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# Characteristics of Atmospheric PM<sub>2.5</sub> Composition during the Implementation of Stringent Pollution Control Measures in Shanghai for the 2016 G20 Summit (HIGHLIGHTS)

- > Effectiveness of the pollution control measures was evaluated for the G20 summit.
- > Concentrations of most of criteria air pollutants reduced whereas O<sub>3</sub> elevated.
- > Impacts of local emissions and regional transport were analyzed at two Supersites.
- $\blacktriangleright$  Secondary aerosols and vehicle exhaust remained the top two sources of PM<sub>2.5</sub>.

#### 25 Abstract

26 To reduce air pollution within a 300 km radius from Hangzhou (the capital city of Zhejiang 27 Province in East China) for the 2016 G20 summit (9/4–9/5), the 14-day (8/24–9/6) stringent 28 pollution control measures were implemented in Shanghai. Changes in atmospheric 29 concentrations during the same 14-day period from 2014 to 2016 were examined at two 30 Supersites, i.e., urban Pudong site (PD) and Dianshan Lake regional site (DSL). Up to 50% 31 reductions were found for PM<sub>2.5</sub>, with 13.1% and 9.7% reductions for SO<sub>2</sub> and NO<sub>2</sub>, 32 respectively. No apparent improvements were found for 8-h average O<sub>3</sub> concentrations. Large reductions were also found for  $SO_4^{2-}(51.4\%)$ ,  $NO_3^{-}(68.8\%)$ , and  $NH_4^{+}(84.4\%)$ , on average. 33 34 Elevated coefficient of divergence values (0.52–0.56) suggested that pollutant sources 35 differed at the two sites. Biomass burning, resuspended dust, combustion, iron and steel 36 industry, sea salt, secondary aerosol, and vehicle exhaust were identified at the DSL site by 37 Positive Matrix Factorization (PMF). Secondary aerosol and vehicle exhaust accounted for 45.7% of PM<sub>2.5</sub> mass, followed 11.2%–13.7% each by industry, resuspended dust, and coal 38 39 and oil combustion.

40 *Keywords*: PM<sub>2.5</sub> composition; pollution control measures; G20 summit; Shanghai

#### 41 **1. Introduction**

42 As a leading voice in China's efforts to improve air quality, Shanghai government has enhanced air quality management to reduce air pollution. The "Clean Air Action of Shanghai 43 44 Municipality (2013–2017)" (Shanghai Municipal Government, 2013) was initiated only one month after the implementation of state Air Pollution Prevention and Control Action Plan 45 46 (The State Council of the People's Republic of China, 2013). Coal-free zones were 47 established in the metropolitan region (i.e., Inner Ring Road) where coal and heavy oil-fired boilers were replaced with clean energy boilers by 2015. In 2014, key industries (e.g., power 48 49 plant, iron and steel, cement and flat glass industries) in Yangtze River Delta (YRD) region 50 (Shanghai Environmental Protection Burea, 2014) were subjected to either initiate control 51 measures or retrofit existing desulfurization, denitration, and dust removal facilities. Industrial emission standards (i.e., smoke and dust  $\leq 10 \text{ mg/m}^3$ , SO<sub>2</sub>  $\leq 35 \text{ mg/m}^3$ , and NO<sub>2</sub>  $\leq$ 52  $50 \text{ mg/m}^3$ ) are expected to be attained prior to the 2020 target schedule. Implementation of 53 54 vehicle (i.e., gasoline and diesel) emission standards has been established in 2014 with 55 complete implementation in 2016. Efforts have also been made to promote low/zero-emission electric vehicles. (Bureau of Statistics of Shanghai, 2014, 2015, 2016; Clean Air Asia, 2017) 56 Reductions in PM<sub>2.5</sub> and O<sub>3</sub> are not apparent over the last three years. As shown in the 57

2014–2016 Air Quality Index (AQI) report (see also in Fig. S1) (Shanghai Environmental 58 59 Monitoing Center, 2017), high frequency of pollution in the number of non-attainment days 60 is wholly attributed to the elevated PM2.5 and O3 concentrations, similar to other 61 representative mega-cities in China (Ministry of Environmental Protection of China, 2016). The days that  $O_3$  as the major pollutant already outnumbered  $PM_{10}$ , making it second status 62 63 only to PM<sub>2.5</sub> in Beijing-Tianjin-Hebei (Jing-Jin-Ji) region (Wang et al., 2017; Wang et al., 2015). O<sub>3</sub> pollution becomes the primary issue in Pearl River Delta (PRD) region (Ling et al., 64 65 2017; Wang et al., 2016).

66 Hangzhou, the capital city of Zhejiang Province, hosted the eleventh international forum 67 for the 20 governments and central bank governors (the G20 summit) on 9/4-9/5 in 2016, which became China's biggest diplomatic event of the year. The neighboring Shanghai, 175 68 69 km northwest of Hangzhou, initiated a "G20 Blue" program over the 14-day period from 8/24 70 to 9/6 with stringent pollution control measures (Shanghai Environmental Protection Bureau, 71 2016). As shown in Table S1, the 14-day control measures include the company shutdown or 72 curtail 255 oil refinery, petrochemical, steel and other industries; reduce 30% of coal-fired 73 boilers and other combustion facilities; stop 101 large construction activities; ban single-hull 74 ferry boats and non-road machinery operations; restrict driving of high emitter (i.e., yellow-75 label) vehicles to downtown; extend public transport services and encourage flexible working 76 schedules. These immediate control actions resulted in a record of low PM<sub>2.5</sub> concentration  $(10.6 \,\mu\text{g/m}^3)$  in the city center. 77

78 The "G20 Blue" program provides the opportunity to investigate changes in air pollutant 79 emissions and ambient air concentrations and compositions. Here, the continuous 14-day 80 (8/24–9/6) sampling and analysis of ground-based meteorology, criteria air pollutant 81 concentrations (PM<sub>2.5</sub>, SO<sub>2</sub>, NO, NO<sub>2</sub>, and O<sub>3</sub>), as well as PM<sub>2.5</sub> chemical speciation were 82 conducted at two Supersites in Shanghai, i.e., urban Pudong site (PD) and Dianshan Lake regional site (DSL). The effectiveness of the implementation of the stringent pollution control 83 84 measures was evaluated during the same 14-day period from 2014 to 2016. Changes in 85 source apportionment of PM<sub>2.5</sub> mass were analyzed among the three years.

86 2. Materials and methods

#### 87 2.1. Supersite characteristics

88 The two Supersites (i.e., PD and DSL in Fig. 1), operated by the Shanghai 89 Environmental Monitoring Center, are situated approximately 44 km apart and designed to 90 characterize urban exposure and pollution transport. The urban-scale PD site (31°13′N, 91 121°32′E) is located in downtown Shanghai (5 km east of urban center, the People's Square);

92 whereas the DSL regional site (31°08′N, 121°05′E) is situated in 7 km east of Dianshan Lake,

93 adjacent to Zhejiang and Jiangsu Provinces.



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Fig. 1. Locations of the Pudong (PD) and Dianshan Lake (DSL) Supersites in Shanghai. The
contour of land use and population density adopted from Shanghai statistical yearbook in
2015 (Bureau of Statistics of Shanghai, 2016).

# 98 2.2. Samples collection and analysis

99 PM<sub>2.5</sub> mass concentrations were determined by a tapered-element oscillating 100 microbalance monitor (TEOM, Thermo FH62C-14). 24-h PM<sub>2.5</sub> samples were collected on 101 quartz-fiber filters (Tissuquartz 2600 QAT, Pallflex membrane filters, USA), using a high-102 volume sampler (Guangzhou Mingye Huanbao Technology Company, China) with a flow 103 rate of 1.0 m<sup>3</sup>/h. The quartz-fiber filters were equilibrated at a constant temperature ( $20\pm1$  °C) 104 and relative humidity (40±1%) environment for 24 h before weighing. Gravitative analyses 105 were conducted using an analytical balance (Mettler Toledo, Switzerland) with a precision of 106 10 µg. Detailed quality control/quality assurance (QC/QA) has been described in the 107 literature (Chow et al., 2015; Collett, 2016; Watson et al., 2017).

Hourly PM<sub>2.5</sub> water-soluble inorganic species (i.e., SO<sub>4</sub><sup>2+</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Cl<sup>-</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, 108 Na<sup>+</sup>, and Mg<sup>2+</sup>) and precursor gases (e.g., SO<sub>2</sub> and NH<sub>3</sub>) were measured by an online 109 MARGA (Monitor for Aerosols and Gases in Ambient Air, Model ADI2080, Metrohm 110 Applikon BV). The MARGA system consists of a steam-jet aerosol collector with a PM<sub>2.5</sub> 111 112 inlet where gases were removed by a wet rotating denuder and ions were captured and 113 dissolved into the supersaturated stream before analyzed by ion chromatograph (Chow and 114 Watson, 2017). Organic and elemental carbon (OC and EC) were acquired with an RT-4 115 analyzer (Sunset Lab, USA). A total of 16 trace elements (i.e., Fe, V, Cr, Mn, Co, Ni, Cu, Zn, 116 Ga, Ge, As, Se, Cd, Au, Ba, Pb) was monitored by a continuous multi-metals monitor 117 (Cooper Xact625) equipped with a reel-to-reel filter tape for nondestructive energy-dispersive 118 X-ray fluorescence (EDXRF) analysis. Due to missing value (> 20 %) (Yuan et al., 2008) for 119 trace elements at the PD site, 336 samples with 26 species at the DSL site were used for the 120 Positive Matrix Factorization (PMF) analysis.

Nitrogen oxides were measured using a chemiluminescence NO-NO2-NOx analyzer 121 (EC 9841B) with a minimum detection limit of  $0.4 \times 10^{-9}$  (volume fraction), while O<sub>3</sub> were 122 123 monitored using a UV spectrophotometry ozone analyzer (EC 9810B) with a minimum detection limit of  $0.4 \times 10^{-9}$ . Meteorological parameters including wind speed and direction, 124 125 temperature, relative humidity, pressure, and rainfall were monitored by an automatic 126 meteorological station (Met Station One Instrument, USA), which are placed 18 m above the 127 ground level on the rooftop of each Supersite. These measurements follows the QC/QA 128 procedure specified in the Technical Guideline of Automatic Stations of Ambient Air Quality 129 in Shanghai, which is based on the national specification HJ/T193-2005 (Ministry of Environmental Protection of China, 2005) and HJ/193-2013 (Ministry of Environmental 130 Protection of China, 2013). 131

# 132 **3. Results and discussion**

#### 133 *3.1 Meteorological conditions and atmospheric pollutant concentrations*

134 Fig. 2a-b show that the daily meteorological phenomena were similar among the three 135 years with a few exception. Temperatures remained moderate (26.8 °C) with low average 136 relative humidity (74%) except for the period of 8/28–8/30 in 2016. Atmospheric pressures 137 were lower during 8/29-9/1 in 2016. Wind speed was remaining low, averaging 1.9 m/s, 138 consistent with the 168-h back trajectories (see also in Fig. S2). The predominant northwesterly winds from 2014 to 2016 were 50.3%, 47.4%, and 44.1% at the PD site, and 139 140 48.8%, 41.6%, and 43.5% at the DSL site, respectively. Therefore, meteorological conditions 141 during the 2016 G20 period were not atypical, indicating that comparisons of air pollutant 142 concentrations can be made among the three years.

In the autumn harvest period, biomass burning can emit more than 60% of trace gases and particles emissions in the YRD region (Zha et al., 2013). Hence, Shanghai usually suffers from the greater impact of the pollutant transport from its adjacent Provinces. More abundant active fires still existed in 2016 from the neighboring regions which somewhat affect the pollution condition in Shanghai (see also in Fig. S3).



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**Fig. 2.** Comparison of meteorological parameters,  $PM_{2.5}$  concentrations (Box-and-Whisker Plot), and trace gaseous pollutants at the PD (a, c, and e) and DSL (b, d, and f) sites during the same period (8/24–9/6) from 2014 to 2016. A Box-and-Whisker plot: the mean (a black sphere), the median (a horizontal line in the box), the 25<sup>th</sup> percentile (the bottom edge of the box), the 75<sup>th</sup> percentile (represented by the top edge of the box), the minimum (the bottom edge of the whisker) and the maximum (the top edge of the whisker).

Fig. 2c–d show higher reductions in  $PM_{2.5}$  during the 2016 G20 period. Daily  $PM_{2.5}$ concentrations during 8/24–8/31 were low, averaging 24.0 µg/m<sup>3</sup> with a minimum of 10.6 µg/m<sup>3</sup> at PD, about 61.0% and 40.8% lower than these found in 2015 and 2014, respectively.

Lower PM<sub>2.5</sub> concentrations were also found for the DSL site during the last week of August 158 in 2016, averaging 31.2  $\mu$ g/m<sup>3</sup> with a minimum of 16.0  $\mu$ g/m<sup>3</sup>, about 66.0% and 48.6% lower 159 than 2015 and 2014, respectively. The prevailing northwestly winds enhanced the diffusion 160 of air pollutants. Prior to the commencement of G20 (9/2-9/3), PM<sub>2.5</sub> concentrations 161 increased to 74.8  $\mu$ g/m<sup>3</sup> at PD and 81.6  $\mu$ g/m<sup>3</sup> at DSL with some decreases on 9/4–9/6 in 162 2016. The DSL reginal site, is surrounded with green and agricultural lands in Fig.1, suffered 163 164 pollutant transport from the neighboring regions with clockwise transport from northwesterly 165 to northeasterly (see also in Fig. S2), contributing to a rise of pollutant concentrations.

166 Gaseous comparison excluded CO as CO concentrations remain below the Chinese 167 National Ambient Air Quality Standards (NAAQS) in recent years (Clean Air Asia, 2017). Not much changes in annual SO<sub>2</sub> concentrations ranged from 18.9  $\mu$ g/m<sup>3</sup> in 2014 to 15.9 168  $\mu g/m^3$  in 2016, below the NAAQS First Standard of 20  $\mu g/m^3$ . Annual NO<sub>2</sub> concentrations 169 170 ranged from 42.1  $\mu$ g/m<sup>3</sup> in 2014 to 40.6  $\mu$ g/m<sup>3</sup> in 2016, close to the NAAQS of 40  $\mu$ g/m<sup>3</sup>. 171 Consequently, no apparent changes in SO<sub>2</sub> were found during the same 14-day from 2014 to 2016 in Fig. 2e–f, with an average of 15.1  $\mu$ g/m<sup>3</sup> in 2014, 21.8  $\mu$ g/m<sup>3</sup> in 2015, and 16.6  $\mu$ g/m<sup>3</sup> 172 in 2016 at the PD site and 14.3  $\mu$ g/m<sup>3</sup> in 2014, 13.0  $\mu$ g/m<sup>3</sup> in 2015, and 12.8  $\mu$ g/m<sup>3</sup> in 2016 at 173 174 the DSL site. Due to regional transport by the prevailing northwesterly winds, concentrations 175 of SO<sub>2</sub> at PD were slightly higher than DSL, while NO<sub>2</sub> at PD was 9.4% higher than that at 176 DSL, reflecting the impact of motor vehicle emissions in urban areas.

177 On the contrary, concentrations of 8-h  $O_3$  elevated to higher instead of dropping during 178 the G20 period, with higher increases found at the PD site as compared to DSL. This is 179 consistent with those reported in recent studies (Li et al., 2018; Wang et al., 2015; Zhao et al., 180 2018), where inhibition of NO<sub>x</sub> on O<sub>3</sub> was more significant in urban areas.



Fig. 3. Diurnal variations of  $PM_{2.5}$ ,  $SO_2$ ,  $NO_2$ , and  $O_3$  at the PD (a) and DSL (b) sites on 9/4 from 2014 to 2016.

184 The diurnal variations on 9/4 (the first day of the 2016 G20 summit) of air pollutants were compared in Fig. 3. It is obvious that rush-hour peaks for PM<sub>2.5</sub>, SO<sub>2</sub>, and NO<sub>2</sub> found in 185 2014 and 2015 were diminished in 2016. PM2.5 and SO2 concentrations remained low 186 187 throughout the day at the PD and DSL sites. Different diurnal patterns were found for NO<sub>2</sub>, 188 peaked at 07:00 and 18:00 LST during the rush-hour traffic at the PD site but remained relatively lower ( $< 40 \ \mu g/m^3$ ) at the DSL site with accommodation found during late evening 189 190 (23:00 LST) to early morning (02:00 LST). Concentrations of ground level O<sub>3</sub> were affected by temperature, namely, high temperature in the daytime was favorable for  $O_3$  formation 191 192 (Xue et al., 2014). Diurnal curves of all monitored pollutants except for O<sub>3</sub> in the G20 period 193 were lower than those found in the previous two years.

194 **3.2** Changes in chemical composition of PM<sub>2.5</sub>

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# 195 3.2.1 Water–soluble inorganic ions and carbonaceous species

In Fig. 4a–b, concentrations of chemical composition of  $PM_{2.5}$  decreased during the 14day period in 2016 and their reductions varied between the PD and DSL sites. Total water-

soluble inorganic ions (TWSIIs) concentrations averaged 9.69  $\mu$ g/m<sup>3</sup> at PD and 13.0  $\mu$ g/m<sup>3</sup> at 198 DSL, which accounted for 56.2% and 52.3% of PM<sub>2.5</sub> mass in the G20 period (see also in Fig. 199 S4). Compared with the previous two years, TWSIIs decreased by at an average of 57.5% and 200 201 44.0% at PD and DSL, respectively. As for main secondary ionic aerosols (SIA), sulfate  $\mathrm{SO_4^{2^-}}$ , nitrate  $\mathrm{NO_3^-}$ , and ammonium  $\mathrm{NH_4^+}$  (SNA), averagely 41.5%, 68.8%, and 76.5% of 202 203 reductions were attained at PD and just 28.9% 29.5%, and 26.0% at DSL. SNA shared 88.3% 204 of TWSIIs at PD and 86.8% at DSL among the three years. Strong correlations between SIA and  $PM_{2.5}$  were observed at the PD (R = 0.87, n = 336, p < 0.01) and DSL (R = 0.82, n = 336, 205 p < 0.01) sites, respectively. These findings denoted that NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> were the 206 207 major ions in PM2.5 and SIA variations were associated with the formation, transportation and 208 removal of PM<sub>2.5</sub>.

No massive reduction in K<sup>+</sup> concentrations was at the PD site from 0.25  $\mu$ g/m<sup>3</sup> to 0.19 209  $\mu$ g/m<sup>3</sup>; whereas banning burning activities contributed to up to 89.6% reductions at the DSL 210 site. Fig. 4b shows apparent reduction of 44.2% for  $NH_4^+$  at the DSL site, suggesting the 211 212 effectiveness of precursor NH<sub>3</sub> emissions control from agricultural activities (Xu et al., 2015; Zhang et al., 2018). However, NH<sub>4</sub><sup>+</sup> exists in compound form through the combination of 213  $SO_4^{2-}$  and  $NO_3^{-}$ . It is possible that the reductions of  $SO_4^{2-}$  and  $NO_3^{-}$  are associated with the 214 215 NH<sub>4</sub><sup>+</sup> decreases (Collett, 2016; Zhang et al., 2018) in the urban environment and the higher concentrations of  $NH_4^+$  at the regional DSL are complicated rather than singly from the local. 216 217 Na<sup>+</sup> concentrations at the PD site were 5 times higher than those at DSL, implying the impact from sea salt at the urban site in Shanghai (Qiao et al., 2016). Reductions of Na<sup>+</sup>, Ca<sup>2+</sup>, 218

and  $Mg^{2+}$  corresponded to dust controls in construction activities and steel sweeping (Xiu et al., 2004; Zhang et al., 2015). Water soluble Ca<sup>2+</sup> accounted for 1.7% to 2.0% of TWSIIs, lower than the 6.8% in Wuhan (Zhang et al., 2015), 4.0% in Zhengzhou (Geng et al., 2013), PD and DSL sites (R = 0.28, n = 336, p < 0.05), suggesting differences in geological composition between the urban and regional areas. Yang et al. (2018) reports high Cl<sup>-</sup>/Na<sup>+</sup> ratios (2.46–5.0) in northern and western China. These ratios were about an order of magnitude higher than those found at PD (0.44) but similar to 2.31 found at DSL. The lower Cl<sup>-</sup>/Na<sup>+</sup> ratio at PD could be ascribed by curtailing coal-oriented consumptions in urban Shanghai (He et al., 2001).



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Fig. 4. Comparison of water-soluble inorganic ions (WSIIs) (a and b), carbonaceous species (c and d), and SOC estimation (e and f) in  $PM_{2.5}$  at the PD and DSL sites during the same period (8/24–9/6) from 2014 to 2016. Pie charts (inset of e and f): changes in the fractions of POC and SOC in OC concentrations at both sites.

Carbonaceous aerosol (TC) is the major component of PM<sub>2.5</sub> and consists of two components, i.e., organic carbon (OC) and elemental carbon (EC). OC could be divided into primary organic carbon (POC) and secondary organic carbon (SOC). POC is emitted directly in the particle-phase and SOC is formed from gas-to-particle conversion in the atmosphere. The presence of SOC can be estimated by the minimum OC/EC ratio (Huang et al., 2012; Zhang et al., 2012):

$$POC = EC \times \left(\frac{OC}{EC}\right)_{min} \tag{1}$$

$$SOC = OC - POC \tag{2}$$

Where (OC/EC)<sub>min</sub> is the minimum OC/EC ratio. The concentrations of OC (POC and SOC) 240 241 and EC, as well as OC/EC ratios of PM<sub>2.5</sub> at the PD and DSL sites are illustrated in Fig. 4c-f. Average TC concentrations were 3.51  $\mu$ g/m<sup>3</sup> and 6.03  $\mu$ g/m<sup>3</sup> during the 14-day period in 242 243 2016 and accounted for 14.5% and 19.3% of PM<sub>2.5</sub> mass (see also in Fig. S4) at PD and DSL, respectively. OC accounted for 71.0%–77.2% of TC, ranging from 7.13  $\mu$ g/m<sup>3</sup> to 7.39  $\mu$ g/m<sup>3</sup> 244 at PD and 1.85  $\mu$ g/m<sup>3</sup> to 2.19  $\mu$ g/m<sup>3</sup> at DSL during the same 14-day period from 2014 to 2016. 245 246 Good correlations were found between OC and  $K^+$  (R = 0.97, n = 336, p < 0.01) at the DSL site, suggesting their common emission sources (Li et al., 2015) and the association with 247 248 fire events (see also in Fig. S3). In Fig. 4c-d, average OC/EC ratios decreased from 3.9 in 249 2014, 3.6 in 2015 to 2.2 in 2016 at the PD site, more pronounced than those found at the DSL 250 site from 2.7 in 2014 to 2.1 in 2016. In Fig. 4e-f, average SOC during the G20 period were 2.7  $\mu$ g/m<sup>3</sup> and 3.6  $\mu$ g/m<sup>3</sup>, accounted for 26.4% and 33.9% of OC at the PD and DSL sites, 251 252 respectively. The SOC/OC ratios shared less variations at DSL (29.7% to 36.2%) than PD (26.4% to 56.6%). PM<sub>2.5</sub> shows higher correlations with SOC (R = 0.85, n = 336, p < 0.01) at 253 254 PD and (R = 0.75, n = 336, p < 0.01) at DSL than with POC (R = 0.53, n = 60, p < 0.01) at PD and (R = 0.66, n = 336, p < 0.01) at DSL. These findings reflect that organic components 255 256 in the atmosphere were related to the formation, transportation and removal of PM<sub>2.5</sub> in

Shanghai (Huang et al., 2012; Liang et al., 2016). Large reductions in SOC/OC ratios at the PD site during the G20 period were more pronounced than those found at DSL. Weak correlations between OC and EC (R = 0.32, n = 336, p < 0.05) at PD and (R = 0.28, n = 336, p < 0.05) at DSL indicate that they derived from different sources (Li et al., 2012; Tao et al., 2014).

#### 262 *3.2.2 Divergence analysis between two sites*

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Coefficient of divergence (CD) can be utilized to characterize the differences of chemical components between the two sampling sites, calculated by the following equation (Zhang and Friedlander, 2000):

$$CD_{AB} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (\frac{C_{iA} - C_{iB}}{C_{iA} + C_{iB}})^2}$$
(3)

Where  $C_{iA}$  and  $C_{iB}$  represent the concentrations of component *i* at site A and site B, respectively. If the CD is approximate to 0.00, the emission sources between the two sites are similar; whereas close to 1.00, the two sites are different (Zhang and Friedlander, 2000). This approach was adopted by (Yang et al., 2002) in Beijing (CD = 0.064) and (Zhang et al., 2015) in Wuhan (CD = 0.098), indicating that the similarity of PM<sub>2.5</sub> compositions. In contrast, the values of CDs between PD and DSL ranged 0.52–0.56 from 2014 to 2016 as shown in Fig. 5., which supports the investigation of the complicated impacts between urban and regional sites.





Fig. 5. Coefficient of divergence (CD) that characterizes the differences of  $PM_{2.5}$  chemical components between PD and DSL during the same period (8/24–9/6) from 2014 to 2016.

# 276 **4. Effectiveness of the pollution control measures**

Annual mean concentrations of  $PM_{2.5}$ ,  $SO_2$ , and  $NO_2$  reduced by 14.1%, 21.9%, and 11.4%, respectively, in 2016 as compared to 2014 and 2015. Though the 14-day pollution control measures imposed during the G20 period, concentrations of  $PM_{2.5}$  mass, carbon, and ions (especially  $SO_4^{2+}$ ,  $NO_3^{-}$ , and  $NH_4^{+}$ ) decreased significantly as compared to the same period in the previous two years shown in Fig.6. Higher number of the air quality attainment days were achieved (see also in Fig. S5). No improvement was found for  $O_3$  at either the PD or DSL sites.

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Fig. 6. Comparison of concentrations of air pollutants ( $PM_{2.5}$  mass, carbon, ions and trace gaseous pollutants) at the PD (a) and DSL (b) sites during the same period (8/24–9/6) from 2014 to 2016.

289 Seven sources were identified by the Positive Matrix Factorization (PMF) model (see also in Fig. S6) for PM<sub>2.5</sub> samples at the DSL site, including biomass burning, resuspended 290 291 dust, high-temperature combustion, iron and steel industry, sea salt, secondary aerosol, and 292 vehicle exhaust. Figs. 7 shows changes in temporal variations in PM<sub>2.5</sub> and source 293 contribution among the three years. The two largest contributors were secondary aerosols and 294 vehicle exhaust among the three years, which accounted for 45.7% of PM<sub>2.5</sub> mass, followed 295 by iron and steel industry (13.7%), resuspended dust (12.3%), and high-temperature 296 combustion from coal and oil boilers (11.2%). Biomass burning ranged from 5.1% to 7.9% of 297 PM<sub>2.5</sub> and was attributed to the prevailing northwesterly transports, also suppoted by the back 298 trajactories (see also in Fig. S2) and active fire detections (see also in Fig. S3).

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Fig. 7. Changes in source apportionment resolved from PM<sub>2.5</sub> samples at the DSL site during
the same period (8/24–9/6) from 2014 to 2016.

# 303 5. Conclusions

304 Implementation of stringent pollution control measures over the 14-day period resulted in large reduction in pollution concentrations. The effectiveness of the pollution control in the 305 306 urban center was more pronounced than regional areas. If the onset of unfavorable meteorological conditions is forecasted, the enhanced reduction measures would be 307 308 implemented several days ahead of pollution episodes. Since the impact of multiple 309 pollutants on PM<sub>2.5</sub> and O<sub>3</sub> concentrations and atmospheric oxidation capacity is nonlinear 310 and complicated, reduction in precursor gases (e.g., SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and VOCs) should be 311 synergistically strengthened for the next stage. However, its long-term impacts on ambient 312 concentration levels need to be further evaluated. A market-based approach needs to be 313 examined to evaluate the cost of an ad-hoc closures of industries. The profit and loss between 314 the environment and business need to be balanced.

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### 322 Appendix A. Supplementary Information

Supplementary Information associated with this article can be found in this section, including: 14-day pollution control measures, distribution of Air Quality Index (AQI) in Shanghai from 2014 to 2016, NOAA HYSPLIT trajectory model results, active fire detection in neighboring regions, fractional abundance of five major species of PM<sub>2.5</sub>, changes in air quality attainment during the G20 period, and source identification PM<sub>2.5</sub> samples using PMF model.

329 **Notes** 

330 The authors declare no competing financial interest.

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### 443 **Figure Captions**

- Fig. 1. Locations of the Pudong (PD) and Dianshan Lake (DSL) Supersites in Shanghai. The
  contour of land use and population density adopted from Shanghai statistical yearbook
  in 2015 (Bureau of Statistics of Shanghai, 2016).
- **Fig. 2.** Comparison of meteorological parameters,  $PM_{2.5}$  concentrations (Box-and-Whisker Plot), and trace gaseous pollutants at the PD (a, c, and e) and DSL (b, d, and f) sites during the same period (8/24–9/6) from 2014 to 2016. A Box-and-Whisker plot: the mean (a black sphere), the median (a horizontal line in the box), the 25<sup>th</sup> percentile (the bottom edge of the box), the 75<sup>th</sup> percentile (represented by the top edge of the box), the minimum (the bottom edge of the whisker) and the maximum (the top edge of the whisker).
- 454 **Fig. 3.** Diurnal variations of  $PM_{2.5}$ ,  $SO_2$ ,  $NO_2$ , and  $O_3$  at the PD (a) and DSL (b) sites on 9/4 455 from 2014 to 2016.
- 456 **Fig. 4.** Comparison of water-soluble inorganic ions (WSIIs) (a and b), carbonaceous species 457 (c and d), and SOC estimation (e and f) in  $PM_{2.5}$  at the PD and DSL sites during the 458 same period (8/24–9/6) from 2014 to 2016. Pie charts (inset of e and f): changes in the 459 fractions of POC and SOC in OC concentrations at both sites.
- Fig. 5. Coefficient of divergence (CD) that characterizes the differences of PM<sub>2.5</sub> chemical components between PD and DSL during the same period (8/24–9/6) from 2014 to 2016.
  Fig. 6. Comparison of concentrations of air pollutants (PM<sub>2.5</sub> mass, carbon, ions and trace gaseous pollutants) at the PD (a) and DSL (b) sites during the same period (8/24–9/6) from 2014 to 2016.
- 465 Fig. 7. Changes in source apportionment resolved from PM<sub>2.5</sub> samples at the DSL site during
  466 the same period (8/24–9/6) from 2014 to 2016.



**Fig.1** 



- 470 Fig. 2





















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