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# Journal Pre-proof

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## Effects of China's Current Air Pollution Prevention and Control Action Plan on Air Pollution Patterns, Health Risks and Mortalities in Beijing 2014-2018

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### Abstract:

Beijing is one of the most polluted cities in the world. However, the “Air Pollution Prevention and Control Action Plan” (APPCAP), introduced since 2013 in China, has created an unprecedented drop in pollution concentrations for five major pollutants, except O<sub>3</sub>, with a significant drop in mortalities across most parts of the city. To assess the effects of APPCAP, air pollution data were collected from 35 sites (divided into four types, namely, urban, suburban, regional background, and traffic) in Beijing, from 2014 to 2018 and analysed. Simultaneously, health-risk based air quality index (HAQI) and district-specific pollution (PM<sub>2.5</sub> and O<sub>3</sub>) attributed mortality were calculated for Beijing. The results show that the annual PM<sub>2.5</sub> concentration exceeded the Chinese national ambient air quality standard Grade II (35 µg/m<sup>3</sup>) in all sites, ranging from 88.5±77.4 µg/m<sup>3</sup> for the suburban site to 98.6±89.0 µg/m<sup>3</sup> for the traffic site in 2014, but was reduced to 50.6±46.6 µg/m<sup>3</sup> for the suburban site, and 56.1±47.0 µg/m<sup>3</sup> for the regional background in 2018. O<sub>3</sub> was another most important pollutant that exceeded the

Grade II standard ( $160 \mu\text{g}/\text{m}^3$ ) for a total of 291 days. It peaked at  $311.6 \mu\text{g}/\text{m}^3$  in 2014 for the urban site and  $290.6 \mu\text{g}/\text{m}^3$  in 2018 in the suburban site. APPCAP led to a significant reduction in  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ ,  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{CO}$  concentrations by 7.4, 8.1, 2.4, 1.9 and  $80 \mu\text{g}/\text{m}^3/\text{year}$  respectively, though  $\text{O}_3$  concentration was increased by  $1.3 \mu\text{g}/\text{m}^3/\text{year}$  during the five-years. HAQI results suggest that during the high pollution days, the more vulnerable groups, such as the children, and the elderly, should take additional precautions, beyond the recommendations currently put forward by Beijing Municipal Environmental Monitoring Center (BJMEMC). In 2014,  $\text{PM}_{2.5}$  and  $\text{O}_3$  attributed to 29,270 and 3,030 deaths respectively, though in 2018 their mortalities were reduced by 5.6% and 18.5% respectively. The highest mortality was observed in Haidian and Chaoyang districts, two of the most densely populated areas in Beijing. Beijing's air quality has seen a dramatic improvement over the five-year period, which can be attributable to the implementation of APPCAP and the central government's determination, with significant drops in the mortalities due to  $\text{PM}_{2.5}$  and  $\text{O}_3$  in parallel. To further improve air quality in Beijing, more stringent regulatory measures should be introduced to control volatile organic compounds (VOCs) and reduce  $\text{O}_3$  concentrations. Consistent air pollution control interventions will be needed to ensure long-term prosperity and environmental sustainability in Beijing, China's most powerful city. This study provides a robust methodology for analyzing air pollution trends, health risks and mortalities in China. The crucial evidence generated forms the basis for the governments in China to introduce location-specific air pollution policy interventions to further reduce air pollution in Beijing and other parts of China. The methodology presented in this study can form the basis for future fine-grained air pollution and health risk study at the city-district level in China.

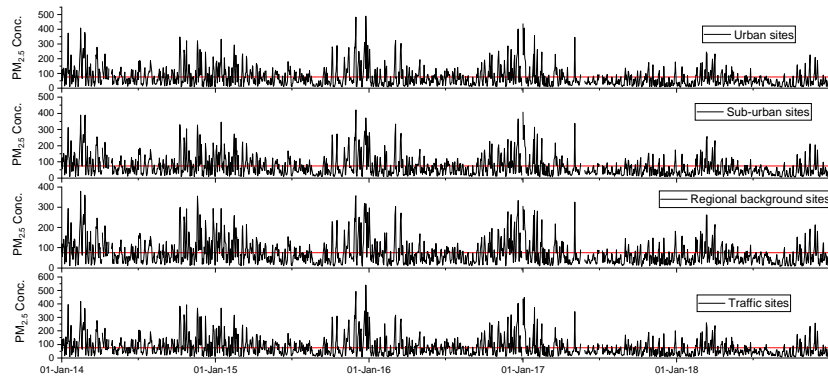
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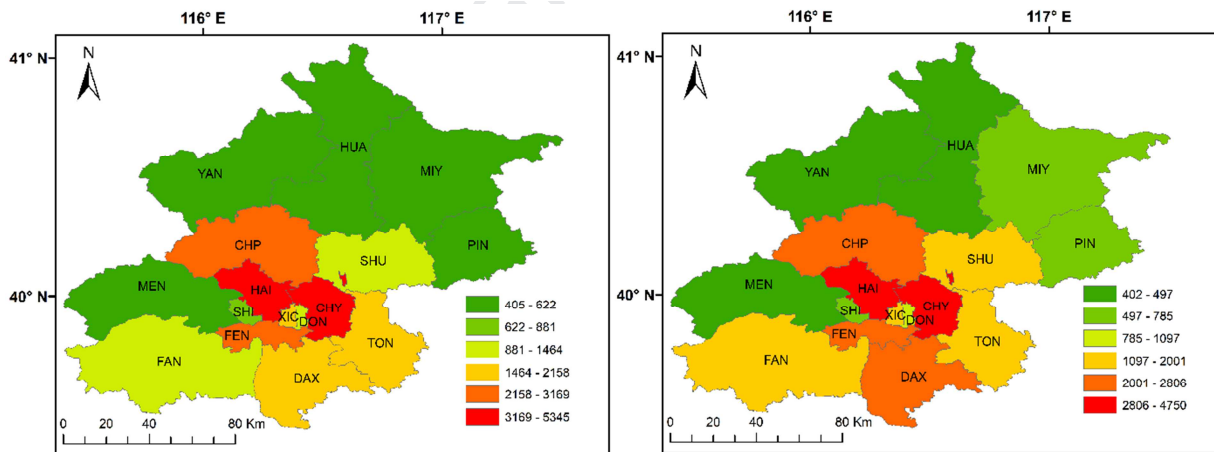
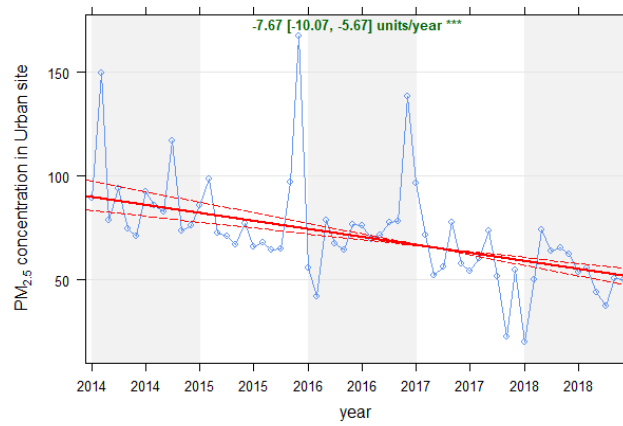
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a



(Top) Time series data of air pollution in four regions in Beijing from 2014 to 2018. (Middle) Trend analysis of deseasonalised time series data. (Bottom) District specific premature death attributed to air pollution

1 **Effects of China's Current Air Pollution Prevention and Control Action Plan**  
2 **on Air Pollution Patterns, Health Risks and Mortalities in Beijing 2014-2018**

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52

### 53 1. Introduction

54 Over the past decades, China has experienced rapid industrialization and economic growth and  
55 become the world's second-largest economy (WB, 2019). Due to air pollution from industries,

56 urbanization and higher energy consumption, the country has to subsequently take up serious  
57 health burden, and associated economic loss (Lelieveld et al., 2015; Zhang et al., 2017; Guan et  
58 al., 2019). The Global Burden of Disease (GBD) study estimated that PM<sub>2.5</sub> exposure led to 852  
59 thousand deaths in China in 2017 (Stanaway et al., 2018) and projected the number of deaths can  
60 have reached 2.3 millions by 2030 and 2.7 millions by 2060 (Xie et al., 2019; OECD, 2016). The  
61 health burden will significantly impact China's economy, costing about 2% in 2030 and 2.6 % in  
62 2060 of China's GDP (OECD, 2016; Xie et al., 2019). After the 2008 Summer Olympics had  
63 been completed in Beijing, air pollution received the highest awareness in China from the public,  
64 decision-makers and academic community. Special focus was put on Beijing, the capital of  
65 China, which suffered from an extremely high level of PM<sub>2.5</sub> pollution (Wang et al., 2010; He et  
66 al., 2016).

67 To overcome the air pollution challenge, China's State Council initiated the "Air Pollution  
68 Prevention and Control Action Plan" (APPCAP) in 2013 which set the targets to reduce air  
69 pollution in most polluted cities across China by 2017 (CSC, 2013). To implement the national  
70 APPCAP and further improve air quality, Beijing Municipal Government additionally  
71 formulated and released the "Beijing Clean Air Action Plan: 2013-2017", which set a target for  
72 the yearly average concentration of PM<sub>2.5</sub> to fall below the threshold of 60 µg/m<sup>3</sup> by 2017  
73 (CCICED, 2013). It is of great interest to the government, policymakers and the overall general  
74 public to know whether or not Beijing's air quality meets the set targets. However, air quality  
75 study in Beijing is complicated by the use of coal for heating and domestic cooking, transport of  
76 air pollutants from neighbouring provinces, adverse meteorological conditions during the winter  
77 and the high oxidizing power that associates with the complex chemical composition (Li et al.,  
78 2017; Lu et al., 2018).

79 Several studies were conducted to investigate the air pollution in Beijing with limited  
80 observational pollutants data, in which the main focus was on the urban site or an entire area  
81 with an average of all monitoring stations data (Wang et al., 2018). Zhang et al., (2018) observed  
82 the decrease of PM<sub>2.5</sub> concentration at the urban and the regional background site by 3.40 and  
83 1.16 µg/m<sup>3</sup>/year from 2014 to 2015, based on a single monitoring station in each location. The  
84 fine particle explosive growth events, caused by the rapid increase in PM<sub>2.5</sub> mass concentration  
85 over a few hours, steadily decreased from 39 events in 2013 to 19 events in 2017 (Liu et al.,  
86 2019), and the number of winter haze days also decreased from 47 days in 2012 to 15 days in

87 2017 (Dang and Liao, 2019). Based on one urban monitoring data, Cheng et al. (2019) reported  
88 that the annual average maximum daily 8-hour average (MDA8-h) O<sub>3</sub> concentration increased  
89 from 61.4 µg/m<sup>3</sup> in 2006 to 96.7 µg/m<sup>3</sup> in 2017 with an increasing rate of 3.55 µg/m<sup>3</sup>/year. The  
90 increasing trend of ozone in Beijing is also reported by Huang et al., (2018). From 2013 to 2015,  
91 the 90<sup>th</sup> percentile of annual average MDA8-h O<sub>3</sub> of Beijing increased from 183.4 to 202.6 µg/m<sup>3</sup>  
92 (BMEPB, 2015). However, during these three years, the annual mean NO<sub>2</sub> concentration  
93 decreased from 56 to 50 µg/m<sup>3</sup> across the urban site (BMEPB, 2015).

94 The Beijing Municipal Environmental Monitoring Center (BJMEMC) empowered the Chinese  
95 citizens by providing the real-time hourly observations data of six air pollutants from 35  
96 monitoring sites to protect their health. Globally, the important indicator to inform the public to  
97 take proper outdoor activities is the Air Quality Index (AQI). However, the AQI neglects the  
98 synergistic health effects of exposure to multiple air pollutants. Employing the Health-risk based  
99 Air Quality Index (HAQI) method associated with exposure to multiple air pollutants and by  
100 comparing the AQI and HAQI, Hu et al., (2015) showed that AQI underestimated the health  
101 risks associated with exposures to multiple pollutants, especially during the extremely high air  
102 pollution days. The air pollution exposure and health impact assessment are state-of-the-art  
103 infrastructures to protect citizens from atmospheric pollution and safeguard their health (Shi et  
104 al., 2019). However, most of these health risk studies were undertaken at the city-level and  
105 underestimated the more fine-grained scale, district-based air pollution control policy  
106 development within Beijing itself. The ground-based air quality measurements serve the best  
107 data for pollution-attributed district-based mortality estimation (Yin et al., 2017; Maji et al.,  
108 2019).

109 The major objective of this study is to examine the temporal and the spatial variation of six  
110 official pollutants based on real-time monitoring data collected at 35 sites across four typical  
111 sites in Beijing, for the period of 2014 - 2018. The second objective includes trend analysis,  
112 based on the Mann-Kendall test. The health risks associated with six official pollutants are  
113 evaluated based on HAQI and the number of district-based pollution-attributed (PM<sub>2.5</sub> and O<sub>3</sub>)  
114 premature deaths is also estimated for 16 districts in Beijing. This study provides a robust  
115 methodology of analyzing air pollution trends, health risks and mortality in China, the crucial  
116 evidence generated forms the basis for policy-makers in China to introduce location-specific air  
117 pollution policy interventions to further reduce air pollution in Beijing. The methodology

118 presented in this study can form the basis for future fine-grained air pollution and health risk  
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## 120 2. Methodology

### 121 2.1. Monitoring sites and data sources

122 Five-year time series (January 2014 to December 2018) of hourly concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>,  
123 O<sub>3</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO for Beijing city was downloaded from 35 monitoring stations operated by  
124 the BJMEMC (<http://beijingair.sinaapp.com/>), which had obtained the data from <http://pm25.in>.  
125 Before 2<sup>nd</sup> April 2014, no data was available for CO, O<sub>3</sub>, SO<sub>2</sub> and NO<sub>2</sub>. According to the  
126 monitoring function, the 35 monitoring stations are divided into 4 categories: the urban  
127 environmental monitoring site (12 stations), the suburban environmental monitoring site (11  
128 stations), the regional background transmission site (control site) (7 stations) and the traffic  
129 pollution monitoring site (5 stations) (BJMEMC, 2019). Each station contains automated  
130 monitoring systems utilized to measure the concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> based on the  
131 National Environmental Protection Standards (NEPS) HJ 655-2013 (MEP, 2013a), and O<sub>3</sub>, SO<sub>2</sub>,  
132 NO<sub>2</sub> and CO based on the NEPS HJ 193-2013 (MEP, 2013b). All four typical sites are listed in  
133 Table S1 and the locations are shown in Fig.1.

134 The potential quality of all the available data was assessed based on the criteria developed in  
135 previous studies (Song et al., 2017; Silver et al., 2018). After screening, if <90% of hourly data  
136 was available for the whole time series, it was removed. Based on the above criteria, all the data  
137 from the Mentougou suburban site was removed. The hourly data was used to calculate daily,  
138 seasonal and yearly averages for the four-type of sites in Beijing. To analyze the five-year time  
139 series for monotonic, linear trends, the Mann-Kendall (M-K) test was used to assess the  
140 significance of trends, and the Theil-Sen estimator was used to calculate the magnitude of the  
141 trend. The R package ‘openair’ was used to perform for air quality data analysis (R Core Team,  
142 2019; Carslaw and Ropkind, 2012). The M-K nonparametric test is utilized to test for a  
143 significant trend. Advantages of the M-K test are: (1) no distributional assumption is made; (2)  
144 no assumption of any specific functional form for the behaviour of the data through time is  
145 made; and (3) the M-K test is resistant to the effects of outlying observations. The results are not  
146 unduly affected by particularly high or low values that occur during the time series.

147 **Fig. 1.** Location of the 35 air quality monitoring sites in Beijing (Tian et al., 2019)

148

## 149 2.2. Health-risk based Air Quality Index (HAQI)

150 Health-risk based air quality indices (HAQI) were proposed in a few studies to include exposure-  
 151 response characteristics (Hu et al., 2015; Shen et al., 2017). To define a HAQI, the total excess  
 152 risk (ER) of exposure to multiple pollutants based on Cairncross's concept (Cairncross et al.,  
 153 2007) was used. In a general form, the excess risk ( $ER_i$ ) for pollutant  $i$  was estimated based on  
 154 the relative risk ( $RR_i$ ) of the pollutant, using the following expression:

$$155 \quad ER_i = RR_i - 1$$

156 (1)

157 where  $RR_i = \exp[\beta_i(Z_i - Z_{i,0})]$ , the exposure-response coefficient ( $\beta_i$ ) describes the increased  
 158 risk of a population associated with a certain health response (such as mortality) when exposed  
 159 to the pollutant  $i$ ,  $Z_i$  is the concentration of the pollutant and  $Z_{i,0}$  is the threshold concentration,  
 160 below which the pollutant demonstrates no adverse health effects.

161 For this study, RR values were chosen from the Chinese studies on long-term exposure to air  
 162 pollution and non-accidental daily mortality of all ages. The RR values are 1.06, 1.0111, 1.0169,  
 163 1.02 and 1.0255 per 10  $\mu\text{g}/\text{m}^3$  increase in concentrations of  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ ,  $\text{SO}_2$  and  $\text{NO}_2$ ,  
 164 respectively, the value is 1.02 per 20  $\mu\text{g}/\text{m}^3$  increase in  $\text{O}_3$  concentration and 1.0255 per one  
 165  $\text{mg}/\text{m}^3$  increase in CO concentration (Yang et al., 2013; Zhang et al., 2017; Turner et al., 2016;  
 166 Atkinson et al., 2018; T. Li et al., 2018).

167 It was assumed that the air pollution concentration below Chinese Ambient Air Quality  
 168 Standards (CAAQS) Grade II, posed little or no health risk. Therefore, the CAAQS 24-hour  
 169 Grade II standard for the six pollutants were used as  $Z_{i,0}$  (<https://cleanairasia.org/node8163/>).  
 170 However, some studies indicated these values could be regarded as the threshold values, as  
 171 below which a zero adverse response would be expected (WHO, 2005).

172 Total ER of all pollutants  $ER_{total}$  is the sum of  $ER_i$  for the individual pollutants as shown in Eq.

173 (2):

$$ER_{total} = \sum_{i=1}^n ER_i = \sum_{i=1}^n (RR_i - 1)$$

175 (2)

176 The equivalent relative risk ( $RR_i^*$ ) and equivalent concentration ( $Z_i^*$ ) is defined as Eq. (3) based  
 177 on the assumption that the  $ER$  of a pollutant  $i$  is equal to  $ER_{total}$  (Hu et al., 2015):

$$ER_{total} = RR_i^* - 1 = \exp[\beta_i(Z_i^* - Z_{i,0})] - 1$$

179 (3)

180 The equivalent concentration of  $i$  ( $Z_i^*$ ), incorporating the health effects from all pollutants was  
 181 used to calculate HAQI based on Eq. (4) and (5):

$$HAQI_i = \frac{(AQI_{high} - AQI_{low})}{(C_{high} - C_{low})} \times (Z_i^* - Z_{i,0}) + AQI_{low}$$

183 (4)

$$HAQI = \max(HAQI_i) \quad i=1 \text{ to } 6$$

185 (5)

186 where  $AQI_{high}$  is the index breakpoint corresponding to  $Z_{high}$ .  $AQI_{low}$  is the index breakpoint  
 187 corresponding to  $Z_{low}$ .  $Z_{high}$  is the concentration breakpoint that is larger than  $Z_i^*$ .  $Z_{low}$  is the  
 188 concentration breakpoint that is smaller than  $Z_i^*$ . The reference concentrations for the pollutants  
 189 in different health categories were provided by the Ministry of Ecology and Environment in  
 190 China (Hu et al., 2015, see Table S2 in Supplementary Material).

### 191 2.3. Health impact assessment

192 To avoid overestimation of mortality attributed to ambient air pollution, two independent  
 193 pollutants  $PM_{2.5}$  and  $O_3$  were used in the present study. In the first stage, the long-term mortality  
 194 attributable to pollutant  $i$  exposure was calculated based on the epidemiological hazard ratio or  
 195 relative risk  $RR_i$ , which could link the pollutant concentration to negative health effects. Then  
 196 pollutant-attributed premature deaths were estimated using the equation (Zheng et al., 2017):

$$197 \quad \Delta Mort_i = [1 - 1/RR_i] \times B_0 \times E_{pop}$$

198 (6)

199 where  $\Delta Mort_i$  is the excess premature death due to pollutant  $i$  exposure for a specific age  
200 group;  $B_0$  is the baseline death rate of a specific health outcome at a specific age and  $E_{pop}$  is the  
201 exposed population number.

202 The Log-Linear model, a commonly used method to estimate the relative risk  $RR_i$  for  $O_3$ , were  
203 drawn from the past (Pascal et al., 2013; Lin et al., 2017). The model is expressed as:

$$204 \quad RR_i(Z) = \exp(\beta_i Z), \quad \text{where } Z = \max(0, Z_i - Z_{i,0})$$

205 (7)

206 where  $Z_i$  represents the ambient concentration of pollutant  $i$  and with  $Z_{i,0}$  represents the  
207 threshold concentration of the pollutant, assuming no health risk association below  $Z_{i,0}$ .  $\beta_i$  is the  
208 exposure-response coefficient for pollutant  $i$ , can be derived from an epidemiological study. To  
209 estimate non-accidental  $O_3$ -attributed mortality,  $RR_i$  value from cohort study by Turner et al.,  
210 (2016) was used. District-specific age-specific population and the baseline mortality rate were  
211 extracted from Beijing Statistical Yearbook (BMBS, 2018). The present study used  $75.2 \mu\text{g}/\text{m}^3$   
212 as a threshold concentration for  $O_3$ , also recommended by Lelieveld et al., (2015).

213 As  $PM_{2.5}$  did not follow the LL exposure-response model, the integrated exposure-response  
214 (IER) model was developed by Burnett et al. (2014) for relative risk estimation. The cause-  
215 specific  $RR$  was calculated through Eq. (8);

$$216 \quad RR_{IER}(Z_a) = \begin{cases} 1 + \alpha(1 - \exp^{-\gamma(Z_a - Z_0)^\delta}), & \text{if } Z_a > Z_0 \\ 1, & \text{else} \end{cases} \quad (8)$$

217 where  $Z_a$  is the annual average ambient  $PM_{2.5}$  concentration;  $Z_0$  is the threshold concentration  
218 below which no additional health impacts are calculated; and  $\alpha$ ,  $\gamma$  and  $\delta$  are the parameters used  
219 to describe the different shapes of the relative risk curve among various diseases (Burnett et al.,  
220 2014). The theoretical minimum risk exposure level of  $2.4 \mu\text{g}/\text{m}^3$  is used for the health risk  
221 assessment. The age and cause-specific baseline death rates for 2014 to 2017 were downloaded  
222 from the GBD study (GBD, 2019). Following the GBD approach, premature deaths due to five

223 causes (IHD, Stroke, COPD, Lung Cancer and ALRI) attributed to PM<sub>2.5</sub> exposure were  
224 calculated for the age group  $\geq 25$  years and  $<5$  years and for O<sub>3</sub>-related non-accidental mortality,  
225 the age group  $\geq 25$  years were selected.

### 226 3. Results and discussion

#### 227 3.1. Annual and seasonal variation of air pollutants

228 Table 1 summarizes the annual average concentration of PM<sub>2.5</sub>, O<sub>3</sub>, NO<sub>2</sub> and SO<sub>2</sub> in four typical  
229 sites in Beijing. The annual PM<sub>2.5</sub> concentrations exceeded the Chinese national ambient air  
230 quality standards (CNAAQs) Grade II (35  $\mu\text{g}/\text{m}^3$ ) in all sites in 2014 to 2018 and varied from  
231 88.5 $\pm$ 77.4  $\mu\text{g}/\text{m}^3$  (suburban) to 98.6 $\pm$ 89.0  $\mu\text{g}/\text{m}^3$  (traffic) in 2014, although the range decreased  
232 to 50.6 $\pm$ 46.6  $\mu\text{g}/\text{m}^3$  (suburban) to 56.1 $\pm$ 47.0  $\mu\text{g}/\text{m}^3$  (regional background) in 2018. The average  
233 seasonal PM<sub>2.5</sub> concentration was in the order of winter > autumn > spring > summer in the year  
234 2014-2017 (Table 1). In the winter on average higher PM<sub>2.5</sub> was observed in the regional  
235 background (104.8  $\mu\text{g}/\text{m}^3$ ) and traffic site (105.4  $\mu\text{g}/\text{m}^3$ ) and in the summer on average lower  
236 PM<sub>2.5</sub> is observed in the suburban site (54.3  $\mu\text{g}/\text{m}^3$ ) and regional background site (54.2  $\mu\text{g}/\text{m}^3$ ).  
237 In 2018, the average PM<sub>2.5</sub> concentration in the spring is higher than winter in all sites due to  
238 smog events in March 2018 (Mullin, 2018). The average PM<sub>2.5</sub> concentration in the spring was  
239 69.2-73.5  $\mu\text{g}/\text{m}^3$  and 73.7-55.7  $\mu\text{g}/\text{m}^3$  in the winter in 2018. Predominantly, high PM<sub>2.5</sub> in the  
240 winter in Beijing was mainly attributed to the adverse meteorological conditions like low  
241 temperature and lower boundary layer height, less precipitation and weaker wind and solid fuel  
242 (coal) combustion for indoor heating (Che et al., 2014; Liu et al., 2019).

243 The annual average of MDA8-h ozone concentrations (AMDA8-h) was 112.8 $\pm$ 65.8  $\mu\text{g}/\text{m}^3$ ,  
244 115 $\pm$ 63.5  $\mu\text{g}/\text{m}^3$ , 117 $\pm$ 62.6  $\mu\text{g}/\text{m}^3$  and 83.6 $\pm$ 50.3  $\mu\text{g}/\text{m}^3$  in the urban, the suburban, the regional  
245 background and the traffic sites in 2014. AMDA8-h O<sub>3</sub> was almost stable from 2015 to 2017 in  
246 Beijing (ranging from 94.6 $\pm$ 60.5  $\mu\text{g}/\text{m}^3$  in 2015 to 94.8 $\pm$ 57.1  $\mu\text{g}/\text{m}^3$  in 2017) and further  
247 increased to 82.8 $\pm$ 50.5  $\mu\text{g}/\text{m}^3$  (traffic) to 105.4 $\pm$ 60.9  $\mu\text{g}/\text{m}^3$  (suburban) in 2018. O<sub>3</sub> concentration  
248 was a little higher in suburban area than others and the lowest was observed in the traffic site. In  
249 the traffic site, the dilution of O<sub>3</sub> is due to the higher photochemical reactions with NO<sub>2</sub> and CO,  
250 exhaust from vehicles. The seasonal O<sub>3</sub> concentration was in the order of  
251 summer > spring > autumn > winter in all the years. In 2014, the average daily maximum 8-h  
252 (MDA8-h) ozone was 125.9  $\mu\text{g}/\text{m}^3$  (traffic) to 167.7  $\mu\text{g}/\text{m}^3$  (suburban) in the summer and 39.9



253  $\mu\text{g}/\text{m}^3$  (traffic) to  $46.8 \mu\text{g}/\text{m}^3$  (suburban) in the winter. The MDA8-h ozone has a range from  
 254  $133.1 \mu\text{g}/\text{m}^3$  (traffic) to  $167.2 \mu\text{g}/\text{m}^3$  (suburban) in the summer and  $45.7 \mu\text{g}/\text{m}^3$  (traffic) to  $60.6$   
 255  $\mu\text{g}/\text{m}^3$  (background site) in the winter in 2018 (Table 1). The 90<sup>th</sup> percentile of annual MDA8-h  
 256 ozone concentrations in Beijing was quite high, at  $204.5 \mu\text{g}/\text{m}^3$  in 2014, and  $197.1 \mu\text{g}/\text{m}^3$  in  
 257 2018, which exceeds Grade II Standard ( $160 \mu\text{g}/\text{m}^3$ ) by 23.2%. Lu et al., (2019) pointed out that  
 258 the ozone concentration in the summer could be attributed to high temperature and low-humidity  
 259 conditions, inducing an increase of biogenic volatile organic compounds emissions and enhanced  
 260 ozone production rate.

261 The annual average  $\text{NO}_2$  concentration exceeded the CNAAQs Grade II ( $40 \mu\text{g}/\text{m}^3$ ) in all the  
 262 years in the urban (range:  $55.9 \pm 30.8 \mu\text{g}/\text{m}^3$  in 2014 to  $43.6 \pm 26.2 \mu\text{g}/\text{m}^3$  in 2018) and the traffic  
 263 (range:  $82.1 \pm 32.7 \mu\text{g}/\text{m}^3$  in 2014 to  $64.4 \pm 28.7 \mu\text{g}/\text{m}^3$  in 2018) sites, whilst  $\text{NO}_2$  never exceeded  
 264 the standard in regional background site (range:  $37.9 \pm 19.8 \mu\text{g}/\text{m}^3$  in 2014 to  $32.5 \pm 17.6 \mu\text{g}/\text{m}^3$  in  
 265 2018). In traffic site, relatively high  $\text{NO}_2$  was observed due to  $\text{NO}_x$  exhaust from vehicles.  
 266 Generally,  $\text{NO}_2$  concentration is higher in the winter and lower in the summer season in all site  
 267 (Table 1). The annual average  $\text{SO}_2$  concentration never exceeded even the CNAAQs Grade I ( $20$   
 268  $\mu\text{g}/\text{m}^3$ ) and gradually decreased from  $12.3$ - $16.0 \mu\text{g}/\text{m}^3$  in 2014 to  $5.9$ - $7.6 \mu\text{g}/\text{m}^3$  in the year 2018.

269 **Table 1.** Annual average pollution concentrations from 2014 to 2018 in four-type of sites in  
 270 Beijing (US, SUB, RBS and TS represent the urban site, the suburban site, the  
 271 regional background site and the traffic site; concentration unit is in  $\mu\text{g}/\text{m}^3$ )

272

### 273 3.2. Daily variation

274 The daily average  $\text{PM}_{2.5}$ , MDA8-h  $\text{O}_3$ ,  $\text{NO}_2$  and  $\text{SO}_2$  concentration ( $\mu\text{g}/\text{m}^3$ ) from 2014 to 2018  
 275 across four typical sites is shown in Fig.2a.  $\text{PM}_{2.5}$  shows a similar trend across all four typical  
 276 sites and most of the highest peaks were observed from January to February.  $\text{PM}_{2.5}$  values  
 277 remained flat from May to August, then they gradually increased from October to December. In  
 278 2014, from January-February, the daily  $\text{PM}_{2.5}$  concentration dropped gently from a maximum of  
 279  $378.6$ - $419.4 \mu\text{g}/\text{m}^3$  to a minimum of  $12.2$ - $14.6 \mu\text{g}/\text{m}^3$  in May-August, then it slowly increased to  
 280  $330.6$ - $393.6 \mu\text{g}/\text{m}^3$  during October-December. Whereas in 2018 relatively lower  $\text{PM}_{2.5}$   
 281 concentrations were observed during this time, and the maximum  $\text{PM}_{2.5}$  concentration was  
 282  $168.7$ - $184.3 \mu\text{g}/\text{m}^3$  between January-February, the minimum was  $10.0$ - $12.4 \mu\text{g}/\text{m}^3$  in May-

283 August and it peaked again at 211.6-258.4  $\mu\text{g}/\text{m}^3$  in October-December. The most extreme  
284 events occurred during November 27<sup>th</sup> to December 1<sup>st</sup> and December 19-26<sup>th</sup>, 2015 was related  
285 to the Southeast Asian haze, and the highest  $\text{PM}_{2.5}$  concentration shoots up to 537.0  $\mu\text{g}/\text{m}^3$  on  
286 December 25 (Koplitz et al., 2016; Z. Liu et al., 2019; Dang and Liao, 2019). The Taklimakan  
287 Desert in Northwest China witnesses frequent dust storm events, which bring about significant  
288 impacts on the downstream air quality (Li et al., 2018), and for that reason, the non-seasonal  
289  $\text{PM}_{2.5}$  concentration peak was observed during March to May in Beijing (BBC, 2017). The traffic  
290 site recorded the maximum number of days (190 days) which exceeded Grade II standard of  
291  $\text{PM}_{2.5}$  concentration (75  $\mu\text{g}/\text{m}^3$ ) while suburban site recorded the minimum number of days (169  
292 days) in 2014, and the corresponding number of days decreased to 80 and 71, respectively, in  
293 2018. The total number of days exceeding the Grade II standards for daily average  $\text{PM}_{2.5}$  during  
294 the period 2014-2018 was 626, accounting for 34% of the total number of days. It is important to  
295 note that urban and suburban sites have a total of 49 days (with 18 days in 2014 and 15 days in  
296 2015) and 41 days (with 16 days in 2014 and 12 days in 2015), in which  $\text{PM}_{2.5}$  concentrations  
297 were higher than 250.0  $\mu\text{g}/\text{m}^3$ , the AQI of these days should be considered as highly unhealthy.  
298 Fig.2b shows the MDA8-h  $\text{O}_3$  concentrations across four-type of sites during 2014-2018.  
299 Generally, the monthly variability of ground-level  $\text{O}_3$  concentrations peaked in May to August  
300 (summer) and was the lowest in January, February and December (winter). In 2014, The peak  
301 and valley values of ground-level MDA8-h  $\text{O}_3$  concentrations in the urban site were  
302 311.6  $\mu\text{g}/\text{m}^3$  and 6.0  $\mu\text{g}/\text{m}^3$ , which were reached in May and November, respectively. Whereas  
303 in 2018, the maximum of 290.6  $\mu\text{g}/\text{m}^3$  was reached in the suburban site in June and the  
304 minimum of 7.3  $\mu\text{g}/\text{m}^3$  was reached in the urban site in December. Relatively low MDA8-h  $\text{O}_3$   
305 was observed in the traffic site due to high photochemical reaction rates in higher  $\text{NO}_2$   
306 concentration. The total number of days exceeding Grade II standards (160  $\mu\text{g}/\text{m}^3$ ) for  $\text{O}_3$  was  
307 291 during 2014-2018 (63 days in 2014 and 62 days in 2018), accounting for 16% of the total  
308 number of days.

309 Typically,  $\text{NO}_2$  exhibits U-shape patterns in a year; lower values in May to August and higher  
310 values in January, November and December (Fig.2c). The traffic site showed flatter U-shapes  
311 due to continuous vehicular emission of  $\text{NO}_x$  in the site and the concentration was quite a bit  
312 higher than the other sites. In December 2014, the maximum  $\text{NO}_2$  concentration was 116.5  $\mu\text{g}/\text{m}^3$   
313 (suburban) to 148.3  $\mu\text{g}/\text{m}^3$  (traffic) and decreased to 77.3  $\mu\text{g}/\text{m}^3$  (regional background) to 102.5

314  $\mu\text{g}/\text{m}^3$  (traffic) in December 2018. The highest peak was observed during 1<sup>st</sup> to 6<sup>th</sup> January 2017,  
315  $\text{NO}_2$  concentration reached 146.4-178.9  $\mu\text{g}/\text{m}^3$ . The number of days exceeding Grade II  
316 standards for daily average  $\text{NO}_2$  (80  $\mu\text{g}/\text{m}^3$ ) during the period 2014-2018 was 152, in which the  
317 lowest exceeded days (18 days) was observed in 2018.

318 Substantially,  $\text{SO}_2$  exhibited U-shape patterns in all years; lower values in May to August, and  
319 higher values in January, November and December (Fig.2d) in all sites, with higher values in  
320 traffic sites. A smooth decreasing trends were observed at all sites, from maximum peaks of  
321 74.0-96.2  $\mu\text{g}/\text{m}^3$  in December 2014 to 13.2-18.8  $\mu\text{g}/\text{m}^3$  in December 2018.

322 **Fig. 2.** Daily average pollution concentrations (a:  $\text{PM}_{2.5}$ ; b: MDA8-h  $\text{O}_3$ ; c:  $\text{NO}_2$ ; d:  $\text{SO}_2$ ) at the  
323 four typical sites in Beijing from 2014 to 2018. (units are  $\mu\text{g}/\text{m}^3$ ) ( $\text{PM}_{2.5}$  extreme events  
324 shown in the box and green horizontal line indicating the Grade II standard).

325

### 326 3.3. Diurnal variation

327 Fig.3a shows the annual diurnal variation of  $\text{PM}_{2.5}$  concentrations across four different typical  
328 sites from 2014 to 2018 in Beijing. The diurnal variation of  $\text{PM}_{2.5}$  concentrations was by and  
329 large characterized by a “W” type double wave. The morning peak occurred around 08:00 to  
330 11:00, and an afternoon valley between 15:00 to 17:00. The peak in the night appeared after  
331 19:00 or midnight and then gradually decreased in the early hours of the morning. In 2014, the  
332 minimum and maximum values of hourly  $\text{PM}_{2.5}$  were 81.9  $\mu\text{g}/\text{m}^3$  and 99.7  $\mu\text{g}/\text{m}^3$  at 16:00 and  
333 23:00 in the urban site, whereas, in the traffic site, the observed values were 87.3  $\mu\text{g}/\text{m}^3$  and  
334 110.2  $\mu\text{g}/\text{m}^3$ , observed at 14:00 and 01:00. As the year progressed, overall  $\text{PM}_{2.5}$  concentration  
335 was decreased and more flat diurnal variation was observed. In 2018, the minimum and  
336 maximum values were 49.8  $\mu\text{g}/\text{m}^3$  (at 07:00 and 16:00) and 56.8  $\mu\text{g}/\text{m}^3$  (at 23:00) in the urban  
337 site, and 50.3  $\mu\text{g}/\text{m}^3$  (at 16:00) and 59.9  $\mu\text{g}/\text{m}^3$  (at 23:00) in the traffic site.

338 The morning and evening peaks were attributable in part to the enhanced human activities during  
339 the rush hours, and the afternoon valley was mainly attributable to a higher atmospheric mixing  
340 layer, which enhanced air pollution diffusion (Martini et al., 2015). It can be observed from Fig.  
341 3b that the diurnal variability showed a strong seasonal variability between the summer and  
342 winter for the year 2014 and 2018. In the summer, the average concentration of pollutants was  
343 higher during the daytime and reduced at night, which contrasted with the situation in the winter.

344 In the summer, a peak was observed between 08:00 to 12:00, though a deep valley occurred  
345 between 07:00 to 15:00 in the winter. This difference was mainly due to the diverse sources of  
346 pollution and their distinct formation mechanisms in different seasons. The air quality in the  
347 summer was more affected by human activities, and during the night, with the decrease in human  
348 activities, the concentration of pollutants dropped to a lower level. The main factors influencing  
349 outdoor air quality during the winter were the transport and diffusion effects of external pollution  
350 sources and the burning of indoor biomass for heating (Liu et al., 2019; Shi et al., 2019). The  
351 impact of human activities was relatively small and superseded by meteorological conditions in  
352 the winter. Therefore, during the night time, the lower atmospheric and stagnant wind conditions  
353 aggravated the accumulation of pollutants and increased the PM<sub>2.5</sub> concentrations (Lang et al.,  
354 2017; Zhang & Cao, 2015). Similar seasonal diurnal variations of PM<sub>2.5</sub> was noted for the year  
355 2014 (Martini et al., 2015).

356 Fig.3c shows the hourly average diurnal variation of O<sub>3</sub> concentrations during 2014-2018,  
357 which was opposite that of the other air pollutants. Ozone concentrations reach a minimum value  
358 before sunrise. Around 09:00, along with increases in solar radiation and temperature,  
359 photochemical reactions become more active. Ozone concentrations begin to increase, and they  
360 peak around 18:00–20:00. The highest peak concentration was observed in Summer. In winter,  
361 the peak concentration of ozone appeared earlier, at around 17:00. From 06:00 to 09:00,  
362 coinciding with the morning peak traffic, the concentration of NO increases rapidly. Solar  
363 radiation is still weak during this period; thus, the concentration of ozone decreases due to  
364 titration with NO ( $O_3 + NO \rightarrow NO_2 + O_2$ ). Between 21:00 and 23:00, the concentration of ozone  
365 decreases rapidly due to the decrease in solar radiation and titration with NO during evening  
366 peak traffic. In the urban site in 2018, the minimum and maximum values of hourly average  
367 O<sub>3</sub> concentration were 29.4  $\mu\text{g}/\text{m}^3$  and 95.2  $\mu\text{g}/\text{m}^3$ , observed at 8.00 and 19:00, respectively.  
368 In the traffic site in 2018, the minimum and maximum values were 20.9  $\mu\text{g}/\text{m}^3$  and  
369 75.5  $\mu\text{g}/\text{m}^3$ , observed at 9:00 and 19:00, respectively. The diurnal variations of O<sub>3</sub> in the  
370 summer and winter has undergone a huge shape metamorphosis and the hourly concentration  
371 on summer days was significantly higher than those in the winter (Fig.3d). In 2018, the minimum  
372 and maximum O<sub>3</sub> in the summer in the urban site were 45.0  $\mu\text{g}/\text{m}^3$  and 153.8  $\mu\text{g}/\text{m}^3$  and, in the  
373 winter, 27.3  $\mu\text{g}/\text{m}^3$  and 50.9  $\mu\text{g}/\text{m}^3$ , respectively. Overall, the amplitude of the variation in  
374 ozone concentration was increased in 5-years.

375 The diurnal variation in NO<sub>2</sub> concentrations can be represented by two peaks (Fig.3e) except in  
376 the regional background sites. The first peak was at 07:00–8:00 and the second one appeared in  
377 20:00–22:00; the second peak was significantly higher than the first one. In the regional  
378 background site, there was a sharp decline at 14:00–15:00 and the peak was reached at 21:00–  
379 22:00. The peak in the morning could be attributed to the rush hour traffic and a peak at night  
380 might be related to NO<sub>2</sub> accumulation caused by relatively unfavourable weather conditions and  
381 high NO emissions (Cheng et al., 2018). Heavy diesel vehicles are allowed to enter the city at  
382 night, resulting in a large amount of NO emissions at night (Sun et al., 2013). The shapes of the  
383 diurnal variations in each site are different and the overall NO<sub>2</sub> concentration decreased across  
384 all the sites from 2014 to 2018. The minimum and maximum values were quite different in the  
385 urban site (28.7 µg/m<sup>3</sup> and 53.1 µg/m<sup>3</sup> in 2018) and traffic site (50.4 µg/m<sup>3</sup> and 72.7 µg/m<sup>3</sup> in  
386 2018). The hourly concentrations of NO<sub>2</sub> during the winter days were significantly higher than  
387 those in the summer (Fig. 3f). This phenomenon might be attributable to the difference in  
388 meteorological conditions. The decrease of NO<sub>2</sub> concentrations in the afternoon was related to  
389 the increase of boundary layer height and an increase in wind speed which results in the dilution  
390 of pollutants. With the decrease in NO<sub>2</sub> and CO concentrations in the afternoon, O<sub>3</sub>  
391 concentrations increased and it was suggested that the maximum O<sub>3</sub> concentration in the  
392 afternoon was mainly due to photochemical reaction under intense solar radiation conditions,  
393 leading to the consumption of NO<sub>2</sub> and CO emissions (Shi et al., 2019). A unimodal shape is  
394 observed for the annual average diurnal variations of SO<sub>2</sub> (Fig.3g). In the urban area, the  
395 minimum hourly average values were observed at 10.8 µg/m<sup>3</sup> and 4.7 µg/m<sup>3</sup> in 2014 and 2018  
396 at early morning 06:00 and the maximum values where 17.9 µg/m<sup>3</sup> and 7.2 µg/m<sup>3</sup> in 2014 and  
397 2018 during 10:00-13:00. The diurnal variation in the winter was much higher than the summer  
398 and each is different in shape (Fig.3h).

399 **Fig. 3.** Diurnal variations of PM<sub>2.5</sub> (a and b), O<sub>3</sub> (c and d), NO<sub>2</sub> (e and f) and SO<sub>2</sub> (g and h) at  
400 four typical sites in Beijing. (a, c, e and g for yearly average, and b, d, f and h for the  
401 seasonal summer and winter variations in 2014 and 2018)

402

403 3.4. The trend of pollutants from 2014 to 2018

404 The Theil-Sen estimator was used to calculate the magnitude of the long-term trend after  
405 accounting for seasonal variations in the data. Fig.4 presents yearly average changes of air  
406 pollutant concentrations at the urban, the suburban, the regional background and the traffic sites  
407 of Beijing from 2014 to 2018. Specifically, average yearly changes were, for PM<sub>2.5</sub> (-7.7, -7.0, -  
408 6.3 and -8.4  $\mu\text{g}/\text{m}^3/\text{year}$  for the urban, the suburban, the regional background and the traffic  
409 sites, respectively), NO<sub>2</sub> (-2.7, -1.7, -1.4 and -3.9  $\mu\text{g}/\text{m}^3/\text{year}$ ), MDA8-h O<sub>3</sub> (1.7, 2.0, -0.9 and  
410 2.5  $\mu\text{g}/\text{m}^3/\text{year}$ ), SO<sub>2</sub> (-2.1, -1.4, -1.9 and -2.2  $\mu\text{g}/\text{m}^3/\text{year}$ ), PM<sub>10</sub> (-9.6, -7.9, -5.1 and -9.7  
411  $\mu\text{g}/\text{m}^3/\text{year}$ ) and CO (-0.07, -0.08, -0.06 and -0.11  $\text{mg}/\text{m}^3/\text{year}$ ). The error on the bar shows the  
412 minimum and maximum yearly change with 95% confidence interval (95% CI). Fig.4 shows that  
413 the air pollution action plan has significantly ameliorated the air quality of Beijing at all four-  
414 type of sites, especially for PM<sub>2.5</sub> and PM<sub>10</sub>, whereas, except for the regional background site,  
415 MDA8-h O<sub>3</sub> increased significantly in all sites. The Action Plan also led to a decrease in SO<sub>2</sub> and  
416 NO<sub>2</sub> but to a lesser extent than that of CO, PM<sub>2.5</sub> and PM<sub>10</sub>, indicating that SO<sub>2</sub> and NO<sub>2</sub> were  
417 significantly affected by other less well-controlled sources. The urban and the traffic sites  
418 showed a bigger decrease in PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub> and SO<sub>2</sub> concentrations in comparison to the  
419 other sites. After accounting for seasonal variations, MDA8-h showed a positive trend, although  
420 annual average MDA8-h O<sub>3</sub> concentration decreased during the study period, this was due to the  
421 increase of summer MDA8-h value.

422 Cheng et al., (2016) reported the increase of MDA8-h O<sub>3</sub> in the urban area by 2.2  $\mu\text{g}/\text{m}^3/\text{year}$ ,  
423 and a slight decrease at background station by 0.9  $\mu\text{g}/\text{m}^3/\text{year}$  from 2004 to 2015. Vu et al.,  
424 (2019) estimated the annual average levels of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub> and CO decreased by 7.4,  
425 7.6, 3.1, 2.5 and 94  $\mu\text{g}/\text{m}^3/\text{year}$ , respectively, whereas the level of O<sub>3</sub> increased by 1.0  
426  $\mu\text{g}/\text{m}^3/\text{year}$  during 2013 to 2017. The higher decreasing rate of PM<sub>2.5</sub> was observed in the urban  
427 areas in North China, by 10  $\mu\text{g}/\text{m}^3/\text{year}$  during 2013-2017 (Li et al., 2020). The ozone increases  
428 in Beijing were largely due to higher background ozone driven by urban heat island effects and  
429 increase in summertime biogenic emissions of VOCs and NO<sub>x</sub> (Lu et al., 2019; Cheng et al.,  
430 2019; Chen et al., 2019). However, Li et al., (2019) argued that the decrease of PM<sub>2.5</sub> contributes  
431 more than the decrease of NO<sub>x</sub> or VOCs emissions to ozone increasing trends in China, and this  
432 is mainly due to aerosol chemistry rather than photolysis.

433

434 **Fig. 4.** The trend of pollutants from 2014 to 2018 (mean values with error bars represent 95%  
435 confidence intervals; unit in  $\mu\text{g}/\text{m}^3/\text{year}$  for  $\text{PM}_{2.5}$ ,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{PM}_{10}$ , and in  
436  $\text{mg}/\text{m}^3/\text{year}$  for CO).

437  
438 The trend of ozone is only positive in Beijing and there are various reasons: (a) The biogenic  
439 VOCs emissions alone enhance surface DMA8 ozone by more than  $29.4 \mu\text{g}/\text{m}^3$  over central-  
440 eastern China in July-August, mainly driven by high temperatures (X. Lu et al., 2019). (b) The  
441 precursor material from long-distance transport has the largest contribution to ambient ozone in  
442 some provinces in China. The emission rate of precursors in Beijing has decreased, however, the  
443 VOCs from nearby areas still make the surface ozone concentration rise (Gao et al., 2019). (c)  
444 Reduced  $\text{PM}_{2.5}$  levels also plausibly cause an increase in  $\text{O}_3$  level due to impacts on ozone  
445 photochemistry and heterogeneous chemistry on aerosol surfaces. Primarily  $\text{PM}_{2.5}$  scavenges the  
446 hydroperoxy ( $\text{HO}_2$ ) and  $\text{NO}_x$  radicals that would otherwise produce  $\text{O}_3$  (Wang et al., 2019). (d)  
447 Also, meteorological conditions were dominant drivers of the ground ozone concentrations, as  
448 the surface temperature. Compared to the previous year's summer of 2017 had hotter and drier  
449 weather conditions, which promoted ozone production and led to higher ozone levels in 2017 in  
450 North China Plain (Ding et al., 2019a).

### 451 3.5. Health-risk-based AQI (HAQI)

452 The exposure-response coefficient characterizes the relationship of pollutants exposure and  
453 corresponding additional mortality health risk due to the six pollutants when their concentrations  
454 are higher than CAAQS Grade II. Although some adverse health impacts may still exist below  
455 the CAAQS Grade II standard, the impact is much smaller (WHO, 2005). Fig.5a shows the  
456 percentage of total average ER values for all pollutants in four regions in Beijing from 2014 to  
457 2018. For the six pollutants,  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  were the two major pollutants that contributed the  
458 highest percentage of total ER,  $\text{PM}_{2.5}$  contributed 71.5% (in the suburban area) to 81.5% (in the  
459 traffic area) and  $\text{PM}_{10}$  contributed 9.9% (background site) to 14.1% (traffic site). The average  
460 contributions by  $\text{O}_3$ ,  $\text{NO}_2$  and CO were 5.4%, 4.2% and 2.6% of total ER, respectively, although  
461 their health effects were also significant. The results reveal that  $\text{PM}_{2.5}$  is the top threat to public  
462 health regarding non-accidental mortality among the pollutants.

463 Each day (from the year 2014 to 2018) is classified into five risk categories based on AQI. The  
464 classification of the category on a certain day could change if based on HAQI (Fig.5b). There is  
465 no misclassification when AQI is less than 100 (AQI-based health days), as HAQI is equal to  
466 AQI. The number of days having good-to-moderate (AQI: 0-100) and hazardous (AQI: > 300)  
467 health risk in regional background site was 974 and 47, and in the traffic site, 894 and 67 days. In  
468 this context, the air quality was healthier in the regional background site than in the traffic site.  
469 For AQI-based light pollution days ( $100 < \text{AQI} < 150$ ), 82, 9.4 and 8.5% of days and 77.5, 13.4  
470 and 9.1% of days would be 'unhealthy for sensitive groups', 'unhealthy' and 'very unhealthy'  
471 pollution based on HAQI values in the urban and the traffic site, respectively. In moderate  
472 pollution days (AQI: 151-200), 39.3, 25.6 and 35.2% of days and 31.7, 10.8 and 57.5% of days  
473 would be 'unhealthy', 'very unhealthy' and 'hazardous' pollution days based on HAQI values in  
474 the urban and the traffic sites, respectively. And in serious days (AQI: 201-300), above 88% of  
475 the days would be 'hazardous' based on HAQI in all sites. In this perspective, the results detect  
476 that the health risks are underestimated based on simple AQI system in many days. The public is  
477 exposed to multiple pollutants on most pollution days, while AQI captures only the single  
478 pollutant with the 'greatest health risk', neglecting the mortality risk of other pollutants. From  
479 2014 to 2018, the average HAQI value decreased around 6% per year.

480 **Fig. 5.** (a) Percentage of the excessive risk of pollutants from 2014 to 2018. (b) Comparison of  
481 the HAQI-classified health risk categories with the AQI-classified categories (with the  
482 average number of days in different ranges; the sum of all year data was included in the  
483 analysis).

484

### 485 3.6. Air pollutants-attributed total premature mortality

486 The  $\text{PM}_{2.5}$  and  $\text{O}_3$  exposure levels vary significantly throughout the year due to both naturogenic  
487 reasons such as meteorology and to anthropogenic emissions like transport. Therefore, based on  
488 the daily data the Monte Carlo simulation of  $\text{PM}_{2.5}$  and  $\text{O}_3$  were used to calculate district wise  
489 pollution concentration in Beijing. Fig.6 (a and b) represents the district wise 5-year annual  
490 average  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentration in Beijing and shows a substantial spatial heterogeneity,  
491 where  $\text{PM}_{2.5}$  pollution was high in the central and southern parts and  $\text{O}_3$  pollution was high in the  
492 northern parts of Beijing. In 2014,  $\text{PM}_{2.5}$  and  $\text{O}_3$  attributed total deaths were 29.27 (95% CI:  
493 18.79–35.4) thousands and 3.03 (95% CI: 1.54–5.9) thousands (Fig.6 c and e). After five years,



494 the estimated number of deaths by PM<sub>2.5</sub> and O<sub>3</sub> decreased by 5.6% and 18.5% (Fig.6 d and f). In  
 495 PM<sub>2.5</sub>-attributed mortality, ischemic heart disease (IHD), cerebrovascular disease (Stroke),  
 496 chronic obstructive pulmonary disease (COPD), lung cancer (LC) among adults (age $\geq$  25 years),  
 497 and acute lower respiratory infection (ALRI) in children (age $\leq$ 5 years) contributed to 37.4, 49.0,  
 498 7.2, 6.3 and 0.2%, respectively. It should be noted in Table 2, that premature death is directly  
 499 proportional to the population size. In Fig.6 (a) shows that PM<sub>2.5</sub> pollution was higher in Daxing,  
 500 Fangshan and Tongzhou districts while the total PM<sub>2.5</sub>-attributed deaths in these districts were  
 501 smaller than those in Haidian, Fengtai and Chaoyang districts due to differences in population  
 502 distribution across these districts (Fig.S12 and Table 2). The highest premature deaths (due to  
 503 PM<sub>2.5</sub> and O<sub>3</sub>) were observed in Haidian (5364 in 2014 and 4653 in 2018) and Chaoyang (5751  
 504 and 5049) and the lowest deaths were observed in Yanqing (484 and 462) and Mentougou (477  
 505 and 461). More details have been reported in Table S4 and Table S5 (Supplementary Material).  
 506 Even though the pollutant concentrations were substantially reduced in these five years, the total  
 507 attributed deaths were not reduced, as the total mortality rate, population and aged population  
 508 ( $\geq$ 25 years) were increased by 5.28, 0.93 and 5.01%, respectively. The cities of Tianjin and  
 509 Hebei in the Beijing-Tianjin-Hebei (BTH) region, showed a 2.35% decrease and a 15.7%  
 510 increase in premature deaths from 2010 to 2015 (Zhu et al., 2019). Zhang et al. (2019) reported a  
 511 17.2% decrease in total non-accidental mortality in China during 2013-2017, whereas Ding et al.,  
 512 (2019b) reported 20.7% decrease of cause-specific mortality during the same period, although  
 513 Wu et al. (2019) observed almost no decrease in the number of PM<sub>2.5</sub>-related premature deaths in  
 514 the Pearl River Delta region from 2006 to 2015.

515 **Fig. 6.** District-specific (a) annual average PM<sub>2.5</sub> concentration ( $\mu\text{g}/\text{m}^3$ ), (b) annual average O<sub>3</sub>  
 516 concentration ( $\mu\text{g}/\text{m}^3$ ) in Beijing from 2014 to 2018, PM<sub>2.5</sub>-attributed premature  
 517 mortality for the year (c) 2014 and (d) 2018, and O<sub>3</sub>-attributed premature mortality for  
 518 the year (c) 2014 and (d) 2018.

519 Note: DAX, MEN, HAI, MIY, YAN, HUA, PIN, SHI, FEN, DON, SHU, FAN, CHA, XIC,  
 520 CHA and TON refer to the districts of Daxing, Mentougou, Haidian, Miyun, Yanqing,  
 521 Huairou, Pinggu, Shijingshan, Fengtai, Dongcheng, Shunyi, Fangshan, Chaoyang,  
 522 Xicheng, Changping and Tongzhou

523

524 **Table 2.** Cause-specific premature deaths attributed to PM<sub>2.5</sub> and O<sub>3</sub> in 16 districts in Beijing

525

526 Very few district-specific health risk assessment works have been done in the past, Yin et al.  
527 (2017) estimated the maximum and minimum  $PM_{2.5}$ -attributed deaths at 1773 in Chaoyang and  
528 113 in Yanqing in 2012. At the city-level,  $PM_{2.5}$ -related reported deaths were 20.9 thousands  
529 (Song et al., 2017) and 25.5 thousands (Li et al., 2018) in 2015, 17.7 thousands (Anenberg et al.,  
530 2019) and 18.2 thousands in 2016 (Maji et al., 2018), 25.8 thousands in 2017 (Ding et al.,  
531 2019b). Ozone-related total reported deaths were 2.98 thousands in 2015 (Xiang et al., 2019) and  
532 2.1 thousands in 2016 (Maji et al., 2019). This difference in estimated pollution attributed deaths  
533 is mainly due to different methodologies for estimating death and ground-level ozone, and  
534 exposure-response coefficient. Most of the past studies used the average mortality rate and age  
535 group population values for China as a whole, which were very different from the district-based  
536 values. For example, in 2017, the average mortality rate in Beijing was 6.2%, while the mortality  
537 rates in Miyun and Changping districts were 9.5% and 3.4%.

#### 538 **4. Policy implications**

539 Air quality improvement is important for the future prosperity and sustainability of China's most  
540 powerful city, Beijing. This improvement in air quality did not happen by chance. It was the  
541 result of an enormous investment of time, resources and political will. In 1998 Beijing declared  
542 war on air pollution. The challenge was to find ways to improve air quality in one of the largest  
543 and fastest-growing cities in the developing world. Air pollution issue in Beijing is partially due  
544 to the energy source, especially the higher use of solid and liquid fuel problems in the current  
545 stage in China. On the one hand, actions of energy conservation and low-carbon energy  
546 transitions to reduce  $CO_2$  emissions also reduce other co-emitted air pollutants, bringing co-  
547 benefits for air quality (Zhang et al., 2016). In this study, we emphasize the effects of past air  
548 pollution control and action plan to improve the current environmental situation in Beijing, even  
549 though  $PM_{2.5}$  concentration did not achieve the national standard and  $O_3$  concentration continued  
550 to increase due to growing VOC emissions in Beijing. The Chinese government should  
551 strengthen the co-governance policy design in the future to achieve maximal effects with  
552 minimal economic loss. In summary, in this study, we provide a comprehensive assessment of  
553 the air quality in urban, suburban and traffic areas in Beijing, and districts level health burden,  
554 which is valuable for policymakers. Now the Beijing authority should develop new district-based  
555 policy, on (1) motor vehicle pollution control by using green transportation, (2) coal-fired

556 pollution control by using green energy, (3) key polluted industries control by strengthening the  
557 air quality standard, (4) fugitive dust pollution control by using urban green infrastructure, and  
558 (5) new technology application in environmental protection, for near-term air quality targets  
559 achievement.

## 560 **5. Conclusion**

561 In this study, data for six official air pollutants are analyzed in 35 monitoring sites (divided into  
562 four types, respectively, the urban, the suburban, the regional background, and the traffic site) in  
563 Beijing based on the data from 2014 to 2018. None of the 35 sites has met the Grade II Chinese  
564 ambient air quality standards for PM<sub>2.5</sub>. The minimum annual average PM<sub>2.5</sub> was observed in the  
565 Badaling monitoring site (68.8 µg/m<sup>3</sup>) in 2014 and the Miyun Reservoir site (45.1 µg/m<sup>3</sup>) in  
566 2018, and the maximum was observed at 125.3 µg/m<sup>3</sup> in 2014 and 72.2 µg/m<sup>3</sup> in 2018 in the  
567 Liuli River site. The highest peak was observed in late December 2015 during the Southeast  
568 Asian haze. During the 626 days or almost half of the five-year studied period, PM<sub>2.5</sub>  
569 concentration had consistently exceeded the Grade II standard. O<sub>3</sub> also exceeded the Grade II  
570 standard for 291 days, which peaks at 311.6 µg/m<sup>3</sup> (in May) in 2014 and 290.6 µg/m<sup>3</sup> (in June)  
571 in 2018. Very high seasonal variations were observed for all six pollutants. The stagnant  
572 meteorological conditions and the use of fossil fuels for indoor heating accounted for the high  
573 PM<sub>2.5</sub> concentration in the winter. The decreasing trends were observed for PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub>,  
574 SO<sub>2</sub> and CO, whereas, O<sub>3</sub> increased during this time. Among the six pollutants, PM<sub>2.5</sub> was the  
575 major threat to public health and the health risks were underestimated based on the current  
576 simple AQI reporting system. From 2014 to 2018, the change of PM<sub>2.5</sub> and O<sub>3</sub> attributed  
577 mortality is low, mainly due to the increase of total mortality rate, the population as well as the  
578 aged population. The diurnal variation can be a good source of information for a person's  
579 exposure assessment during an outdoor visit. Future research can focus on the chemical  
580 composition and source apportionment of fine particles, urban meteorology during haze and  
581 photochemistry for Beijing for a longer time. Furthermore, to improve the quality of life, the  
582 authorities in Beijing may want to impose even more stringent and effective pollutant control  
583 measures, specifically targeting at the most polluted districts in Beijing, such as Chaoyang and  
584 Haidian, with priority given to controlling VOCs and industrial pollution in Beijing and the  
585 nearby urban regions.

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**Table 1.** Annual average pollution concentrations from 2014 to 2018 in four typical sites in Beijing (US, SUB, RBS and TS represent the urban site, the sub-urban site, the regional background site and the traffic site; concentration unit is in  $\mu\text{g}/\text{m}^3$ )

	Sites	2014	2015	2016	2017	2018
PM <sub>2.5</sub>	US	89.8±81.5	83.1±83.2	74.5±72.4	60.3±63.9	52.4±49.0
	SUB	88.5±77.4	80.5±76.1	70.6±66.4	59.0±59.3	50.6±46.6
	RBS	89.3±75.3	81.1±72.0	74.7±64.6	60.3±56.1	56.1±47.0
	TS	98.6±89.0	90.5±91.4	80.7±79.1	65.5±68.7	55.3±52.5
O <sub>3</sub>	US	112.8±65.8	99.6±65.7	97.3±63.4	97.7±60.1	103.3 ±59.6
	SUB	115.0±63.5	99.9±62.9	100.9±62.4	104.1± 61.8	105.4± 60.9
	RBS	117.0±62.6	102.0±62.2	101.4±59.2	97.8±53.4	103.8±54.5
	TS	83.6±50.3	76.8±51.3	78.6±52.4	79.8±52.9	82.8±50.5
NO <sub>2</sub>	US	55.9±30.8	52.2±30.9	51.3±30.0	48.1±28.0	43.6± 26.2
	SUB	45.1±24.2	43.5±25.6	42.8±25.4	42.0±23.6	37.5± 21.5
	RBS	37.9±19.8	37.1±21.6	37.1±20.8	34.9±18.1	32.5±17.6
	TS	82.1±32.7	75.7±33.3	69.8±31.4	67.2±30.2	64.4±28.7
SO <sub>2</sub>	US	13.7±12.9	14.0±14.6	10.5±10.6	8.1±8.8	5.9±4.0
	SUB	12.3 ±12.0	13.4±14.1	10.7±10.2	8.6±8.7	6.0±4.0
	RBS	14.9±11.0	13.4±13.1	10.6±9.2	8.9±8.0	6.3±4.6
	TS	16.0±14.0	17.5±17.0	13.9±12.0	6.3±5.2	7.6±4.9

**Table 2.** Cause-specific premature deaths attributed to PM<sub>2.5</sub> and O<sub>3</sub> in 16 districts in Beijing

Districts	2014						2018					
	PM <sub>2.5</sub>					O <sub>3</sub>	PM <sub>2.5</sub>					O <sub>3</sub>
	IHD	Stroke	COPD	LC	ALRI	Non-accidental	IHD	Stroke	COPD	LC	ALRI	Non-accidental
Daxing	787	1032	180	156	4	205	874	1145	154	134	4	173
Mentougou	150	198	30	26	1	72	154	200	25	22	1	59
Haidian	1832	2410	387	334	9	392	1687	2197	284	246	6	233
Miyun	232	305	45	39	1	172	232	299	37	32	1	151
Yanqing	153	201	29	26	1	71	162	209	26	22	1	42
Huairou	187	246	37	33	1	98	192	247	31	26	1	52
Pinggu	208	274	42	37	1	102	217	283	37	32	1	110
Shijingshan	324	426	68	60	2	104	299	390	52	44	1	100
Fengtai	1161	1524	258	220	6	213	1067	1392	185	159	4	156
Dongcheng	458	602	100	86	2	173	417	545	72	62	2	137
Shunyi	499	656	103	90	2	212	547	712	92	80	2	208
Fangshan	532	696	125	108	3	141	580	759	107	91	2	189
Chaoyang	1967	2580	422	366	10	406	1812	2361	306	265	7	299
Xicheng	651	855	137	120	3	283	591	770	100	86	2	194
Changping	934	1228	187	160	4	231	978	1259	157	135	3	178
Tongzhou	693	908	161	138	4	158	754	989	136	119	3	191

**Highlights:**

- Five years long criteria air pollutants data in Beijing were analyzed.
- The significant reduction of PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO (7.4, 8.1, 2.4, 1.9 and 80 µg/m<sup>3</sup>/ year).
- O<sub>3</sub> concentration increased (1.3 µg/m<sup>3</sup>/year) during the time frame.
- HAQI results suggest that in high pollution days, the sensitive population groups such as children and the elderly should take more stringent.
- From 2014 to 2018, PM<sub>2.5</sub> and O<sub>3</sub> attributed total deaths were decreased by 5.6% and 18.5%.

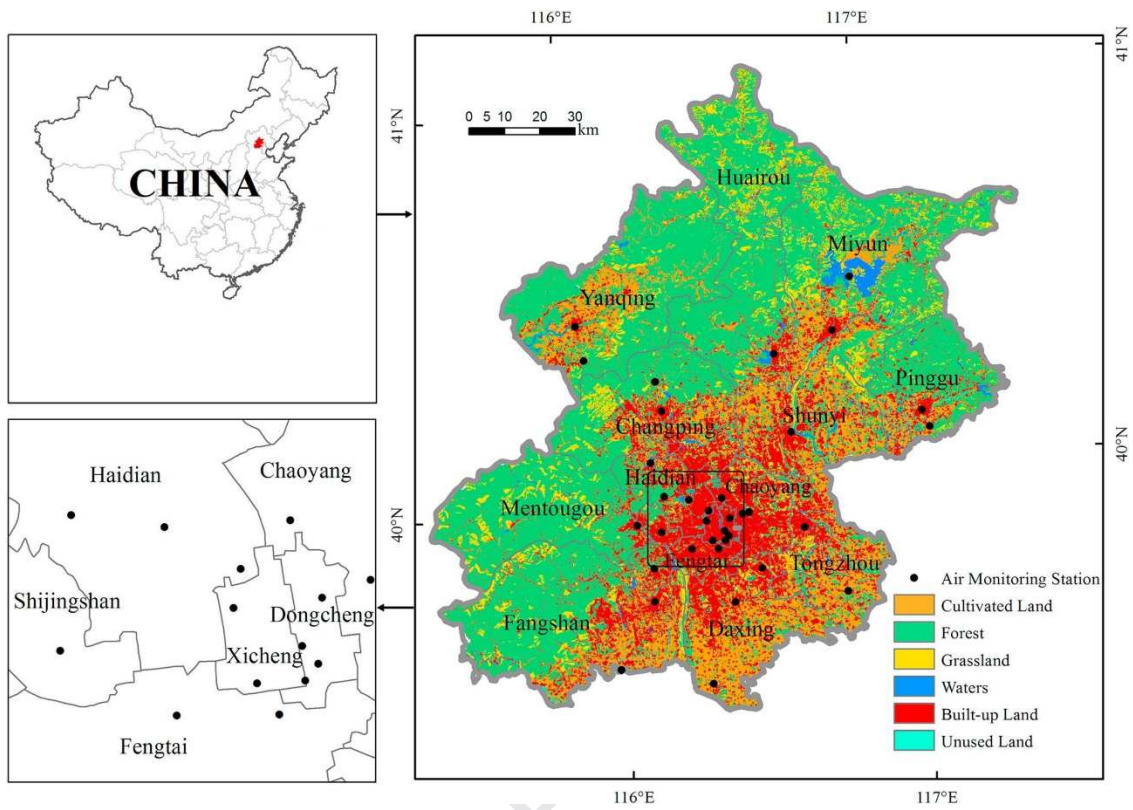
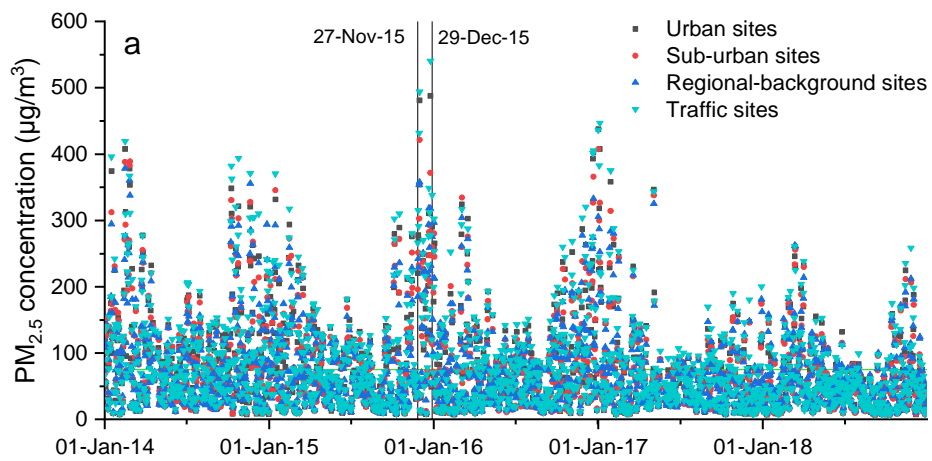
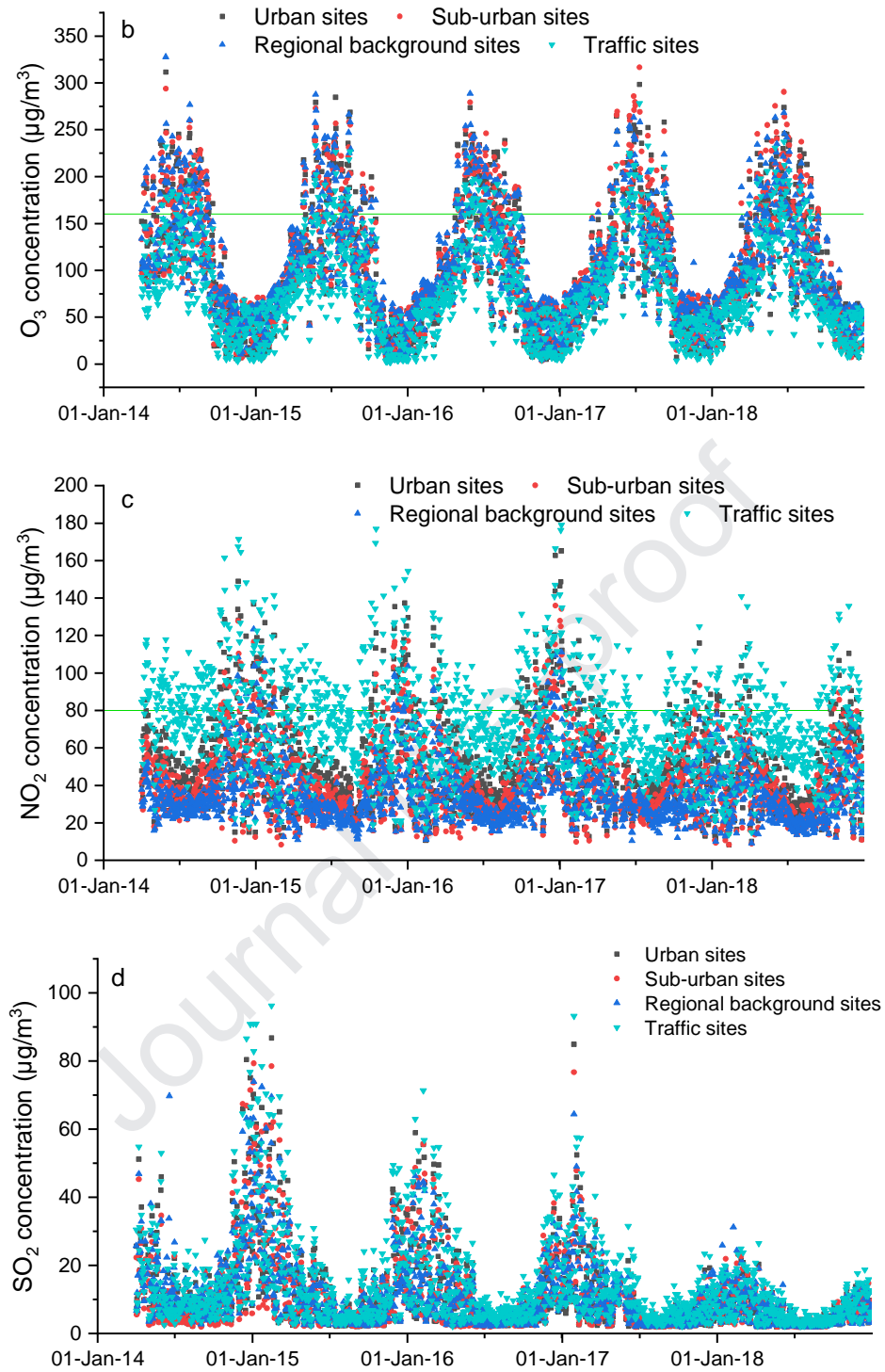
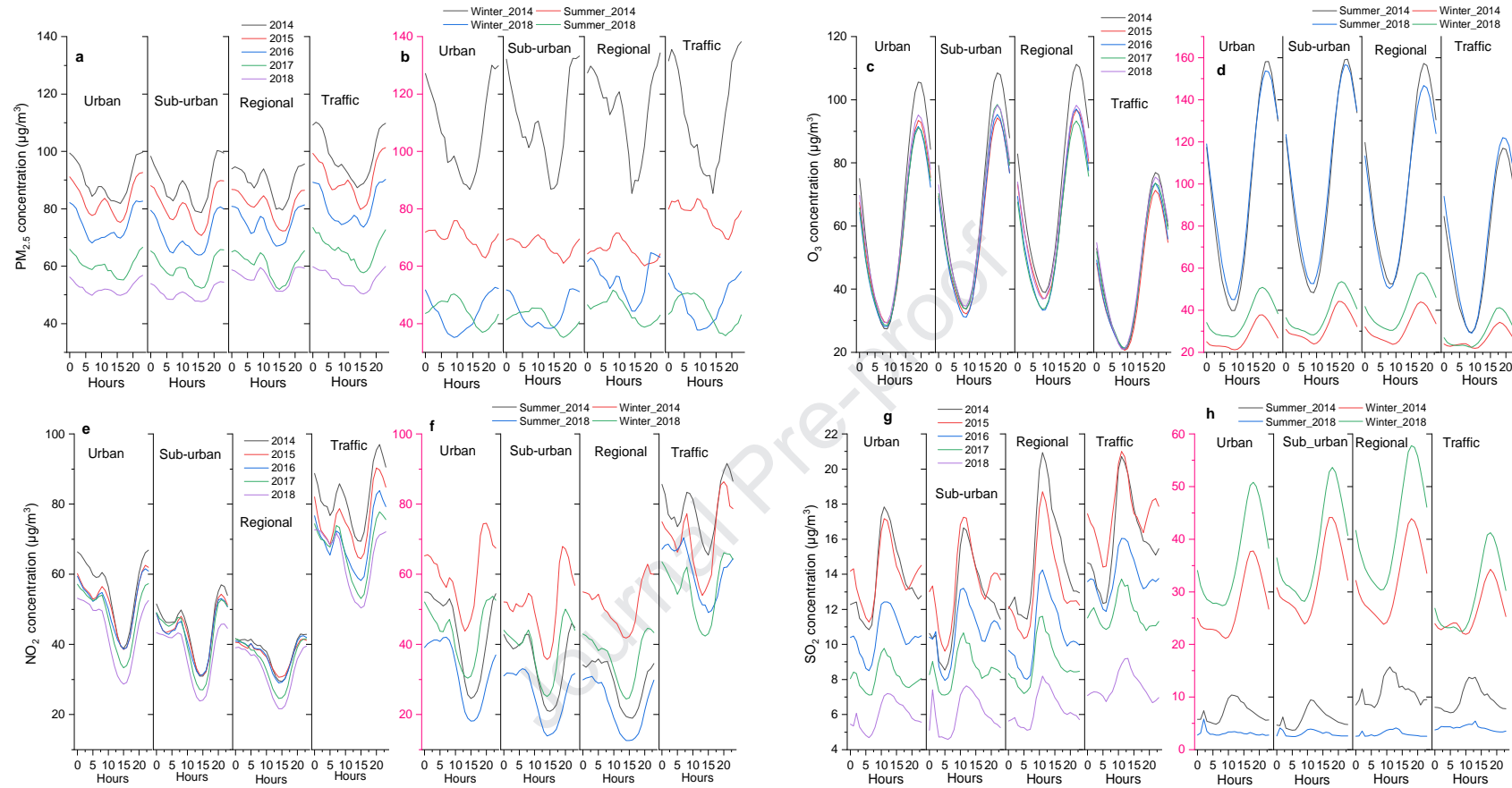


Fig. 1. Location of the 35 air quality monitoring sites in Beijing (Tian et al., 2019)

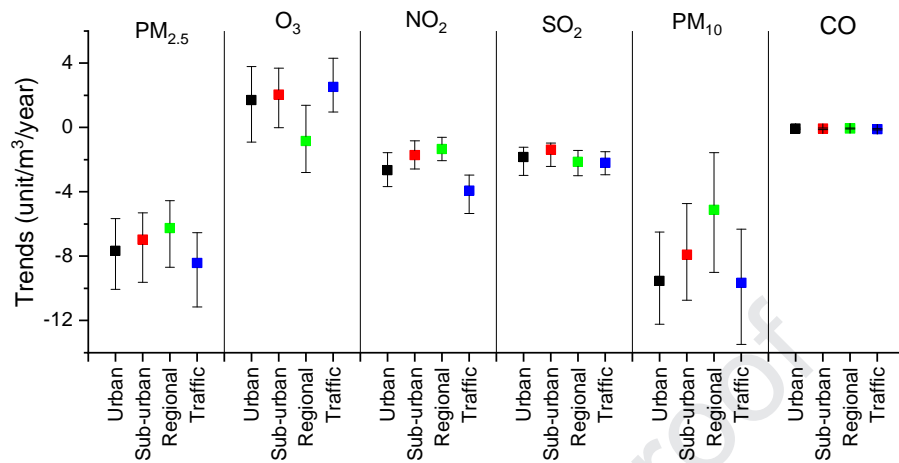




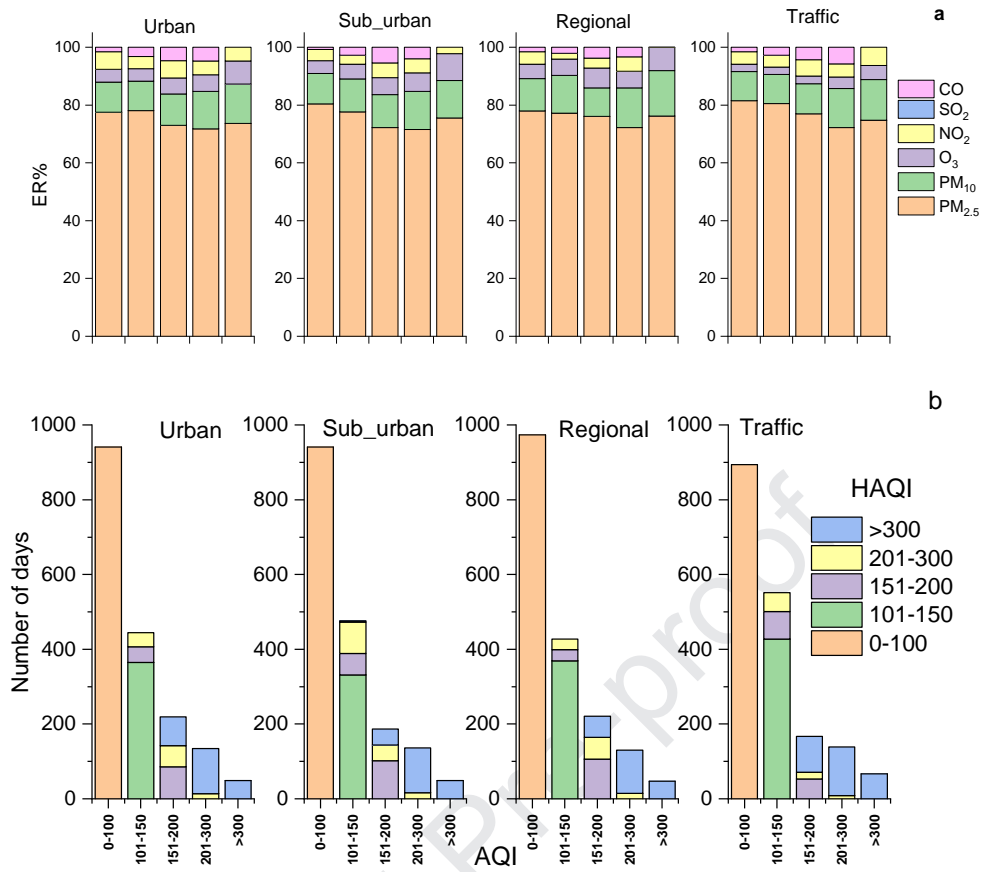
**Fig. 2.** Daily average pollution concentrations (a: PM<sub>2.5</sub>; b: MDA8-h O<sub>3</sub>; c: NO<sub>2</sub>; d: SO<sub>2</sub>) at the four typical sites in Beijing from 2014 to 2018. (units are μg/m<sup>3</sup>) (PM<sub>2.5</sub> extreme events shown in the box and green horizontal line indicating the Grade II standard).



**Fig. 3.** Diurnal variations of PM<sub>2.5</sub> (a and b), O<sub>3</sub> (c and d), NO<sub>2</sub> (e and f) and SO<sub>2</sub> (g and h) at four typical sites in Beijing. (a, c, e and g for yearly average, and b, d, f and h for the seasonal summer and winter variations in 2014 and 2018)

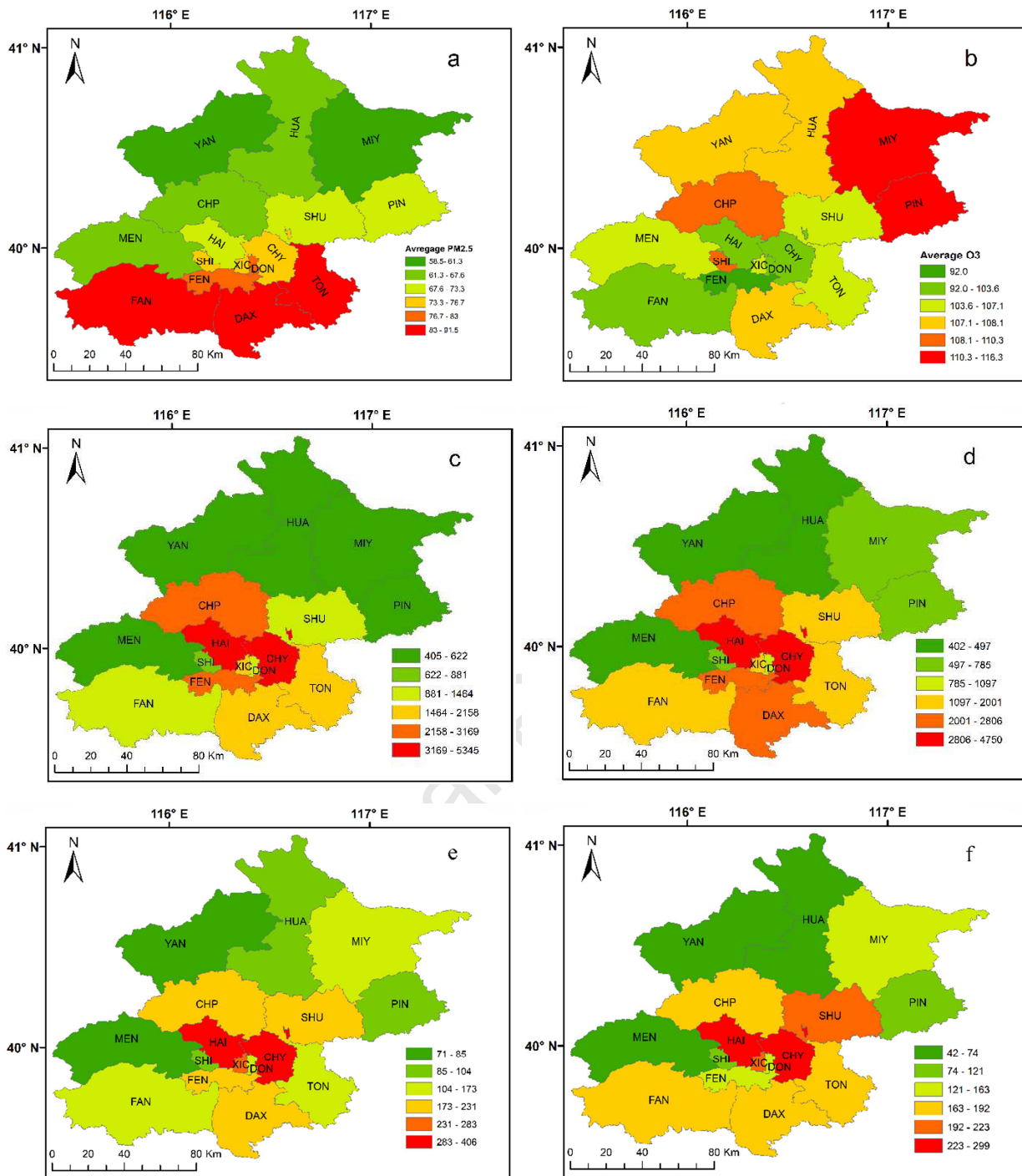


**Fig. 4.** The trends of pollutants from 2014 to 2018 (mean values with error bars represent 95% confidence intervals; the unit for the trends is  $\mu\text{g}/\text{m}^3/\text{year}$  for  $\text{PM}_{2.5}$ ,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{PM}_{10}$ , and  $\text{mg}/\text{m}^3/\text{year}$  for  $\text{CO}$ ).



**Fig. 5.** (a) Percentage of excess risk of the pollutants from 2014 to 2018. (b) Comparison of the HAQI-classified health risk categories with the AQI-classified categories (with the average number of days in different ranges; the sum of all year data was included in the analysis).





**Fig. 6.** District-specific (a) annual average PM<sub>2.5</sub> concentration (μg/m<sup>3</sup>), (b) annual average O<sub>3</sub> concentration (μg/m<sup>3</sup>) in Beijing from 2014 to 2018, PM<sub>2.5</sub>-attributed premature mortality for the year (c) 2014 and (d) 2018, and O<sub>3</sub>-attributed premature mortality for the year (c) 2014 and (d) 2018.

Note: DAX, MEN, HAI, MIY, YAN, HUA, PIN, SHI, FEN, DON, SHU, FAN, CHA, XIC, CHA and TON refer to the districts of Daxing, Mentougou, Haidian, Miyun, Yanqing, Huairou, Pinggu, Shijingshan, Fengtai, Dongcheng, Shunyi, Fangshan, Chaoyang, Xicheng, Changping and Tongzhou

**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.

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CRediT authorship contribution statement:

**K. J. Maji:** Writing - original draft, Conceptualization, Investigation, Formal analysis, Writing - review & editing. **V. O.K. Li:** Writing - original draft, Conceptualization, Supervision, Funding acquisition, Project administration. **J. C.K. Lam:** Writing - original draft, Conceptualization, Supervision, Funding acquisition, Project administration.