# Enhancement of secondary aerosol formation by reduced anthropogenic emissions during Spring Festival 2019 and enlightenment for regional PM<sub>2.5</sub> control in Beijing

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## 1 Abstract

2 A comprehensive field experiment measuring aerosol chemical and physical properties at a suburban site in Beijing around the 2019 Spring Festival was carried out 3 to investigate the impact of reduced anthropogenic emissions on aerosol formation. 4 Sharply reduced sulfur dioxide (SO<sub>2</sub>) and nitrogen dioxide (NO<sub>2</sub>) concentrations during 5 the festival holiday resulted in an unexpected increase in the surface ozone  $(O_3)$ 6 concentration caused by the strong O<sub>3</sub>-titration phenomenon. Simultaneously, the 7 reduced anthropogenic emissions resulted in massive decreases in particle number 8 concentration at all sizes and the mass concentrations of organics and black carbon. 9 However, the mass concentrations of inorganics (especially sulfate) decreased weakly. 10 Detailed analyses of the sulfur oxidation ratio and the nitrogen oxidation ratio suggest 11 that sulfate formation during the holiday could be promoted by enhanced nocturnal 12 aqueous-phase chemical reactions between SO<sub>2</sub> and O<sub>3</sub> under moderate relative 13 humidity (RH) conditions (40 % < RH < 80 %). Daytime photochemical reactions in 14 15 winter in Beijing mainly controlled nitrate formation, which was enhanced a little during the holiday. A regional analysis of air pollution patterns shows that the enhanced 16 formation of secondary aerosols occurred throughout the entire Beijing-Tian-Hebei 17 (BTH) region during the holiday, partly offsetting the decrease in particle matter with 18 an aerodynamic diameter less than 2.5 µm. Our results highlight the necessary control 19 of O<sub>3</sub> formation to reduce secondary pollution in winter under current emission 20 21 conditions.

## 23 1. Introduction

Aerosols consist of liquid and solid particles, and their mixture suspended in the 24 25 atmosphere. The massive increase in aerosol particles caused by human activities in urban areas (e.g., traffic, industrial production, and construction work) can deteriorate 26 air quality to the point of having a detrimental impact on human health (e.g., Chow et 27 al., 2006; Matus et al., 2012; Gao et al., 2017; Zhong et al., 2018; An et al., 2019). 28 Moreover, aerosols can change atmospheric optical and hygroscopic properties, altering 29 the transfer of solar radiation and the development of clouds, thereby changing weather 30 31 and climate in both aerosol source regions and their downstream areas (e.g., Altaratz et 32 al., 2014; R. Zhang et al., 2015; Z. Li et al., 2016, 2019; Y. Wang et al., 2018, 2019b; Jin et al., 2020). 33

With the rapid economic development and urbanization in recent decades in China, 34 the scales of many cities have expanded quickly along with sharply increased 35 populations in urban areas, especially in the three most economically developed regions 36 37 (the Beijing-Tianjin-Hebei (BTH) metropolitan region, the Yangtze River Delta, and the Pearl River Delta). As a result, air pollution has become a severe problem in these 38 39 megacity regions (e.g., Chan and Yao, 2008; Han et al., 2014; Zhong et al., 2018). On 40 some heavy haze days, the mass concentration of particulate matter with an aerodynamic diameter of less than 2.5 µm (PM<sub>2.5</sub>) dramatically increased from tens to 41 hundreds of micrograms per cubic meter in several hours (Guo et al., 2014; Sun et al., 42 43 2016a).

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Over the past a few years, many emission control measures have been taken in

45	China to mitigate air pollution. As a response, the mass concentration of $PM_{2.5}$ has
46	decreased in most cities in China since 2013, especially in the BTH region (Q. Zhang
47	et al., 2019; Vu et al., 2019; Zhai et al., 2019). Organics and black carbon (BC)
48	concentrations largely decreased during these years thanks to the reduction in coal
49	combustion and biomass burning (H. Li et al., 2019a; Xu et al., 2019). Simultaneously,
50	the mass concentrations of inorganics (mainly sulfate, nitrate, and ammonium) also
51	decreased due to the reduction in their gaseous precursors (especially sulfur dioxide, or
52	SO <sub>2</sub> ). However, the mass fraction of inorganics increased by more than 10 $\%$ during
53	these years (H. Li et al., 2019a; Y. Wang et al., 2019a), implying the enhancement of
54	secondary inorganic aerosol (SIA) formation, which partly counteracted the decrease
55	in PM <sub>2.5</sub> . All these variations would change aerosol physicochemical properties. For
56	example, Xie et al. (2020) found that the aerosol pH increased as $PM_{2.5}$ decreased in the
57	past few years in urban Beijing due to the enhanced mass ratio of nitrate to sulfate. As
58	a possible consequence of the elevated aerosol pH, the dissolved ozone $(O_3)$ in particles
59	would play a more important role to SIA formation, especially for sulfate formation
60	(Seinfeld and Pandis, 2016). Therefore, the major chemical processes during haze
61	events and the control target should be re-evaluated. Elaborating the secondary aerosol
62	formation mechanism under current emission conditions is important for taking more
63	proper measures to control PM <sub>2.5</sub> in the future.

Some studies have argued that controlling emissions of nitrogen oxides (NO<sub>x</sub>) is important because nitrate in  $PM_{2.5}$  has had the weakest decrease relative to other chemical species over the past several years (Q. Zhang et al., 2019; F. Zhang et al.,

2020). The transformation of  $NO_x$  to nitrate is closely related to atmospheric oxidation 67 processes (Seinfeld and Pandis, 2016). Surface ozone (O<sub>3</sub>) is an important secondary 68 69 gaseous pollutant and oxidizing agent in the atmosphere. Recent studies have found that a reduction in  $PM_{2.5}$  resulted in an increase in the O<sub>3</sub> volume mixing ratio ([O<sub>3</sub>]) at 70 71 a rate of 3.3 ppbv per annum during the summer of the past few years in the BTH region (K. Li et al., 2019, 2020). The increased [O<sub>3</sub>] can enhance the atmospheric oxidation 72 capacity, thereby promoting the formation of secondary aerosols in summer (T. Wang 73 et al., 2017). However, less emphasis has been placed on the variation in [O<sub>3</sub>] in winter. 74 75 The formation of O<sub>3</sub> and its effect on secondary aerosol formation in a cold environment is thus unclear. 76

Some special events held in China have provided unique opportunities to 77 78 investigate the impact of human activities on air quality by taking advantage of unusual changes associated with short-term, drastic measures implemented by the Chinese 79 government to reduce anthropogenic emissions, such as the 2008 Summer Olympic 80 81 Games (T. Wang et al., 2010; Guo et al., 2013), the 2014 Asia-Pacific Economic Cooperation (Sun et al., 2016b), the 2015 China Victory Day parade (Y. Wang et al., 82 83 2017; Zhao et al., 2017), and the 2016 G20 Summit (H. Li et al., 2019b). The annual Spring Festival holiday is also a special occasion when the vast majority of the 84 population stops working for 2 to 4 weeks (Tan et al., 2009; Y. Zhang et al., 2016; C. 85 Wang et al., 2017). Investigating the impact of changes in anthropogenic emissions on 86 87 gaseous pollutants and aerosol formation during these special occasions may provide useful guidance on more scientifically sound measures to take to control PM<sub>2.5</sub>. 88

A comprehensive aerosol field experiment at a suburban site near the 5th Ring 89 Road in the Daxing District of Beijing was carried out for more than two years, 90 91 including the 2019 Spring Festival. Beijing was one of the three top cities in China with the largest migrating population during the 2019 Spring Festival holiday 92 (https://cloud.tencent.com/developer/news/393324). In addition, fireworks were 93 prohibited throughout the Beijing metropolitan region within the 6th Ring of the Beijing 94 Beltway. The intensity of anthropogenic emissions was thus much weaker than usual 95 during this holiday. Our measurements around this period of the field campaign are thus 96 97 ideal for investigating the impact of reduced anthropogenic emissions on surface O<sub>3</sub> and aerosol formation. 98

99 This paper is structured as follows. Section 2 describes the experiment site and the 100 measurement data used in this study. Section 3 presents the results and discussion, 101 mainly concerning the impact of reduced anthropogenic emissions during the holiday 102 on the variations in trace gases and aerosol chemical species in Beijing and the BTH 103 region. Section 4 presents the conclusions and their implications.

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## 105 2. Experiment site and measurement data

A comprehensive field experiment measuring aerosol physical and chemical properties was conducted from August 2017 to October 2019 at a suburban site in southern Beijing (Fig. 1). Note that this study only employs measurements made around the 2019 Spring Festival from 16 January to 17 February. This site (39.81°N, 116.48°E) is the test center for meteorological instruments constructed by the China Meteorological Administration (CMA). It is surrounded by Beijing's 5th Ring Road, industrial parks, and residential communities (Fig. S1). Aerosol chemical and physical properties in this area are thus mainly anthropogenic, varying considerably around the time of the festival in response to the full cycle of industrial activities as the majority of people stopped and resumed working. This provides an opportunity to investigate the impact of reduced anthropogenic emissions on surface O<sub>3</sub> and aerosol formation processes in winter.

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Figure 1. Map showing the Beijing-Tianjin-Hebei region in China and the location of
the experiment site. The colored background shows the terrain height (unit: m above
sea level).

Table 1 lists the instruments used in this campaign. A scanning mobility particle 124 sizer (SMPS) and an aerodynamic particle sizer (APS) measured the aerosol particle 125 126 number size distribution (PNSD) from 10 nm to 20 µm. The SMPS consists of a differential mobility analyzer (model 3081, TSI Inc.) and a condensation particle 127 128 counter (model 3772, TSI Inc.). The aerodynamic diameter measured by the APS can be converted to the Stokes diameter through division by the square root of the aerosol 129 density. The aerosol density in this study was calculated following the method of Zhao 130 et al. (2017), using measured aerosol chemical composition information. An aerosol 131 chemical speciation monitor (ACSM) equipped with a PM<sub>2.5</sub> lens system, a capture 132 vaporizer, and a quadrupole mass spectrometer was used to measure mass 133 concentrations of non-refractory aerosol chemical species in PM<sub>2.5</sub>, including organics 134 (Org), nitrate  $(NO_3^-)$ , sulfate  $(SO_4^{2-})$ , ammonium  $(NH_4^+)$ , and chlorine (Chl) (Peck et al., 135 2016; Xu et al., 2017; Y. Zhang et al., 2017). A seven-wavelength aethalometer (model 136 137 AE-33, Magee Scientific Corp.) with a PM<sub>2.5</sub> cyclone in the sample inlet was used to 138 retrieve the mass concentration of BC.

In addition to the above aerosol measurements, meteorological parameters were observed by the CMA at the experiment site. The Chinese Ministry of Ecology and Environment network and Beijing Municipal Environmental Monitoring Center (http://106.37.208.233:20035/ and http://www.bjmemc.com.cn/) provided PM<sub>2.5</sub> and trace gas (sulfur dioxide (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), carbon monoxide (CO), and O<sub>3</sub>) measurements made in different locations of the BTH region. Yizhuang in Beijing is the nearest station to the experiment site (about 3.0 km to the southeast, Fig. S1). The

146 total mass concentrations of measured non-refractory aerosol chemical species and BC

147 mass concentrations in PM2.5 show good consistency with the PM2.5 mass

- 148 concentrations obtained from the Yizhuang station (Fig. S2).
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150 **Table 1**. Aerosol instruments used in this campaign and their observed parameters and

151 manufacturer information.

Instrument Measured Parameters		Manufacturer	Model	Time Resolution
SMPS	Particle number size distribution (10–550 nm)	TSI	3938	5 min
APS	Particle number size distribution (0.5–20 μm)	TSI	3321	5 min
ACSM	Mass concentrations of non-refractory aerosol chemical species in PM <sub>2.5</sub>	Aerodyne	Q-ACSM	15 min
Aethalometer	Mass concentration of black carbon	Magee	AE-33	5 min

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## 153 **3. Results and Discussion**

#### 154 **3.1. Basic meteorological and environmental characteristics**

155 While the official Spring Festival holiday was from 4 February to 10 February 2019, many people left before 3 February and came back after the Lantern Festival (19 156 February). In this study, we regarded the days from 16 January to 2 February as the 157 polluted (POL) period with high anthropogenic emissions, more representative of 158 ordinary conditions, and the days from 3 February to 17 February as the background 159 (BG) period with low anthropogenic emissions. Figure 2 shows the time series of 160 meteorological parameters, trace gas volume mixing ratios, and aerosol properties 161 162 during the two periods.



Figure 2. Time series of (a) ambient temperature (*T*) and relative humidity (RH), (b) wind direction (WD) and speed (WS), (c) volume mixing ratios of trace gases  $[O_3, SO_2, NO_2 \text{ and } O_x (O_3+NO_2)]$ , (d) the aerosol particle number size distribution measured by the SMPS, and (e) mass concentrations of aerosol chemical species in PM<sub>2.5</sub> measured by the ACSM and the AE-33. The trace gas information was from the Yizhuang station, and the others were observed at the experiment site in Beijing (16 January to 17 February 2019).

Ambient temperature (*T*) and relative humidity (RH) have clear diurnal cycles (Fig. 2a). The average *T* and RH during the BG period were slightly lower ( $-3.3\pm3.4$  versus 0.2±4.2°C) and higher ( $33.2\pm20.1$  versus 25.8±17.6 %) than those during the POL period, respectively. This was caused by several short-term light snowfall events that

occurred on 6, 12, and 14 February during the BG period. Figures 2b and S3 display similar wind patterns during the POL and BG periods, i.e., wind patterns that changed periodically. The prevailing, strong northerly winds during the two periods were beneficial to dispersing pollutants in Beijing (Sun et al., 2016b; Y. Wang et al., 2017), and thus no heavy haze episodes occurred during these periods. Overall, the meteorological parameters were similar during the POL and BG periods.

Figure 2c illustrates that the volume mixing ratios of SO<sub>2</sub> and NO<sub>2</sub> ([SO<sub>2</sub>] and 184 [NO<sub>2</sub>]) during the BG period were lower than those during the POL period, suggesting 185 186 less gaseous pollutants from anthropogenic emissions during the BG period. In addition, [O<sub>3</sub>] remained at a high level for several days during the BG period but not 187 during the POL period. The average [O<sub>3</sub>] increased by 77.4 % during the BG period 188 189 compared with the POL period (46.2±18.9 versus 26.1±22.2 ppbv). The percent change in [O<sub>3</sub>] due to the "holiday effect" during this field campaign is much higher than that 190 reported in other regions of China (K. Huang et al., 2012; C. Wang et al., 2017; S. Wang 191 192 et al., 2019). Figure 2c also shows a weak variation in  $O_x$  ( $O_3 + NO_2$ ) from the POL period to the BG period, indicating that strong O<sub>3</sub>-titration appeared during the Spring 193 Festival 2019. 194

Many bursts of fine particles (Fig. 2d) occurring mainly during rush hours or at night were observed during the POL period. This is likely related to the substantial increases in gasoline or diesel vehicles on two nearby roads at these times. Zhu et al. (2017) found that efficient nucleation and partitioning of gaseous species from on-road vehicles can promote new particle formation in the wintertime. However, this phenomenon occurred much less frequently during the BG period, likely because of the
massive reduction in on-road vehicles. The few short-term bursts of fine particles
during the BG period occurred during the daytime, presumably because of enhanced
nucleation by photochemical processes.

204 The aerosol chemical species in PM<sub>2.5</sub> also differed during the POL and BG periods (Fig. 2e). During the POL period, the mass concentrations of aerosol chemical species 205 readily accumulated, especially the organics  $(m_{\text{org}})$  with rapid increases at night. The 206 mass concentration of BC  $(m_{BC})$  also clearly increased, likely associated with an 207 208 increase in heavy-duty diesel vehicles and a decrease in the nocturnal planetary boundary layer at night (Y. Wang et al., 2017; Zhao et al., 2017; Z. Li et al., 2017). 209 210 However, the increases in  $m_{\text{org}}$  and  $m_{\text{BC}}$  at night during the BG period were not as strong 211 as those during the POL period. The mass concentration of nitrate  $(m_{NO3})$  largely decreased during the BG period, while there was a weak variation in the mass 212 concentration of sulfate  $(m_{SO4})$ . 213

In summary, distinct differences existed in all observed trace gases and aerosol chemical and physical parameters during the POL and BG periods. However, the meteorological parameters (wind direction and speed, ambient temperature, and RH) and weather regimes were similar during these two periods. This helps to single out the impact of reduced anthropogenic emissions on trace gases and aerosol formation processes.

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## 221 **3.2.** Impact of reduced anthropogenic emissions on aerosol formation processes

222	The average $PM_{2.5}$ mass concentrations were 46.3 and 22.5 $\mu$ g/m <sup>3</sup> during the POL
223	and BG periods, respectively. Figure 3 illustrates the average PNSD and aerosol
224	chemical species in PM <sub>2.5</sub> during the two periods. The particle number concentrations
225	at all sizes were much higher during the POL period than during the BG period,
226	especially for ultrafine particles (with diameters, or $D_{\rm p}$ , < 100 nm). The diurnal
227	variation in PNSD during the POL period shown in Fig. 4a suggests that aerosol
228	particles with $D_p < 50$ nm burst during rush hours and in the nighttime. The total particle
229	number concentration (N) remained greater than $30,000 \text{ cm}^{-3}$ at these times. However,
230	during the BG period, the number concentration of ultrafine particles only increased
231	weakly during rush hours or nucleation times. $N$ was always less than 20,000 cm <sup>-3</sup> on
232	all days during the BG period (Fig. 4b), probably linked with the reduction in on-road
233	vehicles during the holiday. As shown in Table 2, the ratio of BG to POL 10-50 nm
234	particle number concentrations ( $N_{10-50 \text{ nm}}$ ) (0.47) is much smaller than the ratios for
235	larger particles (0.78 for $N_{50-100 \text{ nm}}$ and 0.67 for $N_{>100 \text{ nm}}$ ). These all demonstrate the
236	strong impact of reduced anthropogenic emissions on aerosol number concentrations,
237	especially for nucleation-mode and small Aitken-mode particles.



Figure 3. Average aerosol particle number size distributions (red and blue curves) and mass fractions of aerosol chemical species in  $PM_{2.5}$  (pie charts with red and blue outlines) during the POL (in red) and BG (in blue) periods.

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Figure 4. Diurnal variations in aerosol particle number size distribution (colored
background) and total aerosol number concentration (*N*, shown as grey curves) during
the (a) POL and (b) BG periods.

249	<b>Table 2.</b> Summary of the average aerosol number concentration $(N)$ in different size
250	ranges, volume mixing ratios of trace gases, mass concentrations of $PM_{2.5}$ and different
251	aerosol chemical species, sulfur oxidation ratios (SOR), and nitrogen oxidation ratios
252	(NOR) during the POL and BG periods and their ratios.

	$N_{10-50 \text{ nm}}$	N50-100 nm	<i>N</i> >100 nm	$SO_2$	NO <sub>2</sub>	O3	PM <sub>2.5</sub>
	(cm <sup>-3</sup> )	(cm <sup>-3</sup> )	(cm <sup>-3</sup> )	(ppbv)	(ppbv)	(ppbv)	$(\mu g/m^3)$
POL	20,861±19,935	3,946±2,544	3,888±2,757	8.31±6.35	51.96±27.35	26.06±22.24	46.32±39.05
BG	9,837±8,493	3,071±1,478	2,600±2,223	4.85±3.83	21.87±13.99	46.23±18.86	22.52±20.28
BG/POL	0.47	0.78	0.67	0.58	0.42	1.77	0.49
	MOrg	<i>m</i> NO3	mso4	<i>m</i> NH4	твс	SOD	NOD
_	(µg/m <sup>3</sup> )	$(\mu g/m^3)$	$(\mu g/m^3)$	$(\mu g/m^3)$	$(\mu g/m^3)$	SOK	NOK
POL	23.55±19.58	8.25±7.91	4.59±6.20	3.96±3.83	$5.05 \pm 4.51$	0.27±0.17	$0.09 \pm 0.08$
BG	10.17±9.13	4.09±4.25	$3.82 \pm 4.08$	2.18±2.14	1.91±1.74	0.32±0.18	$0.10{\pm}0.08$
BG/POL	0.43	0.50	0.83	0.55	0.38	1.19	1.11

<sup>253</sup> 

Table 2 also indicates that the mass concentrations (m) of aerosol chemical species 254 in  $PM_{2.5}$  were much less during the BG period than during the POL period . The m 255 related to primary emissions ( $m_{\text{org}}$  and  $m_{\text{BC}}$ ) decreased by more than 50 %, but the *m* of 256 secondary inorganic aerosols (SIA, including nitrate, sulfate, and ammonium) slightly 257 decreased, especially sulfate (m<sub>SO4</sub> decreased by only 17 %). The pie charts in Fig. 3 258 show significant differences in the aerosol chemical species of PM<sub>2.5</sub> during the two 259 260 periods. The mass fractions of Org and BC were lower during the BG period (45 % and 8 %, respectively) than during the POL period (51 % and 11 %, respectively). By 261 contrast, the mass fraction of SIA was higher during the BG period (45 %) than during 262 the POL period (37 %). This indicates that the strongly reduced 263 anthropogenic emissions during the holiday caused sharp decreases in primary aerosols 264 but not secondary aerosols. The sulfur oxidation ratio (SOR) and nitrogen oxidation 265 266 ratio (NOR) are usually calculated to study the transformation of secondary aerosols

267	(Sun et al., 2006; Y. Li et al., 2017). SOR (NOR) is defined as the ratio of the molar
268	concentration of sulfate (nitrate) to the total molar concentration of sulfate (nitrate) and
269	SO <sub>2</sub> (NO <sub>2</sub> ). Table 2 shows that SOR and NOR were higher during the BG period than
270	during the POL period, suggesting that the formation of secondary inorganics was
271	enhanced during the BG period. Figure 5 shows that most large $PM_{2.5}$ mass
272	concentrations (> 100 $\mu$ g/m <sup>3</sup> ) during the POL period occurred along with low RH (<
273	40 %) and low SIA mass fractions, indicating the important contribution of primary
274	emissions to the accumulation of $PM_{2.5}$ in a polluted environment. However, large
275	$PM_{2.5}$ mass concentrations (> 50 µg/m <sup>3</sup> ) during the BG period mainly appeared under
276	moderate RH ( $40 < RH < 80$ %) and high SIA mass fraction conditions, likely caused
277	by enhanced aqueous-phase chemical reactions during this period.



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Figure 5. The variation in secondary inorganic aerosols (SIA) mass fraction in PM<sub>2.5</sub>
as a function of ambient relative humidity during the POL (in red) and BG (in blue)
periods. The different circle sizes denote different PM<sub>2.5</sub> mass concentrations.

284 The diurnal variation in [O<sub>3</sub>] (Fig. 6a) shows more accumulated O<sub>3</sub> during the BG period than during the POL period at any time of the day. In particular, [O<sub>3</sub>] at night 285 was two times higher during the BG period than during the POL period (Fig. 6b). 286 Distinct diurnal variations in SOR were found during the two periods (Fig. 6c). The 287 higher SOR at night during the BG period (Fig. 6c) indicates the enhanced 288 transformation of SO<sub>2</sub> to sulfate, likely related to nocturnal aqueous-phase chemical 289 reactions due to the elevated ambient RH at night (Fig. S4). Figure S5 indicates that 290 SOR increased following an increase in ALWC when the ambient RH was higher than 291

292	$\sim$ 40 % during the BG period. Moreover, Fig. 6d shows that the diurnal variation in the
293	SOR ratio during the two periods was similar to that of the [O <sub>3</sub> ] ratio. This suggests that
294	sulfate formation during the holiday was likely enhanced by nocturnal aqueous-phase
295	chemical reactions between $SO_2$ and $O_3$ . This is consistent with the study of Fang et al.
296	(2019), which found that ambient RH and the O <sub>3</sub> concentration are two prerequisites
297	for rapid sulfate formation via aqueous-phase oxidation reactions. This result highlights
298	that controlling O <sub>3</sub> formation under current emission conditions in winter in Beijing is
299	key to further reducing the formation of sulfate and implies that the high
300	underestimation of sulfate at night in models (Miao et al., 2020) could be caused by an
301	inaccurate simulation of [O <sub>3</sub> ]. The higher daytime NOR (Fig. 6e) than nighttime NOR
302	during the two periods illustrates that the formation of nitrate was mainly controlled by
303	photochemical reactions in winter. Figure 6f shows that the larger NOR difference (the
304	higher NOR ratio) during rush hours during the two periods likely occurred because a
305	mass of emitted NO <sub>x</sub> during rush hours could not be transformed to nitrate during the
306	POL period. Figure 6e and 6f also suggests nitrate formation was somewhat enhanced
307	during the holiday likely due to enhanced daytime photochemical reactions.



Figure 6. Diurnal variations in (a and b) O<sub>3</sub> volume mixing ratio and its ratio, (c and
d) sulfur oxidation ratio (SOR) and its ratio, and (e and f) nitrogen oxidation ratio
(NOR) and its ratio during the BG and POL periods. The ratio of a quantity is that
quantity during the BG period divided by that quantity during the POL period.



## **3.3.** Impact of reduced anthropogenic emissions on regional air pollution







Figure 7. Percent changes in trace gas volume mixing ratios (NO<sub>2</sub>, SO<sub>2</sub>, CO, and O<sub>3</sub>),

the PM<sub>2.5</sub> mass concentration, and the PM<sub>2.5</sub>/CO ratio during the BG period relative to the POL period:  $100 \times \left(\frac{[BG]-[POL]}{[POL]}\right)$  in the Beijing-Tianjin-Hebei (BTH) region.

The PM<sub>2.5</sub> mass concentration decreased at a much lower rate relative to the decrease in [NO<sub>2</sub>] and [SO<sub>2</sub>] in most cities during the BG period (Fig. 7e), while it

Xingtai, and Handan). The ratio PM <sub>2.5</sub> /CO is an indicator of aerosol second formation to primary emissions. An increase in PM <sub>2.5</sub> /CO was found dur period at all cities except for three coastal cities (Tangshan, Tianjin, and revealing the regional enhancement of secondary aerosol formation durin holiday. The weak decrease in PM <sub>2.5</sub> /CO at the three coastal cities was li	ondary ring the BG Cangzhou), ng the kely due to
formation to primary emissions. An increase in PM <sub>2.5</sub> /CO was found dur period at all cities except for three coastal cities (Tangshan, Tianjin, and revealing the regional enhancement of secondary aerosol formation durin holiday. The weak decrease in PM <sub>2.5</sub> /CO at the three coastal cities was li	ring the BG Cangzhou), ng the kely due to
<ul> <li>period at all cities except for three coastal cities (Tangshan, Tianjin, and</li> <li>revealing the regional enhancement of secondary aerosol formation durin</li> <li>holiday. The weak decrease in PM<sub>2.5</sub>/CO at the three coastal cities was li</li> </ul>	Cangzhou), ng the kely due to
<ul> <li>revealing the regional enhancement of secondary aerosol formation during</li> <li>holiday. The weak decrease in PM<sub>2.5</sub>/CO at the three coastal cities was li</li> </ul>	ng the kely due to
holiday. The weak decrease in $PM_{2.5}/CO$ at the three coastal cities was li	kely due to
342 the influence of mixed sea flows.	
343 The regional analysis of air pollution assumes that the findings from	1 Beijing
344 presented in section 3.2 are applicable to the entire BTH region. Regiona	ally reduced
345 anthropogenic emissions resulted in sharply decreased gaseous pollutant	is and
increased O <sub>3</sub> . The enhanced formation of secondary aerosols likely due t	to enhanced
347 aqueous chemical reactions involving O <sub>3</sub> could counteract the decrease i	n PM <sub>2.5</sub> mass
348 concentration. There are two possible reasons explaining the high $[O_3]$ d	uring the
holiday: (1) the reduced gaseous precursors ( $NO_x$ and $SO_2$ ) weakened th	e
350 consumption of O <sub>3</sub> , and (2) O <sub>3</sub> formation in the BTH region is likely vol	atile organic
351 compound (VOC)-controlled under current emission conditions, therefore	re the
reduction in $NO_x$ would lead to higher [O <sub>3</sub> ]. All these results demonstrat	e that it is
353 important to reduce $O_3$ formation to further control $PM_{2.5}$ in winter in the	e BTH region.
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356	
357 4. Conclusions and Implications	

358 In recent years, the mass concentration of particulate matter with an aerodynamic

diameter of less than 2.5 µm (PM<sub>2.5</sub>) has shown a general decreasing trend, presumably 359 due to the series of emission reduction measures taken in China in an attempt to improve 360 361 air quality. However, haze pollution episodes still occur from time to time, including during some special events when primary emissions reduced drastically, such as the 362 Chinese New Year holiday and even during the COVID-19 lockdown when 363 anthropogenic activities diminished dramatically (X. Huang et al., 2020). We conjecture 364 that reductions through primary emissions may be offset by increases in the formation 365 of secondary aerosols. 366

367 To test this, we examined the secondary aerosol formation mechanism in a comprehensive field experiment conducted in Beijing. Aerosol and meteorological 368 measurements were made for more than two years, but data around the 2019 Chinese 369 370 Spring Festival from 16 January to 17 February were employed in this study to single out the impact of emission reductions due to the holiday. The study period was divided 371 into polluted (POL) and background (BG) periods, with high and low anthropogenic 372 373 emissions before and during the festival holiday, respectively. Investigated were the impacts of reduced anthropogenic emissions on trace gases and PM2.5 under similar 374 375 meteorological conditions.

The average  $PM_{2.5}$  mass concentrations were 46.3 and 22.5 µg/m<sup>3</sup> during the POL and BG periods, respectively, with no heavy haze events occurring. The average aerosol particle number size distribution shows that the reduced anthropogenic emissions during the holiday led to decreased aerosol number concentrations at all sizes, especially in the nucleation and Aitken modes (mobility diameters less than 50 nm).

Simultaneously, the reduced anthropogenic emissions resulted in decreases in the 381 volume mixing ratios of SO<sub>2</sub> and NO<sub>2</sub> and an unexpected increase in the volume mixing 382 383 ratio of  $O_3$  [O<sub>3</sub>] during the BG period caused by the appeared O<sub>3</sub>-titration phenomenon. The analysis of the aerosol chemical species in PM<sub>2.5</sub> demonstrates that the large 384 decreases in organics and black carbon mass concentrations during the BG period were 385 likely caused by the large decrease in on-road vehicles. Moreover, the mass 386 concentration of nitrate also decreased while that of sulfate decreased much less during 387 the BG period. Comparisons of the sulfur oxidation ratio (SOR) and the nitrogen 388 389 oxidation ratio (NOR) during the two periods imply that the transformation of gaseous precursors to secondary inorganics (especially the transformation of SO<sub>2</sub> to sulfate) was 390 promoted during the BG period, likely due to the enhanced aqueous chemical reactions 391 392 involving the dissolved O<sub>3</sub>. The diurnal variation in the SOR ratio between the BG and POL periods was similar to that of the [O<sub>3</sub>] ratio, illustrating that sulfate formation was 393 promoted by the enhanced nocturnal aqueous-phase chemical reactions between SO<sub>2</sub> 394 and  $O_3$  under moderate relative humidity (RH) conditions (40 % < RH < 80 %). The 395 higher NOR in the daytime during the two periods points out that the formation of 396 397 nitrate was mainly controlled by photochemical reactions and weakly affected by the increase in [O<sub>3</sub>]. 398

This study also investigated the impact of reduced anthropogenic emissions on regional air pollution patterns during the holiday. The variation trends of trace gases in most cities in the Beijing-Tian-Hebei (BTH) region were similar to those in Beijing, indicating the regional influence of reduced anthropogenic emissions on the volume 403 mixing ratios of trace gases during the holiday. The weak  $PM_{2.5}$  variation and the 404 increased  $PM_{2.5}/CO$  ratio (an indicator of aerosol secondary formation to primary 405 emissions) during the BG period both suggest that the enhanced formation of secondary 406 aerosols offset the regional decrease in  $PM_{2.5}$  during the holiday.

Our findings provide evidence that decreases in anthropogenic emissions can promote the formation of secondary inorganics due to the enhancement of aqueous reactions likely caused by the increased  $[O_3]$ . This result implies that haze mitigation in north China needs a coordinated and balanced strategy for controlling multiple pollutants.

412 413

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*Data availability.* Data from the Chinese Ministry of Ecology and Environment network and Beijing Municipal Environmental Monitoring Center can be downloaded from the websites given in the main text. The measurement data from the field experiment used in this study are available from the first author upon request (yuyingwang@nuist.edu.cn).

425

426 *Author contributions*. ZL and PY designed the field experiment. YW and ZL

427 conceived the study and led the overall scientific questions. YW, QW, and XJ

428	processed the measurement data and prepared this paper. MC copyedited the article.
429	Other co-authors participated in the implementation of this experiment and the
430	discussion of this paper.
431	
432	Competing interests. The authors declare that they have no conflict of interest.
433	
434	References
40E	
435	Alternate O. Komm, I. Domon, I. A. and Himsh, E. Davierry, Claudiania antice hereare also accurling
430	Altaratz, O., Koren, I., Remer, L. A., and Hirsch, E.: Review: Cloud invigoration by aerosols—coupling
437	between microphysics and dynamics, Atmos. Res., $140$ , $38-60$ ,
430	An Z Huang P Zhang P Tio X Li G Cao I Zhou W Shi Z Han X Gu Z and li X:
439	An, Z., Huang, K., Zhang, K., He, X., Li, G., Cao, J., Zhou, W., Shi, Z., Han, T., Gu, Z., and Ji, T.,
440 1/1	Natl Acad Sci U.S.A. 116 8657 https://doi.org/10.1073/pnas.1900125116.2019
112	Chan C K and Yao X : Air pollution in magacities in China Atmos Environ $42$ 1-42
442	https://doi.org/10.1016/i.atmoseny.2007.09.003.2008
444	Chow, J. C., Watson, J. G., Mauderly, J. L., Costa, D. L., Wyzga, R. E., Vedal, S., Hidy, G. M., Altshuler,
445	S. L., Marrack, D., Heuss, J. M., Wolff, G. T., Arden Pope III, C., and Dockery, D. W.: Health effects
446	of fine particulate air pollution: lines that connect. J. Air Waste Manage., 56, 1368–1380.
447	https://doi.org/10.1080/10473289.2006.10464545, 2006.
448	Fang, Y., Ye, C., Wang, J., Wu, Y., Hu, M., Lin, W., Xu, F., and Zhu, T.: Relative humidity and O <sub>3</sub>
449	concentration as two prerequisites for sulfate formation, Atmos. Chem. Phys., 19, 12,295–12,307,
450	https://doi.org/10.5194/acp-19-12295-2019, 2019.
451	Gao, J., Woodward, A., Vardoulakis, S., Kovats, S., Wilkinson, P., Li, L., Xu, L., Li, J., Yang, J., Li, J.,
452	Cao, L., Liu, X., Wu, H., and Liu, Q.: Haze, public health and mitigation measures in China: a review
453	of the current evidence for further policy response, Sci. Total Environ., 578, 148-157,
454	https://doi.org/10.1016/j.scitotenv.2016.10.231, 2017.
455	Guo, S., Hu, M., Guo, Q., Zhang, X., Schauer, J. J., and Zhang, R.: Quantitative evaluation of emission
456	controls on primary and secondary organic aerosol sources during Beijing 2008 Olympics, Atmos.
457	Chem. Phys., 13, 8303-8314, https://doi.org/10.5194/acp-13-8303-2013, 2013.
458	Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng, L.,
459	Molina, M. J., and Zhang, R.: Elucidating severe urban haze formation in China, Proc. Natl. Acad.
460	Sci. U.S.A., 111, 17373, https://doi.org/10.1073/pnas.1419604111, 2014.
461	Han, L., Zhou, W., Li, W., and Li, L.: Impact of urbanization level on urban air quality: a case of fine
462	particles $(PM_{2.5})$ in Chinese cities, Environ. Pollut., 194, 163–170,
463	https://doi.org/10.1016/j.envpol.2014.07.022, 2014.
464	Huang, K., Zhuang, G., Lin, Y., Wang, Q., Fu, J. S., Zhang, R., Li, J., Deng, C., and Fu, Q.: Impact of
465	anthropogenic emission on air quality over a megacity revealed from an intensive atmospheric

- 466 campaign during the Chinese Spring Festival, Atmos. Chem. Phys., 12, 11,631–11,645,
   467 https://doi.org/10.5194/acp-12-11631-2012, 2012.
- Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X.,
  Xu, Z., Chen, L., Li, Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q.,
  Liu, B., Chai, F., Davis, S. J., Zhang, Q. and He, K.: Enhanced secondary pollution offset reduction
  of primary emissions during COVID-19 lockdown in China, National Science Review,

472 https://doi.org/ 10.1093/nsr/nwaa137, 2020.

- Jin, X., Wang, Y., Li, Z., Zhang, F., Xu, W., Sun, Y., Fan, X., Chen, G., Wu, H., Ren, J., Wang, Q., and
  Cribb, M.: Significant contribution of organics to aerosol liquid water content in winter in Beijing,
  China, Atmos. Chem. Phys., 20, 901–914, https://doi.org/10.5194/acp-20-901-2020, 2020.
- Li, H., Cheng, J., Zhang, Q., Zheng, B., Zhang, Y., Zheng, G., and He, K.: Rapid transition in winter
  aerosol composition in Beijing from 2014 to 2017: response to clean air actions, Atmos. Chem. Phys.,
  19, 11,485–11,499, https://doi.org/10.5194/acp-19-11485-2019, 2019a.
- Li, H., Wang, D., Cui, L., Gao, Y., Huo, J., Wang, X., Zhang, Z., Tan, Y., Huang, Y., Cao, J., Chow, J.
  C., Lee, S., and Fu, Q.: Characteristics of atmospheric PM<sub>2.5</sub> composition during the implementation of stringent pollution control measures in shanghai for the 2016 G20 summit, Sci. Total Environ., 648, 1121–1129, https://doi.org/10.1016/j.scitotenv.2018.08.219, 2019b.
- Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., and Bates, K. H.: Anthropogenic drivers of 2013–
  2017 trends in summer surface ozone in China, P. Natl. Acad. Sci. U.S.A., 116, 422–427,
  https://doi.org/10.1073/pnas.1812168116, 2019.
- Li, K., Jacob, D. J., Shen, L., Lu, X., De Smedt, I., and Liao, H.: 2013–2019 increases of surface ozone
  pollution in China: anthropogenic and meteorological influences, Atmos. Chem. Phys. Discuss., 2020,
  1–18, https://doi.org/10.5194/acp-2020-298, 2020.
- Li, Y. J., Sun, Y., Zhang, Q., Li, X., Li, M., Zhou, Z., and Chan, C. K.: Real-time chemical
  characterization of atmospheric particulate matter in China: a review, Atmos. Environ., 158, 270–304,
  https://doi.org/10.1016/j.atmosenv.2017.02.027, 2017.
- Li, Z., Lau, W. K. M., Ramanathan, V., Wu, G., Ding, Y., Manoj, M. G., Liu, J., Qian, Y., Li, J., Zhou,
  T., Fan, J., Rosenfeld, D., Ming, Y., Wang, Y., Huang, J., Wang, B., Xu, X., Lee, S. S., Cribb, M.,
- Zhang, F., Yang, X., Zhao, C., Takemura, T., Wang, K., Xia, X., Yin, Y., Zhang, H., Guo, J., Zhai, P.
  M., Sugimoto, N., Babu, S. S., and Brasseur, G. P.: Aerosol and monsoon climate interactions over
  Asia, Rev. Geophys., 54, 866–929, https://doi.org/10.1002/2015RG000500, 2016.
- Li, Z., Guo, J., Ding, A., Liao, H., Liu, J., Sun, Y., Wang, T., Xue, H., Zhang, H., and Zhu, B.: Aerosol
  and boundary-layer interactions and impact on air quality, Natl. Sci. Rev., 4, 810–833,
  https://doi.org/10.1093/nsr/nwx117, 2017.
- 500 Li, Z., Wang, Y., Guo, J., Zhao, C., Cribb, M. C., Dong, X., Fan, J., Gong, D., Huang, J., Jiang, M., Jiang,
- 501 Y., Lee, S. S., Li, H., Li, J., Liu, J., Qian, Y., Rosenfeld, D., Shan, S., Sun, Y., Wang, H., Xin, J., Yan,
- X., Yang, X., Yang, X., Zhang, F., and Zheng, Y.: East Asian Study of Tropospheric Aerosols and
  their Impact on Regional Clouds, Precipitation, and Climate (EAST-AIRCPC), J. Geophys. Res.
- 504 Atmos., 124, 13,026–13,054, https://doi.org/10.1029/2019JD030758, 2019.
- Matus, K., Nam, K., Selin, N. E., Lamsal, L. N., Reilly, J. M., and Paltsev, S.: Health damages from air
  pollution in China, Global Environ. Change, 22, 55–66,
  https://doi.org/10.1016/j.gloenvcha.2011.08.006, 2012.
- 508 Miao, R., Chen, Q., Zheng, Y., Cheng, X., Sun, Y., Palmer, P. I., Shrivastava, M., Guo, J., Zhang, Q.,
- Liu, Y., Tan, Z., Ma, X., Chen, S., Zeng, L., Lu, K., and Zhang, Y.: Model bias in simulating major

- 510 chemical components of  $PM_{2.5}$  in China, Atmos. Chem. Phys. Discuss., 2020, 1–33, 511 https://doi.org/10.5194/acp-2020-76, 2020.
- Peck, J., Gonzalez, L. A., Williams, L. R., Xu, W., Croteau, P. L., Timko, M. T., Jayne, J. T., Worsnop,
  D. R., Miake-Lye, R. C., and Smith, K. A.: Development of an aerosol mass spectrometer lens system
- 514 for PM<sub>2.5</sub>, Aerosol Sci. Tech., 50, 781–789, https://doi.org/10.1080/02786826.2016.1190444, 2016.
- Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: from air pollution to climate
  change, edited, John Wiley & Sons, 2016.
- Sun, Y., Zhuang, G., Tang, A., Wang, Y., and An, Z.: Chemical characteristics of PM<sub>2.5</sub> and PM<sub>10</sub> in
  haze-fog episodes in Beijing, Environ. Sci. Technol., 40, 3148–3155,
  https://doi.org/10.1021/es051533g, 2006.
- Sun, Y., Chen, C., Zhang, Y., Xu, W., Zhou, L., Cheng, X., Zheng, H., Ji, D., Li, J., Tang, X., Fu, P., and
  Wang, Z.: Rapid formation and evolution of an extreme haze episode in Northern China during winter
  2015, Sci. Rep.-UK, 6, https://doi.org/10.1038/srep27151, 2016a.
- 523 Sun, Y., Wang, Z., Wild, O., Xu, W., Chen, C., Fu, P., Du, W., Zhou, L., Zhang, O., Han, T., Wang, Q., 524 Pan, X., Zheng, H., Li, J., Guo, X., Liu, J., and Worsnop, D. R.: "APEC Blue" : secondary aerosol 525 reductions from emission controls in Beijing, Sci. Rep.-UK, 6, 20668, 526 https://doi.org/10.1038/srep20668, 2016b.
- Tan, P., Chou, C., Liang, J., Chou, C. C. K., and Shiu, C.: Air pollution "holiday effect" resulting
  from the Chinese New Year, Atmos. Environ., 43, 2114–2124,
  https://doi.org/10.1016/j.atmosenv.2009.01.037, 2009.
- Vu, D., Gao, S., Berte, T., Kacarab, M., Yao, Q., Vafai, K., and Asa-Awuku, A.: External and internal
  cloud condensation nuclei (CCN) mixtures: controlled laboratory studies of varying mixing states,
  Atmos. Meas. Tech., 12, 4277–4289, https://doi.org/10.5194/amt-12-4277-2019, 2019.
- Wang, C., Huang, X. F., Zhu, Q., Cao, L. M., Zhang, B., and He, L. Y.: Differentiating local and regional
  sources of Chinese urban air pollution based on the effect of the Spring Festival, Atmos. Chem. Phys.,
  17, 9103–9114, https://doi.org/10.5194/acp-17-9103-2017, 2017.
- 536 Wang, S., Yu, R., Shen, H., Wang, S., Hu, Q., Cui, J., Yan, Y., Huang, H., and Hu, G.: Chemical 537 characteristics, sources, and formation mechanisms of PM<sub>2.5</sub> before and during the Spring Festival in 538 coastal city in Southeast China, Environ. Pollut., 251, 442-452, а 539 https://doi.org/10.1016/j.envpol.2019.04.050, 2019.
- Wang, T., Nie, W., Gao, J., Xue, L. K., Gao, X. M., Wang, X. F., Qiu, J., Poon, C. N., Meinardi, S.,
  Blake, D., Wang, S. L., Ding, A. J., Chai, F. H., Zhang, Q. Z., and Wang, W. X.: Air quality during
  the 2008 Beijing Olympics: secondary pollutants and regional impact, Atmos. Chem. Phys., 10, 7603–
  7615, https://doi.org/10.5194/acp-10-7603-2010, 2010.
- Wang, T., Xue, L., Brimblecombe, P., Lam, Y. F., Li, L., and Zhang, L.: Ozone pollution in China: a
  review of concentrations, meteorological influences, chemical precursors, and effects, Sci. Total
  Environ., 575, 1582–1596, https://doi.org/10.1016/j.scitotenv.2016.10.081, 2017.
- Wang, Y., Zhang, F., Li, Z., Tan, H., Xu, H., Ren, J., Zhao, J., Du, W., and Sun, Y.: Enhanced
  hydrophobicity and volatility of submicron aerosols under severe emission control conditions in
  Beijing, Atmos. Chem. Phys., 17, 5239–5251, https://doi.org/10.5194/acp-17-5239-2017, 2017.
- Wang, Y., Li, Z., Zhang, Y., Du, W., Zhang, F., Tan, H., Xu, H., Fan, T., Jin, X., Fan, X., Dong, Z.,
  Wang, Q., and Sun, Y.: Characterization of aerosol hygroscopicity, mixing state, and CCN activity at
  a suburban site in the central North China Plain, Atmos. Chem. Phys., 18, 11,739–11,752,
  https://doi.org/10.5194/acp-18-11739-2018, 2018.

- Wang, Y., Chen, J., Wang, Q., Qin, Q., Ye, J., Han, Y., Li, L., Zhen, W., Zhi, Q., Zhang, Y., and Cao,
  J.: Increased secondary aerosol contribution and possible processing on polluted winter days in China,
  Environ. Int., 127, 78–84, https://doi.org/10.1016/j.envint.2019.03.021, 2019a.
- Wang, Y., Li, Z., Zhang, R., Jin, X., Xu, W., Fan, X., Wu, H., Zhang, F., Sun, Y., Wang, Q., Cribb, M.,
  and Hu, D.: Distinct ultrafine- and accumulation-mode particle properties in clean and polluted urban
  environments, Geophys. Res. Lett., 46, 10,918–10,925, 10.1029/2019GL084047, 2019b.
- Xie, Y., Wang, G., Wang, X., Chen, J., Chen, Y., Tang, G., Wang, L., Ge, S., Xue, G., Wang, Y., and
  Gao, J.: Nitrate-dominated PM<sub>2.5</sub> and elevation of particle pH observed in urban Beijing during the
  winter of 2017, Atmos. Chem. Phys., 20, 5019-5033, 10.5194/acp-20-5019-2020, 2020.
- Xu, W., Croteau, P., Williams, L., Canagaratna, M., Onasch, T., Cross, E., Zhang, X., Robinson, W., 563 564 Worsnop, D., and Jayne, J.: Laboratory characterization of an aerosol chemical speciation monitor 565 with  $PM_{2.5}$ measurement capability, Aerosol Sci. Tech., 51, 69-83, 566 https://doi.org/10.1080/02786826.2016.1241859, 2017.
- Xu, W., Sun, Y., Wang, Q., Zhao, J., Wang, J., Ge, X., Xie, C., Zhou, W., Du, W., Li, J., Fu, P., Wang,
  Z., Worsnop, D. R., and Coe, H.: Changes in aerosol chemistry from 2014 to 2016 in winter in Beijing:
  insights from high-resolution aerosol mass spectrometry, J. Geophys. Res. Atmos., 124, 1132–1147,
  https://doi.org/10.1029/2018JD029245, 2019.
- Zhai, S., Jacob, D. J., Wang, X., Shen, L., Li, K., Zhang, Y., Gui, K., Zhao, T., and Liao, H.: Fine
  particulate matter (PM<sub>2.5</sub>) trends in China, 2013–2018: separating contributions from anthropogenic
  emissions and meteorology, Atmos. Chem. Phys., 19, 11,031–11,041, https://doi.org/10.5194/acp19-11031-2019, 2019.
- Zhang, F., Wang, Y., Peng, J., Chen, L., Sun, Y., Duan, L., Ge, X., Li, Y., Zhao, J., Liu, C., Zhang, X.,
  Zhang, G., Pan, Y., Wang, Y., Zhang, A. L., Ji, Y., Wang, G., Hu, M., Molina, M. J., and Zhang, R.:
  An unexpected catalyst dominates formation and radiative forcing of regional haze, Proc. Natl. Acad.
  Sci. U.S.A., 117, 3960, https://doi.org/10.1073/pnas.1919343117, 2020.
- 579 Zhang, Q., Zheng, Y., Tong, D., Shao, M., Wang, S., Zhang, Y., Xu, X., Wang, J., He, H., Liu, W., Ding,
- 580 Y., Lei, Y., Li, J., Wang, Z., Zhang, X., Wang, Y., Cheng, J., Liu, Y., Shi, Q., Yan, L., Geng, G.,
  581 Hong, C., Li, M., Liu, F., Zheng, B., Cao, J., Ding, A., Gao, J., Fu, Q., Huo, J., Liu, B., Liu, Z., Yang,
- 582 F., He, K., and Hao, J.: Drivers of improved PM<sub>2.5</sub> air quality in China from 2013 to 2017, P. Natl.
  583 Acad. Sci. U.S.A., 116, 24,463–24,469, https://doi.org/10.1073/pnas.1907956116, 2019.
- Zhang, R., Wang, G., Guo, S., Zamora, M. L., Ying, Q., Lin, Y., Wang, W., Hu, M., and Wang, Y.:
  Formation of urban fine particulate matter, Chem. Rev., 115, 3803–3855, https://doi.org/10.1021/acs.chemrev.5b00067, 2015.
- Zhang, Y., Sun, Y., Du, W., Wang, Q., Chen, C., Han, T., Lin, J., Zhao, J., Xu, W., Gao, J., Li, J., Fu, P.,
  Wang, Z., and Han, Y.: Response of aerosol composition to different emission scenarios in Beijing,
  China, Sci. Total Environ., 571, 902–908, https://doi.org/10.1016/j.scitotenv.2016.07.073, 2016.
- Zhang, Y., Tang, L., Croteau, P. L., Favez, O., Sun, Y., Canagaratna, M. R., Wang, Z., Couvidat, F.,
  Albinet, A., Zhang, H., Sciare, J., Prévôt, A. S. H., Jayne, J. T., and Worsnop, D. R.: Field
  characterization of the PM<sub>2.5</sub> aerosol chemical speciation monitor: insights into the composition,
  sources, and processes of fine particles in eastern China, Atmos. Chem. Phys., 17, 14,501–14,517,
  https://doi.org/10.5194/acp-17-14501-2017, 2017.
- Zhao, J., Du, W., Zhang, Y., Wang, Q., Chen, C., Xu, W., Han, T., Wang, Y., Fu, P., Wang, Z., Li, Z.,
  and Sun, Y.: Insights into aerosol chemistry during the 2015 China Victory Day parade: results from
- 597 simultaneous measurements at ground level and 260 m in Beijing, Atmos. Chem. Phys., 17, 3215-

- 598 3232, https://doi.org/10.5194/acp-17-3215-2017, 2017.
- 599 Zhong, S., Qian, Y., Sarangi, C., Zhao, C., Leung, R., Wang, H., Yan, H., Yang, T., and Yang, B.:
- 600 Urbanization effect on winter haze in the Yangtze River Delta Region of China, Geophys. Res. Lett.,
  601 45, 6710–6718, https://doi.org/10.1029/2018GL077239, 2018.
- 602 Zhu, Y., Yan, C., Zhang, R., Wang, Z., Zheng, M., Gao, H., Gao, Y., and Yao, X.: Simultaneous
- 603 measurements of new particle formation at 1-s time resolution at a street site and a rooftop site, Atmos.
- 604 Chem. Phys., 17, 9469–9484, https://doi.org/10.5194/acp-17-9469-2017, 2017.