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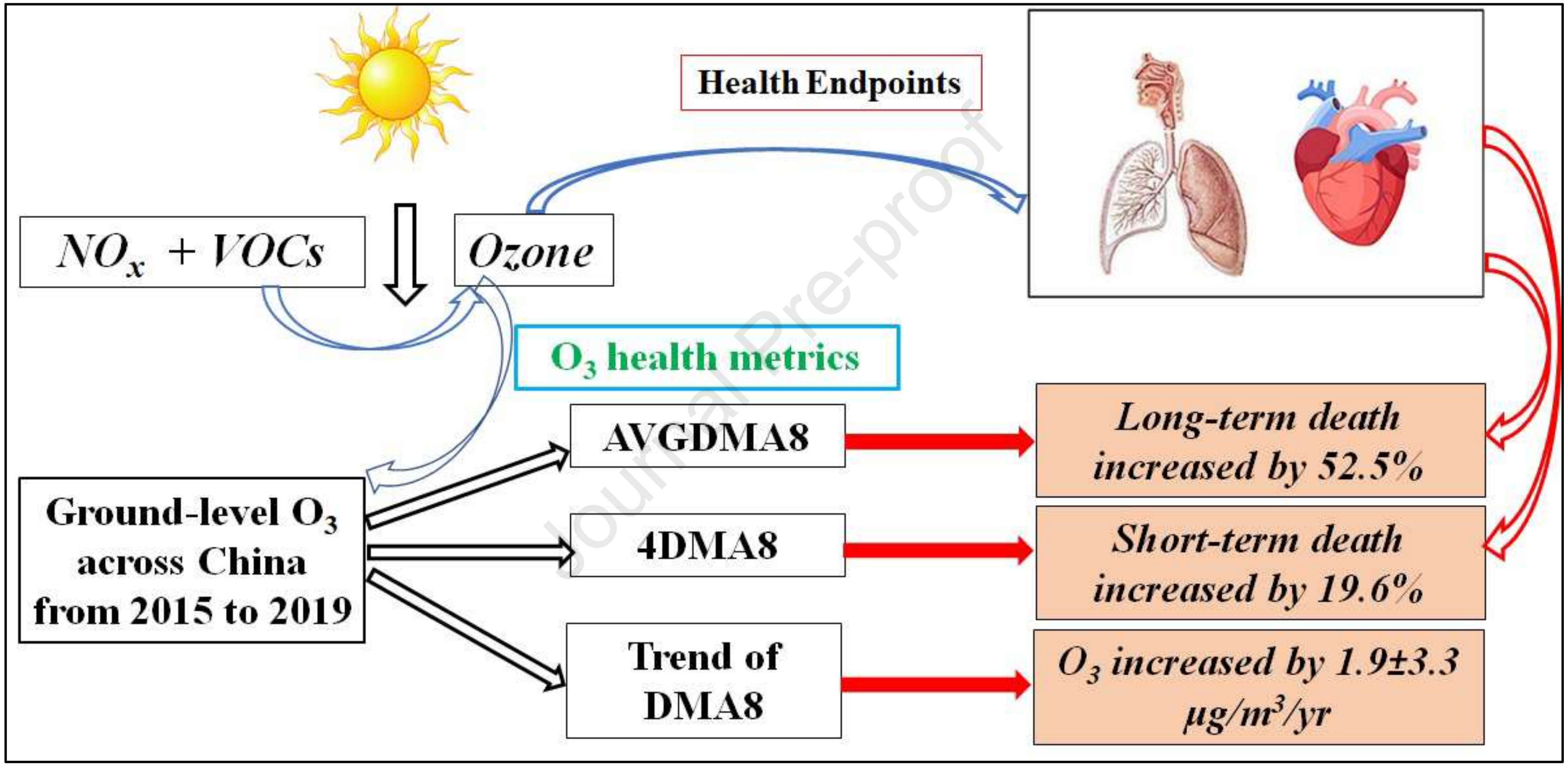
Health Endpoints
NOx + VOCs
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AVGDMA8
4DMA8
Trend of DMA8
Ground-level O3 across China from 2015 to 2019
Long-term death increased by 52.5%
Short-term death increased by 19.6%
O3 increased by 1.9±3.3 μg/m3/yr

Continuous increases of surface ozone and associated premature mortality growth in China during 2015-2019

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Abstract:

Ambient ozone (O_3) pollution has become a big issue in China. Recent studies have linked long- and short-term O_3 exposure to several public health risks. In this study, we (1) characterize the long-term and short-term O_3-attributed health metric in China from 2015-2019; (2) estimate the surface O_3 trends; and (3) quantify the long-term and short-term health impacts (i.e. all-cause, cardiovascular and respiratory mortality) in 350 urban Chinese cities. In these 5-years, the national annual average of daily maximum 8h average (AVGDMA8) O_3 concentrations and warm-season (April−September) 4^{th} highest daily maximum 8h average (4DMA8) O_3 concentrations increased from 74.0±15.5 μg/m^3 (mean±standard deviation) to 82.3±12.0 μg/m^3 and 167±37.0 μg/m^3 to 174±30.0 μg/m^3 respectively. During this period, the DMA8 O_3 concentration increased by 1.9±3.3 μg/m^3/yr across China, with over 70% of the monitoring sites showing a positive upward trend and 19.4% with trends >5 μg/m^3/yr. The estimated long-term all-cause, cardiovascular and respiratory premature mortalities attributable to AVGDMA8 O_3 exposure in 350 Chinese cities were 181,000 (95% CI: 91,500-352,000), 112,000 (95% CI: 38,100-214,000) and 33,800 (95% CI: 0-71,400) in 2019, showing increases of 52.5%, 52.9% and 54.6% respectively compared to 2015 levels. Similarly, short-term all-cause, cardiovascular and respiratory premature mortalities attributed to ambient 4DMA8 O_3 exposure were 156,000 (95% CI: 85,300-227,000), 73,500 (95% CI: 27,500-119,000) and 28,600 (95% CI: 14,500-42,800) in 2019, increases of 19.6%, 19.8% and 21.2% respectively compared to 2015. The results of this study are important in ascertaining the e□ectiveness of recent emission control measures and to identify the areas that require urgent attention.

Keywords: Ozone pollution; Spatiotemporal distribution; Health; Long-term mortality; Short-term mortality, China

1. Introduction

Exposure to ozone (O_3), which is a strong oxidant, is harmful to human health and agricultural production. O_3 is also one of the greenhouse gases, which plays an important role in global climate change (Avnery et al., 2011; Tai and Val Martin, 2017; Emberson et al., 2018; Archer et al., 2019). For these reasons, environmental scientists and regulatory agencies have paid close attention to O_3 in recent years (Orru et al., 2013; Hong et al., 2019). In urban regions, tropospheric O_3 formation occurs by photochemical oxidation of volatile organic compounds (VOCs) and carbon monoxide (CO) in the presence of nitrogen oxides (NO_x) and sunlight (Lu et al., 2019). Major anthropogenic sources of VOCs and NO_x (i.e. O_3 precursors) are motor vehicle exhaust, industrial emissions and chemical solvents (Zhang et al., 2019). The relatively long lifetime of O_3 in the troposphere (approximately 20 days) and photochemical production at regional scales make ground-level O_3 a continental and hemispheric-scale pollutant (Fowler et al., 2020).

In China, along with economic development and urbanization, air pollutants have become a common factor endangering health, causing the government to take intervention measures (Wang et al., 2017). An air pollution control plan was initiated in 2013, which mainly focused on reducing $PM_{2.5}$ (particulate matter with aerodynamic diameter <2.5μm) in the most polluted cities (Zhang et al., 2019). As a result, in recent years China has suffered from severe O_3 episodes and a continuous increase of surface O_3 concentration at the rate of 2-4 $\mu g/m^3$/yr from 2013 to 2019 (Lu et al., 2020).

High tropospheric O_3 concentrations are largely caused by anthropogenic and natural emissions of precursors and meteorological influences mainly in summer (Han et al., 2020). In 2015, high annual daily maximum 8h average (DMA8) O_3 concentrations were reported in major Chinese cities (87.9±13.5 $\mu g/m^3$) and concentrations exceeding 120 $\mu g/m^3$ were frequently observed in the three megacity cluster regions, Beijing–Tianjin–Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) (Kuerban et al., 2020). From 2013 to 2017, the annual mean of the 90^{th} percentile of DMA8 O_3 concentrations in 74 Chinese cities increased from 139 $\mu g/m^3$ (range: 72-190 $\mu g/m^3$) to 167 $\mu g/m^3$ (range: 117-218 $\mu g/m^3$) (Ministry of Ecology and Environment of China, 2017). In 2018, 34.6% of 338 Chinese cities were exposed to 160-217 $\mu g/m^3$ 90^{th} percentile of DMA8 O_3 (Ministry of Ecology and Environment of China, 2018). The 4^{th} highest DMA8 (4DMA8) O_3 values during the warm-season (April-September) over the Chinese sites were 172 ±

28.8 μg/m^3 and high 4DMA8 O_3 values (above 200 μg/m^3) were widely observed in the BTH, YRD, and PRD regions during 2013-2017 (Lu et al., 2018).

Both long- and short-term O_3 exposure causes adverse human health effects. Most of the studies in China have so far focused on health impacts attributed to long-term exposure, but short-term O_3 exposure at high concentrations in the summer season also significantly impacts on human health, so cannot be overlooked (Bell et al., 2014; Tian et al., 2018; Raza et al., 2018). The short-term premature mortality attributed to high 4DMA8 O_3 is thought to have significantly contributed to the total mortality in China (Liang et al., 2019), as the number of people exposed above the 4DMA8 O_3 threshold was very high (Zhan et al., 2018).

The real-time ground-level O_3 data in China have been available online from the China National Environmental Monitoring Centre (CNEMC) (http://www.cnemc.cn/) since 2013, enabling a new opportunity to understand the heterogeneity of ground-level O_3 across China and its short- and long-term health impacts on the Chinese population. This study uses ground O_3 observations between 2015 and 2019. First, we characterize the long-term and short-term O_3 health metrics in China. Second, the nonparametric linear trend for DMA8 metrics during 2015–2019 are analyzed. Third, we estimate the changes in premature mortality attributable to long-term and short-term O_3 exposure in Chinese cities.

2. Methodology

2.1. Ground-level O_3 data

Hourly O_3 data from 2015 to 2019 were obtained from http://beijingair.sinaapp.com/, which provides ratified air quality data across mainland China. The data were available from 1497 monitoring stations in 2015 and 1633 monitoring stations in 2019 (Fig.S1). The quality of all the available data was controlled based on the criteria developed in previous studies (Silver et al., 2018; Xu et al., 2020) (section S1.1.). The O_3 data from 1312 monitoring stations passed the quality standard to be used in this study, covering 350 cities across 31 provinces in China.

The Tropospheric O_3 Assessment Report (TOAR) (Xu et al., 2020) defines 12 metrics to characterize O_3 pollution and its impacts on climate, human health and vegetation. In these metrics, summer average daily maximum 1-h (6DMA1), summer average daily maximum 8-h (6DMA8) and annual average DMA8 (AVGDMA8) O_3 statistics are used to study long-term (chronic) O_3-exposure. 4DMA8 (4th highest DMA8 in summer, approximately 98th percentile), NDGT70 (total number of days with DMA8 > 140 μg/m^3) and SOMO35 metrics (annual sum of daily DMA8 > 70 μg/m^3) are used to study short-term (acute) O_3-exposure (Fleming et al., 2018). The TOAR metrics for the summer period in the Northern hemisphere cover 6 months from April to September. The 4DMA8 reflecting the high end of the ozone distribution over summertime. In this study, we characterized AVGDMA8 and 4DMA8 O_3 metrics and used for corresponding long- and short-term health risk analysis. The O_3 metrics at city levels are calculated by averaging available monitoring site data within each a city.

2.2. Trend analysis

To identify nonparametric monotonic linear trends, the Theil-Sen estimator is used to calculate the magnitude of the trends with de-seasonalised data, while the Mann-Kendall test assesses the significance of trends (threshold of $p < 0.05$) (Lefohn et al., 2018). The trend was analysed in RStudio version 3.6.0 with a series of R packages including “openair”, “tidyverse”, “lubridate” and “dplyr” (R Core Team, 2019; Carslaw, 2019). The “openair” is specifically developed for analysing air quality data.

2.3. Premature mortalities attributed to O_3 exposure

This study estimates short-term and long-term all-cause, cardiovascular and respiratory mortalities attributable to ambient O_3-exposure at 350 urban Chinese cities from 2015-2019. It uses the log-linear exposure-response function, described in the studies by Stanaway et al. (2018) and Seltzer et al. (2018) as:

$$\Delta C = \begin{cases} 0 & if\ [O_3] \le TMREL \\ [O_3] - TMREL & f\ [O_3] > TMREL \end{cases} \quad (1)$$

$$\beta = \ln(RR) / \Delta X \quad (2)$$

$$\Delta Mort = (1 - \exp^{-\beta \Delta C}).\ D_0.\ P_c \quad (3)$$

TMREL is the theoretical minimum risk exposure level. For long-term exposure, $[O_3]$ is the annual mean concentration and ΔC is the estimated annual mean O_3-exposure relative to TMREL. For short-term exposure, $[O_3]$ is the daily concentration and ΔC is the cumulative results of daily mean O_3-exposure relative to TMREL. β is the exposure-response factor derived from the reported relative risk (RR), which links incremental changes in O_3-exposure ΔX (20 μg/m^3 in AVGDMA8 and 10 μg/m^3 in 4DMA8 metric). D_0 is the cause-specific death rate, obtained from the GHDx database (http://ghdx.healthdata.org/gbd-results-tool, available for 2015-2017. 2017 values are used thereafter). P_c is the population (≥30 years) at an individual city and $\Delta Mort$ is the estimated number of cause-specific mortalities at an individual city. Detailed city-level age-specific population data were obtained from the National Bureau of Statistics of the People's Republic of China (NBSC, 2019).

TMREL is 53.4 μg/m^3 for long-term all-cause mortality (Turner et al., 2016) and 53.6 μg/m^3 for long-term cardiovascular and respiratory mortality (Lim et al., 2019). These values are the minimum O_3 concentration in this long-term epidemiological study. For short-term mortality, TMREL of 70 μg/m^3 is used, as recommended in the HRAPIE project (WHO, 2013). Additionally, the Chinese Ambient Air Quality Standards (CAAQS) Grade I for O_3 (100 μg/m^3) – the same as the WHO air quality guideline for O_3 – and null concentration (0 μg/m^3) were also selected as the threshold for sensitivity analysis. In our long-term O_3-exposure health risk study, we estimated an exposure-response coefficient (β) for all-cause mortality based on the study by Turner et al. (2016), and cardiovascular and respiratory mortality from Lim et al. (2019) – both of which are long-term epidemiological studies. For our short-term mortality study, β was estimated for all-cause and cardiovascular mortality from Yin et al. (2017), a large epidemiological study in China, while respiratory mortality was estimated from a large meta-analysis (Dong et al., 2016) (Table S1). To compare observations or metrics reported in units of ppb, we used a conversion factor of 1 ppb = 2 μg/m^3 at a reference temperature and standard pressure of 20 °C and 1013.25 hPa respectively (Lefohn et al., 2018).

3. Results

3.1. Distribution of AVGDMA8 O_3

The nationwide AVGDMA8 O_3 concentrations at 1312 stations for 2015 to 2019 are shown in Fig.1. In 2015, the AVGDMA8 O_3 concentrations ranged from 26.0 μg/m^3 (Sunny city, Liaoning province) to 116 μg/m^3 (Zaozhuang city, Shandong province), with the national mean 74.0±16 μg/m^3 (mean±standard deviation) (Fig.1a). Nationwide AVGDMA8 O_3 concentrations increased in 2016 (77.5±13 μg/m^3), 2017 (84.2±13 μg/m^3) and 2018 (84.4±12 μg/m^3) (Fig.1b-d). In 2019, the national AVGDMA8 O_3 value slightly decreased to 82.3±12 μg/m^3, although it had a wide range, from 49 μg/m^3 (Guangyuan, Sichuan) to 115 μg/m^3 (Jincheng, Shanxi) (Fig.1e). Among 1312 monitoring sites, 63 sites in 2015, 168 sites in 2017 and 88 sites in 2019 exceeded the CAAQS Grade I of 100 μg/m^3 for O_3. The number of cities exceeding the Grade II standard (160 μg/m^3) increased from 11 in 2015 to 38 in 2018, then decreased to 17 in 2019. During these five years, the cities with high AVGDMA8 O_3 values were Alxa League (Inner Mongolia) (109 μg/m^3), Haibei (Qinghai) (104 μg/m^3), Yingkou (Liaoning) (102 μg/m^3), Dongying (Shandong) (102 μg/m^3) and Weifang (Shandong) (100 μg/m^3). Within five years, AVGDMA8 O_3 increased by >20 μg/m^3 in 57 cities and by >5 μg/m^3 in 200 cities. Higher O_3 increases were observed in Jincheng (Shanxi), Chuzhou (Anhui), Laizhou (Shandong), Wuhu (Anhui) and Suzhou (Anhui) (59.8, 57.7, 55.5, 45.3 and 40.7 μg/m^3 respectively). In the megacity-cluster, BTH, PRD and YRD region, AVGDMA8 O_3 values increased from 76.4±13 μg/m^3, 71.7±11 μg/m^3 and 88.2±8.7 μg/m^3 in 2015 to 90.0±5.7 μg/m^3, 83.3±6.0 μg/m^3 and 89.3±5.1 μg/m^3 in 2019. In 2015, there were 86.7 and 11.9 days per monitoring site that exceeded the Grade I and Grade II 8-hr CAAQS. In 2018, those values increased to 112 and 17.6 days, while in 2019 they reduced slightly to 108 and 17 days. From 2015-19 in total, about 511 days per monitoring station exceeded the Grade I standard. The mean DMA8 O_3 concentration over China peaks in summer due to stronger solar radiation and lower humidity, although the patterns were very diﬀerent in three megacity cluster regions. The mean DMA8 O_3 concentrations in BTH and YRD regions peaked in June (2015: 120±23 μg/m^3; 2019: 158±17 μg/m^3) and May (2015: 113±11 μg/m^3; 2019: 124±8.4 μg/m^3) respectively, while the highest value in the PRD region was observed in October (2015: 96±17 μg/m^3; 2019: 125±13 μg/m^3). This diﬀerence is due to the variability in the arrival of the Asian summer monsoon, which brings more cloud, clean marine air and strong convection currents, all unfavourable factors for O_3 production and accumulation (Li et al., 2018). As a result, during the pre- and post-monsoon seasons, O_3 concentration over the YRD and PRD regions generally decrease (Han et al., 2020).

3.2. Distribution of 4DMA8 O_3

The national-level 4DMA8 O_3 concentrations from 2015 to 2019 at 1312 monitoring stations are shown in Fig. 2. The 4DMA8 value represents the severity of surface O_3 pollution, focusing on the high end of the O_3 distribution, most likely caused by local emissions. The mean 4DMA8 O_3 values across all monitoring stations was 167±37 μg/m^3 (range: 51.6-455 μg/m^3) in 2015 (Fig.2a), increasing to 183±37 μg/m^3 (99.0-322 μg/m^3) in 2017 (Fig.2c), then decreasing to 174±30 μg/m^3 (94.1-262 μg/m^3) in 2019 (Fig.2e). During 2015-2019, the bottom-level 4DMA8 O_3 values increased, whereas the upper-level values decreased, while the overall 4DMA8 values increased at a rate of 8% per year. High 4DMA8 O_3 concentrations, above 200 μg/m^3, were widely observed in the BTH (215±22 μg/m^3) and YRD (202±19 μg/m^3) regions. 254 sites in 2015, 450 sites in 2017 and 307 sites in 2019 exceeded 200 μg/m^3. 764 sites showed a positive increase in 4DMA8, of which 98 sites showed an increase of >50 μg/m^3 during 2015-2019. At city-level, the highest 4DMA8 O_3 values were observed in Yingkou (Liaoning) (235±14 μg/m^3), Dongying (Shandong) (229±9.4 μg/m^3), Zibo (Shandong) (226±6.2 μg/m^3), Baoding (Hebei) (224±25 μg/m^3), Huludao (Liaoning) (224±21 μg/m^3) and Beijing (224±81 μg/m^3) during the five-year study period. Out of the 31 provinces, three provinces in 2015, nine in 2017 and seven in 2019 exceeded the 4DMA8 threshold of 200 μg/m^3, with most of these provinces located on the coastal and inland region in Eastern and Central China (e.g. Beijing, Shandong, Liaoning).

3.3. Trends of DMA8 O_3

Fig.3a shows the change of AVGDMA8 O_3 concentrations from 2015 to 2019, while Fig.3b shows the absolute trends of DMA8 O_3 concentrations with regional heterogeneity. Across China, DMA8 O_3 concentrations increased at a rate of 1.9±3.3 μg/m^3/yr with higher rates in some provinces, including Tianjin (8.2±3.5 μg/m^3/yr), Anhui (5.8±3.5 μg/m^3/yr), Shanxi (5.2±4.6 μg/m^3/yr) and Fujian (4.1±2.8 μg/m^3/yr). Slower negative trends were observed in Shanghai (-0.3±1.7 μg/m^3/yr), Zhejiang (-0.4±2.0 μg/m^3/yr) and Jilin (-1.5±2.1 μg/m^3/yr). The positive trends in Beijing, Guangzhou, Chongqing and Shenzhen were observed at 0.8±1.6 μg/m^3/yr, 2.6±2.5 μg/m^3/yr, 2.5±0.3 μg/m^3/yr and 2.1±3.2 μg/m^3/yr respectively. Overall, 70.0% (931 stations) of all monitoring stations showed a positive trend, with 19.4% showing the trend at a rate of >5 μg/m^3/yr. 27.8% stations (mostly located in Shanghai, Zhejiang and Jilin) showed negative trends. This is an important finding, which will help in developing an effective O_3 control policy in China.

3.4. Long-term premature mortality attributed to O_3

Long-term O_3 exposure has been shown to escalate all-cause nonaccidental, respiratory and cardiovascular premature deaths among adults (≥30 years). In 2015, about 832 million people

(64.6% of the total population in China) were exposed to AVGDMA8 O_3 concentrations above 70 μg/m^3. The level of exposed population increased to 1172 million (90.7%) in 2017 and 1117 million (86%) in 2019. The all cause-specific mortality figures, estimated by this study, in 350 cities in China from 2015 to 2019 are shown in Fig. 4 and Figs. S4-S5. The detailed data of estimated O_3-related long-term mortalities in 31 provinces are reported in Table S4. The log-linear model estimated national all-cause deaths at 119,000 [95% Confidence Interval (CI): 60,000-231,000] in 2015 and 181,000 (95% CI: 91,500-352,000) in 2019, when 53.4 μg/m^3 is used as the threshold value (Table 1). In 2015, the estimated national number of cardiovascular deaths was 73,000 (95% CI: 24,800-141,000), while respiratory deaths were 21,900 (95% CI: 0-46,400). These figures increased in 2019 to 112,000 (95% CI: 38,100-214,000) and 33,800 (95% CI: 0-71,400), when O_3 concentrations rolled back from the threshold value of 53.6 μg/m^3. This study estimates that cardiovascular and respiratory-related deaths accounted for 61.6% and 18.6% of all-cause mortality. From 2015-2019, the all-cause, cardiovascular and respiratory-related deaths attributed to O_3-exposure increased by 52.5%, 52.9% and 54.6% respectively. The increase in O_3 concentrations and population (size + age) was responsible for 38% and 7.9% increases in all-cause premature deaths. All-cause, cardiovascular and respiratory deaths increased by 2.4%, 2.6% and 3.8% respectively due to increases in the baseline death rate. The cities with the highest five-year average O_3-attributed all-cause deaths were observed in the high population regions of Shanghai [4,600 (95% CI: 2,300-8,900)], Beijing [2,900 (95% CI: 1,500-5,700)], Weifang (Shandong) [1,900 (95% CI: 1,000-3,700)], Linyi (Shandong) [1,800 (95% CI: 1,000-3,600)] and Baoding (Hebei) [1,8 (95% CI: 1,000-3,500)]. The provinces with >5% increase in all-cause mortality were Shandong (11.5%), Jiangsu (9.6%), Henan (9.3%), Guangdong (7%) and Hebei (6.7%), highlighting the need to focus on these regions for O_3 pollution control (Table 1).

The estimation of premature deaths attributed to O_3 is sensitive to the threshold value used in the model. The average all-cause mortality was 456,000 (95% CI: 233,000-864,000) when the threshold was 0 μg/m^3 and this figure was reduced to 1,000 (95% CI: 500-2,000) when the threshold was set to 100 μg/m^3. The estimated average cardiovascular and respiratory mortalities were 279,000 (95% CI: 98,000-517,000) and 83,000 (95% CI: 0-166,000) per year respectively, at a threshold of 0 μg/m^3. The corresponding values were very low, 600 (95% CI: 200-1,200) and 200 (95% CI: 0-400) per year when the threshold was 100 μg/m^3, as the average population exposed to grater than 100 μg/m^3 AVGDMA8 O_3 concentrations was only 80 million (Table S2).

3.4. Short-term premature mortality attributed to O_3

In this study, we estimated that about 923 million people (71% of the total population) are exposed to five-year mean 4DMA8 O_3 concentrations greater than 160 μg/m^3 and about 345 million people (27%) are exposed to 4DMA8 O_3 concentrations above 200 μg/m^3. The city-specific and cause-specific short-term mortality estimates are shown in Fig. 5 and Figs. S6-S7. The detailed data of short-term mortalities of all-cause, cardiovascular and respiratory in 31 provinces in 2015 and 2019 are reported in Table 1 and Table S2. In China, the short-term all-cause premature mortality attributed to ambient 4DMA8 O_3 exposure increased by 19.6%, from 131,000 (95% CI: 71,300-190,000) in 2015 to 156,000 (95% CI: 850,300-227,000) in 2019. The highest number of deaths was estimated in 2017 [163,000 (95% CI: 88,800-236,000)]. Short-term cardiovascular and respiratory mortality increased by 19.8%, from 41,400 (95% CI: 23,000-99,000) to 73,500 (95% CI: 27,500-119,000) and 21.2%, from 23,600 (95% CI: 11,900-35,300) to 28,600 (95% CI: 14,500-42,800) respectively, during the study period.

At the provincial level, higher numbers of all-cause premature deaths were observed in Shandong (10%) [2015: 13,300; 2019: 15,400], Jiangsu (8.7%) [2015: 11,800; 2019: 13,500] and Henan (8.4%) [2015: 10,400; 2019: 13,000]. A large percentage increase in premature deaths was estimated in Anhui (65.8%), Chongqing (56.8%), Fujian (42.6%), and Tianjin, Jiangxi, Shanxi, Hebei and Hunan provinces (30%-39%). The mortality burden decreased in Xinjiang, Jilin and Inner Mongolia provinces (-15.7%, -6.6% and -1.4% respectively), and increased slightly over Ningxia and Guangxi provinces (0.2% and 1.8%). In 2015, the highest all-cause mortality was observed in four cities: Shanghai [3,500 (95% CI: 1,900-5,000)], Beijing [3,300 (95% CI: 1,800-4,800)], Baoding [95% CI: 1,600 (800-2,200)] and Tianjin [95% CI: 1,500 (800-2,200)]. By 2019, Shanghai [3,700 (95% CI: 2,000-5,400)], Beijing [3,600 (95% CI: 2,000-5,200)], Baoding [2,000 (95% CI: 1,100-2,900)] and Tianjin [2,100 (95% CI: 1,100-3,000)] still had the highest number of O_3-attributed short-term all-cause mortality.

At the 0 μg/m^3 4DMA8 O_3 concentration threshold, the average estimated number of premature deaths by all-causes, cardiovascular are respiratory disease were 244,000 (95% CI: 134,000-352,000), 115,000 (95% CI: 43,000-184,000) and 44,000 (95% CI: 22,000-65,000) per year, respectively. The corresponding values were 108,000 (95% CI: 59,000-156,000), 51,000 (95% CI: 19,000-82,000) and 20,000 (95% CI: 10,000-30,000) respectively, when 100 μg/m^3 4DMA8 O_3

concentrations were selected as the threshold. Further details about premature deaths at 0 and 100 μg/m^3 thresholds from 2015 to 2019 are reported in Table S2.

4. Discussion

This study has characterized the trends in DMA8, AVGDMA8 and 4DMA8 O_3 concentrations in China during 2015-2019. Then AVGDMA8 and 4DMA8 O_3 concentrations were used to estimate the premature mortality attributable to long-term and short-term O_3 exposures. The study found that the O_3 concentrations and corresponding premature deaths have increased significantly in most provinces in China. This severity of O_3 pollution raises a new challenge in China, where the focus so far has been to control the $PM_{2.5}$ concentrations. There are various explanations for the failure to reduce O_3 concentrations in different provinces in China, (a) Recent bottom-up emission estimations and satellite formaldehyde observations have indicated increasing anthropogenic VOCs in eastern China, causing a positive trend in O_3 concentration (M. Li et al., 2019). (b) The long-distance transport of O_3 precursors is the largest contributor to ground O_3 in the Tibetan Plateau, BTH, YRD and PRD regions (Liu and Wang, 2020). (c) $PM_{2.5}$ acts as a scavenger of hydroperoxy (HO_2) and NO_x radicals, which contribute to the formation of O_3. The aim of reducing $PM_{2.5}$ concentration has led to an increase in these radicals, causing an increase in O_3 concentrations (K. Li et al., 2019).

The substantial heterogeneous increase in O_3 across China is consistent with the findings of previous studies (Silver et al., 2018; Wu et al., 2019; Ma et al., 2016). The heterogeneous distribution is mainly due to (a) O_3 being a secondary pollutant, mainly produced by a series of photochemical reactions between its precursors (NO_x and VOCs). The relationship between O_3 and its precursor is generally nonlinear. The O_3-NO_x-VOCs sensitivity relationship determines the types of O_3 pollution in different regions. In brief, when the concentration of NO_x in the atmosphere is high, the generation of O_3 is controlled by VOCs, however, when the VOCs concentration in the atmosphere is high, O_3 generation is controlled by NO_x (Yang et al., 2020). (b) In most cities, including Beijing, Tianjin, Shanghai and Guangzhou, O_3 generation is VOCs-sensitive, mainly because human intervention in urban districts have greatly affected the emissions of precursors. Industry and transportation caused a large amount of NO_x emissions, and the titration effect suppressed the increase in the O_3 concentration in urban areas (Wang et al., 2019; Lu et al., 2018). (c) In different regions, meteorological factors have heterogeneous effects on O_3 generation, especially in eastern China (Wang et al., 2017; Han et al., 2020).

Air pollution-attributed deaths can be categorised into- long-term effects, short-term effects and mixed-effects. For mixed-effects, air pollution may have played a role both in increasing the decedent's underlying susceptibility or frailty and in triggering the event. For example, patients with chronic bronchitis enhanced by long-term air pollution exposure may be hospitalized with an acute, air pollution-related exacerbation of their illness, leading to death shortly afterwards. The cohort-based effect estimates capture the full number of deaths across all three types of air pollution-attributable cases. However, deaths due to short-term "acute" advancement of death (short-term and mixed-effects) cannot be disentangled from deaths due to air pollution-enhanced chronic morbidity (long-term effects). The percentage of mixed-effects have not quantified in past studies (Kunzli, 2001; Giani et al., 2020). In the past, only a few studies quantified the O_3-related premature mortalities in China, and most were focused on the long-term premature deaths. During 2015-2019 in China, 1288 million people were exposed annually to 4DMA8 O_3 concentration greater than 100 μg/m^3 (WHO air quality guideline). However, only 80 million people were exposed to above 100 μg/m^3 AVGDMA8 O_3 concentration. Therefore, premature death due to short-term O_3 exposure cannot be ignored.

The threshold values and health endpoints adopted from the several existing epidemiological studies varied, resulting in a broad range in the estimated mortality values. The use of different health-related O_3 metrics in long-term and short-term exposure play an important role in health risk studies (Liu et al., 2018). At a threshold of 0 μg/m^3 (null concentration), long-term premature mortality plays an important factor due to the high exposure-response coefficient for long-term mortality, whereas at the higher threshold value (100 μg/m^3), short-term premature mortality is the dominant factor because of the greater number of exposed people above that threshold (Table S2).

In the present study, the adopted relative risk for all-cause long-term mortality is higher than the used relative risk for all-cause short-term mortality – although, in 2015, the estimated short-term all-cause mortality was higher than the estimated long-term all-cause mortality (Table 1). The probable explanation is that in 2015, the exposed population that experienced O_3-concentration above the selected threshold value was higher in short-term mortality (1285 million) than in long-term mortality (1225 million), and for short-term mortality, the estimated mean ΔC was 97 μg/m^3 whereas for long-term mortality, mean ΔC was 20 μg/m^3 (Eq. 1). In 2019, the long-term mortality value crossed the short-term mortality. This was because (a) during the study period, the mean AVGDMA8 O_3 concentration increased by 11%, and mean 4DMA8 values increased by only 4%. (b) In the megacity-cluster (the most populous area) of BTH and PRD and YRD regions, AVGDMA8 O_3 values increased by 18, 16 and 1%, whereas 4DMA8 O_3 concentration increased by 9, 13 and -1%, respectively. (c) The exposed population above the threshold concentration increased by 6% for long-term mortality but only by 1% for short-term mortality.

The past studies that make estimates premature deaths using different forms of O_3 concentration data (i.e. ground-level monitoring data, chemical transport models (CTM), satellite data) gives different results (Ghude et al., 2016; Feng et al., 2019; Seltzer et al., 2018). Malley et al. (2017) and Chowdhury et al. (2020) estimated the highest long-term respiratory mortality, 316,000 in 2010 and 230,000 in 2015, respectively. Both studies selected the same threshold value (53.4 $\mu g/m^3$) and health metric (AVGDMA8) and adopted the identical respiratory mortality-related risk factor from the epidemiological study by Turner et al. (2016). Despite the increasing O_3 concentration in China, the estimated respiratory premature deaths were different, mainly due to the use of different CTM (GEOS-Chem model and ECHAM/MESSy model) to estimated surface O_3 concentration, and different use of the baseline mortality rate. With similar parametric values, and based on the ground-level monitoring data, Seltzer et al. (2018) reported 200,000 respiratory deaths in 2015. In the present study, we estimated 82,800 (95% CI: 58,000-105,000) premature respiratory deaths in five-years average using the Turner et al. (2016) study(Table S3). Lin et al. (2018) and Liu et al. (2018) selected 75.2 $\mu g/m^3$ as the threshold and 6DMA1 as the health metrics and estimated the O_3-related COPD mortality to be 89,400 in 2014 and 71,900 in 2015. Both studies used WRF-CMAQ to simulate the ground O_3 with a resolution of 36 km×36 km and respiratory mortality-related relative risk from an epidemiolocal study by Jerrett et al. (2009). Even so, the values were different, mainly due to the higher simulated ground level 6DMA1 O_3 concentration in 2014 [150 $\mu g/m^3$, Lin et al., (2018)] compared to 2015 [108 $\mu g/m^3$, Liu et al., (2018)].

Multiple recent studies in China have indicated a consistent association with all-cause mortality and have provided evidence for associating respiratory and cardiovascular mortality with short-term exposure to higher O_3 concentrations (Yin et al., 2017; Lei et al., 2019). Liang et al. (2019) reported 160,000, 54,000 and 27,000 all-cause, cardiovascular and respiratory mortality respectively in China in 2016. Yao et al. (2020) estimated 310,000, 170,000 and 45,700 all-cause, cardiovascular and respiratory mortality respectively in 2017. Significantly higher estimated mortality figures in the Yao et al. (2020) study was mainly due to the use of 0 μg/m^3 as a threshold value, although past clinical studies have reported no respiratory symptoms below 70 μg/m^3 and the evidence for linearity does not extend to zero (US EPA, 2013). Using 70 μg/m^3 as the threshold and SOMO35 as the health metric, Feng et al. (2019) estimated 74,000 short-term respiratory deaths in 2015.

Compared with the previous studies, the key strength of our study lies in the fact that it is the first to estimate long-term and short-term O_3-attributed all-cause, cardiovascular and respiratory-related premature mortalities in 350 Chinese cities based on ground-level measurements during 2015-2019. However, there are some limitations. First, we used a long-term O_3-exposure-associated risk factor derived from a US cohort study, but the risk factor estimates for the US population may not apply to China due to the differences in factors like race, education background, marital status, dietary conditions, alcohol consumption, cigarette-smoking status, socioeconomic status and body mass index (BMI). That said, the range of ambient DMA8 O_3 concentration for urban China is similar to that observed in the USA by Turner et al. (2016) and Lim et al. (2019) study. Second, it may be argued that the relative risks of O_3 exposure may vary from city to city in China, whereas we used a constant value across all regions in our study. City- or region-specific relative risks in China are so far unavailable. Third, the study mainly focused on urban areas, but rural populations are also exposed to long-distance transported O_3 (Seltzer et al., 2018). Finally, we used a log-linear concentration-response function for the quantitative health impact assessment study, although we are aware that linear functions have been used in previous studies, which could provide different estimates (Liu et al., 2018).

5. Conclusions

To the best of our knowledge, this is the first study to evaluate the health burden attributable to long-term and short-term exposure to ambient O_3 in China at the national level from 2015-2019. Decades of hard work and effective air quality management strategies and practices have seen significant improvements in $PM_{2.5}$ levels across China, although several O_3 episodes and upward trending concentrations have largely gone unnoticed. This study found that for 2015-2019, daily 8 h maximum average (DMA8) O_3 concentrations continually increased across China at a rate of 1.9±3.3 μg/m^3/yr. This is estimated to cause approximately 163,000 (95% CI: 82,200-316,000), 100,000 (95% CI: 34,200-192,000) and 30,200 (95% CI: 0-63,800) premature long-term all-cause, cardiovascular and respiratory deaths attributed to O_3. Exposure to O_3 during 2015-2019 has been estimated to cause an additional 149,000 (95% CI: 81,300-216,000), 70,100 (95% CI: 26,200-113,000) and 27,000 (95% CI: 13,700-40,600) short-term all-cause, cardiovascular and respiratory deaths in 350 cities in China. We also found that some highly populated provinces showed a faster increase in O_3 levels and these provinces could be targeted to adopt a series of strict air pollution control measures to reduce the public health and economic burdens. To further improve air quality in China, we suggest increasing the focus on the control of O_3 precursor emissions from the chemical and solvent industries. Consistent air pollution control interventions will be needed to ensure long-term prosperity and environmental sustainability in China. The development of more city-specific, province-specific and long-term epidemiological studies with specific demographic characteristics will help to quantify health impacts more accurately.

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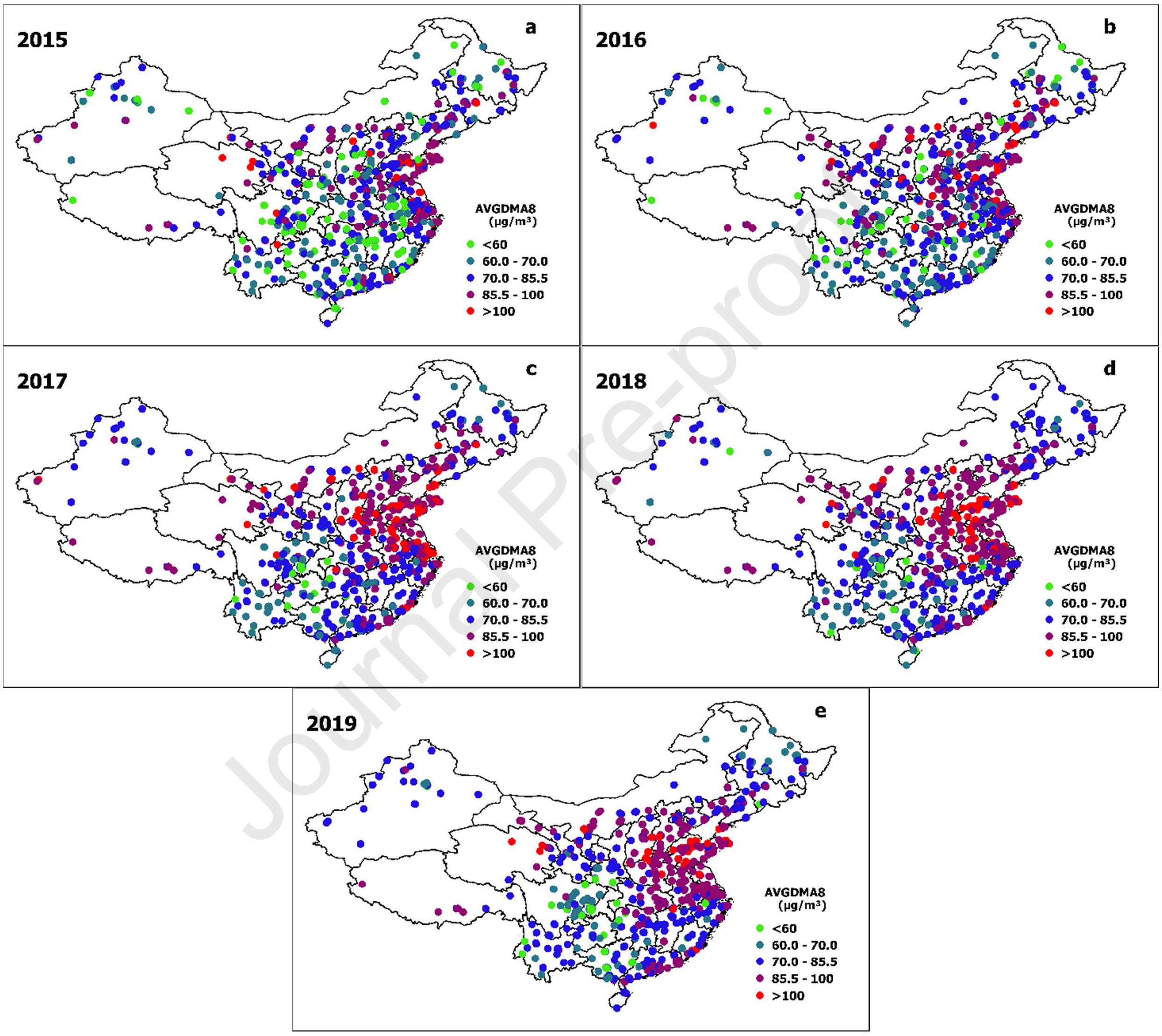

Fig.1. Monitoring site-specific spatiotemporal distributions of annual averaged daily 8-h maximum average (AVGDMA8) ozone (μg/m^3) in China in 2015−2019

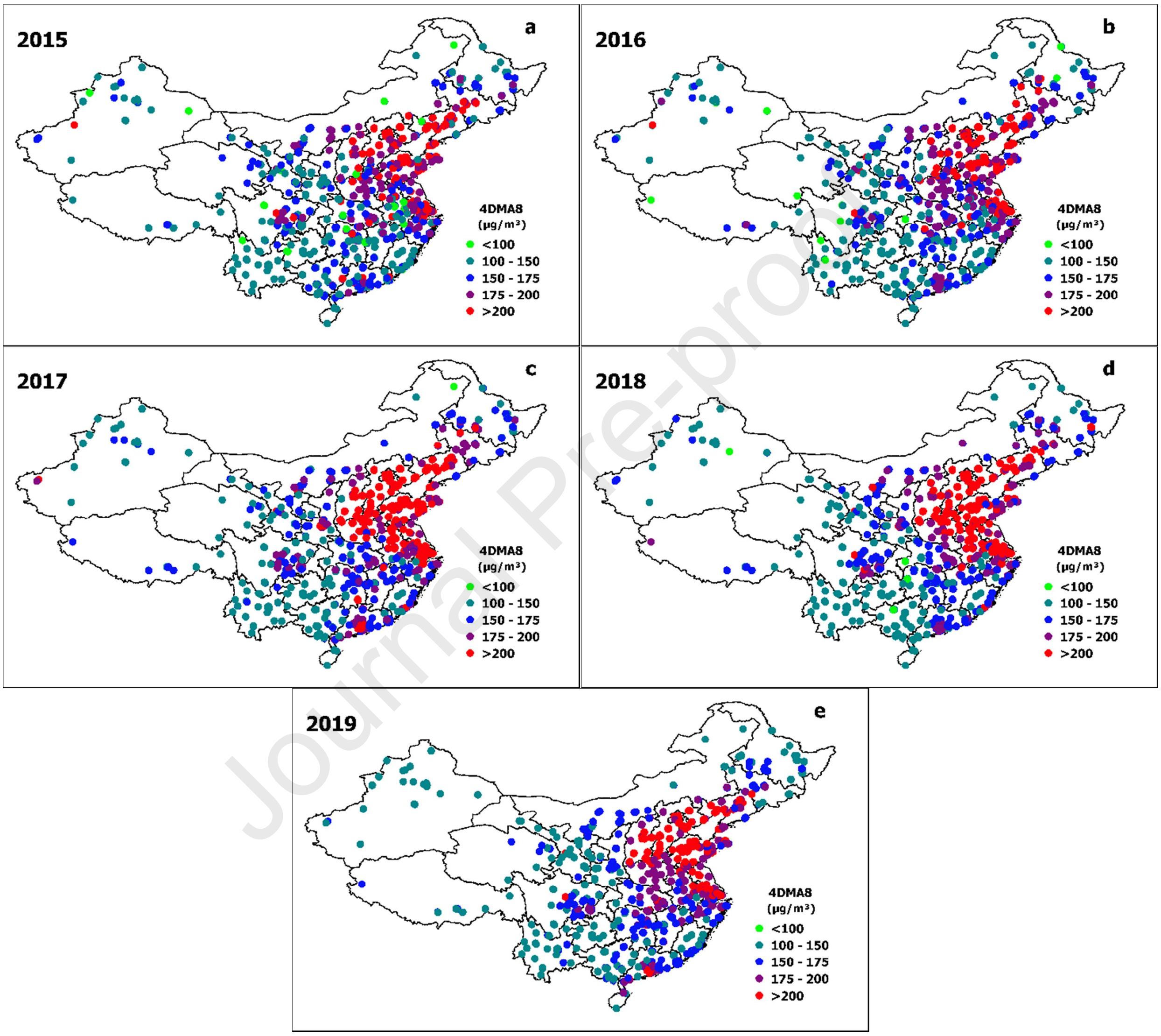

Fig. 2. Monitoring site-specific spatiotemporal distributions of April−September (warm-season) averaged 4th highest daily 8-h maximum average (4DMA8) ozone (μg/m^3) in China in 2015−2019

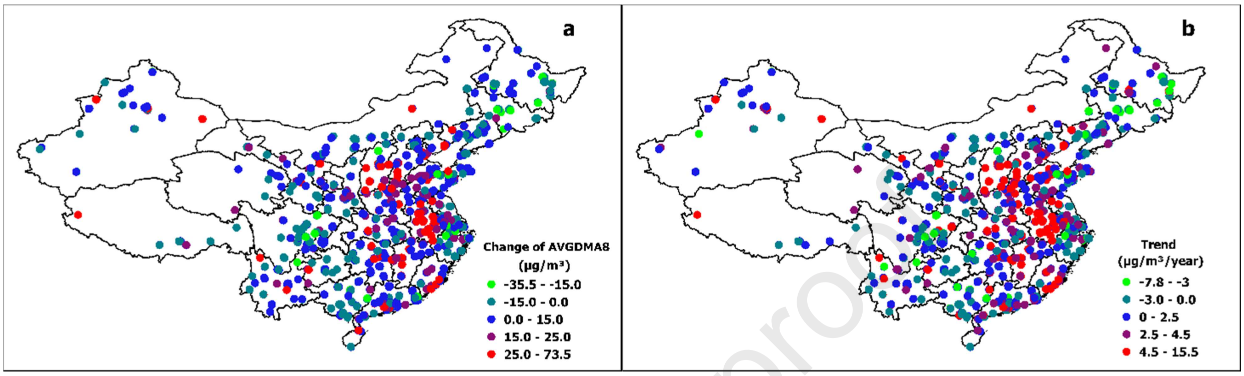

Fig. 3. Monitoring site-specific (a) change of annual averaged daily 8-h maximum average (AVGDMA8) ozone (μg/m^3) (2015-2019) (b) trend of daily 8-h maximum average (DMA8) (μg/m^3/year) during 2015-2019

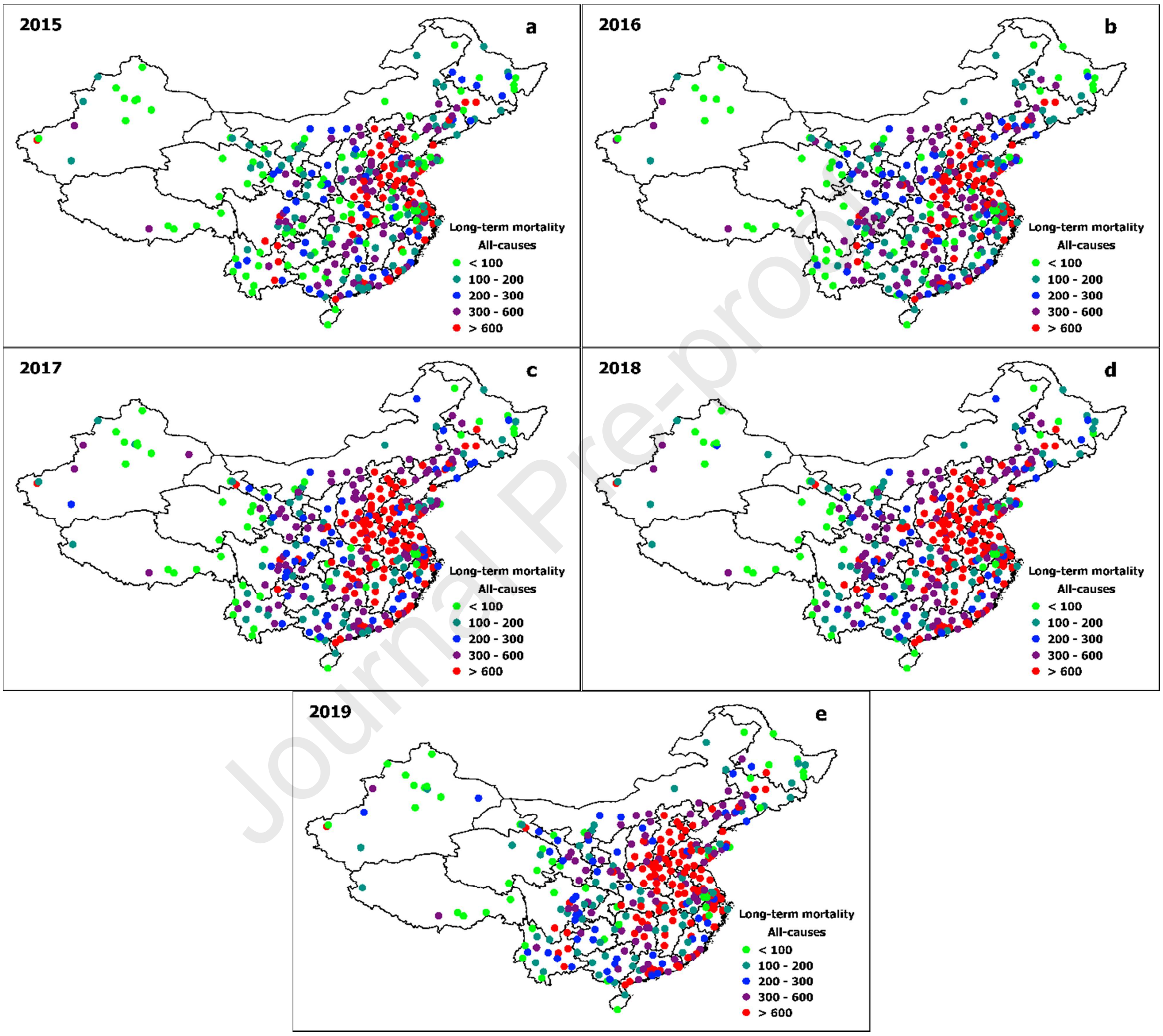

Fig. 4. City-specific spatial distribution of the long-term premature deaths in all-cause in China from 2015 to 2019

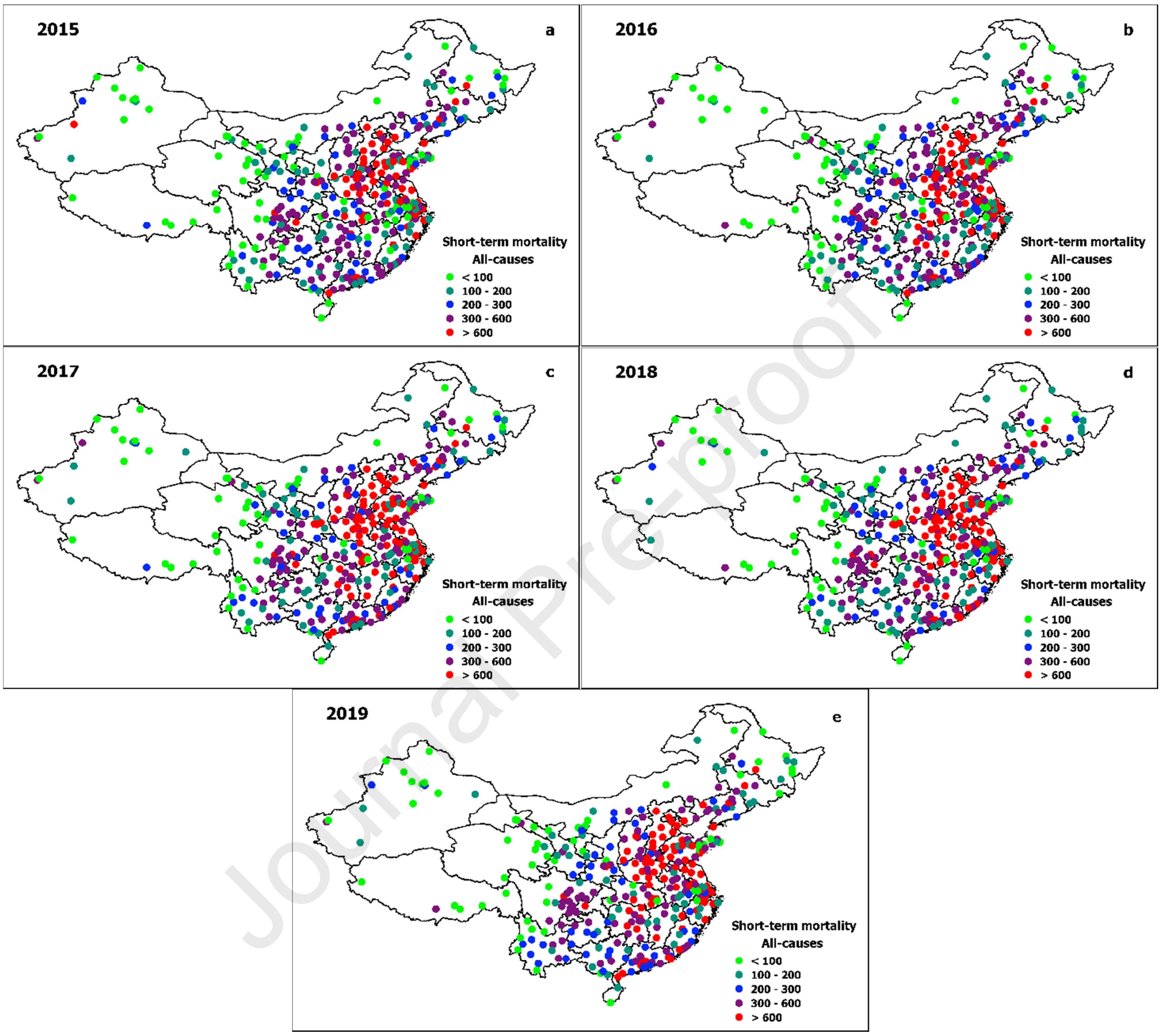

Fig. 5. City-specific spatial distribution of the short-term premature deaths in all-cause in China from 2015 to 2019

Table 1. The estimated cause-specific premature deaths attributed to long-term and short-term exposure to ozone in China and province with high premature deaths.

Region	Year	Long-term premature mortality ($\times 10^3$/yr) (95% CI)			Short-term premature mortality ($\times 10^3$/yr) (95% CI)		
		All-cause	Cardiovascular	Respiratory	All-cause	Cardiovascular	Respiratory
China	2015	119 (59.9-231)	72.9 (24.8-141)	21.9 (0-46.4)	131 (71.3-190)	61.4 (23.0-99.0)	23.6 (11.9-35.3)
	2016	140 (70.9-274)	86.4 (29.5-167)	25.8 (0-54.6)	138 (74.9-199)	64.7 (24.2-104)	24.6 (12.5-37.0)
	2017	183 (92.8-356)	113 (38.6-217)	34.3 (0-72.2)	163 (88.8-236)	76.5 (28.7-123)	29.7 (15.1-44.4)
	2018	189 (95.9-368)	117 (39.9-224)	35.4 (0-74.4)	158 (86.1-229)	74.3 (27.8-120)	28.8 (14.6-43.2)
	2019	181 (91.5-352)	112 (38.1-214)	33.8 (0-71.4)	156 (85.3-227)	73.5 (27.5-119)	28.6 (14.5-42.8)
Shandong	Average 2015-19	18.6 (9.5-36.2)	11.5 (3.9-21.9)	3.5 (0-7.2)	14.9 (8.1-21.6)	7.0 (2.6-11.3)	2.7 (1.4-4.0)
Jiangsu	Average 2015-19	15.6 (7.9-30.3)	9.6 (3.3-18.4)	2.9 (0-6.1)	12.9 (7.1-18.7)	6.1 (2.3-9.8)	2.3 (1.2-3.5)
Henan	Average 2015-19	15.0 (7.6-29.3)	9.3 (3.2-17.8)	2.8 (0-5.9)	12.5 (6.8-18.1	5.9 (2.2-9.5)	2.3 (1.2-3.4)
Guangdong	Average 2015-19	11.4 (5.8-22.4)	7.0 (2.4-13.7)	2.1 (0-4.5)	10.7 (5.8-15.5)	5.0 (1.9-8.1)	1.9 (1-2.9)
Hebei	Average 2015-19	10.9 (5.5-21.3)	6.7 (2.3-12.9)	2.0 (0-4.3)	11.3 (6.2-16.3)	5.3 (2.0-8.5)	2.0 (1.0-3.0)

Table 1. The estimated cause-specific premature deaths attributed to long-term and short-term exposure to ozone in China and province with high premature deaths.

Region	Year	Long-term premature mortality ($\times 10^3$/yr) (95% CI)			Short-term premature mortality ($\times 10^3$/yr) (95% CI)		
		All-cause	Cardiovascular	Respiratory	All-cause	Cardiovascular	Respiratory
China	2015	119 (59.9-231)	72.9 (24.8-141)	21.9 (0-46.4)	131 (71.3-190)	61.4 (23.0-99.0)	23.6 (11.9-35.3)
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Highlights

- Ozone (O_3) trend and corresponding health burden in China are analyzed from 2015-2019.
- O_3 concentrations have increased by 1.9±3.3 μg/m^3/year during the study period.
- The O_3-attributed long-term all-cause of deaths have increased by 52.5%.
- The O_3-attributed short-term all-cause of deaths have increased by 19.6%.
- Monitoring site-area specific control policies should be tailored to specific issues.

Author contributions:

KJM: Conceptualization, Methodology, Formal analysis, Data curation, Writing - original draft.
AN: Writing - review & editing, Visualization, Supervision.

Declaration of competing 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.