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1 **Long-term Characterization of Roadside Air Pollutants in Urban Beijing and**  
2 **Associated Public Health Implications**

3 Xuefang Wu<sup>1</sup>, Tuan V. Vu<sup>3</sup>, Roy M. Harrison<sup>4,5</sup>, Jing Yan<sup>1</sup>, Xiaohan Hu<sup>6</sup>, Yangyang Cui<sup>1</sup>, Aijun  
4 Shi<sup>7</sup>, Xinyu Liu<sup>1</sup>, Yan Shen<sup>1</sup>, Gen Zhang<sup>2\*</sup>, Yifeng Xue<sup>1\*</sup>

5 <sup>1</sup>National Engineering Research Center of Urban Environmental Pollution Control, Beijing  
6 Municipal Research Institute of Environmental Protection, Beijing 100037, China

7 <sup>2</sup>State Key Laboratory of Severe Weather and Key Laboratory for Atmospheric Chemistry of the  
8 China Meteorological Administration (CMA), Institute of Atmospheric Composition, Chinese  
9 Academy of Meteorological Sciences (CAMS), Beijing, 100081, China

10 <sup>3</sup>School of Public Health, Imperial College London, London, United Kingdom

11 <sup>4</sup>Division of Environmental Health and Risk Management, School of Geography, Earth and  
12 Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, United  
13 Kingdom

14 <sup>5</sup>Department of Environmental Sciences/Centre of Excellence in Environmental Studies, King  
15 Abdulaziz University, PO Box 80203, Jeddah, 21589, Saudi Arabia

16 <sup>6</sup>Beijing Pollution Source Management Affairs Center, Beijing 100089, China

17 <sup>7</sup>Beijing Vehicle Emission Management Affair Centre, Beijing 102612, China

18 \* Corresponding authors. Email: zhanggen@cma.gov.cn (Gen Zhang), xueyifeng@cee.cn  
19 (Yifeng Xue)

20 **Abstract:**

21 Road traffic constitutes a major source of air pollutants in urban Beijing, which are responsible  
22 for substantial premature mortality. A series of policies and regulations has led to appreciable  
23 traffic emission reductions in recent decades. To shed light on long-term (2014-2020) roadside  
24 air pollution and assess the efficacy of traffic control measures and their effects on public health,  
25 this study quantitatively evaluated changes in the concentrations of six key air pollutants (PM<sub>2.5</sub>,  
26 PM<sub>10</sub>, NO<sub>2</sub>, SO<sub>2</sub>, CO and O<sub>3</sub>) measured at 5 roadside and 12 urban background monitoring  
27 stations in Beijing. We found that the annual mean concentrations of these air pollutants were  
28 remarkably reduced by 47% to 71% from 2014 to 2020, while the concurrent ozone  
29 concentration increased by 17.4%. In addition, we observed reductions in the roadside  
30 increments in PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO of 54.8%, 29.8%, 20.6%, and 59.1%, respectively,  
31 indicating the high effectiveness of new vehicle standard (China V and VI) implementation in  
32 Beijing. The premature deaths due to traffic emissions were estimated to be 8379 and 1908 cases  
33 in 2014 and 2020, respectively. The impact of NO<sub>2</sub> from road traffic relative to PM<sub>2.5</sub> on  
34 premature mortality was comparable to that of traffic-related PM<sub>2.5</sub> emissions. The public health  
35 effect of SO<sub>2</sub> originating from traffic was markedly lower than that of PM<sub>2.5</sub>. The results  
36 indicated that a reduction in traffic-related NO<sub>2</sub> could likely yield the greatest benefits for public  
37 health.

38 **Keywords:** traffic emissions; roadside increment; traffic control policy; PM<sub>2.5</sub>; NO<sub>2</sub>; health  
39 effects

## 40 **1. Introduction**

41 The emissions originating from road traffic are widely acknowledged as a major source of  
42 air pollutants in urban areas, and human exposure to air pollutants stemming from road traffic  
43 emissions has been demonstrated to generate harmful effects on health (Beelen et al., 2008,  
44 Rissler et al., 2012, Pant and Harrison, 2013). Traffic emissions make up a significant fraction of  
45 fine particulates (PM<sub>2.5</sub>) in Beijing, accounting for 5-12% of primary PM<sub>2.5</sub> (Srivastava et al.,  
46 2021, Xu et al., 2021), directly emitted by the tailpipes from 2016 to 2017. Road traffic also  
47 contributes to secondary particles via gas-to-particle transformation (Pant and Harrison, 2013,  
48 Harrison et al., 2021b). In addition, non-exhaust emissions play a significant role in coarse and  
49 fine particles, arising from tyre wear, brake wear, and re-suspension of dust, contributing 4-13%  
50 to PM<sub>2.5</sub> in Beijing (Wu et al., 2014, Harrison et al., 2021a, Srivastava et al., 2021). Kam et al.  
51 (2012) reported that road vehicular emissions heavily affect small particles, including both  
52 primary exhaust particles and secondary species, while the coarse fraction was mostly affected  
53 by re-suspension of dust. Additionally, motor vehicles release a very large amount of gaseous  
54 pollutants into the atmosphere in Beijing (Cai et al., 2017). According to the Multi-resolution  
55 Emission Inventory for China (MEIC, 2018), the transportation sector emitted approximately 6  
56 kt a<sup>-1</sup>, 97 kt a<sup>-1</sup> and 566 kt a<sup>-1</sup> of SO<sub>2</sub>, NO<sub>x</sub> and CO, respectively, from 2013 to 2017, accounting  
57 for 13%, 40%, 37% of their total primary emissions, respectively. Moreover, a large amount of  
58 volatile organic compounds (VOCs) released from vehicle exhaust emissions could contribute to  
59 the formation of secondary organic aerosols and ozone, thus enhancing severe haze events in  
60 winter (Huang et al., 2014, Li et al., 2017, Harrison et al., 2021a).

61 To better understand the impact of air pollutants originating from vehicles upon the  
62 atmospheric environment and associated health effects, it is important to evaluate the  
63 contribution of road traffic emissions and to compare the air quality at different observation sites.  
64 Numerous studies have reported the adoption of paired-site data to estimate the contribution  
65 from traffic emissions to air pollutants, for example, Harrison and Beddows, (2017) and Wang et  
66 al., (2010) estimated the increment due to traffic emissions by subtracting the concentration of  
67 air pollutants at urban background monitoring sites from that at proximate roadside sites. For  
68 instance, Harrison et al. (2021b) reported that the PM<sub>2.5</sub> roadside traffic increment reached  
69 approximately 5 µg m<sup>-3</sup> in Beijing during 2016 to 2018, which is similar to that in Paris and  
70 Hong Kong. Chen et al. (2018) collected hourly air pollutant data at four traffic stations in

71 Beijing during 2014 to 2017 and concluded that roadside environments with heavy traffic are  
72 highly polluted and should be improved. The characteristics of short-term roadside air pollutants  
73 were reported by Zhang et al. (2019b) but just focused on one selected road in fall and winter.  
74 Despite reports of the characteristics of air pollutants at traffic sites in Beijing, few studies have  
75 explored their adverse effects on human health in this city.

76 Traffic-related air pollutant exposure exerts significant impact upon public health  
77 worldwide among urban residents (Forehead and Huynh, 2018, Lelieveld et al., 2015). For  
78 example, the traffic intensity on the nearest road to a given residential area exhibited a clear  
79 association with mortality in a long-term study of a Dutch cohort (Beelen et al., 2008). The air  
80 pollutants emitted from road vehicles adversely affected pregnancy outcomes, including a lower  
81 birthweight and cardiovascular defects (Chen et al., 2008, Jacobs et al., 2017, Jiang et al., 2017).  
82 Furthermore, due to their small median diameters, road traffic-generated aerosols are able to  
83 penetrate deeply into the human respiratory system, thereby causing harmful health outcomes  
84 such as asthma attacks, chronic bronchitis, and lung cancers, hence increasing morbidity and  
85 mortality (Künzli et al., 2000, Vu et al., 2018). Yi et al., (2017) estimated the association  
86 between traffic-related air pollutants and allergic diseases in children in Seoul (South Korea) and  
87 found that a higher risk for children to develop atopic eczema. As a traffic-related air pollutant,  
88 NO<sub>2</sub> has received much attention due to its long-term effects upon all-cause mortality (Huangfu  
89 and Atkinson, 2020, Huang et al., 2021, Brimblecombe et al., 2021). In Beijing, the number of  
90 annual premature deaths attributed to traffic-related PM<sub>2.5</sub> exposure was estimated to be 4435  
91 based upon weekday concentrations and 3462 based upon weekend day pollution levels (Tong et  
92 al., 2020).

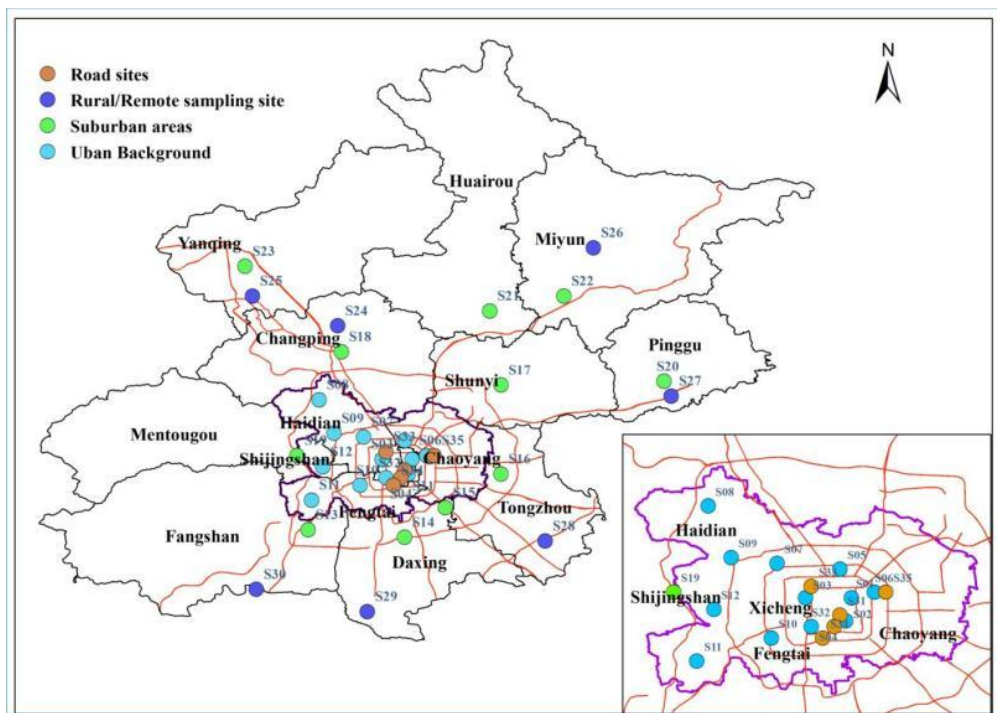
93 Similar to other metropolitan areas, Beijing has implemented a series of policies and  
94 regulations encompassing traffic emission control to reduce roadside air pollution over the past  
95 decade, such as the Regulations of the Beijing Municipality on Atmospheric Pollution  
96 Prevention and Control, and Regulations of the Beijing Municipality on Atmospheric Pollution  
97 Prevention and Control from Motor Vehicles and Non-road Mobile machinery (GBM, 2018;  
98 BTMB, 2020). These policies mainly focus on the implementation of new vehicle emission  
99 standards and fuel standards, elimination of aging vehicles, promotion of clean-energy vehicles,  
100 and adjustment of the fleet structure and transport system by increasing public transport  
101 infrastructure.

102 The aim of this study was to give a comprehensive insight into roadside air pollution from  
103 2014 to 2020 and to assess the effects of different vehicle types due to stricter traffic policies.  
104 Additionally, the adverse effects of air pollution exposure on human health were evaluated. This  
105 work firstly presents the long-term trends of air pollutants at five roadside monitoring stations to  
106 recognize the effects of sampling environments with various vehicle volumes and types.  
107 Secondly, roadside increments of air pollutants were estimated based on measured data retrieved  
108 from roadside and urban background monitoring stations by a paired-site study. Finally, the  
109 public health impacts of traffic-related PM<sub>2.5</sub>, NO<sub>2</sub> and SO<sub>2</sub> emissions were estimated and are  
110 discussed.

## 111 **2. Materials and methods**

### 112 **2.1. Data selection and analysis**

113 Since 2013, the concentrations of six major air pollutants (PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub>, SO<sub>2</sub>, CO and  
114 O<sub>3</sub>) in Beijing have been observed at 35 air quality monitoring stations with automated  
115 monitoring systems and are presented on the website of the Beijing Municipal Ecological and  
116 Environmental Monitoring Center (<http://www.bjmemc.com.cn/>). These monitoring stations  
117 include 12 urban background sites, 11 suburban sites, 7 rural sites and 5 traffic sites (as shown in  
118 Figure 1). In this study, the datasets of the traffic and urban background sites during 2014 to  
119 2020 were downloaded from <http://beijingair.sinaapp.com>. The hourly concentrations of PM<sub>2.5</sub>  
120 and PM<sub>10</sub> covered the period from January 1 2014 to December 31 2020 and the values of the  
121 other components covered April 1 2014 to December 31 2020. The concentration levels  
122 exceeding three times the median value over all similar characterized sites were suspected as  
123 outlier data points. Those data which lasted for less than three hours and exceeded the previous  
124 concentrations by more than tenfold were deleted. These were few, and the effect on mean  
125 concentrations was negligible.



**Figure 1.** Map of the 35 air quality monitoring stations in Beijing. The orange lines represent the main highways in Beijing.

Based on the characteristics of the monitoring sites with various vehicle volumes and types, the five traffic sites were divided into three groups (Harrison et al., 2021b). The first group contains Qianmen station, Yongdingmen Inner Street station and Xizhimen North Street station, which represent inner city streets with a majority of light-duty gasoline vehicles. Secondly, the South 3<sup>rd</sup> Ring Road station represents a typical arterial highway with a high loading of diesel trucks. There are normally several bus stations near the monitoring station and the road width is 60 m with a high vehicle volume. Thirdly, the East 4<sup>th</sup> Ring Road station locates near a highway with a high loading of diesel vehicles due to several nearby large warehouses. This station is located along the East 4<sup>th</sup> Ring Road with a surrounding environment comprising parks and business areas, and there is a high vehicle volume with a 67 m road width.

Roadside increments were calculated by subtracting the concentrations of air pollutants at urban background monitoring stations from those observed at the roadside monitoring sites. This difference represents the influence of traffic in an urban area above the air quality from the urban background. It was calculated as follows.

$$\Delta C_i = C_{i, road} - C_{i, urban} \quad (1)$$

where  $\Delta C_i$  is the road increment of type  $i$  air pollutant;  $C_{i, road}$  is the pollutant  $i$  concentration at

145 the roadside monitoring site; and  $C_{i, urban}$  is the pollutant  $i$  concentration at the urban background  
146 site.

147 For an individual site,  $\Delta C_i$  was calculated by subtracting the air pollutant level at nearby  
148 urban background site from the air pollutant concentration at the traffic site. For the whole of  
149 Beijing, the  $\Delta C_i$  was averaged by the roadside increments of air pollutant  $i$  at five traffic  
150 monitoring sites.

## 151 **2.2. Calculations of the health impacts attributable to the traffic-related air pollutants**

152 Many studies have applied various methods to calculate the impact of long-term exposure to  
153 fine particles, nitrogen dioxide, sulphur dioxide and ozone on all-cause premature mortality  
154 (Burnett et al., 2014; Cohen et al., 2017; Jerrett et al., 2009; Turner et al., 2016; Yin et al., 2017).  
155 The attributable mortality or disability adjusted life years (DALYs) have been estimated based  
156 on detailed estimates of the exposure concentration and application of exposure-response  
157 functions. In this study, we estimated the premature mortality and acute morbidity attributable to  
158 traffic-related  $PM_{2.5}$  via the exposure-response (ER) function. Moreover, we calculated the  
159 relative public health effects of pairs of air pollutants based on the relative risk of the attributable  
160 mortality as derived from previous studies in China.

161 The concentrations of  $PM_{2.5}$  from a dispersion simulation on urban roads in Beijing were  
162 estimated by the urban air pollution dispersion and chemistry model ADMS-Urban, based on the  
163  $PM_{2.5}$  emission inventory of exhaust vehicles. ADMS-Urban is a quasi-Gaussian pollution  
164 dispersion and chemistry model that has been widely applied for assessment of emission control  
165 strategies and simulation of the dispersion of air pollutants (e.g. Biggart et al., 2020; Cui et al.,  
166 2022).

167 On the basis of the population data (<https://www.worldpop.org/geodata/summary?id=3487>  
168 3), mortality and morbidity baseline (National Health Commission of China, 2015 & 2021) in for  
169 each year, the acute morbidity and premature deaths attributable to traffic-related  $PM_{2.5}$  exposur  
170 e were calculated using a Poisson regression model as follow.

$$171 P_{tra} = P_{total} \times \frac{EXP(\beta \times C_{tra}) - 1}{EXP(\beta \times C_{tra})} \times I_0 \quad (2)$$

172 where  $P_{tra}$  is the number of premature deaths due to traffic-related  $PM_{2.5}$ ;  $P_{total}$  represents the total  
173 population in Beijing;  $\beta$  is the beta coefficient from a log-linear model indicating changes in all-  
174 cause mortality and respiratory morbidity per unit concentration, which is 5.4‰ and 5‰,  
175 respectively, from the previous studies (Fang et al., 2016; Tong et al., 2020);  $C_{tra}$  refers to the



176 exposure concentration of population-weighted traffic-related PM<sub>2.5</sub>. I<sub>0</sub> denotes the baseline  
177 mortality and morbidity rates in Beijing.

178 In order to assess the relative public health effect of NO<sub>2</sub>, SO<sub>2</sub> and PM<sub>2.5</sub>, we followed the  
179 methods of Harrison and Beddows (2017), who first considered two pollutants in the same model  
180 and compared the effects of PM<sub>2.5</sub> and NO<sub>2</sub> originating from traffic on the premature mortality in  
181 London. They applied the Hazard Ratio coefficients for PM<sub>2.5</sub> and NO<sub>2</sub> that were recommended  
182 by the World Health Organization (WHO) Health Risks of Air Pollution in Europe (HRAPIE)  
183 project to assess the mortality impact of traffic-related NO<sub>2</sub> and PM<sub>2.5</sub>, which increased from  
184 2010 to 2015. We collected the Hazard Ratios for air pollutants, thereby obtaining possible  
185 results from previous studies in China, for each 10 µg m<sup>-3</sup> increment in China, respectively,  
186 which is described in detail in the Supplementary. The Hazard Ratios for annual mean PM<sub>2.5</sub>,  
187 NO<sub>2</sub> and SO<sub>2</sub> exposure and natural-cause mortality were 1.11 (95% CI: 1.08-1.14), 1.06 (95% CI:  
188 1.01-1.04) and 1.105 (95% CI: 1.022-1.195).

189 The relative health impact on the all-cause mortality was calculated as follows.

$$190 \quad RHI_{i,j,r} = \frac{\Delta C_{i,r}}{\Delta C_{j,r}} \times \frac{MI_i - 1}{MI_j - 1} \quad (3)$$

191 where RHI<sub>i,j,r</sub> is the relative health impact of air pollutants i and j in year r; ΔC<sub>i,r</sub> and ΔC<sub>j,r</sub> are  
192 the road increments of air pollutants i and j, respectively, in year r; and MI<sub>i</sub> and MI<sub>j</sub> are the  
193 Hazard Ratios per unit mass, which represent the all-cause mortality impact of air pollutants i  
194 and j. In this study, i refers to the NO<sub>2</sub> or SO<sub>2</sub>, and j represents PM<sub>2.5</sub>, and r is the year from 2014  
195 to 2020.

196 Here we just focused the relative health effect of the traffic-related air pollutants, such as  
197 PM<sub>2.5</sub>, NO<sub>2</sub>, and SO<sub>2</sub> based on their road incremental concentration of pollutants. For O<sub>3</sub>, it is  
198 mostly titrated by the fresh NO emitted from road traffic at roadside, so the road traffic  
199 emissions are a sink for ozone, and hence the roadside increment is negative for O<sub>3</sub> (See Figure  
200 2). The corresponding public health risk of ozone with other air pollutants is significantly  
201 reduced due to the titration effect of NO at roadside. Therefore, we did not include ozone in our  
202 assessment of health impacts.

### 203 **3. Results**

#### 204 **3.1. Long-term trends of the air pollutants in the roadside environment of urban Beijing**

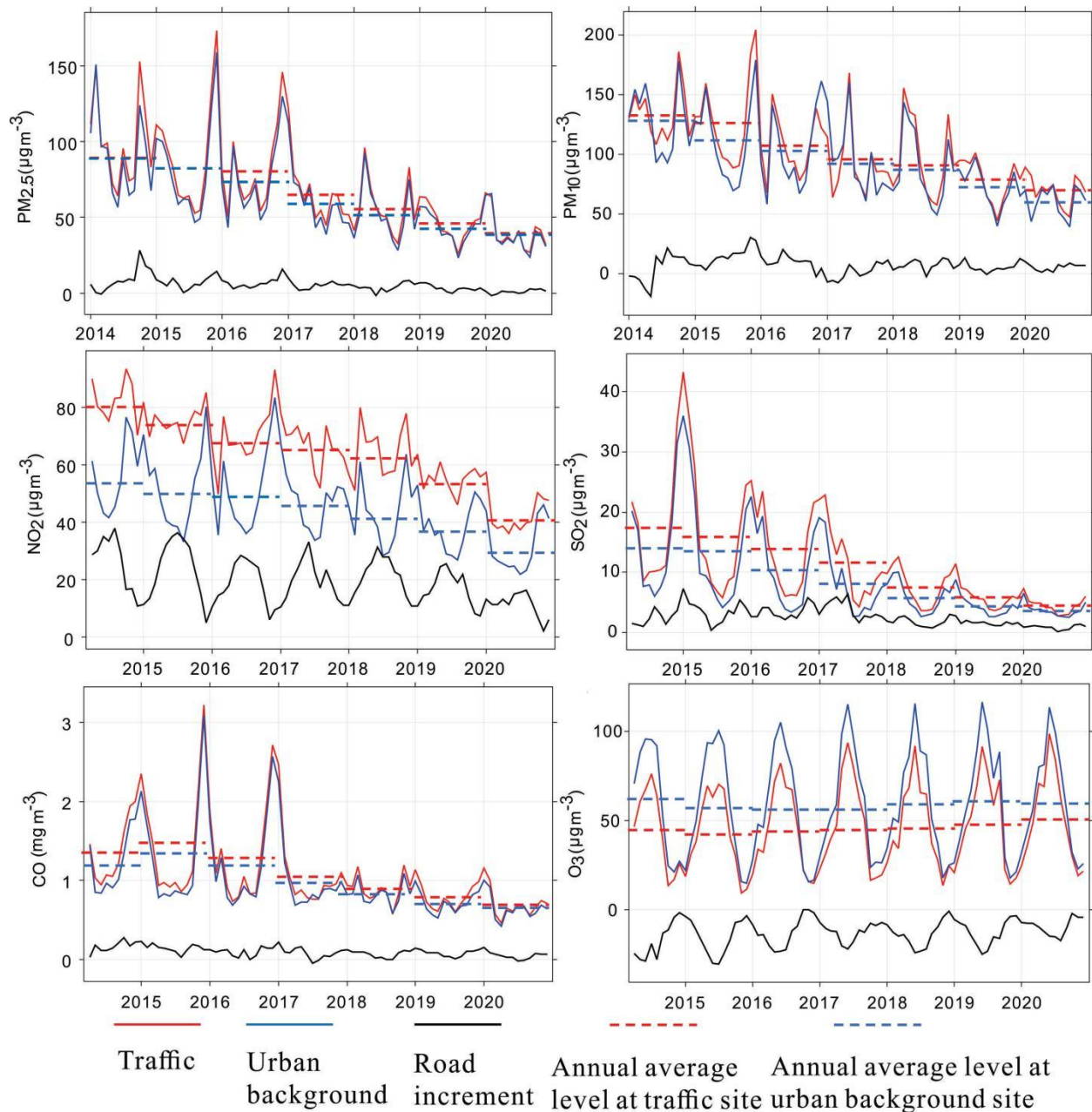
205 The average mass concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub>, CO and O<sub>3</sub> from 2014 to 2020  
206 at five roadside monitoring sites were 101.1 µg m<sup>-3</sup>, 68.3 µg m<sup>-3</sup>, 65.3 µg m<sup>-3</sup>, 11.0 µg m<sup>-3</sup>, 1.1

207 mg m<sup>-3</sup>, and 46.1 µg m<sup>-3</sup>, respectively. These levels were higher than those at urban background  
 208 sites, except for ozone, which were 94.4 µg m<sup>-3</sup>, 63.0 µg m<sup>-3</sup>, 45.8 µg m<sup>-3</sup>, 8.6 µg m<sup>-3</sup>, 1.0 mg m<sup>-3</sup>,  
 209 and 59.2 µg m<sup>-3</sup>, respectively. The spatial pattern indicated that the NO<sub>2</sub> level in the city was  
 210 largely affected by traffic emissions, but the PM<sub>2.5</sub> level was largely affected by a large  
 211 background from regional transport of air pollution (Figure S1 and S2). This result reflected the  
 212 fact that traffic is a significant source for the air pollutants in the city, which was verified by the  
 213 significant correlations of NO<sub>2</sub> and other air pollutants (Figure S3). As shown in Table 1 and  
 214 Figure 2, the observations demonstrate that air pollutant concentrations have decreased  
 215 significantly by 47% to 71% between 2014 and 2020. For example, the mean value of PM<sub>2.5</sub> in  
 216 2020 was almost 40% lower than that observed in 2014. Moreover, there was a remarkable  
 217 decrease for other air pollutants, such as 4.4-20.0% a<sup>-1</sup> for PM<sub>10</sub>, 2.4-22.9% a<sup>-1</sup> for NO<sub>2</sub>, 10.2-  
 218 39.7% a<sup>-1</sup> for SO<sub>2</sub>, and 10.4-18.2% a<sup>-1</sup> for CO. On the contrary, ozone increased by 17.4% during  
 219 this period. The gradual decrease of these air pollutants is primarily attributed to the  
 220 implementation of a series of policies and regulations on traffic emission control and  
 221 management (Vu et al., 2019). Whereas Vu et al. (2019) used the Random Forest machine  
 222 learning algorithm to reduce the impact of both seasonal and weather variations in this study, the  
 223 trends have not been deweathered as this had only modest effects and the current dataset is  
 224 longer, giving less scope for influences from inter-annual variability of weather. It should also be  
 225 noted that although data are reported for 2020, these are likely to be anomalous due to the  
 226 significant effect of the COVID-19 lockdown (Cao et al., 2021, Shi et al., 2021), which is  
 227 comparable with the findings at Wuhan, Daegu and Tokyo during this period (Ma et al., 2020).  
 228 **Table 1.** Concentration of the air pollutants (mg m<sup>-3</sup> for CO and µg m<sup>-3</sup> for others) and the  
 229 decrease in concentration relative to the previous year (% a<sup>-1</sup>) during 2014-2020.

Year	PM <sub>2.5</sub>	PM <sub>10</sub>	NO <sub>2</sub>	SO <sub>2</sub>	CO	O <sub>3</sub>
	Conc.% decrease	Conc.% decrease	Conc.% decrease	Conc.% decrease	Conc.% decrease	Conc.% decrease
2014	98.2	132.7	82.0	16.1	1.4	45.9
2015	90.4	8.0	126.9	4.4	75.6	7.8
2016	80.8	10.6	106.8	15.8	69.8	7.7
2017	67.6	16.3	91.8	14.1	68.1	2.4
2018	55.7	17.6	94.5	-3.0	64.5	5.3
2019	46.1	17.3	75.6	20.0	55.4	14.0
2020	39.9	13.4	66.6	11.9	42.7	22.9
					4.5	23.9
					0.7	11.3
						51.5
						-6.5

230 Conc. means concentration.

231 Figure 2 further presents the seasonal variation in the monthly values of air pollutants at  
232 traffic sites and urban background sites. Except for ozone, the traffic sites attained higher values  
233 than those at urban background sites in most months. PM<sub>2.5</sub> concentrations were higher in winter  
234 (traffic sites: 83.2  $\mu\text{g m}^{-3}$ ; urban background sites: 76.8  $\mu\text{g m}^{-3}$ ) and lower in summer (traffic  
235 sites: 52.6  $\mu\text{g m}^{-3}$ ; urban background sites: 49  $\mu\text{g m}^{-3}$ ), followed by those in fall and spring. This  
236 result corresponds with the findings in urban Beijing obtained by Xu and Zhang (2020). SO<sub>2</sub> and  
237 CO trends were similar to that of the PM<sub>2.5</sub> concentration. The highest PM<sub>10</sub> concentrations  
238 (traffic sites: 117.7  $\mu\text{g m}^{-3}$ ; urban background sites: 114.2  $\mu\text{g m}^{-3}$ ) were observed during spring,  
239 followed by winter, fall, and summer. The highest concentration of PM<sub>10</sub> in spring may be  
240 attributable to the significant contribution of Asian dust that is transported from North China  
241 during spring (Liu et al., 2014). There is a minor seasonal fluctuation for NO<sub>2</sub> at the traffic sites  
242 (range: 60.8 to 68.5  $\mu\text{g m}^{-3}$ ), but the NO<sub>2</sub> data appeared to indicate higher values in winter and  
243 fall, at approximately 52  $\mu\text{g m}^{-3}$ , and a lower level in summer (33.9  $\mu\text{g m}^{-3}$ ) at the urban  
244 background sites. In contrast, the highest level of ozone was found in summer.



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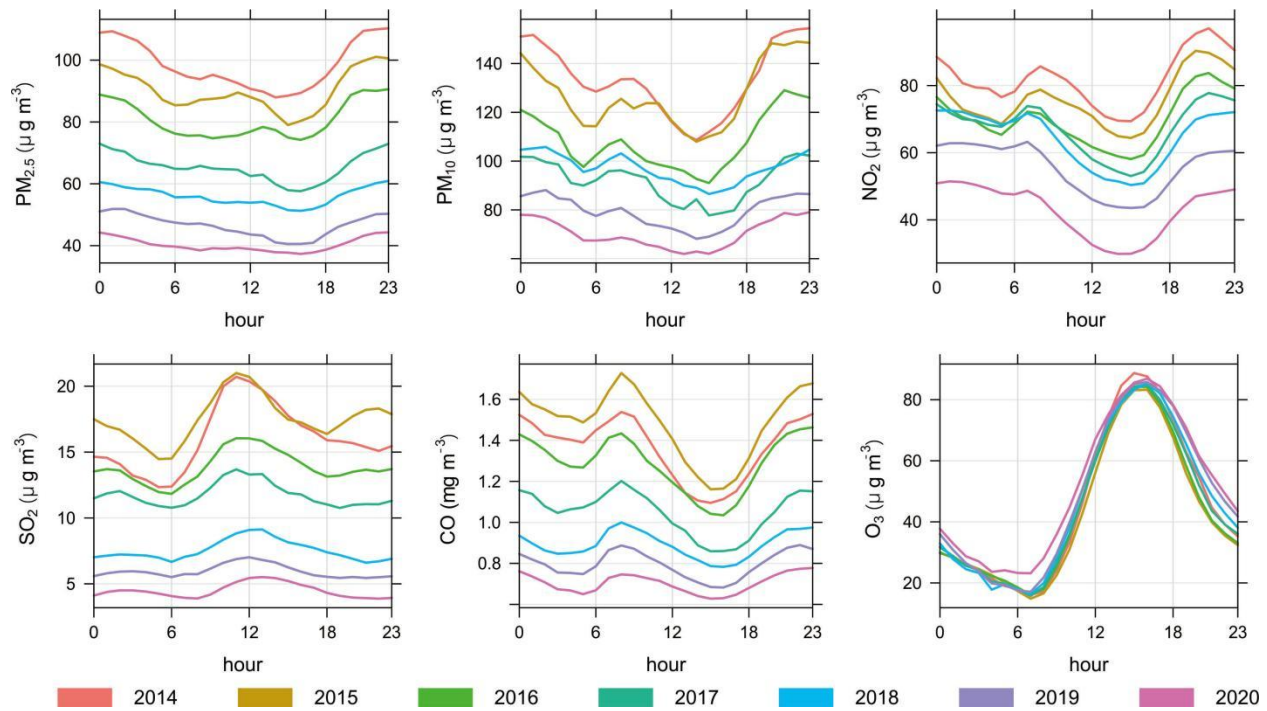
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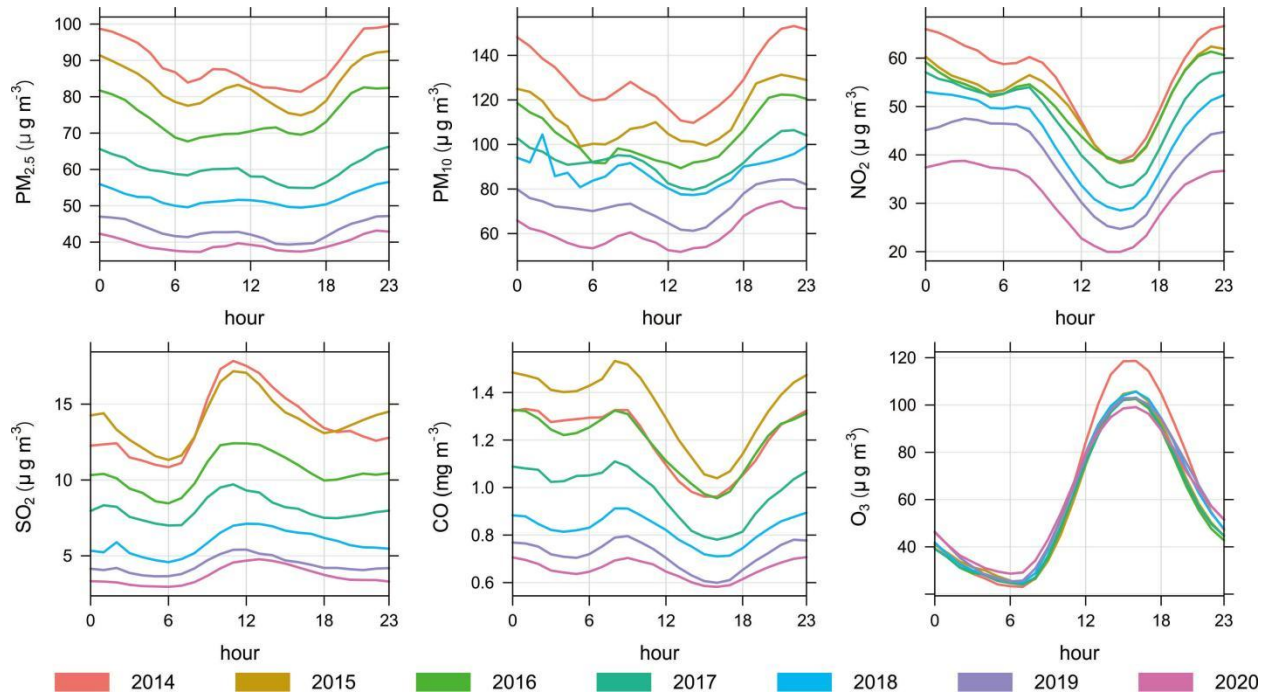
**Figure 2.** Monthly mean concentration trends of air pollutants at the traffic sites and urban background monitoring sites, and the roadside increment. The pollutant values at the traffic sites are calculated as the average value over the five traffic monitoring sites. The pollutant levels at urban background sites are calculated as the average value from twelve urban background monitoring sites

Diurnal variations of air pollutants at traffic sites and urban background sites during 2014 to 2020 are shown in Figures 3 and 4, respectively. Generally, higher levels of air pollutants were observed at nighttime rather than in daytime, indicating that mixed boundary layers and low

254 temperature highly influence the daily patterns of pollutants (Zhong et al., 2017). The highest  
 255 NO<sub>2</sub> concentration at the traffic sites was observed earlier than that at urban background sites. A  
 256 weak peak was observed in the morning (7:00-9:00 am) which is a typical high emission period  
 257 due to the traffic rush hour. The lowest values of these air pollutants were observed during 5:00  
 258 to 6:00 am and 12:00 to 16:00 pm. On the contrary, the peak of concentrations for SO<sub>2</sub> and ozone  
 259 occurred during daytime. Additionally, the difference between the highest and lowest values at  
 260 traffic sites was approximately 22% greater than that at urban background sites, suggesting that  
 261 the NO<sub>2</sub> concentration in the ambient environment was significantly affected by traffic.



262  
 263 **Figure 3.** Diurnal variation of components at traffic sites during 2014 to 2020.

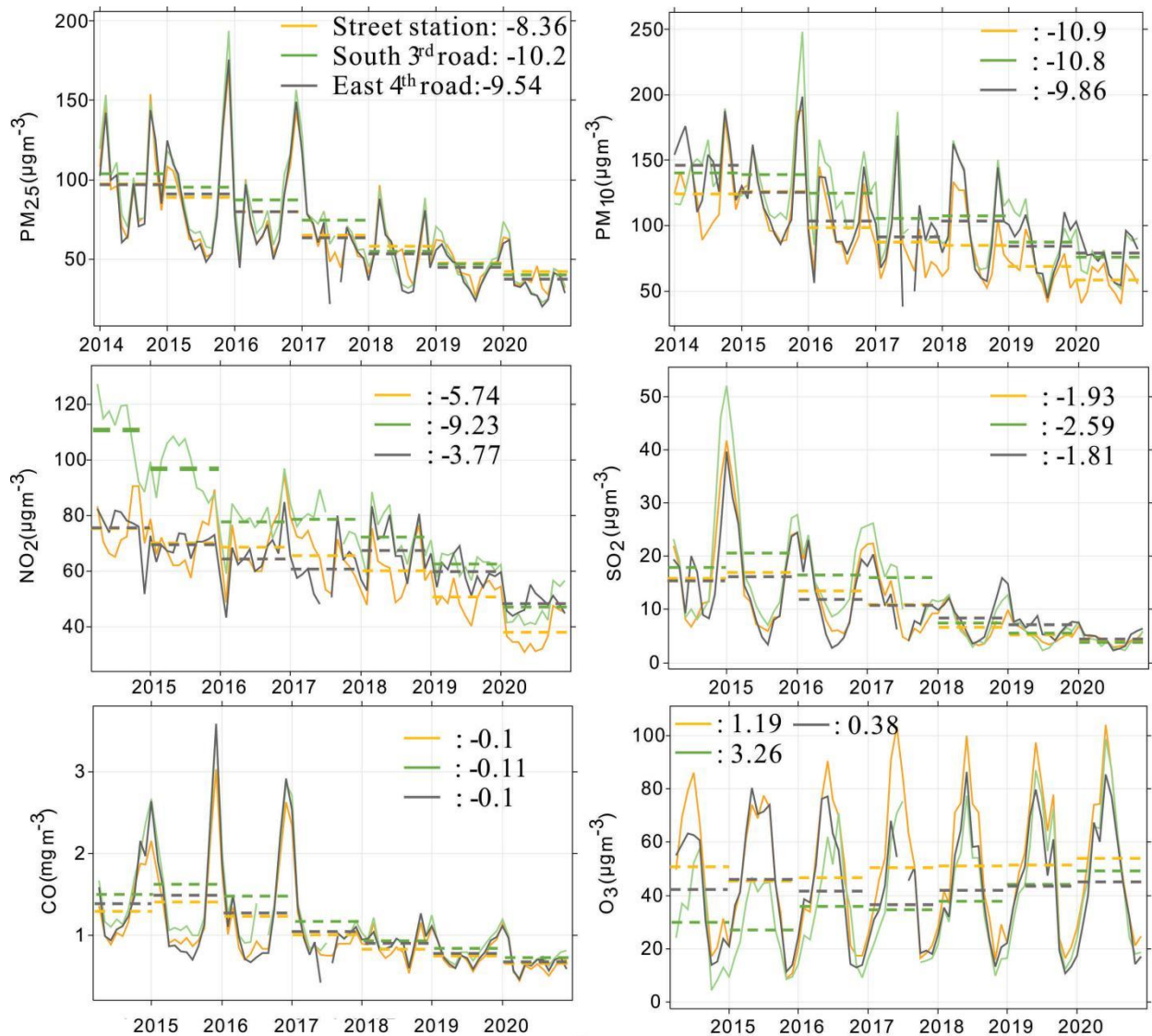


264  
265 **Figure 4.** Diurnal variation of components at urban background sites during 2014 to 2020.

266 **3.2. Comparison of different traffic monitoring sites**

267 The air pollutant levels at the South 3<sup>rd</sup> Ring Road station were higher than those at the  
 268 Street stations and the East 4<sup>th</sup> Ring Road station during 2014 to 2020 as presented in Figure 5.  
 269 For instance, the PM<sub>2.5</sub> concentrations at the South 3<sup>rd</sup> Ring Road were 5.9 to 9.9 µg m<sup>-3</sup> higher  
 270 than those in the Street stations and 3.8 to 8.2 µg m<sup>-3</sup> higher than those at the East 4<sup>th</sup> Ring Road  
 271 during 2014 to 2017. On the contrary, PM<sub>2.5</sub> values at these stations were similar during 2018 to  
 272 2020, which showed a similar trend with the SO<sub>2</sub>. PM<sub>2.5</sub> and SO<sub>2</sub> concentrations at these three  
 273 groups of sites indicated a statistically significant difference (p<0.001) between street sites and  
 274 South 3<sup>rd</sup> and East 4<sup>th</sup> Ring Road stations. The other air pollutants at these sites also exhibited a  
 275 statistically significant difference (p<0.001). Based on the Theil-Sen estimator, the overall trend  
 276 of air pollutants at the South 3<sup>rd</sup> Ring Road station, as shown in Figure 5, decreased more than  
 277 those at the other stations every year. The values of PM<sub>10</sub> at the South 3<sup>rd</sup> Ring Road were also  
 278 mostly higher than that at Street stations (11.0 to 26.8 µg m<sup>-3</sup> higher), but were comparable to  
 279 that on the 4<sup>th</sup> Ring Road. This result suggests that non-exhaust emissions, including road surface  
 280 and resuspended dust, and wear of the brakes and tyres, contributed to coarse particles. This is  
 281 attributed to the larger number of heavy duty vehicles on highways than that on the urban streets,  
 282 which produce high non-exhaust emissions. NO<sub>2</sub> was mainly influenced by vehicles, and was the  
 283 highest in the South 3<sup>rd</sup> Ring Road (109.8 to 47.6 µg m<sup>-3</sup>). There is a clearly decreasing trend for

284 the NO<sub>2</sub> level from 2014 to 2020 at these stations, indicative of the remarkable achievement in  
 285 Beijing's traffic air quality improvement after implementing a series of regulations on traffic  
 286 emission control from 2013. Regarding CO, the South 3<sup>rd</sup> Ring Road also had CO concentrations  
 287 0.05 to 0.3 mg m<sup>-3</sup> higher than that at Street stations and the 4<sup>th</sup> Ring Road. This probably results  
 288 from the higher traffic volume. However, the concentration of O<sub>3</sub> was highest in the Street  
 289 stations (45.8 to 54.2 μg m<sup>-3</sup>), followed by the 4<sup>th</sup> Ring road (32.9 to 46.7 μg m<sup>-3</sup>) and South 3<sup>rd</sup>  
 290 Ring road (27.7 to 49.5 μg m<sup>-3</sup>). The O<sub>3</sub> values were higher in summer and lower in winter, and  
 291 show a gradual increase from 2014 to 2020.



292 --- AAC at street station --- AAC at South 3<sup>rd</sup> road - - - - - AAC at East 4<sup>th</sup> road  
 293 **Figure 5.** Trend of the monthly mean pollutant concentration at traffic monitoring stations. The  
 294 numbers shown in the figure represent the overall trend of air pollutant level (μg m<sup>-3</sup> a<sup>-1</sup>) by

295 Theil-sen estimator. AAC refers to the annual average concentration.

296 **3.3. Long-term trends of roadside increment of air pollutants.**

297 The roadside increments and their percentage increases above urban background  
 298 concentrations of air pollutants in Beijing for the period of 2014 to 2020 are summarized in  
 299 Table 2 and Figure 6. The highest annual mean roadside increment when expressed as a  
 300 concentration was found for CO (95.7  $\mu\text{g m}^{-3}$ ), followed by NO<sub>2</sub> (19.5  $\mu\text{g m}^{-3}$ ), PM<sub>2.5</sub> (5.3  $\mu\text{g m}^{-3}$ )  
 301 and SO<sub>2</sub> (2.4  $\mu\text{g m}^{-3}$ ), which is lower than those observed in London and Hong Kong during  
 302 2016 to 2018 which is also a metropolis with a high vehicle volumes and has implemented many  
 303 policies to reduce the traffic-related air pollutants (Harrison et al., 2021). It clearly indicates that  
 304 vehicle exhaust is a significant source of CO and NO<sub>2</sub> in urban Beijing. PM<sub>2.5</sub> roadside  
 305 increments experienced an obvious decrease from 2014 (9.4  $\mu\text{g m}^{-3}$ ) to 2020 (0.9  $\mu\text{g m}^{-3}$ ), and  
 306 the percentage increase above urban background values by traffic decreased from 16.8% in 2014  
 307 to 3.5% in 2020. In the case of NO<sub>2</sub>, the increments due to traffic and the percentage increment  
 308 above urban background presented a smaller change during this period compared with the PM<sub>2.5</sub>  
 309 and CO. It appears that the upgrades to the vehicle fleet over this period had greater benefits for  
 310 PM<sub>2.5</sub> emissions than for oxides of nitrogen, although this may be an artefact of the non-linear  
 311 relationship between NO<sub>x</sub> and NO<sub>2</sub>. Measurements at a roadside site in London show little  
 312 change in NO<sub>2</sub> despite a substantial reduction in NO<sub>x</sub> (Krecl et al., 2021) The roadside  
 313 increments of PM<sub>2.5</sub>, NO<sub>2</sub> and SO<sub>2</sub> were approximately 30-50% higher at the South 3<sup>rd</sup> road than  
 314 those at the East 4<sup>th</sup> road and Street sites during 2014 to 2017, but the levels of air pollutant at  
 315 these sites presented similar concentrations during 2018 to 2020.

316 **Table 2.** Roadside increment ( $\Delta C$ ) ( $\mu\text{g m}^{-3}$ ) and percentage increment of roadside concentration  
 317 of air pollutants above that of urban background concentration from 2014 to 2020 (% increase).

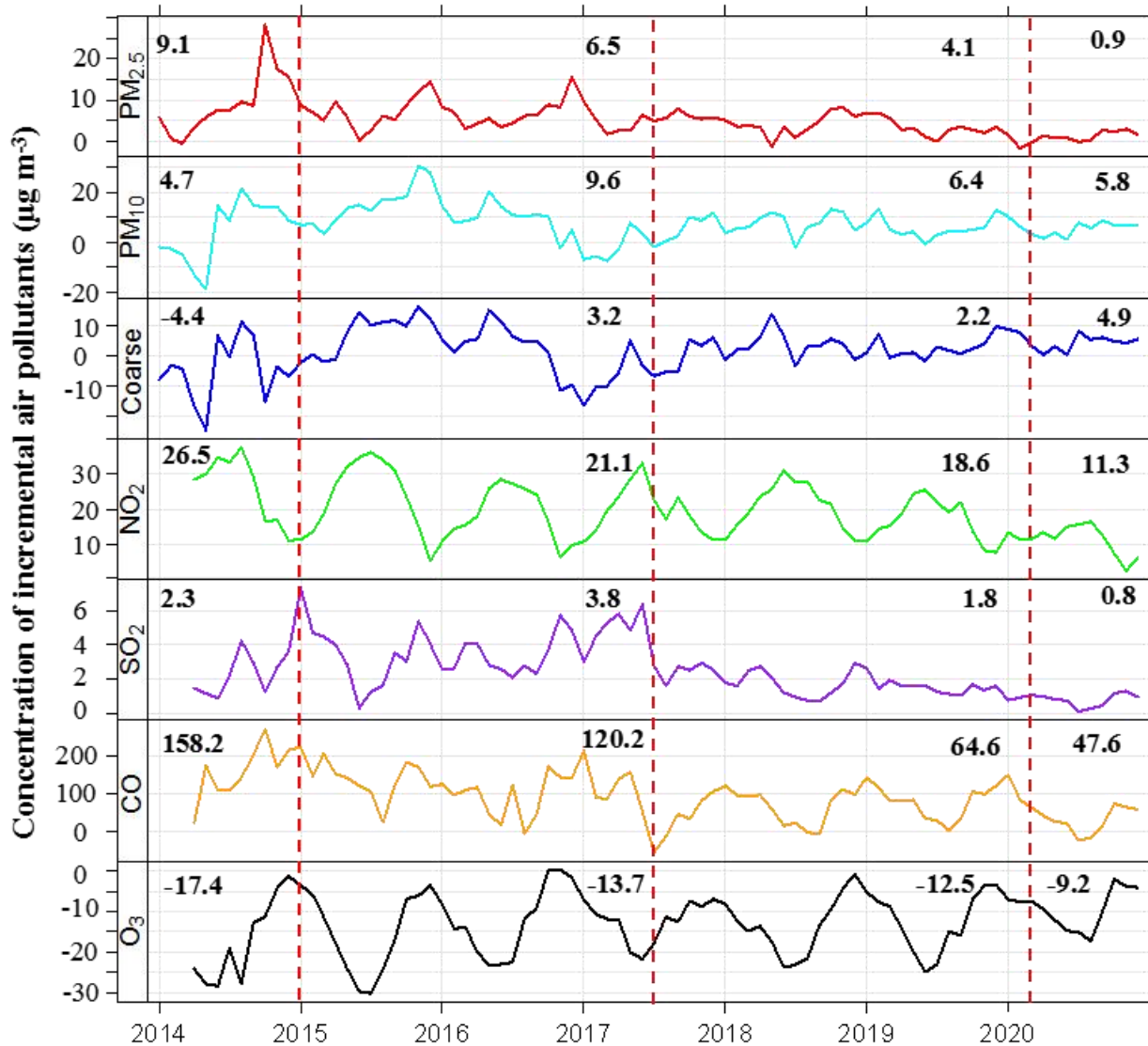
Year	PM <sub>2.5</sub>		PM <sub>10</sub>		NO <sub>2</sub>		SO <sub>2</sub>		CO		O <sub>3</sub>	
	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase	Conc.	% increase
2014	9.4	16.8	3.6	6.5	26.4	68.4	2.3	35.7	164.2	15.5	-17.2	-20.9
2015	7.1	12.7	14.4	20.9	23.7	67.1	3.5	41.1	143.4	13.2	-15.1	-29.4
2016	6.7	13.1	10.3	16.6	18.6	51.5	3.3	51.0	95	9.5	-12.3	-11.9
2017	5.4	12.8	2.3	7.2	19.2	56.3	3.7	73.9	72.1	9.4	-12.0	-26.7
2018	4.0	12.5	7.5	16.0	20.8	70.2	1.7	39.4	65.9	9.5	-13.5	-21.5
2019	3.4	10.1	5.8	15.5	16.7	66.4	1.6	47.8	78.6	12.0	-12.3	-23.2
2020	0.9	3.5	5.8	23.6	11.3	53.5	0.8	28.8	47.6	8.0	-9.2	-15.2



318 Conc. denotes the concentration.

319  $(\% \text{ increase} = (\Delta C_i / C_{i, \text{urban}}) \times 100$

320 Average roadside increments during 2014-2020 accounted for 7.7% and 7.2% of the total  
321 mass concentrations of PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, at the traffic sites. The highest increment of  
322 fine particles was found in fall and winter, which could be attributed to poorer dispersion of  
323 primary emissions. There is an opposite trend in the temporal variation of NO<sub>2</sub> and O<sub>3</sub> roadside  
324 increments, as shown in Figure 6. Higher roadside increments of NO<sub>2</sub> were found in summer, but  
325 the roadside increment of O<sub>3</sub> reached a peak in winter. The difference between the monthly  
326 variation of road increments of O<sub>3</sub> and NO<sub>2</sub> probably results from the complex reactions between  
327 NO<sub>2</sub>-NO<sub>x</sub>-O<sub>3</sub> yielding the different concentrations of secondary NO<sub>2</sub> and O<sub>3</sub> depending upon the  
328 occurrence of NO emitted from road vehicle exhaust and photochemistry. Figure S4 shows the  
329 polar plots of roadside increment of air pollutants by year. It seems that the decrease in the case  
330 of PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub> and CO in Beijing was linked to prohibition of trucks and old vehicles from  
331 entering the city within the Sixth Ring Road during daytime since 2014. It is confirmed by a  
332 distinct peak of NO<sub>2</sub> in the late evening as shown in Figure 3. Similarly, the temporal trend of  
333 roadside increment of SO<sub>2</sub> also had an obvious decrease from 2018.



334

335 **Figure 6.** Trend of the roadside increment in Beijing during 2014-2020. The numbers in the  
 336 figure indicate the road increments during different periods under the various policies.

337 **4. Discussion**

338 **4.1 Discussion on the improvement of air quality at traffic sites**

339 The concentrations of air pollutants in both the roadside and urban environment exhibited a  
 340 substantial decrease from 2014 to 2020 in Beijing, clearly indicative of the effectiveness of air  
 341 pollution control measures. Road traffic contributed to a notable roadside increment of NO<sub>2</sub>  
 342 above the urban background, and the NO<sub>2</sub> at roadside sites declined the most at the South 3<sup>rd</sup>  
 343 Ring Road and Street stations, and NO<sub>2</sub> at East 4<sup>th</sup> had a slightly decrease during this period.  
 344 This may be the result of the low emission zone program from 2016 in central urban Beijing. The

345 gradual decreases of PM<sub>2.5</sub>, PM<sub>10</sub>, CO, and SO<sub>2</sub> in the roadside atmosphere were not only driven  
346 by the reduction of emissions from vehicles, but also caused by the reduction of other  
347 combustion sources, such as industry and power plants (Zhang et al., 2019a). Ozone is a  
348 secondary pollutant that is affected by several factors. The dominant factor is the NO<sub>x</sub>-O<sub>3</sub>  
349 photochemical steady state, as the reduction of traffic-related NO has led to increases in O<sub>3</sub> due  
350 to reduced O<sub>3</sub> titration (Shi et al., 2021). PCA and cluster analysis showed that low wind speed,  
351 high temperature and relative humidity promote the accumulation of O<sub>3</sub> (Table S1 and Figure S5)  
352 (Guo et al., 2017, Xue et al., 2014).

353 Roadside increments as calculated using a paired-site approach at different monitoring sites  
354 in Beijing could be subject to artefacts if other sources influence the measuring site. Long-term  
355 datasets and consideration of multiple sites, however, should minimise this effect. Due to the  
356 characteristics of the different monitoring sites and different traffic volumes, the roadside  
357 increments measured at the five sites represent a range of behaviours, but all show a consistent  
358 pattern. The results reported above indicate that roadside increments of PM<sub>2.5</sub> diminished  
359 appreciably from 2014 to 2020 in Beijing, which could be explained by the reduced primary  
360 emissions of fine particles and associated gaseous precursors originating from vehicles. In the  
361 case of NO<sub>2</sub>, the roadside increment declined slightly and showed a similar trend before 2019  
362 with a higher level in summer and lower level in winter. It is likely that NO<sub>x</sub> declined more than  
363 NO<sub>2</sub>, but the data are not available to verify this.

364 The downward trends of the PM<sub>2.5</sub>, SO<sub>2</sub> and CO concentrations in Figure 2 showed a  
365 breakpoint from 2017, and the rate of decline increased to 16.3%, 10.3% and 17.3% comparing  
366 with 2016. This phenomenon is likely attributable to the implementation of a new China VI fuel  
367 standard in January 2017 and the prohibition of China I and II standard light-duty vehicles from  
368 entering the central zone within the Fifth Ring Road (which includes the traffic roadside  
369 monitoring sites) after February 2017. Additionally, the implementation of the China V standard  
370 for all spark ignition vehicles in July 2015 and the increased electrification of the vehicle fleet in  
371 Beijing contributed to the decline of PM<sub>2.5</sub> and CO. As shown in Tables S2 and S3, the  
372 proportion of China V vehicles in Beijing reached around 50% from 2017, and the emission  
373 limits of China V and VI for PM and CO originating from the vehicles were reduced, and ultra-  
374 low sulphur fuels were adopted for all vehicles from 2017 in the cities surrounding Beijing.  
375 Meanwhile, China set up a no-coal zone in cities around Beijing from November 2017, which

376 may also have contributed to the reduction of the PM<sub>2.5</sub> and SO<sub>2</sub> in roadside air, but also in the  
377 urban background. Based on Figure 6, the roadside increments of PM<sub>2.5</sub>, NO<sub>2</sub> and CO could be  
378 divided into four periods: the first period of 01/2014-01/2015, the second period of 01/2015-  
379 06/2017 (implementation of China V standard), the third period of 07/2017-12/2019  
380 (introduction of China VI vehicle and China VI fuel standard), and the period after 2020 which  
381 was affected by the COVID-19 lockdown and prohibition of China III diesel trucks from  
382 entering Beijing. The introduction of these policies is already having a very significant benefit  
383 for air quality. For example, roadside increments of PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO during the third  
384 period were 4.1 µg m<sup>-3</sup>, 18.6 µg m<sup>-3</sup>, 1.8 µg m<sup>-3</sup> and 64.6 µg m<sup>-3</sup>, versus those of 6.5 µg m<sup>-3</sup>, 21.1  
385 µg m<sup>-3</sup>, 3.8 µg m<sup>-3</sup> and 120.2 µg m<sup>-3</sup> during second period, respectively. It reveals that the  
386 implementation of the new vehicle standards (Sun et al., 2021) was highly effective in terms of  
387 reduction of the roadside increments for CO (59.1%), PM<sub>2.5</sub> (54.8%), NO<sub>2</sub> (29.8%), SO<sub>2</sub> (20.6%).  
388 The change for the NO<sub>2</sub> was relatively limited, possibly due to the non-linearity between NO<sub>x</sub>  
389 and NO<sub>2</sub> concentrations. These control policies were found not to affect the roadside increment  
390 of coarse particles (PM<sub>2.5-10</sub>) (see Figure 6) which arise largely from non-exhaust sources  
391 (Thorpe and Harrison, 2008) and were not subject to additional controls. On the other hand, the  
392 decrease of air pollutants from 2020 was due to the reduction of traffic flow (decrease by 7.5%  
393 with 2019) in Beijing due to the COVID-19 lockdown.

#### 394 **4.2. Relative public health impacts of traffic-related PM<sub>2.5</sub>, NO<sub>2</sub> and SO<sub>2</sub>**

395 In Beijing, traffic emissions have significantly contributed to the air pollutants that have an  
396 influence on public health (Lei et al., 2012; Tong et al., 2020). Based on the ADMS simulation,  
397 the annual average PM<sub>2.5</sub> population weighted concentration from vehicle emissions is 8.0 µg m<sup>-3</sup>  
398 and 1.56 µg m<sup>-3</sup> in 2014 and 2020, respectively (Figure 7), which is similar to the PM<sub>2.5</sub> level  
399 estimated from the paired-site method and comparable with the results simulated by Tong et al.  
400 (2020). The annual attributable premature mortality due to traffic-related PM<sub>2.5</sub> was calculated by  
401 the ER function, which was 8379 cases (CI: 3686, 7586) in 2014, while an obvious decrease in  
402 health effect occurred in 2020 with 1908 (CI: 1217, 2533) cases due to much reduced PM<sub>2.5</sub>  
403 emissions from vehicles. Among the total premature deaths, the number within the Fifth Ring  
404 Road accounted for 58.5% in 2014, which were 688, 1219, 1388 and 1608 cases within the 2<sup>nd</sup>  
405 Ring Road, 2<sup>nd</sup> - 3<sup>rd</sup>, 3<sup>rd</sup> - 4<sup>th</sup>, and 4<sup>th</sup> -5<sup>th</sup> Ring Road, respectively, resulting from the lower  
406 population in the city center. By comparison, the proportion of premature deaths due to traffic-

407 related PM<sub>2.5</sub> within the Fifth Ring Road slightly decreased to 50% in 2020. For the case of the  
408 acute morbidity from respiratory diseases due to PM<sub>2.5</sub>, the incidence in 2014 with 461479 cases  
409 was higher than that in 2020 with 93045, illustrating another aspect of the severe public health  
410 burden of traffic-related PM<sub>2.5</sub>. As mentioned above, a series of policies have been implemented  
411 to reduce the emissions of air pollutants from vehicles from 2013, resulting in the reduction of  
412 health burden.

413 In addition, the relative public health impacts of traffic-related PM<sub>2.5</sub> NO<sub>2</sub> and SO<sub>2</sub> were  
414 approximated according to their effect on all-cause mortality based on an exposure-response  
415 function during different time periods. The ratio of premature mortality impact per unit mass for  
416 NO<sub>2</sub> and PM<sub>2.5</sub> calculated from the Hazard Ratios is 0.27. The ratio of mortality impact for SO<sub>2</sub>  
417 and PM<sub>2.5</sub> is 0.95 for a unit mass of each. When the relative impact of two traffic-related  
418 pollutants is to be estimated, the relative concentrations of these two pollutants arising from  
419 traffic should be taken into account, as in equation (3). As a result, the ratio of all-cause mortality  
420 owing to road traffic-related NO<sub>2</sub> relative to PM<sub>2.5</sub> ranged from 0.76 to 0.97 in 2017, and reached  
421 1.32 in 2019, which is presented in Figure S6. Meanwhile, the ratio for mortality caused by the  
422 SO<sub>2</sub> relative to PM<sub>2.5</sub> is 0.23 in 2014, and 0.44-0.65 during 2015-2019. This analysis may  
423 underestimate the effects of NO<sub>2</sub> as it uses a ratio of NO<sub>2</sub>/PM<sub>2.5</sub> derived from roadside  
424 measurements, whereas the NO<sub>2</sub>/NO<sub>x</sub> ratio typically increases with greater dilution away from  
425 roadside, an effect explicitly accounted for by Harrison and Beddows (2017) who had NO<sub>x</sub> data  
426 as well as NO<sub>2</sub> from roadside and background sites. It indicates that the premature mortality  
427 impact of traffic-related NO<sub>2</sub> is now comparable to, or may exceed the impact of PM<sub>2.5</sub> from  
428 traffic. The influence of traffic-related SO<sub>2</sub> upon mortality was markedly less. As shown in  
429 Figure S7, the traffic-related nitrogen dioxide and sulphur dioxide relative to PM<sub>2.5</sub> at street  
430 stations yielded a similar relative impact to that at the South 3<sup>rd</sup> and East 4<sup>th</sup> Ring Road site  
431 before 2018, indicating that the effect of NO<sub>2</sub> on premature mortality was of a similar magnitude  
432 to that of PM<sub>2.5</sub>, but greater than that of SO<sub>2</sub>. However, the trend changed afterwards. The effect  
433 of NO<sub>2</sub> and SO<sub>2</sub> at the East 4<sup>th</sup> Ring Road on public health relative to PM<sub>2.5</sub> increased faster than  
434 at the other two sites, which was due to the prohibition of diesel trucks entering the inner city.

435 Although strict control measures have been applied to vehicle emissions, the impact of air  
436 pollutants on human health has a significant variation according to the vehicle types and fuel. Shi  
437 et al. (2021) concluded that the decline in NO<sub>2</sub> concentrations attributable to the COVID-19

lockdown which introduced severe restrictions on road traffic was not as notable as expected in China. This is consistent with the result from Grange et al. (2017), who analysed  $\text{NO}_x$  and  $\text{NO}_2$  hourly concentrations spanning 130 million hours retrieved from roadside monitoring stations in Europe and found that the roadside  $\text{NO}_2$  level declined due to reduced emissions of  $\text{NO}_x$  much less than expected due to the increasing usage of diesel vehicles in Europe. According to the Emissions Inventory in 2021 (Table S5), 96.5% of  $\text{PM}_{2.5}$  vehicle exhaust emissions and 74% of  $\text{NO}_x$  emissions were attributable to diesel vehicles in Beijing.

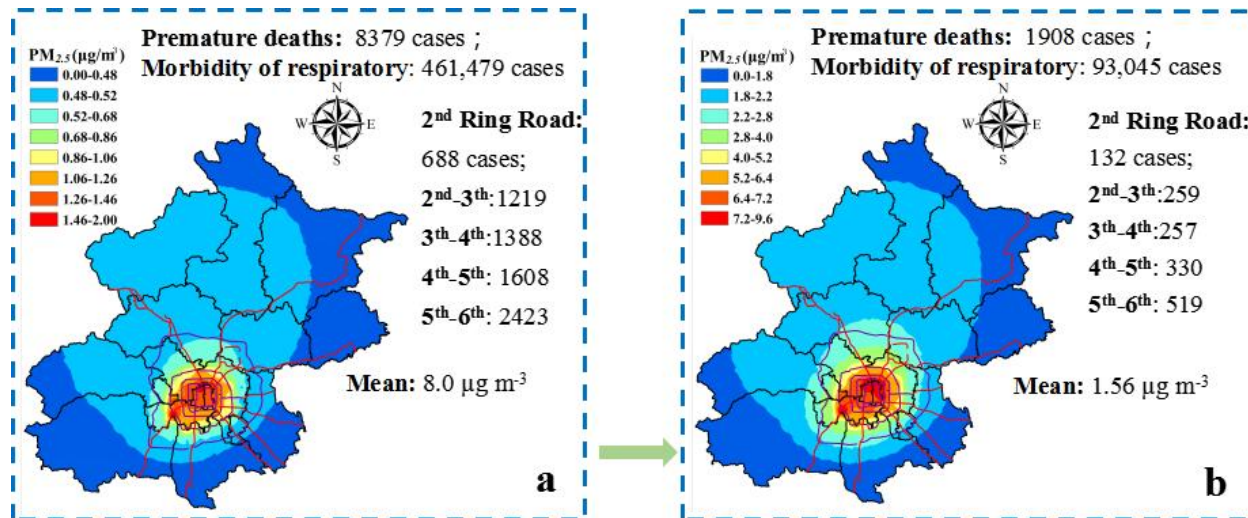


Figure 7 The spatial distribution of simulated  $\text{PM}_{2.5}$  concentration from vehicles in 2014 (a) and 2020 (b). The numbers in the figure refer to the annual average  $\text{PM}_{2.5}$  and the number of premature deaths in various zones.

## 5. Conclusion

In order to evaluate the characteristics of traffic-related air pollutants and consequent health impacts, this study collected hourly mass concentrations of  $\text{PM}_{2.5}$ ,  $\text{PM}_{10}$ ,  $\text{NO}_2$ ,  $\text{SO}_2$ ,  $\text{CO}$  and  $\text{O}_3$  from 5 traffic sites and 12 urban background sites in Beijing during 2014 to 2020. The roadside increments of air pollutants were obtained using a paired-site approach. As a result, the annual mean air pollutant concentrations are found to have diminished significantly by 47% to 71% from 2014 to 2020, specifically 4.4-20.0 %  $\text{a}^{-1}$  for  $\text{PM}_{10}$ , 2.4-22.9 %  $\text{a}^{-1}$  for  $\text{NO}_2$ , 10.2-39.7 %  $\text{a}^{-1}$  for  $\text{SO}_2$ , and 10.4-18.2 %  $\text{a}^{-1}$  for  $\text{CO}$ . On the contrary, the concentration of ozone increased by 17.4% during this period. In terms of the diurnal variation of air pollutants, a higher level of air pollutants was observed at nighttime than those at daytime due to a shallower mixed boundary layer and temperature. The air pollutant levels at South 3<sup>rd</sup> Ring Road station were higher than

460 those at the street stations and East 4<sup>th</sup> Ring Road station during 2014 to 2020, which could be  
461 affected by the air mass passage through the area south of Beijing with its dense industry.

462 The roadside increments and the percentage increase above urban background values of  
463 PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub> and CO decreased from 2014 to 2020, revealing that the implementation of new  
464 vehicle standards (China V and VI) led to reductions of air pollutants in the roadside  
465 environment, except for PM<sub>10</sub> and O<sub>3</sub>. Based on ADMS simulated PM<sub>2.5</sub> dispersion model  
466 concentrations from exhaust vehicle emissions, the premature deaths due to traffic-related PM<sub>2.5</sub>  
467 were estimated to be 8379 and 1908 cases in 2014 and 2020, respectively. The number of PM<sub>2.5</sub>-  
468 induced acute cases of respiratory diseases were 461479 cases in 2014 and 93045 cases in 2020,  
469 illustrating a more severe public health burden of traffic-related PM<sub>2.5</sub> than that reflected by  
470 mortality alone. The relative public health impacts of PM<sub>2.5</sub>, NO<sub>2</sub> and SO<sub>2</sub> were estimated  
471 roughly from their influence on all-cause mortality. It was found that the effect of NO<sub>2</sub> from road  
472 traffic on premature mortality was of a similar magnitude to that of PM<sub>2.5</sub>, and greater than that  
473 of SO<sub>2</sub>. It indicates that further reduction of NO<sub>x</sub> emitted from traffic will likely have substantial  
474 benefits for public health.

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