula $W126 = \sum_i w_i C_i$, where C_i denotes the hourly ozone concentration in ppb for the 12 h period from 08:00 to 19:59 local time and w_i is the weighting index defined as $w_i = \frac{1}{1 + M \times \exp(-AC_i/1000)}$, where $M = 4403$ and $A = 126$	April–September
exceedance (day)	number of days with the ozone concentration exceeding the Chinese grade II national air quality standard, defined as MDA8 > 160 $\mu g\ m^{-3}$ or MDA1 > 200 $\mu g\ m^{-3}$ over residential, industrial, and rural areas	annual

[a]More details of the calculation methods are provided in Table S1.

maximum 8 h average (MDA8) ozone level in 30 of the 74 major cities exceeding 200 $\mu g\ m^{-3}$ (the grade II national air quality standard for protection of residential areas).[14] Previous studies from field observations and satellite retrievals have indicated severe ozone pollution in China; e.g., observed hourly maximum ozone concentrations in China frequently exceeded 150 ppb.[15,16] These values are comparable to or higher than the present-day annual fourth highest MDA8 (4MDA8, approximately 98th percentile) ozone values in the Los Angeles Basin (~110 ppb),[8] an area with the most severe ozone pollution in the United States. In contrast to the general decreasing ozone levels in the United States and Europe,[5] available surface ozone observations have shown significant positive trends in China since 1990.[17−21] Despite the increasing level of attention, the severity of ozone pollution in China compared with that in other industrialized regions of the world has not been quantified on the basis of extensive ozone monitoring.

This study aims to understand the current status of observed surface ozone pollution in China from a global perspective by comparing Chinese ozone data to ozone observations in other industrialized regions of the Northern Hemisphere, as revealed by ozone metrics relevant to human health and crop/ecosystem productivity. The data come from China's recently available nationwide surface ozone measurements and from the Tropospheric Ozone Assessment Report (TOAR, http://www.igacproject.org/activities/TOAR) organized by the International Global Atmospheric Chemistry Project (IGAC).[22] We also analyze changes in ozone pollution in China over the past 5 years based on the different ozone exposure metrics.

2. MATERIALS AND METHODS

2.1. The China National Environmental Monitoring Center (CNEMC) Network. Surface air pollutants in mainland China are monitored by the CNEMC of the Ministry of Environmental Protection in China (MEPC), and data are reported hourly (http://106.37.208.233:20035/). This nationwide observational network, designed for monitoring urban and suburban air pollution in mainland China, became operational in 2013 in 74 major cities, and by 2017, it included 1597 nonrural sites covering 454 cities (Figure S1). These measurements now document the air quality in Chinese cities and have been applied in recent studies.[23−26] In this study, we use the hourly measurements of ozone, carbon monoxide (CO), nitrogen dioxide (NO_2), sulfur dioxide (SO_2), and fine particulate matter ($PM_{2.5}$) from 2013 to 2017. Quality controls for all species have been applied following previous studies[23] to remove data outliers (Supporting Information).

2.2. TOAR Surface Ozone Database. We use the TOAR surface ozone data set (accessed from https://doi.org/10.1594/PANGAEA.876108)[27] for a global comparison of ground-level ozone. The TOAR surface ozone database includes ozone statistics and metrics relevant to studies on climate, human health, and vegetation exposure, calculated from hourly ozone measurements at more than 9000 monitoring sites around the world since the 1970s.[22] Procedures for data harmonization, quality control, and metric calculation have been described by Schultz et al.[22] Here we analyze the latest 5 year (2010−2014) aggregated ozone metrics as well as the long-term (1980−2014) yearly ozone metrics from the TOAR database. Recent reports suggest that levels of ozone or its precursors in the United States and Europe have remained relatively stable or slightly decreased after 2014.[28,29] We use the warm-season (April−September) metrics, except for SOMO35 (sum of ozone means over 35 ppb) that is aggregated annually. While the TOAR database includes thousands of observational sites in Europe, the United States, Japan, and Korea, it has data from only 32 sites located in China (half are in Hong Kong). Our analysis of the

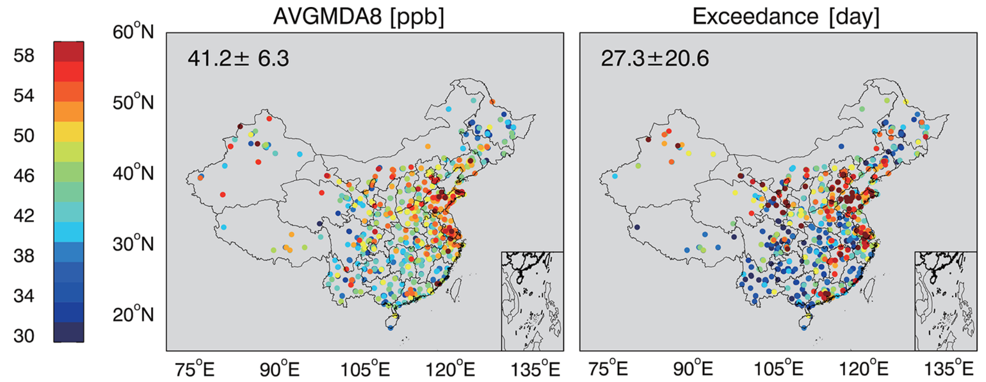

Figure 1. Distribution of annual mean MDA8 (left) and ozone air quality exceedance (right) averaged over 2013−2017. Only sites with at least 3 year measurements are included. Mean values and standard deviations over the sites are shown in the insets.

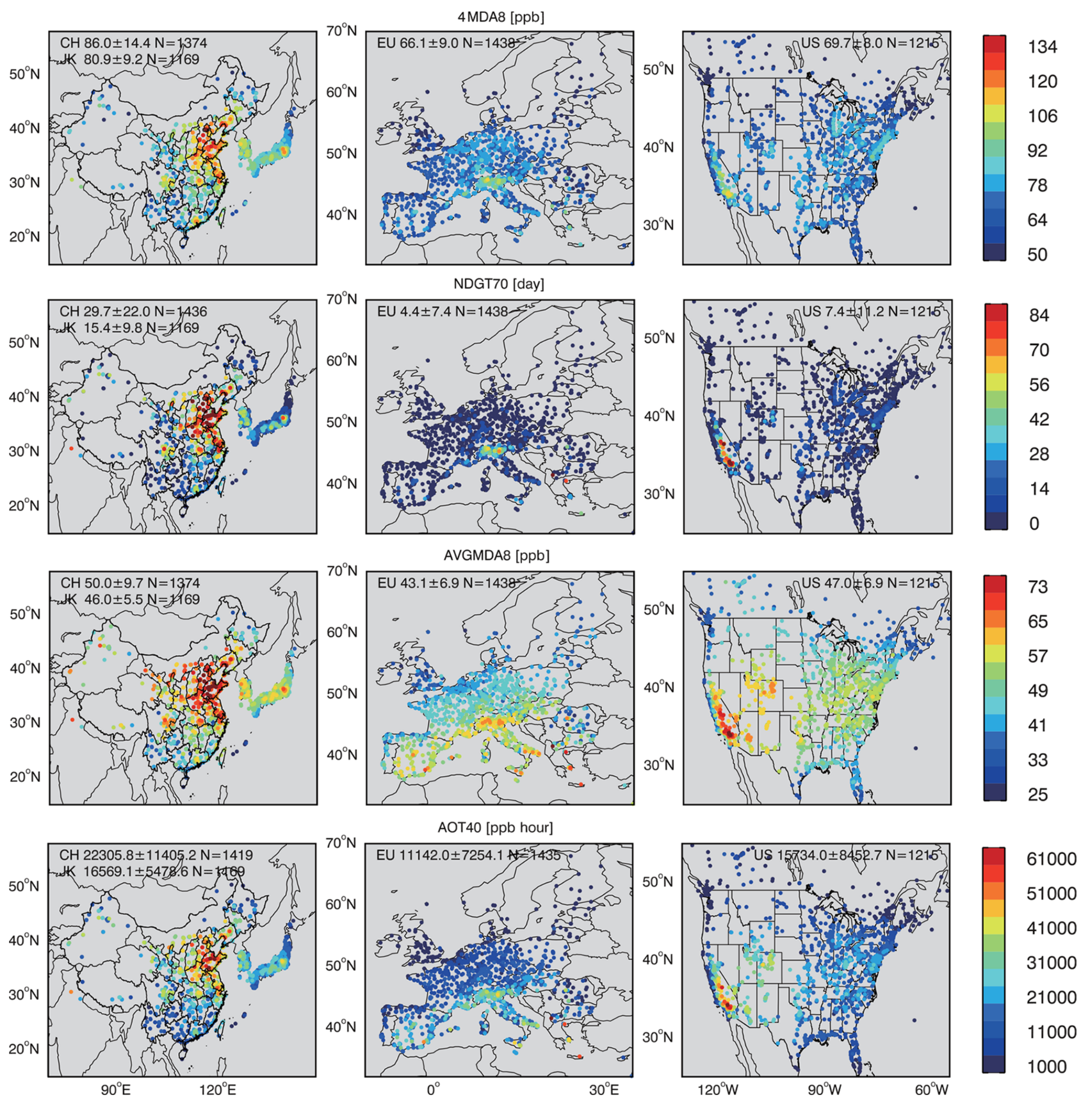

Figure 2. Comparison of April−September ozone metrics (4MDA8, NDGT70, AVGMDA8, and AOT40) between China (including CNEMC sites and TOAR Chinese sites) and other industrialized regions [Japan and Korea (JK), Europe (EU), and the United States]. Only sites with at least 3 year measurements are included. Values shows in insets are the regional means and standard deviations averaged over the N sites. The differences between China and other industrialized regions are statistically significant ($p < 0.05$) based on analysis of variance tests for all four metrics.

CNEMC observations (not included in the TOAR surface ozone database) thus fills this gap.

2.3. Ozone Metrics for Human Health and Crop/Ecosystem Exposure. Table 1 lists the 11 ozone metrics analyzed in this study. These metrics include standard statistics

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such as median, 98th percentile (Perc98), MDA8, and daytime average (DTAvg), as well as metrics for assessing human health and ecosystem exposure impacts.[30] MDA8 is a widely used daily ozone metric for air quality regulation and human health impact studies in many regions such as the United States, Europe, and China. The response of human health to ozone exposure is controlled by ozone level, exposure duration frequency, and physical activity.[4,30] Following TOAR, ozone exposure is evaluated in our study by short-term human exposure metrics such as daily maximum 1 h (MDA1) and MDA8, and the total number of days with MDA8 values of >70 ppb (NDGT70), or by metrics that consider the cumulative exposure (e.g., annual SOMO35). Risk estimates from epidemiological studies based on these exposure metrics are summarized in ref 4 and have recently been applied in China.[31] Ozone damage to vegetation is cumulative and can be disproportionately larger for high ozone concentrations.[30] Here we use the AOT40 and W126 metrics that correlate with crop yields[32,33] as indicators of vegetation exposure. We calculated all metrics based on hourly CNEMC measurements following the TOAR definitions, procedures, and data completeness requirements (Table S1). We also calculated the annual total numbers of days with the ozone level exceeding the Chinese grade II (Exceedance) ozone air quality standard (Table 1).

3. RESULTS AND DISCUSSION

3.1. Spatiotemporal Distribution of Ozone Air Quality across Mainland China. Figure 1 shows the spatial distribution of annual mean MDA8 (annual AVGMDA8) in Chinese cities averaged for 2013−2017. MDA8 and MDA1 are also used to determine exceedances of the national ozone air quality standard (Table 1) in China. The annual AVGMDA8 and AVGMDA1 ozone values averaged for the Chinese sites are 41.2 ± 6.3 ppb (87.9 ± 13.5 μg m^{-3}) and 48.2 ± 7.2 ppb (103.6 ± 15.4 μg m^{-3}), respectively. During 2013−2017, there were >27 days per year that exceeded the grade II air quality standard, averaged over the monitoring sites. Spatially, ozone hot spots (annual AVGMDA8 > 50 ppb and exceedance > 40 days) extend across eastern China, especially in the North China Plain (NCP) and the Yangtze River Delta (YRD), mainly induced by anthropogenic sources producing high levels of ozone precursors. Severe ozone pollution also occurs in some western cities, e.g., in the central Gansu Province where exceedance is >80 days, comparable to that in the eastern regions. This is likely due to unique topography (valley basins in mountainous regions) coupled with high local ozone precursor emissions from the petrochemical industry and vehicles.[34]

The mean MDA8 ozone level over China peaks in summer due to strong photochemistry, similar to other regions at northern midlatitudes, but the patterns vary among different regions (Figures S2 and S3). MDA8 ozone levels in the YRD and NCP peak in May (60 ppb for the regional average) and June (68 ppb), respectively, while the averaged MDA8 value in the Pearl River Delta (PRD) is highest in October (57 ppb). This difference is due to the timing of the Asian summer monsoon, which brings cloudy and rainy weather, marine air, and strong deep convection, all unfavorable factors for ozone chemical production and accumulation.[35,36] As a consequence, ozone pollution over the YRD and PRD generally decreases during the summer monsoon and peaks before its arrival or after its retreat.

3.2. Comparison with Other Northern Midlatitude Regions. Figure 2 and Figure S4 compare different human health and vegetation exposure metrics for warm-season surface ozone in China to those in other industrialized regions, including Japan and South Korea (JK), Europe (EU), and the United States (collectively termed JKEU hereafter). We use the nonrural sites (Supporting Information) from the TOAR database. We show that warm-season median and mean DTAvg ozone metric values averaged for China (30.2 ± 8.1 and 41.6 ± 8.6 ppb, respectively) are comparable to those of other regions, e.g., median values of 29.0 ppb over JK, 31.0 ppb over EU, and 33.5 ppb over the United States (Figure S4). Active local anthropogenic emissions and photochemistry lead to high DTAvg ozone levels in East Asia and California.[37,38] High median and DTAvg values also occur at high elevations such as western China, the European Alps, and the western United States, reflecting the typical increase in the ozone level with altitude.[39−42]

The severity of Chinese surface ozone pollution becomes obvious when comparing metrics such as 4MDA8 and Perc98, which focus on the high end of the ozone distribution and are likely caused by local pollution episodes. As shown in Figure 2 and Figure S4, 4MDA8 and Perc98 ozone values averaged over the Chinese sites are 86.0 ± 14.4 and 80.7 ± 14.1 ppb, respectively, approximately 20−25% higher than the averages of European and U.S. sites. High 4MDA8 ozone values of >100 ppb are widely observed in the NCP, YRD, and PRD (Figure 2). High ozone days also occur much more frequently in China, as indicated by the NDGT70 metric (Figure 2). The NDGT70 value averages 29.7 ± 22.0 days over the Chinese sites, approximately twice the value in JK, a factor of 6 higher than the value in the EU, and a factor of 3 higher than the value in the United States. NDGT70 values of >70 days are common in eastern China, while in other regions, only a few sites in California reach such a high frequency of exceedance.

In addition to NDGT70, we compare two other health exposure metrics (AVGMDA8 in Figure 2 and SOMO35 in Figure S4). The national warm-season mean AVGMDA8 value in China (50 ppb) is 6.3−16% higher than those found in JKEU (43.1−47.0 ppb), while the mean annual SOMO35 value in China [(4.3 ± 1.7) × 10^3 ppb day] is 25−95% higher than the regional mean SOMO35 values of 2.2−3.4 × 10^3 ppb day in JKEU cities. The AVGMDA8 and SOMO35 values are particularly high in populous eastern China (Table S2), e.g., AVGMDA8 > 55 ppb in the NCP, and SOMO35 > 5.5 × 10^3 ppb day in the YRD. To the best of our knowledge, no other region of the world, with extensive ozone monitoring, has such a large population exposed to such severe and frequent ozone pollution episodes.

Comparisons of vegetation exposure metrics also indicate the potential for greater ozone-induced plant damage in China. The AOT40 (Figure 2) and W126 (Figure S4) metrics in China are (2.2 ± 1.1) × 10^4 and (2.9 ± 1.7) × 10^4 ppb h, which are 35−100 and 50−170% higher than the JKEU regional means, respectively. W126 shows a larger difference due to its weighting toward higher ozone levels. High AOT40 (>4 × 10^4 ppb h) and W126 (>6 × 10^4 ppb h) values are widespread in eastern and central China. Previous field surveys at limited locations and coarse-resolution modeling studies (e.g., two degrees) estimated that ozone pollution in China could decrease the wheat yield by 6.4−14.9% and the annual net primary production by ∼14%.[33,43] These studies focused on ozone pollution before 2010, and thus, we may expect even

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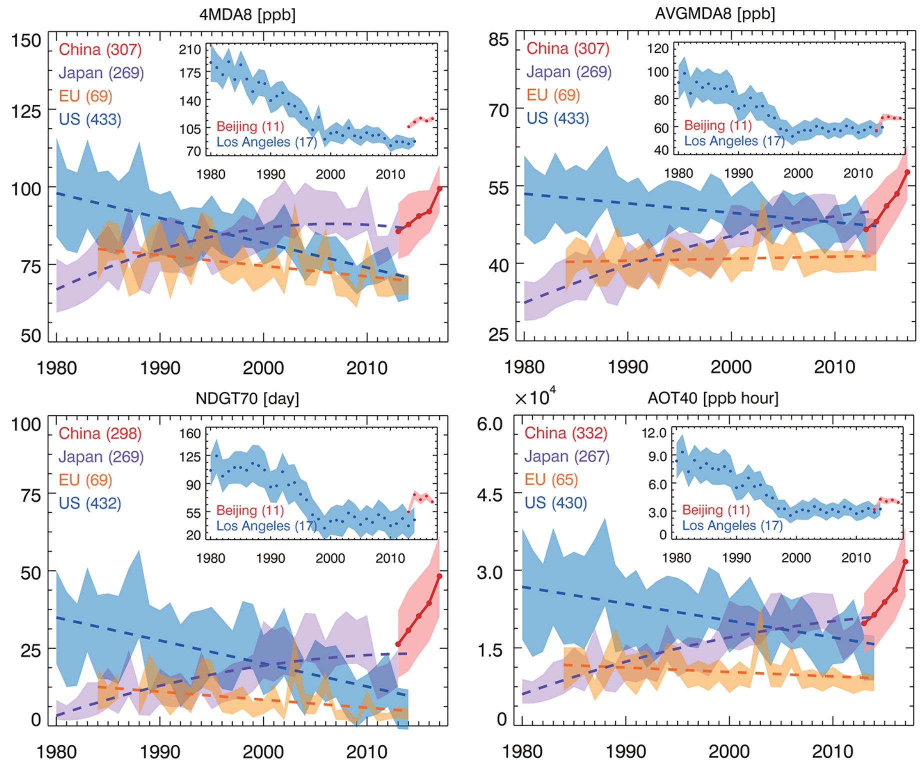

Figure 3. Evolution of urban surface ozone levels in China (red), Japan (purple), Europe (orange), and the United States (blue) from 1980 to 2017. Also shown are the ozone time series in Beijing (red) and Los Angeles (blue). For Japan, the EU, and the United States, only urban sites with available records for more than 25 years (1980–2014) are considered. For China, sites in 74 major cities (Figure S1) with continuous observations in 2013–2017 are considered. Shaded areas represent the range of mean values ± the 50% standard deviation for each region. Numbers of sites are shown in parentheses. The dashed lines denote the linear fitting curves for Europe and the United States and the parabolic fitting curve for Japan.

stronger effects for the more recent conditions recorded by the nationwide network.

3.3. Changes in Surface Ozone Pollution over China in 2013–2017. The 5 year CNEMC measurements allow us to examine the interannual variability of surface ozone pollution over China. Here we focus on the sites in 74 major Chinese cities (Figure S1) where continuous observations from 2013 to 2017 are available.

Figure 3 shows the evolution of different ozone metrics for the Chinese sites over 2013–2017 compared with those in Japan, Europe, and the United States over 1980–2014. Figure S5 shows interannual variations of additional ozone metrics averaged over these Chinese sites during 2013–2017. We find that all ozone metrics show continuous increases over the 5 years in China. Averaged over the 74 cities, the DTAvg, 4MDA8, and Perc98 values increased at rates of 3.7–6.2% year^{-1}, while the ozone exposure metrics SOMO35, AOT40, and W126 increased at higher rates (11.9–15.3% year^{-1}). The increases are statistically significant in 2016–2017 relative to 2013–2014 based on analysis of variance tests (Table S4) and are consistent with previous studies of long-term ozone increases at a limited number of sites across China.[17−21] Even though the 5 year period is too short to derive statistically robust trend estimates, these results indicate an increasing severity of human and crop/ecosystem ozone exposure across China. We find the largest increases in ozone exposure in eastern and central China, especially in the NCP and YRD, overlap with the most populous areas (Figure S6). From a historical perspective, present-day ozone levels in the major Chinese cities are comparable to or even higher than the 1980 levels in the United States when emission controls were just beginning to have an impact on reducing ozone levels (Figure 3).

The Chinese State Council implemented the Action Plan on Air Pollution Prevention and Control in September 2013, resulting in nationwide reductions in ambient SO_2, CO, NO_2, and $PM_{2.5}$ levels, as shown in Figure S7. The emerging severity of ozone pollution in China raises a new challenge to current emission control actions. Previous observational and modeling analyses showed that ozone chemical production in the NCP, YRD, and PRD regions is likely VOC-sensitive or mixed-sensitive.[16,44−49] Recent bottom-up emission estimates and satellite formaldehyde observations indicated increasing VOC levels in eastern China that can be attributed to anthropogenic sources.[20,50] For those VOC-sensitive regions, either decreasing NO_x levels or increasing VOCs levels could potentially enhance ozone pollution. Therefore, VOC controls could be explored as a possible control strategy. Reduced $PM_{2.5}$ levels over 2013–2017 may also cause an increase in the level of ozone due to impacts on ozone photochemistry and heterogeneous chemistry on aerosol surfaces.[51] In addition, the interannual variability of surface ozone can be influenced by meteorological conditions. The summer of 2017 had hotter and drier weather conditions compared to those of previous years (Figure S8), which favored ozone production and led to higher ozone levels. Further studies are needed to better quantify the contributions of emission changes and weather variability to recent trends in surface ozone levels over China.

We conclude that China has become a global hot spot of present-day surface ozone pollution, and human and vegetation exposure in China is greater than in JKEU. Although statistics such as median and mean DTAvg, which

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focus on the midrange of the ozone distribution, are comparable to those of JKEU, the magnitude and frequency of high-ozone events are much greater in China. While high-ozone pollution also occurs in some other developing regions, such as India in the presummer monsoon season (April and May)[36] and Mexico City,[52] the lack of accessible ozone observations prevents us from comparing those regions to China and JKEU. China's surface ozone air quality deteriorated in 2016–2017 compared to 2013–2014, indicating that current emission control measures have not been effective for reducing ozone air pollution.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.8b00366.

CNEMC data quality control (SI 1), station type in the TOAR database (SI 2), calculation of the ozone metrics (Table S1), ozone metric values over the NCP, YRD, and PRD (Table S2), ozone metric values over China, Japan and South Korea, Europe, and the United States (Table S3), analysis of variance test of year on year ozone increase (Table S4), site locations (Figure S1), annual cycle of MDA8 over China (Figure S2), seasonal and spatial distribution of MDA8 over China (Figure S3), additional ozone metrics over China, Europe, and the United States (Figure S4), interannual variability of ozone metrics (Figures S5 and S6), interannual variations of national mean NO_2, CO, SO_2, and $PM_{2.5}$ levels (Figure S7), and surface temperature and relative humidity anomalies in the summer of 2017 (Figure S8) (PDF)

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Notes

The authors declare no competing financial interest.

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