Substantial reductions in ambient PAHs pollution and lives saved as a co-benefit of effective long-term PM2.5 pollution controls

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1 **Substantial reductions in ambient PAHs pollution and lives saved as a co-benefit of effective**

2 **long-term PM$_{2.5}$ pollution controls**

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Abstract:

Under great efforts in fighting against serious haze problem of China since 2013, decreasing of air pollutants especially for fine particles ($PM_{2.5}$) has been revealed for several key regions. This study tried to answer whether the reduction of $PM_{2.5}$-bound polycyclic aromatic hydrocarbons (PAHs) was coincident with $PM_{2.5}$ because of long-term pollution control measures (PCM), and to assess source-oriented health risks associated with inhalation exposure to PAHs. Field measurements were carried out before and after the publishing of local air pollution protection plan for Nanjing, a mega-city in east China. Results indicated that the air quality was substantially improving, with a significant reduction in annual average $PM_{2.5}$ by 34%, and moreover, $PM_{2.5}$-bound PAHs significantly reduced by 63% ($p<0.001$). The remarkable reduction was mainly attributable to the change of emission sources, compared to the influence of atmospheric circulation patterns, surface meteorological conditions, and atmospheric chemical reaction. Four PAHs sources including coal combustion (CC), petroleum and oil burning (PO), wood burning (WB) and vehicle emission (VE) were identified. On an annual basis, contributions to ambient $PM_{2.5}$-PAHs from WB, PO, CC and VE sources in the period before the action of control measures were 2.26, 2.20, 1.96 and 5.62 ng m^{-3}, respectively. They reduced to 1.09, 0.37, 1.31 and 1.77 ng m^{-3} for the four source types, with the reduction percentages as 51, 83, 33 and 68%, respectively. The estimated reduction in lifetime lung cancer risk was around 61%. The study that firstly assessed the health effects of PAHs reduction as a co-benefit raised by air PCM sustained for a long period is believed to be applicable and referential for other mega-cities around the world for assessing the benefits of PCM.

1. Introduction

China is one of the regions holding the highest atmospheric fine particle ($PM_{2.5}$) concentrations of the world (Donkelaar et al., 2015; Cheng et al., 2016). The Chinese government has been trying to mitigate air pollution for nearly thirty years since the 1980s (Feng and Liao, 2016). In 2012, when the new ambient air quality standard was released, $PM_{2.5}$ pollution has attracted worldwide attention for both scientists and decision makers, especially after the extremely severe and long-lasting haze pollution event occurred in January 2013 (Fu and Chen, 2017; Wang et al., 2017a). In September 2013, the national "Action Plan on Atmospheric Pollution Prevention and Control (APPC)" was promoted, in which the main reduction measures for $PM_{2.5}$ emphasized mainly on coal combustion, industrial manufacturing processes, urban/suburban fugitive dust, cooking activities, vehicle exhaust and fuel quality. It was predicted the implementation of these measures could reduce $PM_{2.5}$ emission by 30% in 2017 relative to 2012 in the Beijing-Tianjin-Hebei region (Cai et al., 2017). It is important and of growing interests to assess the effectiveness of pollution control measures (PCM) on air quality, either for a long-term period (Chen et al., 2016a; Wang et al., 2017a, b), short-term serious air pollution episodes (Xu et al., 2016a; Wang et al., 2017c), or during specific short-term mega-events like APEC (Guo et al., 2016; Wang et al., 2017d). However, to our knowledge, few researchers concerned the role of PCM in potential reductions of toxic chemical components in $PM_{2.5}$, such as heavy metals (Chen et al., 2016b; Li et al., 2016b), black carbon (BC) (Chen et al., 2016c)/elemental carbon (EC) (Wang et al., 2017b) and polycyclic aromatic hydrocarbons (PAHs) (Wang et al., 2011; Xu et al., 2013), which have significant health impacts.

PAHs are a group of organic contaminants formed in nearly all in-complete combustion processes and can be long-range transported in the air (Shrivastava et al., 2017). Carcinogenic and teratogenic PAHs are associated with many adverse health outcomes like increased levels of oxidative stress (Bae et al., 2010), gastroschisis (Lupo et al., 2012), ischaemic heart disease (Burstyn et al., 2005), systemic inflammation (Delfino et al, 2010), adverse health symptoms in survivors of myocardial infarctions (Kraus et al, 2011) and children's cognitive development (Edwards et al., 2010), obesity (Scinicariello and Buser, 2014) and behavior development (Perera et al., 2012). Since the implementation of APPC from 2013 of China (totally ten types of measures including thirty-five sub-items were published in APPC), sources of $PM_{2.5}$ changed significantly.

90   For PCM of APPC, coal combustion, industrial processes, cooking activities, biomass

91   burning, and vehicle emissions are in fact the key sources for atmospheric PAHs (Zhang and

92   Tao, 2009). Therefore, it is believed that PAHs sources may exhibit notable changes as well.

93   It is essential to access to what extent the PAHs concentrations and associated health risk that

94   could be reduced when these control measures for $PM_{2.5}$ are implemented at a certain city or

95   region, and it is interesting to clarify whether the decreasing trends of $PM_{2.5}$ and PAHs are

96   same or not. Investigation into the main sources leading to the decreasing of PAHs

97   concentrations and risks and identification roles of influencing factors like meteorological

98   conditions, photochemical reaction and source changes in the variations of PAHs are valuable

99   for the evaluation of PCM. It is also informative for the future researches and policymaking.

100   Since 2013, the four-year solid efforts to reduce source emissions of air pollutants

101   especially for $PM_{2.5}$ in Nanjing (an industrial city and one of the central megacities located in

102   the Yangtze River Delta (YRD) region) provided a unique opportunity to study the variation

103   in $PM_{2.5}$-bound PAHs concentrations, as well as the associated risks. A campaign from 2013

104   to 2016 covered two distinguished periods of March 2013-February 2014 (Period 1) and

105   November 2015-July 2016 (Period 2), that was before and after the AR-APPC

106   (Administrative Rules of Nanjing for further strengthening the APPC) in Nanjing. The

107   AR-APPC was proposed at February 2014 and issued from April 2015. In the APPC of

108   Nanjing published of 2014, six types of control measure were proposed, including optimizing

109   the industrial structure and layout, improving energy structure, strengthening industrial

110   pollution prevention and control, developing green transportation, control fugitive dust, and

111   control pollution from agricultural and human activities (**Table S1**). The initial target of this

112   AR-APPC is to reduce the annual mass concentration of $PM_{2.5}$ in Nanjing by 7%, 13% and

113   20% in 2015, 2016 and 2017 relative to that in 2013, respectively.

114   The main objectives of this paper are: (1) to evaluate the reduction of air pollutants and

115   PAHs in Nanjing; (2) to identify the variation in PAHs concentrations, markers, chemical

116   degradation and sources; (3) to interpret the reasons for PAHs variation in view of source

117   emission and meteorological conditions; and (4) to quantify the potential health risk

118   reductions of PAHs and allocated it to various sources. To our knowledge, this is the first and

119   maybe the only research to evaluate the PAHs concentrations of the four years since 2013 in

4

120 China. It is helpful to expand the understanding of co-benefits of PCM for $PM_{2.5}$, which has

121 important enlightenment for regional or city-level assessment of air quality improvement.

122 **2. Method**

123 **2.1 Site description and $PM_{2.5}$ sampling**

124   Ambient $PM_{2.5}$ samples were collected on top of a seven-floor building (~20 m above the

125 ground) of Jiangsu Academy of Environmental Sciences. In Period 1, samples were collected

126 during March, June, and September in 2013 and January in 2014, to represent four seasons of

127 spring, summer, autumn and winter. In each season, the sampling lasted for about one week. The

128 sampling flow was ~16.7 L min^{-1} (Wuhan Tianhong Ltd., China). In Period 2, $PM_{2.5}$ samples were

129 collected on a rooftop of Jiangsu Environmental Monitoring Center (JEC, a six-floor building,

130 about 15 m above the ground) at a flow rate of ~100 L min^{-1} (TH-150C, Wuhan Tianhong Ltd.,

131 China) (Kong et al., 2015a, 2015b) during November of 2015 and January, April and July of 2016.

132 The sampling lasted for about 7-14 days in each season. The two sampling sites are 0.5 km away

133 in the distance, at two close blocks. It is a typical traffic/residential region of urban Nanjing, with

134 residential communities surrounded. There is a steel factory about 15 km to the north of the site

135 and two chemical industry parks about 20 km to the north and northwest. Within the distance from

136 20 km of the sampling sites, some power plants and industrial plants existed, mostly concentrated

137 in the north, west and south direction. Location of the sampling sites and surrounding major

138 sources are shown in **Figure. 1**.

139   Each day, the $PM_{2.5}$ sampling lasted for ~24 h (starting from about 08:00) for both the two

140 periods, with quartz fiber filters used. Filters were sealed in aluminum foil bags and weighted by a

141 microbalance (Ohaus Discovery DV214CD) with the balance under controlled environment and

142 stored under -20 °C until laboratory analysis. The detailed treatment method of the filters was

143 listed in Kong et al. (2015a, 2015b).

144 **2.2 Meteorological and air quality data acquisition**

145   The meteorological parameters including ambient temperature (T), relative humidity (RH),

146 wind speed (W_S) and visibility (V_S) were obtained from the Nanjing Meteorological Bureau. The

147 precipitation information was collected from http://www.wunderground.com/. The daily averaged

148 mass concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3 were collected from the supersite on

149 the rooftop of JEC. The site is the same location for the $PM_{2.5}$ sampling site at Period 2. Organic

5

150 carbon (OC) and elemental carbon (EC) were monitored by Sunset Laboratory

151 Semi-Continuous OC/EC Carbon Aerosol Analyzer (Sunset-OCEC, RT-4) in Jiangsu

152 Environmental Center and the corresponding data for sampling periods were provided. For

153 assessing the air quality variation in Nanjing, the daily averaged mass concentrations of

154 $PM_{2.5}$, PM_{10}, SO_2, NO_2 and CO for nine air quality monitoring sites (the nine sites can be

155 found at http://106.37.208.233:20035/) were provided from 1/January/2013 to

156 30/December/2016. The 8-hours moving average value of O_3 was used for each day.

157 **2.3 Laboratory PAHs analysis and quality controls**

158   Laboratory PAHs analysis was the same as that described in Kong et al. (2015b).

159 Briefly, filters were extracted ultrasonically with dichloromethane, concentrated using a

160 rotary evaporator, and then transferred to a silica gel for cleanup and purification. The elutes

161 were finally concentrated to ~1 mL under a gentle nitrogen stream and then spiked with

162 internal standards prior to instrument analysis. PAHs analyzed by a trace 2000 GC-MS

163 (Thermo Finnigan, USA) operated in selected ion monitoring (SIM) model. Totally 18 PAHs

164 were detected including naphthalene (NaP, 2-ring), acenaphthylene (Acy, 3-ring),

165 acenaphthene (Ace, 3-ring), fluorene (Fl, 3-ring), phenanthrene (Phe, 3-ring), anthracene

166 (Ant, 4-ring), fluoranthene (Flu, 4-ring), pyrene (Pyr, 4-ring), benzo[a]anthracene (BaA,

167 4-ring), chrysene (Chr, 4-ring), benzo[b]fluoranthene (BbF, 5-ring), benzo[k]fluoranthene

168 (BkF, 5-ring), benzo[a]pyrene(BaP, 5-ring), Benzo(e)pyrene (BeP, 5-ring),

169 dibenz[a,h]anthracene (DBA, 6-ring), indeno[1,2,3-cd]pyrene (InP, 6-ring),

170 benzo[ghi]perylene (BghiP, 6-ring) and coronene (Cor, 7-ring). 2 and 3-ring PAHs are

171 attributed to low molecular weight PAHs (LPAHs). 4-ring PAHs belong to medium

172 molecular weight PAHs (MPAHs) and high molecular weight PAHs (HPAHs) contain 5-, 6-

173 and 7-ring PAHs. Two types of PAHs are also paid attention, named as combustion-derived

174 PAHs (COMPAHs), including Flu, Pyr, Chr, BbF, BkF, BaA, BeP, BaP, InP and BghiP and

175 carcinogenic PAHs (C-PAHs) including Chr, BaA, BbF, BkF, BaP, InP and DBA (Kong et

176 al., 2015b; Wang et al., 2015).

177   For quality assurance and quality control, field blank and laboratory blank filters were

178 treated following the same procedure in regular samples. Quantification of PAHs was done

179 by the retention times and peak areas of the calibration standards. Internal standard method

180 was used. Reserve liquid (1000 mg/L) including Perylene-d12, Chrysene-d12, Acenaphthene-d12,

181 Naphthalene-d8 and Phenanthrene-d10 was diluted by n-hexane to 20 mg/L and was stored below

182 4°C. The recovery test was performed by spiking known amounts of a mixture of PAHs and then

183 the spiked filter was treated the same way as mentioned above. The recoveries of each PAHs were

184 in 81%-93%, and the relative standard deviation was less than 10%. The detection limits for the 18

185 kinds of PAHs ranged between 3.0-10.0 ng. Results reported were subtracted from blanks, but not

186 corrected by recoveries.

187 **2.4 Back trajectory and mixing layer height calculation**

188 Backward air mass trajectories (72 h) with the starting height of 500 m are calculated using

189 NOAA Air Resource Lab HYSPLIT 4.8 model, driven by the GDAS meteorological dataset

190 (1°×1°) (Wang et al., 2016b; Ye et al., 2017). To achieve the sub-sets of trajectories, the backward

191 trajectories computed every six hour (00:00, 06:00, 12:00 and 18:00) each day were clustered.

192 Clustering process is listed in Hysplit User's Guide-Version 4. On the NOAA's READY Archived

193 Meteorology online calculating program (http://ready.arl.noaa.gov/READYamet.php), the mixing

194 layer height (MLH) was calculated every three hour per day.

195 **2.5 PAHs source apportionment**

196 Positive matrix factorization (PMF) model is adopted to investigate PAHs sources. As a

197 multivariate factor analysis tool, it decomposes a matrix of speciated sample data into factor

198 contribution matrix (G) and factor profile matrix (F), and is expressed as follows:

199
$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (1)$$

200 where x_{ij} is the concentrations of jth PAHs species in the ith sample; g_{ik} is the contribution of

201 the kth source to the ith sample; f_{kj} is the mass fraction of the jth compound from the kth source; e_{ij}

202 is the residual for each sample/species. US Environmental Protection Agency PMF version 5.0

203 was adopted. Detailed PMF running description was listed in Supplementary file S1 **(including**

204 **Figure S1, Table S2-S4)**.

205 **2.6 Potential source contribution function and concentration weighted trajectory**

206 Potential source contribution function (PSCF) and concentration weighted trajectory (CWT)

207 were adopted to evaluate the influence of regional PAHs sources and were calculated with the

208   software Traj-Stat, based on the daily source contributions and backward trajectories.

209   Weighted-PSCF (WPSCF) values were calculated by multiplying PSCF values with a

210   weighting function. A concentration-weighted-trajectory (CWT) model was introduced to

211   add additional information on source contribution levels of different potential regions.

212   WPSCF and weighted CWT (WCWT) were adopted for discussion. The detailed description

213   of PSCF and CWT calculation was listed in **Supplementary file S2**.

214   **2.7 Health risk assessment**

215   Potential health risks of PAHs via inhalational exposure have been widely characterized

216   by calculating incremental lifetime cancer risk (ILCR) as:

217    $$ILCR = UR_{BaP} \times BaP_{eq} \qquad (8)$$

218    $$BaP_{eq} = \sum_{i=1}^{n}(C_i \times TEF_i) \qquad (9)$$

219   where UR_{BaP} is the unit cancer risk factor of BaP, as 8.7×10^{-5} per ng m^{-3} (Callén et al.,

220   2014; Chen et al., 2016a; Yu et al., 2016; Pereira et al., 2017); C_i is the concentration of

221   PAHs species (ng m^{-3}); $BaPeq$ is the BaP equivalent concentration (ng m^{-3}), which is

222   calculated by multiplying the C_i with their corresponding toxic equivalent factor (TEF_i). The

223   TEF_i was selected as 0.001 for NaP, Acy, Ace, Fl, Phe, Flu and Pyr, 0.01 for Ant, Chr, BeP

224   and BghiP, 0.1 for BaA, BbF, BkF, and InP, and 1 for BaP and DBA (Khan et al., 2015; Zhu

225   et al., 2015; Chen et al., 2017).

226   **3 Results and discussion**

227   **3.1 Criteria air pollutants and meteorological conditions before and after the AR-APPC**

228   In **Table 1**, there were no significant differences in average ambient temperature, Ws,

229   RH and MLH between the two periods, while visibility significantly decreased by 34%

230   ($p<0.001$) in Period 2. The average concentrations of PM$_{2.5}$, PM$_{10}$ and O$_3$ for the Period 2

231   significantly reduced by 34% ($p<0.001$), 34% ($p<0.001$) and 18% ($p<0.05$), respectively, in

232   comparison with those for Period 1 (**Table 2**). There were no significant changes of the other

233   criteria gaseous pollutants like CO, SO$_2$, and NO$_2$. From the evaluation report on air quality

234   published by Ministry of Environmental Protection of China, reduction of PM$_{2.5}$ could be

235   clearly found for YRD region from 2013 to 2016 (**Figure S2**). As expected, the average

236   annual mass concentrations of PM$_{2.5}$, PM$_{10}$, SO$_2$, and NO$_2$ in Nanjing were all decreasing

237   gradually from 2013 to 2016, by 38%, 38%, 50% and 20%, respectively (**Figure 2**). It has

238   successfully achieved the goal of 20% reduction for $PM_{2.5}$ proposed in the AR-APPC. The

239   reduction rate of $PM_{2.5}$ for the whole Nanjing was similar with that for the given site and sampling

240   periods of this study. Though the decreasing rate was high to 59% for the $PM_{2.5}$ in the winter

241   season of the two periods, $PM_{2.5}$ concentration in winter was still highest as 72.5 ± 37.7 μg m^{-3} in

242   Period 2. It indicated that the current regulations for winter should be enforced more rigorously,

243   exactly and beneficially.

244   **3.2 Comparison of ambient PAHs before and after the AR-APPC**

245   As shown in **Figure. 3**, the overall averaged PAHs concentrations were significantly reduced

246   by 63% (*p*<0.001) at Period 2, higher than that of 34% for $PM_{2.5}$ mass concentrations. The

247   averaged COMPAHs and C-PAHs exhibited concentration reductions of 55% and 60% (*p*<0.001).

248   Significant reductions were also observed for the averaged concentrations of each PAHs

249   individual, ranging from 45% to 87% (*p*<0.01). To eliminate the influence of meteorological

250   conditions, PAHs mass concentration was normalized by $PM_{2.5}$ concentration (Anastasopoulos et

251   al., 2012; Kong et al., 2015b; Wang et al., 2015) and Ws (Wang et al., 2016a). Ratios of

252   PAHs/$PM_{2.5}$ and PAHs/Ws were statistically lower for Period 2, with the decreasing rates of 82%

253   (*p*<0.01) and 57% (*p*<0.001), respectively. This confirmed the substantial decrease of PAHs for

254   Period 2 and the decreasing rate of PAHs was larger than that of $PM_{2.5}$. It indicated that the PCM

255   for $PM_{2.5}$ had played a positive role in PAHs reduction. Now the annual averaged PAHs

256   concentrations of Nanjing (4.61 ± 3.29 ng m^{-3}) were comparable to the reported levels in

257   Thessaloniki (4.66 ng m^{-3}) (Saffari et al., 2013), Hong Kong (4.59 ng m^{-3}) (Ma et al., 2016) and

258   Shanghai (6.49 ng m^{-3}) (Wang et al., 2016b) and were lower than most of those in other cities as

259   compiled in **Table S5**.

260   As the most carcinogenic PAHs, the averaged concentrations of BaP decreased by 57%

261   (P<0.001) for Period 2 when compared with that of Period 1 (**Table 3**). The ambient BaP in urban

262   Nanjing was continuously decreasing during the last two decades. The averaged BaP in 2001 was

263   8.05 ng m^{-3} (September, an urban site) (Yang et al., 2005), 4.17 ng m^{-3} in 2001/2002 (five urban

264   sites, whole year) (Wang et al., 2006a), 3.83 ng m^{-3} in 2004 (summer and winter, an urban site)

265   (Wang et al., 2007), 3.73 ng m^{-3} in 2009/2010 (from November 2009 to July 2010, one urban and

266   one suburban site) (He et al., 2014), 0.65 ng m^{-3} in 2013/2014 (this study) and 0.27 ng m^{-3} in

267 2015/2016 (this study), respectively. The annual averaged guideline value of BaP

268 recommended by the World Health Organization and European Union was 1 ng m^{-3}

269 (Ravindra et al., 2008; Wang et al., 2017d). The annual BaP concentration of Nanjing is now

270 below the guideline value. From 2001/2002 to 2009/2010, the variation of BaP is little,

271 decreased by 10.5%. While from 2009/2010 to 2013/2014, obvious decreasing by 83% was

272 found. We can deduce that the reduction of PAHs may be a longer-term trend in the first

273 decade. While after 2009/2010, the reduction rate accelerated. There should be other forcing

274 favored it, which should be the proposed PCM for air pollutants after 2013. The effective

275 PAHs reduction along with the air pollution control strategies during short-term mega-events

276 was also reported (Wang et al., 2011; Xu et al., 2013; Li et al., 2016c).

277 From Table 3, clear seasonal variations were observed, with highest PAHs

278 concentrations in winter and lowest concentrations in summer. The winter/summer ratios of

279 PAHs were averaged as 3.3 and 4.7 for Period 1 and Period 2, respectively. PAHs/PM$_{2.5}$

280 exhibited the similar winter and summer variation. This seasonal variation pattern has been

281 widely reported in former studies (Marchand et al., 2004; Wang et al., 2006b; Akyüz and

282 Çabuk, 2008; Guo et al., 2009; Martellini et al., 2012; Mancilla et al., 2016; Yu et al., 2016).

283 Following reasons could explain the seasonal variations in PAHs in this study: (1) additional

284 emissions in winter from residential heating with coal and wood as fuels in North China

285 (Wang et al., 2006b; Chen et al., 2016a; Lin et al., 2015; Lv et al., 2016), which can transport

286 and affect air quality of Nanjing (detailed discussion in Section 3.4.2 and 3.4.3); (2) reduced

287 atmospheric dispersion due to inversion and lower MLH in winter (as 312±124 and 445±187

288 m for winter of the two periods, respectively); (3) enhanced partition to particles under lower

289 temperature; (4) less pronounced photochemical degradation processes in winter as low

290 ambient temperature (discussion in Section 3.3.2) (Marchand et al., 2004; Chen et al., 2016a).

291 The change exited in not only total PAHs mass concentration but also the PAHs

292 composition profile. It can be observed that the overall averaged mass percentage of LPAHs

293 (2 and 3 ring PAHs) decreased from 25% to 14% (decreased by 44%, *p*<0.001), the overall

294 averaged MPAHs (4 ring PAHs) increased from 22% to 32% (increased by 44%, *p*<0.001)

295 and the HPAHs (5, 6 and 7 ring PAHs) varied a little bit (from 53% to 54%) **(Figure 3)**.

296 Generally, higher LPAHs concentration was related to non-combusted petroleum emission

297 and HPAHs were mainly from fossil fuel combustions (He et al., 2014). It can be inferred that

298 petroleum emission of PAHs reduced after the AR-APPC.

299 **3.3 Difference in meteorological conditions, atmospheric chemical degradation and sources**

300 **between the two periods**

301 **3.3.1 The role of circulation patterns, surface meteorological parameters and long-range**

302 **transportation**

303   The circulation patterns can influence the air quality through transport pathways and

304 determine local meteorology (Xu et al., 2016a). The circulation patterns (seasonal mean

305 geopotential height and wind vectors at 10 m above sea level) for four seasons of the two periods

306 were illustrated in **Figure S3**. At first glance, the circulation patterns of the two periods were

307 similar for all the four seasons. In autumn and winter, Nanjing was under control of Siberian high

308 pressure and it was stronger for Period 2 than that of Period 1, which was the typical features of

309 East Asian winter (Kim et al., 2016). The main backward trajectory clusters of the four seasons

310 were also similar for the two periods (**Figure S4**), with north and northwest trajectories dominated

311 in winter and autumn. Thus, the air pollutants at Nanjing were easily affected by the upstream

312 transport at the northwest direction and the impact was more profound in Period 2 at the two

313 seasons. It should be noted that in later autumn and winter period, the North China area exhibits

314 high coal consumptions (**Figure S5**) and high emission densities of many air pollutants from

315 biomass burning (Zhou et al., 2017). Meanwhile, weak negative correlations were found between

316 PAHs and temperature and MLH, especially for higher rings PAHs (**Table S6, Figure S6**). As

317 shown in Figure S6, high rings PAHs also exhibited weak positive correlations (most P values

318 lower than 0.01) with SO_2, NO_2, OC, and EC, that originated mainly from combustion sources

319 (Thornhill et al., 2008; Mancilla et al., 2016). All these can verify the higher PAHs concentrations

320 in winter. Furthermore, above analysis highlighted the importance of coal and biomass burning in

321 north China at cold periods on affecting the PAHs concentrations in Nanjing. For yearly variation,

322 as there were no significant differences in temperature and MLH between the two periods, the role

323 of them in explaining PAHs reduction may be limited.

324 **3.3.2 The role of atmospheric chemical degradation and particle coatings**

325   During the atmospheric transportation, PAHs compounds are exposed to oxidants, such as O_3,

326 OH, NO_2 and nitric acid, which degrade PAHs and thereby change the PAHs compositions and

327 concentrations (Thornhill et al., 2008; Dvorská et al., 2011; Gao et al., 2011) and the reaction

328 rate is depended on the particle compositions and meteorological factors. BaP/BeP ratio is

329 often used as an indicator of the aging process, as BaP degradation is typically faster than

330 BeP in the air (Ravindra et al., 2008; Pinxteren et al., 2009). In this study, BaP/BeP ratio

331 significantly increased from 0.45±0.15 (Period 1) to 0.70±0.15 (Period 2) ($p<0.001$). This

332 may suggest that PAHs at Period 1 were more aged than those of Period 2, which was

333 consistent with a higher O_3 concentration of Period 1. This may further indicate that the

334 difference in the "initial" ambient PAHs level between the two periods could be even higher

335 than the reduction calculated based on observed concentration now.

336 In some previous studies, PAHs/EC ratio was suggested to indicate the photochemical

337 degradation of PAHs. Kong et al. (2012) systematically investigated the emission of PAHs

338 and inorganic chemical components in $PM_{2.5}$ of various sources and the PAHs/EC ratios for

339 different sources were shown in **Figure S7**. For all the sources except for gasoline vehicle

340 emission (PAHs/EC ratio as 0.19), the PAHs/EC ratios ranged between 7.2-422 ng μg^{-1},

341 which were much higher than the ratios of ambient air of this study (3.31±2.34 for Period 1

342 and 1.20±0.72 for Period 2), further verifying the aging of PAHs. The PAHs/EC ratio is

343 typically low under high O_3 concentrations and high temperature due to stronger

344 photo-degradation of PAHs (**Figure S8**). In this study, although the O_3 concentrations were

345 significantly decreased at Period 2, the PAHs/EC values did not exhibit higher values as

346 expected. Conversely, it was significantly lower than that of Period 1 (P<0.001). This appears

347 to say that PAHs from the Period 2 were more aged compared to those in Period 1. One

348 reason is that source profiles were believed to be changed from the Period 1 to Period 2, thus

349 a direct comparison of the overall average PAHs/EC ratios may be inappropriate here to

350 indicate the aging degree between the two studied periods. Decreased emission of PAHs from

351 oil evaporation (which do not contribute to EC) as pollution control at Period 2 can lead to

352 lower PAHs/EC ratio, and increased relatively contributions from vehicle emissions would

353 lower the ratio as well. In fact, even though the sources were not changed or similar, a simple

354 comparison based solely on the PAHs/EC ratio and/or their dependence on ambient O_3 may

355 be biased and associated with high uncertainty, because: (1) O_3 can only partly explain the

356 PAHs losses and the role of OH and other oxidants (like NO_2) in PAHs photolysis can also

357   dominate the PAHs losses (Marchand et al., 2004); (2) the coating effects of fresh particles with

358   secondary aerosols which can shield PAHs from chemical degradation (Thornhill et al., 2008;

359   Shrivastava et al., 2017). Thornhill et al. (2008) used a criterion of $(PAHs+10)/BC>11$ ng μg^{-1} to

360   identify data points representing uncoated particles. In this study, the values of $(PAHs+10)/EC$

361   were 6.16 ± 2.77 and 4.00 ± 1.19 ng μg^{-1} for Period 1 and Period 2, respectively, indicating the

362   particle phase PAHs were both coated by secondary aerosols for the two periods.

363   Therefore, it is believed that the measured PAHs for both the two periods were aged.

364   However, it is difficult to quantitatively evaluate and compare the degree of photochemical aging

365   in the present study. Considering the comparable O_3 and NO_2 concentrations and similar

366   meteorological conditions of the two periods, it is believed the photochemical degradation played

367   a limited role in explaining the significant reduction of averaged mass concentrations of PAHs

368   from Period 1 to Period 2 in this study.

369   3.3.3 The role of pollution control measures

370   In China, the PCM is initially to reduce the regular air pollutants of CO, SO_2, NO_x, $PM_{2.5}$,

371   PM_{10}, and VOCs. The higher reduction rate of $PM_{2.5}$ associated PAHs than $PM_{2.5}$ itself observed

372   in this study indicated that effective controls could have a positive influence on ambient PAHs

373   pollution. According to the report of Nanjing Environmental Protection Bureau, the main sources

374   of $PM_{2.5}$ in Nanjing were coal combustion (27.4%), industrial production (19.0%), vehicle

375   emission (24.6%), fugitive dust (14.1%) and other sources (14.9%) in 2014

376   (http://jsnews2.jschina.com.cn/system/2015/04/30/024548067.shtml). A series of emission sources

377   have been controlled or improved after 2015 (**Table S7**). The total coal consumption was

378   controlled to below 30 million tons. The gasoline, gasoline oil and solvent oil consumption

379   amount decreased by 15.6%, 8.6% and 54.1% in 2015 when compared with those in 2013. The use

380   of clean energy like coal gas and natural gas increased by 24.3% and 7.4%, respectively.

381   Meanwhile, many improving measures were adopted for coal and oil burning, industrial processes

382   and domestic activities, etc. These all lead to the reduction of $PM_{2.5}$ and also associated PAHs.

383   Sources of $PM_{2.5}$ could be more complex than those of PAHs. The significant weak positive

384   correlations between high ring PAHs (4-7 rings) and SO_2, NO_2, OC and EC (Figure S6) indicating

385   that there may exist similar sources of PAHs with the precursors and key components of $PM_{2.5}$.

386   However, their sources were not the same. No significant correlations were found for 2 and 3 ring

387   PAHs with SO_2, NO_2, OC, and EC, suggesting different sources or formation pathways.

388   Therefore, the control measures of $PM_{2.5}$ and its precursors may not play the same role for

389   PAHs reduction of different rings, which can explain the difference in reduction rate of $PM_{2.5}$

390   (34%) and PAHs (63%) in this study.

391   PAHs markers are powerful to identify and attribute emission sources (Mancilla et al.,

392   2016). Typical PAH markers were Chr, BkF, Flu, Pyr, BaA and BaP for coal burning

393   (Bourotte et al., 2005; Ravindra et al., 2008; Teixeira et al., 2015); Acy, Ant, Phe, Flu, Pyr,

394   Chr, BbF and BkF for wood burning (Marchand et al., 2004; Bourotte et al., 2005; Teixeira et

395   al., 2015); BghiP, Cor and InP for gasoline emission (Pinxteren et al., 2009; He et al., 2014;

396   Shen et al., 2014; Pereira et al., 2017); Flu, Pyr, Chr, BbF, BkF and BeP for diesel emission

397   (Marchand et al., 2004; Bourotte et al., 2005; He et al., 2014; Mancilla et al., 2016); Pyr, BaP

398   and BaA for natural gas combustion (Bourotte et al., 2005) and Pyr, Flu and Phe for

399   incineration (Ravindra et al., 2008). We summed the concentrations of these markers and

400   adjusted to $PM_{2.5}$ mass concentration for each source type of the two periods. The calculated

401   ratios were significantly reduced for sources including wood burning (Acy, Ant, Phe, Flu, Pyr,

402   Chr, BbF and BkF) ($p<0.05$), gasoline vehicle emission (BghiP, Cor and InP) ($p<0.01$) and

403   diesel vehicle emission (Flu, Pyr, Chr, BbF, BkF and BeP) ($p<0.05$). This indicated that the

404   PCM played an effective role in reducing the contributions of wood burning and vehicle

405   emission to PAHs in Nanjing. The open biomass burning was strictly monitored and

406   forbidden in recent years. For coal combustion, many efforts have been made mainly for

407   industrial activities in China, while the pollutants control from domestic coal burning was

408   still limited, which can contribute 10.7% of PAHs emission of China (Zhang and Tao, 2009).

409   For vehicle emission, the improved fuel quality (lower sulfur and aromatic content)

410   (Ravindra et al., 2008), the adoption of catalytic converters (Ravindra et al., 2008), the

411   adoption of clean energy vehicles, eliminating the vehicles which cannot meet the higher

412   emission standards and so on all make fundamental contributions to the PAHs reduction. In

413   short, along with the economic and social development, though more energies are consumed,

414   the PAHs concentrations can be effectively reduced with comprehensive pollution control

415   measures for $PM_{2.5}$.

416   To sum up, as the similarity in the circulation patterns and surface meteorological parameters

417   for the two sampling periods, the observed significant reduction in ambient PAHs levels of

418   Nanjing was mainly associated with the source change raised by strict and effective PCM.

419   **3.4 PAHs source apportionment and variations between two periods**

420   **3.4.1 Diagnostic ratios**

421   Diagnostic PAHs ratios are a useful tool to distinguish their sources qualitatively (Guillon et

422   al., 2013). Four widely used ratios including Flu/(Flu+Pyr), BaA/(BaA+Chr), Ant/(Ant+Phe) and

423   InP/(InP+BghiP) (Ravindra et al., 2008; Wang et al., 2011; Lin et al., 2015; Li et al., 2016a;

424   Mancilla et al., 2016; Yu et al., 2016) were adopted in **Figure 4**, with the indicative sources for

425   different values summarized. The values of Flu/(Flu+Pyr) and InP/(InP+BghiP) distributed mainly

426   within 0.5-0.6 and 0.4-0.6, reflecting the marked contributions of coal and biomass burning and

427   petroleum combustion. For BaA/(BaA+Chr), the values ranged between 0.11-0.5, covering the

428   sources of petro genic, coal combustion, vehicle emission, and biomass burning. The values of

429   Ant/(Ant+Phe) (varying in 0.06-0.34) highlighted the importance of wood burning and petroleum.

430   Significant differences were only found for Ant/(Ant+Phe) ratios, decreasing from Period 1

431   (0.20±0.07) to Period 2 (0.14±0.05) (P<0.001). However, the ratios of the two periods still

432   indicated the similar sources. It also proposed the cautions that atmospheric reactivity can modify

433   the atmospheric PAHs levels and thus the ratios between PAHs (Ravindra et al., 2008; Mancilla et

434   al., 2016).

435   **3.4.2 Source apportionment using PMF**

436   To better explain the variation in PAHs sources of the two periods, the PMF modeling was

437   performed with the source profiles and contributions shown in **Figure 5**. Oil combustion was

438   associated with the high concentration of the more volatile PAHs such as Fl, Flu, and Pyr, along

439   with moderate levels of the higher molecular weight PAHs, i.e. BbF and InP (Ravindra et al.,

440   2008). Acy, Ace, Fl, Phe, and Ant were typical markers for volatilization of crude oil and

441   petroleum products (Wang et al., 2016b). Therefore, a mixed source of petroleum and oil burning

442   source (PO) was confirmed in this study. PO was also resolved by Chen et al. (2016c), with high

443   loadings on Flu, Pyr and moderate loadings on Pyr, Chr, BeP, BaP, and BghiP. According to the

444   markers listed in section 3.3.3 and the resolved source profiles, totally four sources were identified

445   as coal combustion (CC), PO, wood burning (WB) and vehicle emission (VE), accounting for

446   18.7%, 26.1%, 15.3% and 39.8% of total PAHs at Period 1, respectively (**Figure 5a**). The

447   corresponding contributions were 24.2%, 15.7%, 24.4% and 35.7%, respectively at Period 2

448   (**Figure 5b**).

449   Overall, the PAHs concentrations contributed from the four types of sources all reduced

450   in Period 2, by 83%, 68%, 51% and 33% for PO, VE, WB, and CC, respectively (**Figure 5c**).

451   The decreased absolute contributions of them reflected the PCM was effective in reducing

452   PAHs from the sources. Though great efforts have been done by the local government for

453   controlling coal and wood/biomass burning in Nanjing, the long-range transport of the two

454   types of sources from North China partly offset these efforts. It can explain the fewer

455   reduction rates of WB and CC when compared with those for PO and VE. From **Table S8**, it

456   can be found that in the cities of North China, the contributions of CC were always at a high

457   level, for example, as 38-40% in Zhengzhou (Wang et al., 2015; Wang et al., 2017b) and 52.1%

458   in Taiyuan (Li et al., 2016a). The contributions of wood/biomass burning at northern Chinese

459   cities ranged in 12% (Zhengzhou) (Wang et al., 2015)-37.1% (Xi'an) (Wang et al., 2016b).

460   The contributions of PO as 27% and 41% (for marine vessels) were reported for coastal sites

461   in Taiwan (Chen et al., 2016a) and Hong Kong (Ma et al., 2016), respectively. The VE

462   contribution to PAHs in this study was similar to that in Shanghai (43%) (Liu et al., 2017).

463   Though the PAHs sources differed in various studies, varying with space and time, the PAHs

464   sources in Nanjing exhibited mixture properties, characterized by contributions from

465   local/regional, inland/coastal and southern/northern of China.

466   Clearly, seasonal variation of the four types of source contributions is illustrated in

467   **Figure 6**. The contributions from WB (5.57±3.21 and 1.86±0.91 ng m^{-3} for Period 1 and

468   Period 2) and CC (3.26±1.49 and 2.81±1.86 ng m^{-3} for Period 1 and Period 2) both exhibited

469   highest values in winter, which could be related to the additional fuels combustion for heating

470   purpose. The vehicle emission also contributed most in winter (9.74±5.36 and 3.04±1.61 ng

471   m^{-3} for Period 1 and Period 2), implying the enhanced emission at cold start mode. The

472   extreme higher contribution of VE at Spring of Period 1 may be related to the lowest

473   temperature at that period as 8.8±3.0 °C. For PO emission, it held slightly higher

474   contributions in summer owing to more evaporation under the higher ambient temperature.

475   The seasonal variation in source contributions all favored well to the seasonal variation of PAHs

476   concentrations as discussed above.

477   **3.4.3 Source region analysis**

478   As the results of WPSCF in **Figure 7**, the source regions with high probabilities of WB were

479   mainly located at the west, northwest and north side of Nanjing and the regions extending to a

480   larger area at Period 2. For CC, the source regions at Period 1 located mainly at the

481   southwest-northeast directions, while they changed to the north and northwest directions at Period

482   2. The variation in source regions for WB and CC highlighted the importance of the domestic coal

483   and wood burning for heating. For VE, the main source regions were on the southeast side,

484   surrounding Nanjing at Period 1 and then changed to the northwest, north and northeastern of

485   China in Period 2. For PO, the source regions at Period 1 were mainly concentrated in the lower

486   reaches of the Yangtze River and the offshore sea area; while in Period 2, the main source regions

487   moved to the middle reaches of the Yangtze River, the coastal area of PRD and near offshore

488   areas. These regions all hold intensive emissions from both ocean (Fan et al., 2016) and inland

489   vessels/ships (Song, 2015). The geographic origins of the four sources were consistent for

490   weighted concentration weighted trajectory (WCWT) analysis (**Figure 8**). Higher contributions of

491   WB and CC were found at Period 2 from north China. Lower level contributions of PO and VE at

492   Period 2 were also verified, owing to the shrinkage of the regions with higher WCWT values.

493   To conclude, after the effective control of air pollutants, the sources of PAHs at Nanjing at

494   Period 2 were mainly influenced by the regional transport of CC and WB from North China

495   especially for domestic use, local+regional transports of ocean and inland vessels/ships and

496   local+regional transports of vehicle emission from North China. It indicated the control of WB,

497   CC and VE should be more strictly in North China. To obtain the local/regional contributions of

498   various sources, a quantitative research is needed based on an accurate emission inventory in the

499   future.

500   **3.5 Reduced health risks of PAHs and source allocation**

501   Through the effective source control, the PAHs obviously reduced from Period 1 to Period 2

502   in Nanjing. It suggested that the human health risks also decreased. By adopting the simple

503   point-estimate approach (Wang et al., 2011), the estimated ILCR for Period 1 and Period 2 were

504   4.13×10^{-4} and 1.07×10^{-5}, respectively (**Figure 9**). Thus, the overall cancer risks due to inhalation

505   exposure to PAHs reduced significantly by 61% for Period 2 ($p<0.001$). However, it still

506   exceeded the acceptance limit of cancer risk (10^{-6}) (Chen et al., 2016a; Lv et al., 2016).

507   Reduced cancer risks were also found for the periods with mega-events, such as the APEC

508   meeting (Xie et al., 2017). It should be noted that uncertainties are inherent in cancer risk

509   assessment, as a lack of knowledge about the factors affecting exposure or toxicity

510   assessment (Hong et al., 2016).

511   To interpret the risk reduction, contributions of the four types of sources were calculated

512   by PMF model as listed in **Figure 10**. CC dominated the contributions to ILCR, similar for

513   the two periods, as about 52%. The contributions to ILCR of PO obviously reduced from

514   23.5% to 4.5% from Period 1 to Period 2; while the contributions of WB and VE increased

515   from 1.75% to 13.7% and from 22.3% to 29.9%. Though still, the dominant sources were

516   coal combustion and vehicle emission, their relative contribution varied in mass

517   concentration and incremental cancer risks. Source pollution controls should not only focus

518   on mass pollution level, but also those having larger toxic and contributing significantly to

519   the health impacts.

520   Health risks associated with exposure to other pollutants like CO, SO_2, NOx, O_3, $PM_{2.5}$

521   and associated toxic components (heavy metals, BC, etc.) are interesting and hot topics. The

522   present study only evaluated risks associated with $PM_{2.5}$-bound PAHs exposure. When taking

523   other pollutants into account, health benefits received from the effective PCMs could be more

524   significant. This is worthy to be investigated in the future, with big datasets (including air

525   pollutants, personal exposure monitoring data, human disease and meteorological parameters)

526   collected and dose-equivalence relationships for different chemicals with health effects

527   clarified.

528   **4 Conclusion**

529   As a non-routine monitored pollutants, PAHs were focused in a four-year study in

530   Nanjing to answer whether the air pollution control measures (PCM) can substantially affect

531   the PAHs concentrations, in view of that PAHs can also be impacted by meteorological

532   parameters and atmospheric chemical reaction. Two sampling campaigns in March

533   2013-January 2014 (Period 1) and November 2015-July 2016 (Period 2) just before and after

534   the local air pollution plan published in April 2014 were completed.

535   In Period 2, the average concentrations of $PM_{2.5}$ significantly reduced by 34% of that for

536   Period 1. The total PAHs concentrations exhibited a significantly higher reduction rate as 63%

537   than $PM_{2.5}$, indicating the PCM for $PM_{2.5}$ have played a positive role in PAHs reduction

538   unexpectedly. The nonlinear reduction of $PM_{2.5}$ and associated PAHs was related to their different

539   sources and the different effects of PCM played on $PM_{2.5}$ and its precursors as well as the different

540   ring of PAHs. The similarity in atmospheric circulation patterns and no obvious differences in

541   meteorological parameters indicated that the main reason for controlling PAHs reduction was the

542   source variation.

543   Diagnostic ratios and PMF modeling identified four sources, as coal combustion (CC),

544   petroleum and oil burning (PO), wood burning (WB) and vehicle emission (VE). Contributions

545   from all these four source types to ambient PAHs had substantially decreased, from 33% to 83%.

546   The reduction percentages were more notable for VE and PO, compared to the reduction

547   percentages in CC and WB. A co-benefit of cancer risk reduction by 61% was obtained. The

548   cancer risk was still higher than 10^{-6}, with CC dominated the contributions as about 52%. In

549   Nanjing, as the dominated contributions of coal burning and vehicle emission to atmospheric

550   PAHs, effective emission mitigation strategies of PAHs should be developed from both local and

551   regional views. Meanwhile, the obvious seasonal variation in PAHs concentrations and source

552   contributions and potential source regions highlighted the importance of further efforts on the

553   reduction of coal and wood burning in North China at a heating period.

554   This study provides useful data and new insights for assessing the effects of air pollution

555   control measures, from the view of human health, not only just from the reductions of routine air

556   pollutants. The outcomes could be important to regional air quality management and decision

557   makers. The analysis and main findings here are also applicable and helpful for other areas around

558   the world.

559

568

569   **Appendix**

570   Materials associated with this manuscript is provided in the supporting file available free of charge

571   via the internet.

572

573   Data are available on request to Shaofei Kong (kongshaofei@cug.edu.cn).

574

575   **Competing interests**

576   The authors declare that there is no conflict of interest.

577

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