

1 **Differentiating local and regional sources of Chinese urban air pollution based on**  
2 **effect of Spring Festival**

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11 **Abstract:** The emission of pollutants is extremely reduced during the annual Chinese Spring Festival  
12 (SF) in Shenzhen, China. During the SF, traffic flow drops by ~50% and the industrial plants are almost  
13 entirely shut down in Shenzhen. To characterize the variation in ambient air pollutants due to the  
14 “Spring Festival effect”, various gaseous and particulate pollutants were measured in real time in urban  
15 Shenzhen over three consecutive winters (2014–2016). The results indicate that the concentrations of  
16 NO<sub>x</sub>, volatile organic compounds (VOCs), black carbon (BC), primary organic aerosols, chloride, and  
17 nitrate in submicron aerosols decrease by 50%–80% during the SF period relative to the non-Spring  
18 Festival periods, regardless of meteorological conditions, which suggests that these pollutants are  
19 mostly emitted or secondarily formed from urban local emissions. The concentration variation of  
20 species mostly from regional or natural sources, however, is found to be much less, such as for bulk  
21 PM<sub>2.5</sub>. More detailed analysis of the Spring Festival effect reveals an urgent need to reduce emissions  
22 of SO<sub>2</sub> and VOCs on a regional scale rather than on an urban scale to reduce urban PM<sub>2.5</sub> in Shenzhen,  
23 which can also produce some use for reference for other megacities in China.

24 **Key words:** Spring Festival effect; local emissions; regional pollution; PM<sub>2.5</sub>; ozone

## 25 **1 Introduction**

26 The rapid economic development and urbanization of China over the recent decades has brought with  
27 it the consequence of severe atmospheric pollution, especially in the key economically developed  
28 regions, such as the Beijing–Tianjin–Hebei region (Sun et al., 2013, 2015; Guo et al., 2014), the  
29 Yangtze River Delta (Huang et al., 2013), and the Pearl River Delta (PRD), as well as their densely  
30 populated megacities (Hagler et al., 2006; Zhang et al., 2008; He et al., 2011). Great efforts have been  
31 made to determine the sources and formation mechanisms of fine particles (PM<sub>2.5</sub>) in these region.  
32 Previous studies indicate that PM<sub>2.5</sub> forms from primary fine particles and through secondary formation  
33 from gaseous precursors (Zhang et al., 2008; Zheng et al., 2009a; Huang et al., 2014), and the sources  
34 of local production and regional transport are both important (Huang et al., 2014; Huang et al., 2006,  
35 2011; Li et al., 2015).

36  
37 The causes of air pollution in urban atmosphere in China are particularly complicated, and bring great  
38 challenges to management strategies for protecting human health (Parrish and Zhu, 2009). To explore  
39 the causes of urban air pollution in China, previous studies have focused on monitoring and comparing  
40 the reduction in emissions during special events, such as the 2008 Beijing Olympic Games (Huang et  
41 al., 2010), the 2010 Guangzhou Asian Games (Xu et al., 2013), the 2014 Asia Pacific Economic  
42 Cooperation conference (APEC) (Chen et al., 2015; Sun et al., 2016; Zhang et al., 2016) and the 2015  
43 China victory day parade (Zhao et al., 2016). During such events, the air quality improved remarkable  
44 because of short-term limitations on traffic and industrial activity (Huang et al., 2010; Wang et al.,  
45 2010; Xu et al., 2013; Sun et al., 2016; Zhao et al., 2016). However, these limitations were temporary,  
46 non-repeatable measures, so the air quality monitoring campaigns cannot be repeated. Actually, a

47 spontaneous reduction in emissions occurs every year in China during the Spring Festival (SF), which  
48 is the single most important holiday in China. During the week-long holiday (in January or February  
49 every year), the urban emission patterns depart significantly from the usual patterns: traffic decreases  
50 in the mega cities because most people are not working, and most of the industries, stores, and  
51 production sites are closed in the city except for the infrastructure (e.g., power plants) that cannot be  
52 shut down (Qin et al., 2004; Feng et al., 2012; Shi et al., 2014). Tan et al. (2009) reported that the  
53 concentrations of NO<sub>x</sub>, CO, NMHC, SO<sub>2</sub>, and PM<sub>10</sub> were lower in the SF periods than in the non-  
54 Spring Festival (NSF) periods in the metropolitan area of Taipei over 1994-2006, while the variation  
55 of O<sub>3</sub> was in a reversed trend. Jiang et al. (2015) found that the ambient concentrations of VOCs had  
56 a sharp decline by ~60% during the SF in Shijiazhuang.

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58 This study focuses on Shenzhen as a special example to evaluate the effect on urban air pollution of  
59 the SF. Shenzhen is in the eastern Pearl River Delta (PRD) and is the fourth largest economic center  
60 in China, with a total residential population of over 10 million and a fleet of civilian vehicles of more  
61 than 3.1 million (Shenzhen Yearbook of Statistics, 2015). Known as the country's city of most floating  
62 population, Shenzhen owns 7.4 million immigrants in 2014, which accounts 70% of the city's total  
63 population (Shenzhen Yearbook of Statistics, 2015). During the SF period, over 50% of the residents  
64 in Shenzhen are used to travel back to their hometowns (<http://sz.gov.cn>). It is reported that the traffic  
65 flow in Shenzhen during the SF of 2016 (Feb 7–13) was only the half before the SF period  
66 (<http://sz.gov.cn>). Additionally, industrial activities are almost totally suspended in Shenzhen during  
67 the SF period. To characterize the air quality during such extreme reductions of anthropogenic  
68 activities during the SF period in Shenzhen, various air pollutants in Shenzhen urban areas were

69 comprehensively and systematically monitored in real time in winter for three consecutive years  
70 (2014–2016). The annual SF in Shenzhen thus provides an excellent spontaneous control experiment  
71 for local emissions, which could provide unique and valuable information regarding the sources of  
72 urban air pollution.

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## 74 **2 Experimental methods**

### 75 **2.1 Monitoring sites and meteorological conditions**

76 The monitoring site (22°36'N, 113°54'E) was on the roof (20 m above ground level) of an academic  
77 building on the campus of Peking University Shenzhen Graduate School (PKUSZ) (Figure S1).  
78 PKUSZ is located in the western urban area of Shenzhen, and there are no significant anthropogenic  
79 pollution sources nearby except a local road ~100 m far from the sampling site. Figure S1 shows that  
80 the sampling area had a middle-level aerosol pollution during the winter in comparison with other  
81 areas in PRD and further areas. A highly resolved temporal and spatial emission inventory for PRD  
82 indicates that the sampling area is characterized by lower SO<sub>2</sub> emissions but higher NO<sub>x</sub> and VOCs  
83 emissions in comparison with other areas in PRD (Zheng et al., 2009b). The sampling schedule ran  
84 roughly from late January to early March over 2014–2016, which includes the official SF holiday  
85 period and the prior and following periods. Our definition of the SF period follows that of the statutory  
86 public holiday calendar in China, and it is continuous seven days in each year. While the seven days  
87 immediately before or after the holidays are actually the transition periods between the holidays and  
88 normal days (called the Tran. periods hereafter), when people begin to move from the city (or their  
89 hometowns) to their hometowns (or the city), the typical non-spring festival (NSF) periods are better  
90 defined as the 7–14 days close to the SF period (called the NSFT period hereafter, where T indicates

91 time similar). The specific dates and the average meteorological parameters are listed in Table 1, and  
92 Figure S2 shows wind rose plots. The data in Table 1 show that the meteorology differs among the SF,  
93 NSFT, and Tran. periods. To control for the influence of meteorology on the evaluation of emissions,  
94 we selected another 7-day period each year when the meteorology is similar to that of the SF period  
95 (called the NSF<sub>M</sub> period hereafter, where M indicates meteorology similar); the detailed parameters  
96 are listed in Table 1 and Figure S2. The meteorological data for the SF period are fairly similar to those  
97 of the NSF<sub>M</sub> period, suggesting similar meteorological conditions.

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114 **Table 1.** Summary of meteorological conditions at sampling site during the SF, NSFT, NSFMM and  
 115 Tran. periods of 2014–2016.

		SF	Tran.	NSFT	NSFM
Data period	2014	Jan 31–Feb 6	Feb 7–Feb 13	Feb 14–Feb 20	Feb 20–Feb 26
	2015	Feb 18–Feb 24	Feb 11–Feb 17	Feb 4–Feb 10	Jan 24–Jan 30
	2016	Feb 7–Feb 13	Feb 14–Feb 20	Feb 21–Feb 27	Feb 27–Mar 4
Meteorological parameters	Temperature (°C)	19.0±4.7	14.1±5.3	14.1±4.0	18.1±3.8
	RH (%)	68.1 ±17.8	69.3±18.4	64.9±16.7	67.4±14.7
	Wind speed (m s <sup>-1</sup> )	0.88 ±0.57	0.81±0.49	0.83±0.48	0.86±0.55
	Dominant wind direction	NW	NW and NE	NW and NE	NW
	Precipitation (mm)	0	0	0	0
	UVA (W m <sup>-2</sup> )	5.4±8.5	2.5±4.3	3.8±6.7	5.0±8.0
	UVB (W m <sup>-2</sup> )	0.24±0.40	0.11±0.25	0.16±0.32	0.22±0.38

## 116 2.2 Instrumentation

117 For the ambient sampling in this study, the measuring instruments were placed in a room on the top  
 118 floor of a four-story teaching building at PKUSZ. A high-sensitivity proton transfer reaction mass  
 119 spectrometer (PTR–MS) (Ionicon Analytik GmbH, Austria) was used to measure the selected volatile

120 organic compounds (VOCs). The PTR–MS measured a total of 25 masses in the selected ion mode at  
121 a time resolution of 30 s. Background checks were done for 30 of every 300 scan cycles with an  
122 activated charcoal trap at 360 °C, which can remove VOCs from the ambient air without changing  
123 water content. The VOCs reported here (Table S1) may be broadly classified into three categories:  
124 oxygenated VOCs [OVOCs: methanol, acetone, methyl ethyl ketone (MEK), acetaldehyde, and acetic  
125 acid], aromatics (benzene, toluene, styrene, C8 and C9 aromatics), and three types of tracers [isoprene,  
126 acetonitrile, and dimethyl sulfide (DMS)]. The PTR–MS was calibrated every 5 to 7 days by using a  
127 TO15 mixture standard (Air Environmental Inc., US) and permeation tubes (Valco Instruments Co.  
128 Inc., US) (de Gouw and Warneke, 2007).

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130 An aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR–ToF–AMS) (Aerodyne  
131 Research, US) was deployed to measure non-refractory PM<sub>1</sub> (NR-PM<sub>1</sub>) (Canagaratna et al., 2007) in  
132 the period 2014–2015 with a time resolution of 4 min. An aerosol chemical speciation monitor (ACSM)  
133 (Aerodyne Research, US) was used in 2016 with a dynamic resolution of 10 min. The detailed  
134 description of the ACSM is available in the recent review (Ng et al., 2011). The HR-ToF-AMS and  
135 ACSM were calibrated every month following the standard protocols (Ng et al., 2011; Jayne et al.,  
136 2000).

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138 An aethalometer (AE–31) (Magee, US) was used for simultaneous detection of refractory black carbon  
139 (BC) with a time resolution of 5 min. In addition, a Scan Mobility Particle Sizer (TSI Inc., US) system  
140 was used to determine the particle number size distribution in the size range 15–615 nm (Stokes  
141 diameter) with a time resolution of 5 min. The stokes diameters of 15–615 nm is converted to

142 aerodynamic diameters of 22–800 nm, and then  $PM_{0.8}$  mass concentration can be calculated with the  
143 particle density assumed according to the AMS measurement results of species.

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145 To measure the  $PM_{2.5}$  mass concentration, we used a Thermo Scientific TEOM 1405–D monitor. The  
146 trace-gas instruments included a 43i sulfur dioxide ( $SO_2$ ) analyzer, a 42i nitric oxide (NO)–nitrogen  
147 dioxide ( $NO_2$ )–nitrogen oxide ( $NO_x$ ) analyzer, a 49i ozone ( $O_3$ ) analyzer, and a 48i carbon monoxide  
148 (CO) analyzer (Thermo Scientific, US). A meteorological station, also located on the roof of the same  
149 building, measured the main meteorological parameters, such as temperature, relative humidity, and  
150 wind speed (see Table 1).

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## 152 **3 Results and Discussion**

### 153 **3.1 The NSF–SF differences for major air pollutants**

154 The results of observations from 2014 to 2016 appear in Figures S3–S5. Figure 1 shows the averaged  
155 percent changes in the concentrations of major air pollutants of the SF periods relative to the two NSF  
156 periods and Tran. period over 2014–2016. Most of fragments of  $m/z$  44 and  $m/z$  57 are the tracer of  
157 oxygenated organic aerosol and the tracer of primary hydrocarbon organic aerosol (Zhang et al., 2005),  
158 respectively, which are measured by AMS. The notation  $O_3$ –8h refers to the average maximum  $O_3$   
159 concentration over a continuous diurnal 8 h and  $PM_{0.8-2.5}$  refers to the difference between the  
160 concentrations of  $PM_{2.5}$  and  $PM_{0.8}$ .

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162 We can divide these air pollutants into three classes based on their percent changes: The group with  
163 the largest drop (hereinafter called “LD”) in concentration includes the aromatics (–50% to –88% for

164 the various species, see Figure S6), OVOCs (−40% to −85% for the various species, see Figure S6),  
165 NO<sub>x</sub>, chloride (Chl), nitrate (NO<sub>3</sub><sup>−</sup>), BC, and m/z 57. The concentrations of these pollutants all  
166 decrease by over 50% during the SF period compared with both the NSF periods. Apparently, the  
167 dominant sources for most of these pollutants are primarily local emissions in the urban scale, such as  
168 combustion sources for BC, m/z 57, and NO<sub>x</sub> (Zhang et al., 2005; Kuhlbusch et al., 1998; Lan et al.,  
169 2011), and vehicle, industrial and solvent use for aromatics (Liu et al., 2008). As detailed in the  
170 following section, the diurnal patterns and relationships with respect to wind speed further confirm the  
171 sources of these pollutants. The dramatic decrease in the ambient concentrations of these species is  
172 consistent with reduction in local anthropogenic activities in Shenzhen during the SF period. The SF  
173 causes a 50% decrease in urban traffic and temporarily closing of almost all local industrial plants. The  
174 nitrate and chloride measured by AMS or ACSM are actually ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) and  
175 ammonium chloride (NH<sub>4</sub>Cl), which are typical secondary air pollutants. These are thought to form  
176 via reversible phase equilibria with gaseous ammonia (NH<sub>3</sub>), nitric acid (HNO<sub>3</sub>), and hydrochloric  
177 acid (HCl) (He et al., 2011; Huang et al., 2011; Zhang et al., 2007). Typically, the formation of NH<sub>4</sub>NO<sub>3</sub>  
178 from NO<sub>x</sub> and the reaction between HCl and NH<sub>3</sub> occur quickly in the atmosphere (Stelson and  
179 Seinfeld, 1982; Baek et al., 2004), suggesting that the concentrations of NO<sub>3</sub><sup>−</sup> and Chl in winter in  
180 Shenzhen depend largely on the emission of precursors such as HCl and NO<sub>x</sub>. Therefore, the  
181 significant decline in the ambient concentrations of NO<sub>3</sub><sup>−</sup> and Chl during the SF period and indicates  
182 that their precursors also have local origins, similar to the case for primary pollutants (this is also  
183 supported by the discussion in the following sections). The huge decline in the ambient concentration  
184 of OVOCs during the SF period shows that the source of these pollutants is (i) mainly from local  
185 emissions, including vehicle and industrial emissions (Schauer et al., 1999; Singh et al., 2001) and (ii)

186 from secondary reactions involving local primary VOCs (Liu et al., 2015). Thus, in the LD group, the  
187 significant reduction in local sources of pollutants strongly impacts the concentration of air pollutants.  
188

189 The pollutants in the next group undergo a medium drop in concentration during the SF period  
190 (hereinafter called “MD”). These are PM<sub>2.5</sub>, NR-PM<sub>1</sub>, PM<sub>0.8</sub>, organic aerosol, m/z 44, sulfate (SO<sub>4</sub><sup>2-</sup>),  
191 ammonium (NH<sub>4</sub><sup>+</sup>), isoprene, acetonitrile, DMS, and carbon monoxide (CO), and their percent change  
192 varies from -20% to -55% when comparing the SF periods to the NSFT and NSF<sub>M</sub> periods. The  
193 species in this group are either typical regional air pollutants mostly from beyond the urban scale, such  
194 as CO, which has a long lifetime and is a tracer for combustion sources, acetonitrile from rural biomass  
195 burning (de Gouw et al., 2003; Le Breton et al., 2013), m/z 44 representing secondary organic aerosols,  
196 SO<sub>4</sub><sup>2-</sup> from SO<sub>2</sub> oxidation (He et al., 2011; Huang et al., 2011), or typical tracers mainly emitted by  
197 natural sources, such as isoprene from vegetation (Guenther et al., 1995) and DMS from marine source  
198 (Dacey and Wakeham, 1986). In winter, the northeastern monsoon prevails in the PRD and transports  
199 significant amounts of various air pollutants from the northern inland, increasing air pollution of the  
200 PRD to the highest levels through the year (Huang et al., 2014). In particular, the small drop in CO  
201 concentration during the SF period puts it in this group and indicates that the contribution to regional  
202 air pollution does not decrease significantly during the SF period. Note that, the significant declines of  
203 the concentrations of isoprene and DMS imply that they have anthropogenic sources, which will be  
204 supported in the following sections. The other air pollutants in this group are the reflection of the  
205 overall effect of the reduction of relevant air pollutants: OA is the whole of the two types of organic  
206 aerosol represented by m/z 44 and m/z 57, NH<sub>4</sub><sup>+</sup> is represented by SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and Chl, and NR-PM<sub>1</sub>  
207 is the sum of all species measured by AMS or ACSM (their average chemical compositions during

208 different periods are shown in Figure S3–S5).

209 The group of pollutants with smallest decrease in concentration (hereinafter called “SD”) includes SO<sub>2</sub>

210 and PM<sub>0.8–2.5</sub>, and O<sub>3</sub> (8h) in the case of comparison with NSF. The magnitude of the average percent

211 change is less than 20% relative to the two NSF periods. It is interesting to note that there was even

212 concentration increase in other O<sub>3</sub>–related cases. The average concentration of SO<sub>2</sub> was only 2.8 ppbv

213 in Shenzhen in 2015 (<http://www.szhec.gov.cn/>), which is much lower than that in Beijing (4.7 ppbv)

214 and elsewhere in China (<http://www.zhb.gov.cn/>). This result is partly attributed to the negligible coal

215 consumption in Shenzhen, which instead relies mainly on natural gas and liquefied petroleum gas

216 (Shenzhen Yearbook of Statistics, 2015). The emission inventory indicates that power plants and

217 international marine container vessels are the dominant source of SO<sub>2</sub> in Shenzhen (Wang et al., 2009;

218 Zheng et al., 2009b). According to official statistics, the Shenzhen port piloted 401, 568, and 521 ships

219 during the SF period in the years 2014–2016, respectively, which is quite similar to numbers for the

220 NSF periods (<http://www.pilot.com.cn/>). As infrastructure, power plants are not fully shut down during

221 SF. On the other hand, a piece of evidence for the regional origin of SO<sub>2</sub> is from the newly established

222 356 m meteorological and environmental monitoring iron tower in Shenzhen. The ambient SO<sub>2</sub>

223 concentrations were similar at the highest platform (ave.=7.4 ppbv@325 m) and the lowest platform

224 (ave.=7.2 ppbv@60 m) during January–February, 2017, indicating that SO<sub>2</sub> was already well mixed in

225 the atmosphere and the local contributions should be minor. In contrast, the concentrations of NO<sub>x</sub>,

226 which belongs to Group LD, had a 56% higher concentration at the lowest platform than at the highest

227 platform (Zhuang, 2017). The small decrease of SO<sub>2</sub> is thus a reasonable result of the stable emissions

228 during the SF periods and the primarily regional origin. The small decline of PM<sub>0.8–2.5</sub> during the SF

229 period suggests that the reduction of more aged particles of larger sizes in PM<sub>2.5</sub> is much lower than

230 fresher particles of smaller size. This can be also confirmed by particle number concentration (PNC)  
231 measurement by SMPS, as shown in Figure 2. The largest difference of the PNC between the SF and  
232 NSFPM periods exists mainly in a smaller size range (20–40 nm), which is recognized as the nucleation  
233 mode or second Aitken mode that represents fresh combustion emission (Ferin et al., 1990). In terms  
234 of chemical composition of  $PM_{0.8-2.5}$ , implications can be found in our previous size distribution  
235 measurement of aerosol chemical composition, using a ten-stage micro orifice uniform deposit  
236 impactor (MOUDI), during the fall to winter in Shenzhen (Lan et al., 2011). The results clearly indicate  
237 that smaller fine particles (e.g., 0.18–0.56  $\mu m$ ) contains relatively more BC ( $BC/SO_4^{2-}=0.83$ ), while  
238 larger fine particles (e.g., 1.0–1.8  $\mu m$ ) contained a higher proportion of  $SO_4^{2-}$  ( $BC/SO_4^{2-}=0.17$ ). The  
239  $SO_4^{2-}$  in  $PM_{2.5}$  in Shenzhen has been well proved to be mostly a regional pollutant, with similar  
240 concentrations at various sites including both urban and rural sites (Huang et al., 2014). Therefore, the  
241 very small decrease of  $PM_{0.8-2.5}$  during SF should be closely related to its enrichment of secondary  
242 regional species like  $SO_4^{2-}$ . Contrary to other pollutants, the concentrations of  $O_3$ , present small  
243 increasing during the SF period (except a little decline when comparing  $O_3$ -8h with the NSFPM period),  
244 which could be attributed to the different drop rates for  $O_3$  precursor species, i.e.  $NO_x$  and VOCs (Qin  
245 et al., 2004), and will be discussed in more detail in section 3.2.

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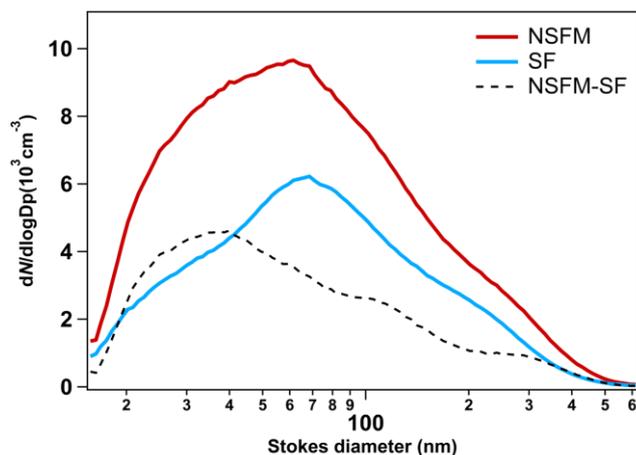
247 The decreasing ratios of various species during SF when compared with the NSFT and NSFPM periods  
248 are similar, which suggests that the meteorological variations might not be the dominant reason for the  
249 species decreasing during SF. This means that the strong decrease in the concentrations of air pollutants  
250 in group LD and MD is mainly due to the abatement of local sources. The larger decline in the SF  
251 period when compared to NSFT than to NSFPM is associated with the lower temperature and stronger

252 winds from the polluted northwest inland of the PRD during the NSFT period. In addition, the effect  
 253 of the SF on the concentrations of the various species is almost identical each year (see Figure S7),  
 254 which further confirms that the pollutant concentrations are determined primarily by the activity of the  
 255 sources. In Figure 1, the percent changes of pollutants of the SF periods relative to the Tran. periods  
 256 are also presented, and it is found that the three-group classification defined above is also applicable,  
 257 while the decrease levels are lower. For example, the average decrease percent of Group LD for the  
 258 Tran. period case is 61%, while those for the NSFT and NSFm cases are 71% and 63%, respectively.  
 259 This result is consistent with the fact that the SF travel of people occurred mostly during the seven  
 260 days before and after the SF holidays (<http://sz.gov.cn>), and thus the city became much emptier even  
 261 in the Tran. periods. In order to make a deeper and valid comparison for revealing the SF effect, the  
 262 following discussion will only take the NSFm periods and SF periods for comparative analysis due to  
 263 their more similar meteorology.



264

265 **Figure 1.** Percent change in concentrations of major air pollutants during the SF period relative to (a)  
266 Tran., (b) NSFT and (c) NSFMT periods averaged over 2014–2016.



267

268 **Figure 2.** Distribution of particle number concentration in the 15–615 nm size range during the SF  
269 and NSFMT periods.

270

### 271 3.2 The diurnal variation of major air pollutants

272 As shown in Figure 3, the diurnal cycles of all LD pollutants (except for the OVOCs) reveal significant  
273 peaks in concentration around 8–9 am in the NSFMT period, which is attributed to the low planetary  
274 boundary layer (PBL) in the morning and local rush hour traffic emissions. The evening rush hour  
275 peak, however, is not apparent for all the species, which is attributed to the higher ambient temperature  
276 and thus the higher PBL at that time than in the morning. During the SF period, the concentrations of  
277 all pollutants are far lower over the entire day. In particular, the rush-hour peaks become much smaller  
278 or disappear altogether, which is consistent with the large reduction in local vehicle emissions during  
279 the SF period. Although the sources of Chl remained uncertain in previous studies (Huang et al., 2011;  
280 Aiken et al., 2008), the maximal reduction (80%) in this pollutant during the morning rush hour during

281 the SF period implies that local traffic emissions account for a significantly fraction of this pollutant  
282 in Shenzhen (Figure 3E). Contrary to other species in this group, the concentration of OVOCs is high  
283 in the daytime and peaks in the morning after the morning rush hour time during the NSFm period  
284 (Figure 3D), suggesting that photochemical production and/or daytime industrial activities may be  
285 important sources of OVOCs. The concentrations of different aromatics and OVOCs usually follow  
286 similar diurnal variations (Figure S8).

287

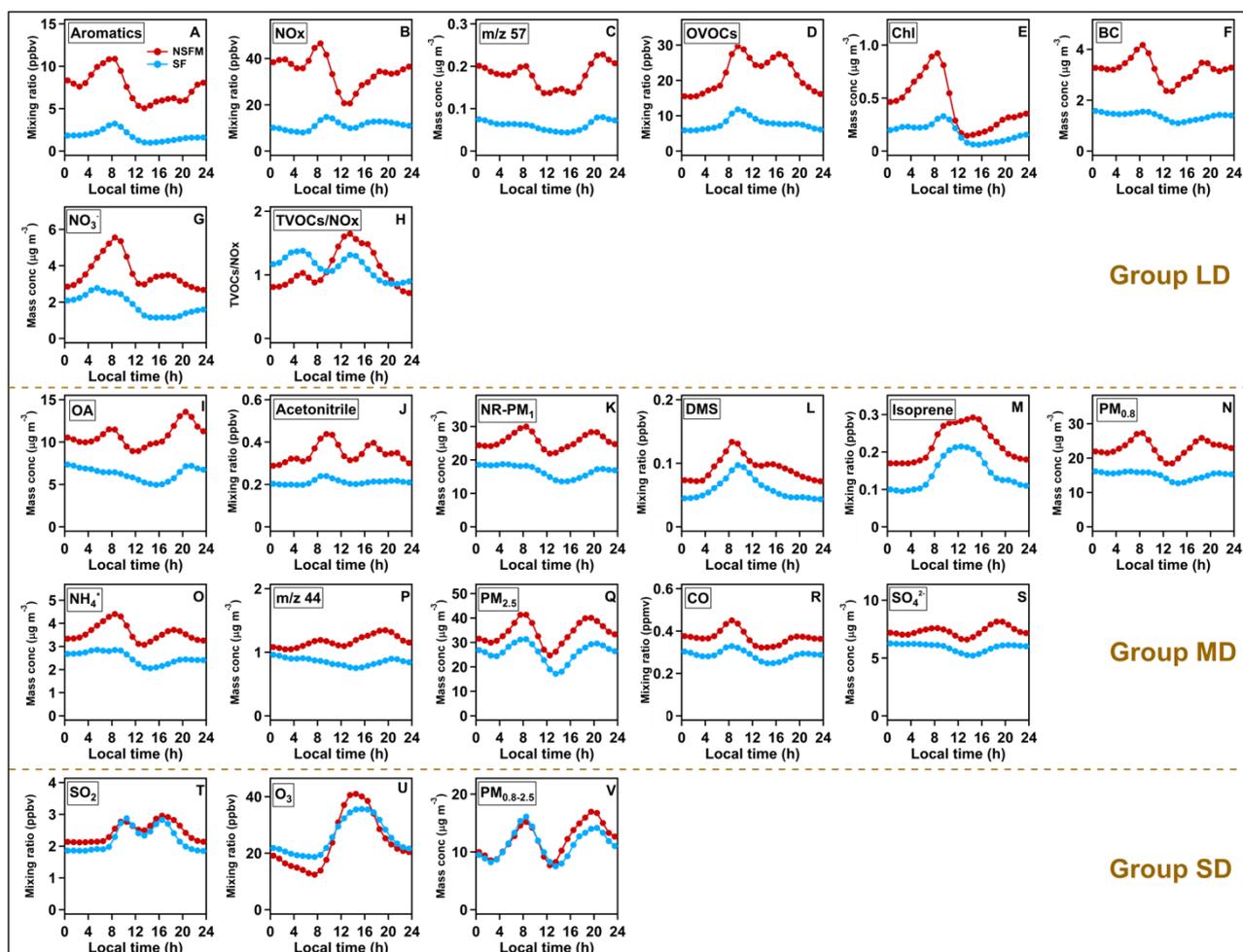
288 The diurnal variations of the MD pollutants are relatively smooth except for two VOCs mainly from  
289 natural sources (isoprene and DMS; see Figures 3L and 3M), which indicates that these pollutants  
290 predominantly come from regional sources and are dispersed more uniformly over a larger scale. The  
291 apparent difference of the diurnal variations of those anthropogenic air pollutants between the SF and  
292 NSFm periods also exists in the rush hours (except for acetonitrile; Figure 3J), however, the reduction  
293 in local sources has a relatively weak effect on the overall concentrations of these pollutants.  
294 Acetonitrile, which is a tracer of biomass burning, is more concentrated during the daytime and its  
295 peak concentration occurs after the rush hours during the NSFm period (Figure 3J), which is similar  
296 to the result obtained for OVOCs and may be attributed to the influence of daytime anthropogenic  
297 activities, for example, industrial biomass boilers. Isoprene is primarily emitted by vegetation as a  
298 function of light and temperature, so the concentration of this pollutant goes through a broad peak that  
299 spans the daytime hours during both the NSFm and SF periods. The percent change in isoprene  
300 concentration between the SF and NSFm periods is approximately  $-40\%$  (Figure 3M), despite the  
301 NSFm and SF periods having similar temperature and solar radiation, which implies that the  
302 contribution of anthropogenic sources to isoprene cannot be overlooked in Shenzhen. Many studies

303 have reported isoprene from vehicle exhaust, especially in cold seasons (Barletta et al., 2005; Borbon  
304 et al., 2001). DMS is reported to be a marine tracer (Dacey et al., 1986), its peak concentration occurs  
305 in the morning during both the NSF<sub>M</sub> and SF periods (Figure 3L), which is presumably related to the  
306 minimal PBL. The concentration of DMS decreases by 30%–50% during the SF period, which reflects  
307 the reduced DMS emissions from anthropogenic sources. As reported in the literature, industrial  
308 activities can make significant emissions of DMS (Schafer et al., 2010).

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310 The diurnal variations of PM<sub>0.8–2.5</sub>, SO<sub>2</sub> and O<sub>3</sub> demonstrated more similar concentrations and trends  
311 in the SF and NSF<sub>M</sub> periods, respectively (Figure 3T–3V). For PM<sub>0.8–2.5</sub>, a small difference is found  
312 in the afternoon, which is supposed to be a result of more aged larger particles formed through stronger  
313 photochemical reactions during the NSF<sub>M</sub> period. Though, slight differences appear in SO<sub>2</sub>  
314 concentration, mainly during the nighttime when the PBL is low. These data suggest a minor role of  
315 local near-ground SO<sub>2</sub> sources, such as vehicles. Although the daytime peak concentration of O<sub>3</sub> during  
316 the NSF<sub>M</sub> period is slightly greater than that during the SF period, this trend reverses from the evening  
317 to the midmorning hours. Similar phenomena have also been observed in other emission-reduction  
318 studies of urban areas (i.e., O<sub>3</sub> concentrations are greater on holidays than on non-holidays) (Qin et al.,  
319 2004; Tan et al., 2009). In addition, O<sub>3</sub> concentrations were higher during the 2008 Beijing Olympic  
320 Games (Chou et al., 2011), during which strict controls were imposed. The lower peak concentration  
321 of O<sub>3</sub> in the afternoon (13:00–16:00 LT) during SF suggests that the large reduction on precursors can  
322 also help mitigate the daytime O<sub>3</sub> concentration. However, the O<sub>3</sub> concentration at night during SF  
323 was higher than that during NSF<sub>M</sub>, which could be attributed to the oxidation reaction with NO of  
324 higher concentrations during NSF<sub>M</sub>, producing a titration effect and thus destroying O<sub>3</sub> (Qin et al.,

325 2004; Tan et al., 2009). As a result, although the reduction in emissions of urban anthropogenic sources  
 326 leads to a large decline of NO<sub>x</sub> and VOCs, this reduction does not mitigate the average ambient O<sub>3</sub>  
 327 concentration, which implies that the concentration ratio VOCs/NO<sub>x</sub> play an important role in  
 328 controlling O<sub>3</sub> concentration.



329  
 330 **Figure 3.** Diurnal variations in concentrations of major air pollutants at PKUSZ site over the SF  
 331 (blue dots) and NSFM (red dots) periods.

332  
 333 **3.3 Influence of wind on observed air pollutants**

334 Wind plays a crucial role in the dilution and transport of air pollution. The wind field patterns are quite

335 similar between the SF and NSFm periods (Figure S2). In general, the concentrations of LD air  
336 pollutants depend strongly on wind speed during the NSFm period, whereas this dependence becomes  
337 much weaker during the SF period (Figure 4). The difference in the concentration of LD air pollutants  
338 (including various aromatics and OVOCs, see Figure S9) between the NSFm and SF periods is  
339 maximal (50%–80%) under conditions of low wind speeds (<1 m/s) because local pollution can more  
340 easily accumulate under these conditions. These results confirm that the concentration of air pollutants  
341 mainly from local sources is strongly reduced during in the SF period.

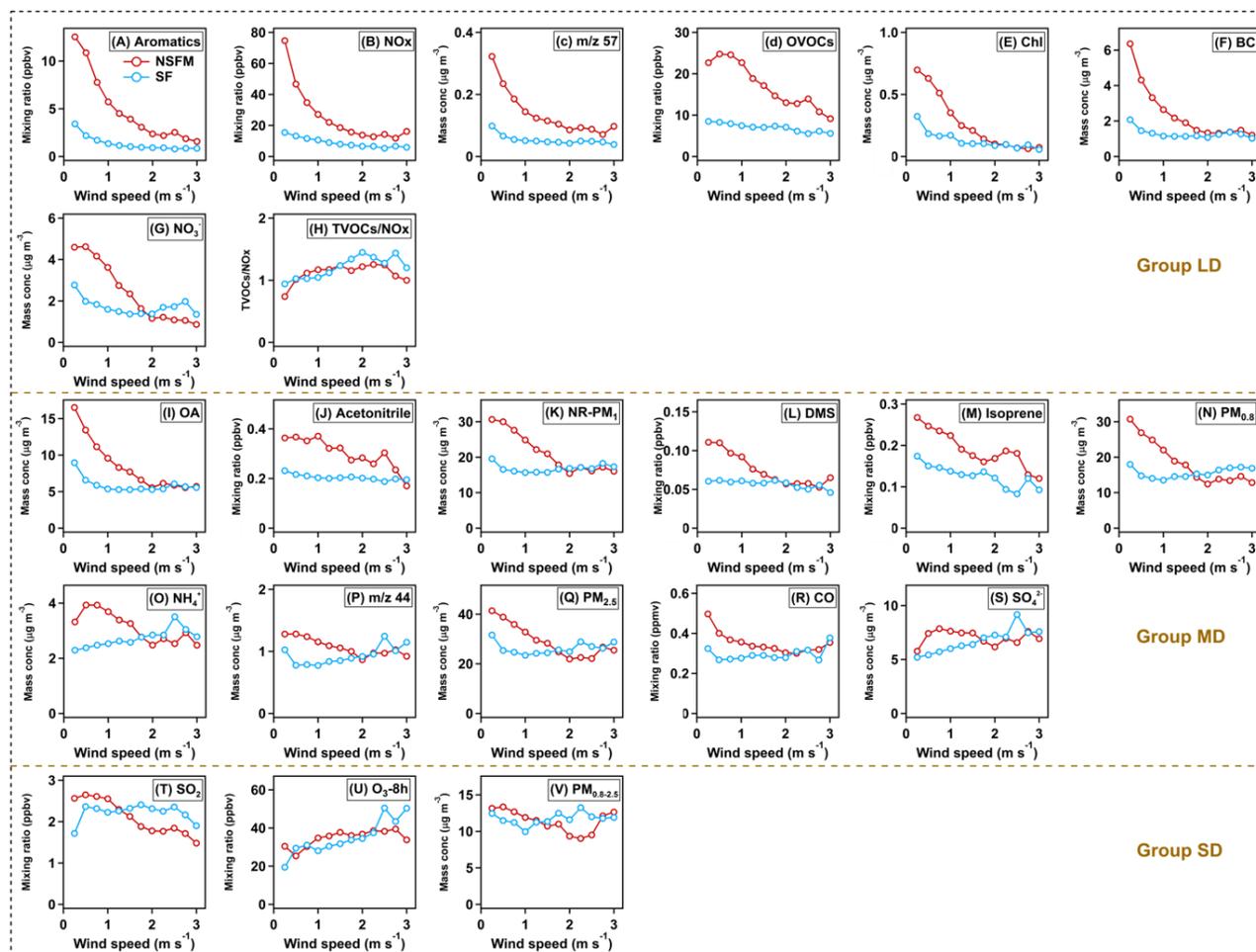
342

343 Compared with the LD pollutants, the concentrations of CO, SO<sub>4</sub><sup>2-</sup>, m/z 44, isoprene, DMS, and  
344 acetonitrile do not vary significantly with wind speed during the NSFm period, providing further  
345 evidence that these pollutants primarily come from regional or natural sources and are consequently  
346 more evenly distributed in the atmosphere.

347

348 In the Group SD, SO<sub>2</sub> is generally little influenced by wind speed during the SF period, while some  
349 higher concentrations appeared under low wind speeds during the NSFm period, indicating again small  
350 contribution of urban local sources to SO<sub>2</sub>. The fluctuation of PM<sub>0.8-2.5</sub> both in the SF and NSFm  
351 periods does not reveal a clear relationship with wind speed, suggesting again it is not a typical locally  
352 emitted air pollutant. The variations of O<sub>3</sub>-8h display the opposite trend to other air pollutants both in  
353 the SF and NSFm periods, growing smoothly as wind speed increases, which could be possibly  
354 attributed to more regional transport and/or the higher VOCs/NO<sub>x</sub> ratio under high wind speeds  
355 (Figure 4H). Note that, when the proportion of regional transport relative to local emission becomes  
356 bigger under higher wind speeds, the concentrations of NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, m/z 44, PM<sub>0.8-2.5</sub>, and O<sub>3</sub>-8h are

357 even slightly higher in the SF period than in the NSFM period, implying that regional photochemical  
 358 production during the SF period is not weakened.



359  
 360 **Figure 4.** Concentrations of major air pollutants as a function of wind speed during the SF and  
 361 NSFM periods.

362

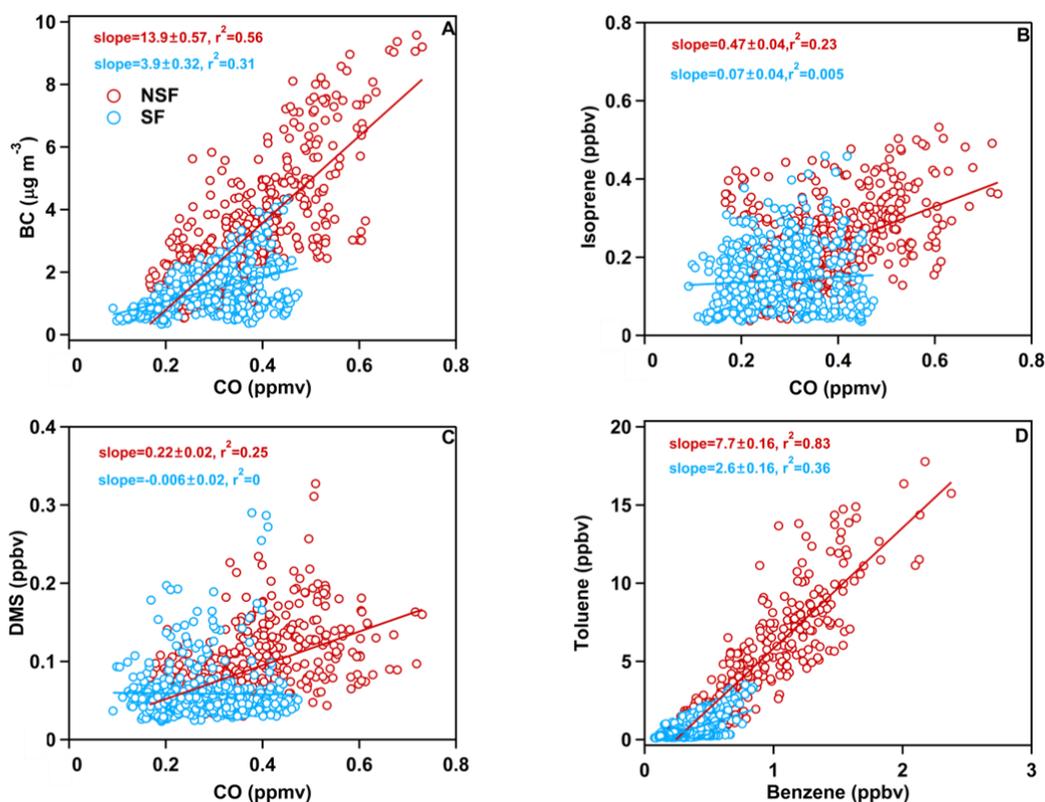
### 363 3.4 Emission ratio analysis

364 Several groups of special correlations were applied to analyze the source characteristics of air  
 365 pollutants in Figure 5. CO and BC are both products of incomplete combustion (Subramanian et al.,  
 366 2010), but gaseous CO can travel farther because of its longer atmospheric lifetime (approximately a

367 month for CO vs a week for BC) (Khalil et al., 1990; Ogren et al., 1983). As shown in Figure 5A, the  
368 correlation coefficient and slope between BC and CO during the NSFPM period ( $r^2 = 0.56$ , slope = 13.9)  
369 is greater than during the SF period ( $r^2 = 0.31$ , slope = 3.9), suggesting that local combustion sources  
370 make a much greater contribution during the NSFPM period, but decline significantly during the SF  
371 period (He et al., 2011). The concentrations of isoprene and DMS, are not correlated with CO during  
372 the SF, whereas their correlation with CO is non-negligible during the NSFPM period (Figures 5B and  
373 5C), suggesting again that these pollutants have an anthropogenic source during the NSFPM period.

374

375 The toluene/benzene ratio can be used to estimate the contribution of traffic emissions (Schneider et  
376 al., 2005). Generally, a value of 1.2–3 is found to be characteristic of vehicular emission in many urban  
377 areas (Nelson et al., 1984; Wang et al., 2002; Araizaga et al., 2013). The lower ratio of toluene to  
378 benzene (ave.=2.6) in the SF period suggests that the dominant source is vehicle emission. This ratio  
379 in the NSFPM period, however, is much higher (ave.=7.7), indicating more complicated sources of  
380 VOCs like huge amount of toluene solvent usage in industrial activities in PRD (Barletta et al., 2005,  
381 2008; Chan et al., 2006). This finding is well consistent with the temporary closure of industrial plants  
382 in the SF period, which leads to little toluene emission.



383

384 **Figure 5.** Correlation between air pollutants (A) BC and CO (B) isoprene and CO, (C) DMS and  
 385 CO, and (D) toluene and benzene during the SF (blue circles) and the NSF (red circles) periods.

386

### 387 3.5 Conclusions

388 This study uses the SF in Shenzhen to investigate how the urban air quality reacts to significant,  
 389 temporary reductions in emission. During the winters of 2014 to 2016, the air quality was observed  
 390 continuously at Peking University Shenzhen Graduate School, from which we obtained the percent  
 391 change in the concentrations of various air pollutants during the SF periods with respect to the  
 392 comparable NSF periods. The analysis of these data shows that, despite meteorological variations, the  
 393 Spring Festival clearly and consistently influences the urban concentrations of various air pollutants.  
 394 The air pollutants can be divided into three groups: the large-decrease (LD) pollutants are those with

395 a percent change in concentration of  $-50\%$  to  $-80\%$  during the SF period and include aromatics,  $\text{NO}_x$ ,  
396  $m/z$  57, OVOCs, Chl, BC, and  $\text{NO}_3^-$ . These results are consistent with the variation in urban emission  
397 sources during the SF, suggesting that these pollutants are mostly directly locally emitted or formed  
398 from secondary reactions between locally emitted pollutants. The medium-decrease (MD) pollutants  
399 are  $\text{PM}_{2.5}$ ,  $\text{NR-PM}_1$ ,  $\text{PM}_{0.8}$ , organic aerosol,  $m/z$  44,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , isoprene, acetonitrile, DMS, and  
400 CO; the concentrations of these pollutants decrease by 20% to 55% during the SF, which indicates that  
401 the extreme reduction in urban emissions during the SF period has limited effect on air pollutants  
402 mostly from regional or natural sources. Finally, the slight-decrease (SD) pollutants include  $\text{SO}_2$ ,  
403  $\text{PM}_{0.8-2.5}$ , and  $\text{O}_3$ . The average percent change in the concentrations of these pollutants during the SF  
404 period is less than 20%, which indicates that a significant reduction in urban emissions does not  
405 significantly affect their concentration. Of particular interest is the origin of  $\text{PM}_{0.8-2.5}$ , which is almost  
406 completely regional.

407

408 The results of this study show that the extreme reductions in urban emissions of Shenzhen only affects  
409 the concentration of smaller fresh particles, such as  $\text{PM}_{0.8}$ , whereas the reduction of  $\text{PM}_{2.5}$  is only  
410 slightly affected because of the weak influence on aged, larger particles such as  $\text{PM}_{0.8-2.5}$ . The  
411 concentrations of  $\text{SO}_4^{2-}$  and secondary organic aerosols are hardly affected by local reductions in  
412 emissions. Therefore, reducing the emissions of  $\text{SO}_2$  and VOCs on a regional scale is critical for  
413 reducing their concentrations and achieving the goal of reducing concentrations of  $\text{PM}_{2.5}$ , at least for  
414 South China. On the other hand,  $\text{O}_3$  has recently become an increasingly important air pollutant in  
415 China, especially in the PRD. However, the large reduction of  $\text{O}_3$  precursors ( $\text{NO}_x$  and VOCs) during  
416 the SF period only lead to small variation of  $\text{O}_3$  concentrations. Consequently, further investigations

417 are required to control not only the emissions of VOCs and NO<sub>x</sub> but also their concentration ratio.

418

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423

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