

Sensitivity Analysis of PM_{2.5}-Bound Total PCDD/Fs-TEQ Content:

In the Case of Wuhu City, China

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Abstract

During 2015-2017, the atmospheric PM_{2.5}, PM_{2.5}/PM₁₀, PCDD/Fs, PCDD/F phase distribution and PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in Wuhu and Bengbu were investigated in this study. In addition, the sensitivity analysis for PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ of Wuhu was also studied. During 2015-2017, the three-year average PM_{2.5} concentration in Wuhu was 53.0 µg m⁻³, and in Bengbu was 61.4 µg m⁻³; the results also showed the annual average PM_{2.5} concentrations of these two cities had declined, but the levels were still far above the WHO annual PM_{2.5} standard (10 µg m⁻³). In addition, in Wuhu, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer (0.166 ng-WHO₂₀₀₅-TEQ g⁻¹) were approximately only 68.8% in magnitude lower than the average value of other three seasons (0.532 ng-WHO₂₀₀₅-TEQ g⁻¹), and that of Bengbu in summer (0.187 ng-WHO₂₀₀₅-TEQ g⁻¹) was approximately 66.7% in magnitude lower than the average value of other three seasons (0.561

23 ng-WHO₂₀₀₅-TEQ g⁻¹). Sensitivity analysis showed that the PCDD/F concentration was the most
24 positively correlated sensitive factor for PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ, and when
25 $\Delta P/P$ was changed from 0% to +50%, $\Delta S/S$ responded from 0% to +106%. The second positively
26 correlated sensitive factor was PM₁₀ concentration, and when $\Delta P/P$ was changed from 0% to +50%,
27 $\Delta S/S$ responded from 0% to +72%. This was followed by atmospheric temperature, and its effect
28 was negatively correlated, when $\Delta P/P$ was changed from -50% to +50%, $\Delta S/S$ responded from +73%
29 to -112%. The last sensitive parameter was PM_{2.5} concentration, with the impact divided into two
30 stages: when $\Delta P/P$ was changed from 0% to +70%, $\Delta S/S$ responded from 0% to +33%, but when
31 $\Delta P/P$ was changed from +70% to +100%, $\Delta S/S$ responded from +33% to +25%. The results of this
32 study provide useful information that can be used to achieve more insights into both atmospheric
33 PM_{2.5} and PCDD/Fs.

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36 **Keywords:** PM_{2.5}; PM₁₀; PCDD/Fs; phase distribution; sensitivity analysis

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46 **Introduction**

47 The particulate matter (PM) and polychlorinated dibenzo-p-dioxins and polychlorinated
48 dibenzofurans (PCDD/Fs) in the ambient air have received great public concern due to their
49 significant correlation with the incidences of pulmonary and cardiac diseases (Schwartz *et al.*,
50 1996; Ito *et al.*, 2006).

51 Particulate matter (PM) is a suspension of solid or liquid particles in the atmosphere, which is
52 a kind of aerosol (Ghosh *et al.*, 2014). According to the aerodynamic diameters of PM, it can be
53 divided into TSP (range from ~0 to 100 μ m), PM₁₀ (range from ~0 to 10 μ m) and PM_{2.5} (range from
54 ~0 to 2.5 μ m) (Lu *et al.*, 2016; Chow *et al.*, 2015). The sources of PM can be natural or
55 anthropogenic, with forest burning, dust storms and volcanic eruptions the main natural sources,
56 while industrial activities, vehicle exhausts and the construction industry are the major
57 anthropogenic sources (Kong *et al.*, 2014; Alghamdi *et al.*, 2015; Bilos *et al.*, 2001). The PM in
58 ambient air is a conglomerate of organic and inorganic carbon, mineral element, nitrates,
59 ammonium, sulfate and so on (Zhu *et al.*, 2017). While the PM emitted into atmosphere by
60 primary sources, the secondary aerosol forms with a specific ratio and environmental conditions
61 (Lee *et al.*, 2016). Previous studies of the chemical characteristics of PM in the atmosphere
62 indicated that it not only had adverse effects on air quality and even global climate, but also
63 impacts on human health due to its particle toxicity (Huang *et al.*, 2014; Wang *et al.*, 2014; Liu *et*
64 *al.*, 2016; Chen *et al.*, 2014).

65 PCDD/Fs are well-known persistent organic pollutants (POPs) and semi-volatile organic
66 compounds (SOCs), which can transport over long distances and interact for long periods in the

67 environment (Wu *et al.*, 2009b; Chen *et al.*, 2014; Lee *et al.*, 2016), as well as bio-accumulate in
68 the fatty tissues and bio-magnify within the food chain (Shih *et al.*, 2009). PCDD/Fs are extremely
69 hazardous chemicals, and ingestion, inhalation and dermal contact are the primary pathways for
70 them to get into human bodies (Shih *et al.*, 2009; Chen *et al.*, 2010), thus posing risks to the
71 immune system, interfering with hormones and even leading to cancer (Lin *et al.*, 2010; Chi *et al.*,
72 2011). PCDD/Fs were detected in the emissions of municipal solid waste incinerators (MSWIs)
73 for the first time (Olie *et al.*, 1977), and since then have become one of the most controversial
74 environmental pollutants. Combustion processes and some industrial activities are the most
75 dominant sources of PCDD/Fs released to the environment, such as power and heating facilities,
76 metal smelting processes and waste incineration (Schuhmacher *et al.*, 2000; Wang *et al.*, 2003; Lin
77 *et al.*, 2007; Hsieh *et al.*, 2009; Chuang *et al.*, 2010, 2011). PCDD/Fs are complex mixture of
78 different congeners, there are 210 possible congeners and 17 of these have been shown to be more
79 toxic, with the 2,3,7,8 positions attached by chlorine atoms, and the toxicities are estimated by the
80 toxic equivalent quantity (TEQ) (Cheruiyot, *et al.*, 2016).

81 In the atmosphere, after being emitted from combustion facilities, the PCDD/Fs are
82 distributed into both gas and particle phases (Li *et al.*, 2008b; Chen *et al.*, 2011a; Kou *et al.*, 2015).
83 Many studies show that the gas-particle partitioning of PCDD/Fs is highly dependent on their
84 vapor pressures, ambient temperatures and other parameters (Wu *et al.*, 2009a; Wang *et al.*, 2010;
85 Cheruiyot *et al.*, 2015). More fractions of PCDD/Fs are volatilized into gas with an increase in
86 temperature (Fiedler, 1996; Oh *et al.*, 2001). The degradation of PCDD/Fs depends on chemical
87 and photochemical reactions, and their removal mainly relies on the atmospheric deposition
88 (Giorgi, 1988; Wu *et al.*, 2009a; Huang *et al.*, 2011a; Mi *et al.*, 2012; Chi *et al.*, 2009).

89 This study investigated the PM_{2.5} concentrations, PM_{2.5}/PM₁₀ ratios, PCDD/F concentrations,
90 gas-particle partitioning, and PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content, while the
91 sensitivity analysis of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents was also studied and
92 discussed.

93

94 **METHODS**

95 Two cities, namely Wuhu (31°33'N, 118°38'E) and Bengbu (32°93'N, 117°34'E) in Anhui
96 province, China, were evaluated in this study. The monthly mean concentrations of both PM_{2.5} and
97 PM₁₀, monthly temperature and precipitation in both cities were obtained from local air quality
98 monitoring stations and the Statistics Yearbook of China.

99 The total PCDD/F concentration was simulated by regression analysis of the PM₁₀
100 concentration. Tang *et al* (2017) reported that there is a high correlation between PM₁₀ values and
101 total PCDD/F mass concentrations. Tang *et al* (2017) included the following two regression
102 equations:

$$103 \quad Y_1 = 0.0138x + 0.0472 \quad (1)$$

$$104 \quad Y_2 = 0.0117x - 0.021 \quad (2)$$

105 Y_1, Y_2 : total PCDD/F concentration (pg m⁻³)

106 X : PM₁₀ concentration in ambient air (μg m⁻³)

107 The final total PCDD/F concentration was the average of Y_1 and Y_2 .

108

109 **Gas-Particle Partitioning**

110 The PCDD/F concentrations in the gas and particle phases, respectively, were calculated by

111 using gas-particle partitioning model as Eq. (3) (Yamasaki *et al.*, 1982; Pankow, 1987; Pankow
112 and Bidleman, 1991, 1992):

113

$$114 \quad K_p = \frac{F/TSP}{A} \quad (3)$$

115 K_p : temperature-dependent partitioning constant ($\text{m}^3 \mu\text{g}^{-1}$);

116 TSP: concentration of total suspended particulate matter, which was multiplied by PM_{10}
117 concentration with 1.24 ($\mu\text{g m}^{-3}$);

118 F: concentration of the compounds of interest bound to particles (pg m^{-3});

119 A: gaseous concentration of the compound of interest (pg m^{-3}).

120 Plotting $\log K_p$ against the logarithm of the subcooled liquid vapor pressure, P_L^0 , gives

$$121 \quad \log K_p = m_r \times \log P_L^0 + b_r \quad (4)$$

122 P_L^0 : subcooled liquid vapor pressure (Pa);

123 m_r : cited slope;

124 b_r : cited y-intercept.

125 Complete datasets on the gas-particle partitioning of PCDD/Fs in Taiwan have been reported
126 (Chao *et al.*, 2004), with the values $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$. These values
127 were used in this study for establishing the partitioning constant (K_p) of PCDD/Fs.

128 A previous study correlated the P_L^0 of PCDD/Fs with gas chromatographic retention
129 indexes (GC-RI) on a nonpolar (DB-5) GC-column using p,p'-DDT as a reference standard. The
130 correlation has been re-developed as follows (Hung *et al.*, 2002).

$$131 \quad \log P_L^0 = \frac{-1.34(RI)}{T} + 1.67 \times 10^{-3}(RI) - \frac{1320}{T} + 8.087 \quad (5)$$

132 RI: gas chromatographic retention indexes developed by Donnelly *et al.* (1987) and Hale *et al.*

133 (1985);

134 T: ambient temperature (K).

135

136 **RESULTS AND DISCUSSION**

137 **1. PM_{2.5} Concentration**

138 The PM_{2.5} concentration not only has a significant correlation with air visibility and human
139 health, but can also reflect the PCDD/F concentration of a region. For the period 2015-2017, the
140 monthly average PM_{2.5} concentrations in the ambient air of Wuhu and Bengbu are shown in Figs.
141 1 (A), 1 (B) and 1 (C). As for Wuhu, over the three years examined, the lowest PM_{2.5}
142 concentration occurred in 2017, at 27.0-99.0 $\mu\text{g m}^{-3}$, and with an average of 48.9 $\mu\text{g m}^{-3}$; followed
143 by 2016, which was in the range of 25.0-90.0 $\mu\text{g m}^{-3}$ and with an average of 53.0 $\mu\text{g m}^{-3}$; and in
144 2015, in the range of 32.0-108.0 $\mu\text{g m}^{-3}$, with an average of 57.3 $\mu\text{g m}^{-3}$. Comparing the annual
145 average PM_{2.5} concentrations, we can see that the highest values occurred in 2015, followed by
146 2016, and the lowest occurred in 2017. The PM_{2.5} annual average concentration, compared to that
147 for 2015 to 2017, was reduced by approximately 14.7%. The tendency of the PM_{2.5} level to
148 decline slowly may be because of social efforts to improve the quality of the environment. As a
149 whole, the PM_{2.5} concentration of the three-year average in Wuhu ranged between 25.0 and 108.0
150 $\mu\text{g m}^{-3}$, with an average of 53.0 $\mu\text{g m}^{-3}$. It also can be seen that even though the air quality of
151 Wuhu was improved significantly, the PM_{2.5} concentrations were still far above the WHO air
152 quality regulated standard (10 $\mu\text{g m}^{-3}$), so more efforts are needed issue.

153 With regard to Bengbu (Figs, 1 (A), 1 (B) and 1 (C)), the lowest monthly average PM_{2.5}
154 concentration also occurred in 2017, which was between 34.0 and 98.0 $\mu\text{g m}^{-3}$, with an average of

155 $61.1 \mu\text{g m}^{-3}$; and those in 2016 ranged from 33.0 to $101.0 \mu\text{g m}^{-3}$ and averaged $59.8 \mu\text{g m}^{-3}$; during
156 2015, the $\text{PM}_{2.5}$ concentration ranged between 47.0 and $88.0 \mu\text{g m}^{-3}$ and averaged $63.2 \mu\text{g m}^{-3}$.
157 These results reveal that from 2015 to 2016 the annual average $\text{PM}_{2.5}$ concentration fell from 63.2
158 to $59.8 \mu\text{g m}^{-3}$, falling by approximately 5.4% . However, the highest values increased by 14.8%
159 from 2015 ($88.0 \mu\text{g m}^{-3}$) to 2016 ($101.0 \mu\text{g m}^{-3}$). As a whole, the $\text{PM}_{2.5}$ concentration for these
160 three years in Wuhu ranged between 33.0 and $101.0 \mu\text{g m}^{-3}$, with an average of $61.4 \mu\text{g m}^{-3}$. This
161 indicates that the $\text{PM}_{2.5}$ level in Wuhu was slightly lower than that in Bengbu. This is probably due
162 to the development of industry with poor air pollution control, as well as more pollutants being
163 emitted from mobile and stationary sources.

164 When regard to the seasonal variations, the four seasons were defined as spring (March, April,
165 May), summer (June, July, August), fall (September, October, November) and winter (January,
166 February and December). For Wuhu, during 2015, the average $\text{PM}_{2.5}$ concentrations in spring,
167 summer, fall and winter were 49.7 , 34.7 , 60.3 and $84.3 \mu\text{g m}^{-3}$, respectively; and those in 2016
168 were 52.3 , 32.0 , 51.0 and $76.7 \mu\text{g m}^{-3}$, respectively; while those in 2017 were 47.7 , 30.3 , 46.7 and
169 $71.0 \mu\text{g m}^{-3}$, respectively. For Bengbu, during 2015, the average $\text{PM}_{2.5}$ concentrations in spring,
170 summer, fall and winter were 57.3 , 48.0 , 69.0 and $78.3 \mu\text{g m}^{-3}$, respectively; and those in 2016
171 were 60.0 , 37.7 , 53.3 and $90.0 \mu\text{g m}^{-3}$, respectively; while those in 2017 were 60.0 , 40.0 , 52.3 and
172 $92.0 \mu\text{g m}^{-3}$, respectively. This indicates that the $\text{PM}_{2.5}$ concentrations varied from season to
173 season, the highest values always occurred in winter and the lowest in summer, while the values
174 for spring were very similar with fall and both in the middle levels. As a whole, the three-year
175 average of $\text{PM}_{2.5}$ concentration of Wuhu in summer ($32.3 \mu\text{g m}^{-3}$) was 58.2% in magnitude lower
176 than that in winter ($77.3 \mu\text{g m}^{-3}$); and the values of Bengbu in summer ($41.9 \mu\text{g m}^{-3}$) was 51.7% in

177 magnitude lower than that in winter ($86.8 \mu\text{g m}^{-3}$).

178 During the cold seasons, cities at a higher latitude have a very low ground temperature, so it
179 is easier for high stability vertical atmospheric convection, to occur, which hinders the dispersion
180 of air pollutants. As a result, an accumulation of $\text{PM}_{2.5}$ concentrations occurs in regions with
181 human activity. In addition, the parts of north China require more coal and other fossil fuel
182 combustion for heating during winter, and thus have more air pollutant emissions. Therefore, the
183 air current blowing from the northern cities, which have quite high $\text{PM}_{2.5}$ concentration levels,
184 may increase the $\text{PM}_{2.5}$ concentrations in both Wuhu and Bengbu, cities located in central China.
185 And in warm season the air temperature is higher and the vertical transport of air current is more
186 violent, which can accelerate the dispersion of air $\text{PM}_{2.5}$. Moreover, in summer both rainfall and
187 wind speed are stronger, enhancing the effects of rainfall scavenging and wind blowing in
188 removing the $\text{PM}_{2.5}$ from the atmosphere.

189

190 **2. $\text{PM}_{2.5}/\text{PM}_{10}$ Ratio**

191 The $\text{PM}_{2.5}/\text{PM}_{10}$ ratio can reflect the contribution of fine particulate matter to the ambient air
192 pollutants, and thus mirror the status of air pollution. The monthly average of $\text{PM}_{2.5}/\text{PM}_{10}$ ratio in
193 the ambient air in Wuhu and Bengbu are presented in Figs. 2 (A), 2 (B) and 2 (C).

194 The annual $\text{PM}_{2.5}/\text{PM}_{10}$ ratios of Wuhu were in the range of 0.53-0.73, with an average of
195 0.67 in 2015, of 0.61-0.74 and with an average of 0.66 in 2016; and of 0.42-0.89 and with an
196 average of 0.54 in 2017. As for Bengbu, the annual $\text{PM}_{2.5}/\text{PM}_{10}$ ratios ranged between 0.61 and
197 0.79 and averaged 0.70 in 2015, ranged between 0.57 and 0.79 and averaged 0.65 in 2016, and
198 between 0.43 and 0.72 and averaged 0.59 in 2017, respectively. The $\text{PM}_{2.5}/\text{PM}_{10}$ ratio of the

199 three-year range in Wuhu was 0.42-0.89, with an average of 0.63, was 0.43-0.79, and with an
200 average of 0.65 in Bengbu. These results show that the annual average of $PM_{2.5}/PM_{10}$ ratios in
201 both Wuhu and Bengbu were all lower than those reported in the Beijing-Tianjin-Hebei region
202 (0.83), the Yangtze River Delta region (0.76) and the Pearl River Delta region (0.74) (Chen *et al.*,
203 2017). This result was also consistent with the conclusion of Zhou *et al.* (2015), which reported
204 there was a strong positive correlation between $PM_{2.5}$ and PM_{10} mass concentration in the
205 atmosphere. In particular, the trend of a decreasing $PM_{2.5}/PM_{10}$ ratio was similar to that seen for
206 the $PM_{2.5}$ concentration.

207 For Wuhu, during 2015, the three highest monthly averages of $PM_{2.5}/PM_{10}$ ratios were 0.73 in
208 November, 0.72 in December and 0.70 in January; while in 2016, the three highest monthly
209 averages of $PM_{2.5}/PM_{10}$ ratios were 0.74 in December, 0.72 in July and 0.72 in January; in 2017
210 the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.89 in December, 0.67 in
211 November and 0.65 in February. However, in 2015, the three lowest monthly averages of the
212 $PM_{2.5}/PM_{10}$ concentration ratios were 0.53 in June, 0.64 in July and 0.66 in September; while in
213 2016, the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ concentration ratios were 0.61 in April,
214 0.62 in September and 0.64 in August; in 2017, the three highest monthly averages of the
215 $PM_{2.5}/PM_{10}$ ratios were 0.42 in April, 0.44 in May and 0.51 in July.

216 For Bengbu, during 2015, the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were
217 0.79 in November, 0.78 in December and 0.76 in February; while in 2016, the three highest
218 monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.57 in September, 0.57 in April and 0.59 in
219 August; in 2017 the three highest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.72 in
220 February, 0.71 in January and 0.68 in December. However, in 2015, the three lowest monthly

221 averages of the $PM_{2.5}/PM_{10}$ ratios were 0.61 in April, 0.64 in June and 0.64 in October; while in
222 2016, the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were 0.57 in September, 0.57 in
223 April and 0.59 in August; in 2017 the three lowest monthly averages of the $PM_{2.5}/PM_{10}$ ratios were
224 0.43 in May, 0.51 in April and 0.55 in June, respectively.

225 In general, it was found that a higher $PM_{2.5}/PM_{10}$ ratio always accompanied a higher $PM_{2.5}$
226 concentration. This demonstrated that $PM_{2.5}$ is the major portion of atmospheric particles. A
227 previous study (Tang *et al.* 2017) reported the atmospheric particles were mostly due to the
228 gas-particle transformation, going through the condensation and flocculation processes, and then
229 the accumulation mode of $PM_{2.5}$. Furthermore, a certain fraction of the $PM_{2.5}$ concentration in the
230 ambient air is due to the resuspension of road dust or entrainment of naked lands. A higher
231 $PM_{2.5}/PM_{10}$ ratio signifies more harmful air pollution to human health, and thus should be a case
232 for more concern.

233

234 3. PCDD/F Concentration

235 Previous studies have shown the strong correlation between PM_{10} and PCDD/F mass
236 concentrations, and Lee *et al.* (2016), Suryani *et al.* (2015) and Huang *et al.* (2011a) reported the
237 correlation coefficients were as high as 0.98, 0.99 and 0.94, respectively. In Tang *et al.* (2017), the
238 monthly concentrations of PCDD/Fs in the ambient air were simulated by PM_{10} using the
239 regression analyses (Wang *et al.*, 2010). Based on the PM_{10} concentrations, the total PCDD/F mass
240 concentrations were calculated using Eq. (1) and Eq. (2).

241 The results shown that in Wuhu, the total PCDD/F mass concentrations were in the range of
242 0.65-1.94 $pg\ m^{-3}$ and with an average of 1.09 $pg\ m^{-3}$ in 2015, of 0.51-1.61 $pg\ m^{-3}$ and with an

243 average of 1.01 pg m^{-3} in 2016, and of $0.63\text{-}1.43 \text{ pg m}^{-3}$ and with an average of 1.07 pg m^{-3} in
244 2017; while in terms of concentrations of toxicity equivalent quantity (TEQ), those ranged
245 between 0.026 and $0.085 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$ in 2015, ranged between 0.021 and 0.070
246 $\text{pg-WHO}_{2005}\text{-TEQ m}^{-3}$ in 2016, and ranged between 0.025 and $0.074 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$ in
247 2017, and the corresponding average values were 0.050 , 0.047 and $0.050 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$,
248 respectively. The time variation results show that both the average of the total PCDD/F mass and
249 total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations decreased slowly, and this may be due to a better
250 control of domestic emissions. Comparing with previous studies, the values of the
251 total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations were all at the same levels of those seen in the
252 Kaohsiung area, ranging between 0.021 and $0.077 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$ and with an average of
253 $0.048 \text{ pg WHO}_{2005}\text{-TEQ m}^{-3}$ for 2014, and between 0.021 and $0.072 \text{ pg WHO}_{2005}\text{-TEQ m}^{-3}$ and
254 with an average of $0.044 \text{ pg WHO}_{2005}\text{-TEQ m}^{-3}$ for 2015 in southern Taiwan (Lee *et al.*, 2016).

255 In Bengbu, the total PCDD/F mass concentrations were in the range of $0.82\text{-}1.53 \text{ pg m}^{-3}$ with
256 an average of 1.17 pg m^{-3} , of $0.73\text{-}1.82 \text{ pg m}^{-3}$ and with an average of 1.17 pg m^{-3} , of $0.71\text{-}1.85 \text{ pg}$
257 m^{-3} and with an average of 1.31 pg m^{-3} in 2015, 2016 and 2017, respectively; while in terms of
258 concentrations of toxicity equivalent quantity ranged between 0.033 and $0.073 \text{ pg-WHO}_{2005}\text{-TEQ}$
259 m^{-3} in 2015, between 0.029 and $0.080 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$ in 2016, ranged between 0.029 and
260 $0.085 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$ in 2017, and the corresponding average values were 0.054 , 0.054 and
261 $0.061 \text{ pg-WHO}_{2005}\text{-TEQ m}^{-3}$, respectively. On the whole, both the average of the total PCDD/F
262 mass and total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations were all at high levels.

263 With regard to the seasonal variations, for Wuhu, during 2015, the average
264 total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in spring, summer, fall and winter were 0.049 ,

265 0.030, 0.054 and 0.068 pg-WHO₂₀₀₅-TEQ m⁻³, respectively; and those in 2016 were 0.054, 0.025,
266 0.048 and 0.060 pg-WHO₂₀₀₅-TEQ m⁻³, respectively; while those in 2017 were 0.054, 0.029, 0.045
267 and 0.056 pg-WHO₂₀₀₅-TEQ m⁻³, respectively. For Bengbu, during 2015, the
268 total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations in spring, summer, fall and winter were 0.059,
269 0.037, 0.060 and 0.059 pg-WHO₂₀₀₅-TEQ m⁻³, respectively; and those in 2016 were 0.063, 0.032,
270 0.053 and 0.070 pg-WHO₂₀₀₅-TEQ m⁻³, respectively; while those in 2017 were 0.079, 0.037, 0.053
271 and 0.074 pg-WHO₂₀₀₅-TEQ m⁻³, respectively. The total PCDD/Fs-WHO₂₀₀₅-TEQ concentration
272 of Wuhu in summer (0.028 pg-WHO₂₀₀₅-TEQ m⁻³) was 48.1% in magnitude lower than the
273 average value of other three seasons (0.054 pg-WHO₂₀₀₅-TEQ m⁻³); and the values of Bengbu in
274 summer (0.035 pg-WHO₂₀₀₅-TEQ m⁻³) was 44.4% of magnitude lower than the average value of
275 other three seasons (0.063 pg-WHO₂₀₀₅-TEQ m⁻³).

276 These results show that the lowest total PCDD/Fs-WHO₂₀₀₅-TEQ concentration of the two
277 cities were all occurred in summer, which means that the levels of particulate matter had a
278 significant effect on the total PCDD/F concentrations. Therefore, a higher total
279 PCDD/Fs-WHO₂₀₀₅-TEQ concentration always accompanied with a higher level of particulate
280 matter. As such, controlling the PM emissions from sources will subsequently result in reductions
281 in ambient dioxin levels.

282

283 **4. Gas-Particle Partitioning of PCDD/Fs**

284 The gas-particle partitioning of PCDD/F plays an important role in the efficiency of the
285 atmospheric wet and dry deposition (Bidleman and Harner, 2000). Several factors are important
286 here, such as the ambient temperature, PCDD/F concentrations, vapor pressure and the

287 atmospheric particulate concentration (Hoff *et al.*,1996). The gas-particle partition was calculated
288 by meteorological data using Eq. (3), Eq. (4) and Eq. (5), and the seasonal gas-particle partitioning
289 of total PCDD/Fs-WHO₂₀₀₅-TEQ in the ambient air of Wuhu and Bengbu during 2015-2017 are
290 shown in Figs.3 (A), 3 (B) and 3 (C).

291 In Wuhu, during 2015, the seasonal average temperatures were 17.3, 26.7, 18.6 and 6.5°C in
292 spring, summer, fall and winter, respectively; in 2016, the seasonal average temperatures of spring,
293 summer, fall and winter were 17.1, 28.3, 19.4 and 6.4°C, respectively; while in 2017, the average
294 temperatures of spring, summer, fall and winter were 17.5, 28.6, 17.6 and 6.6°C, respectively. As
295 for the seasonal variations of gas-particle partitioning, the fractions of gas phase total
296 PCDD/Fs-WHO₂₀₀₅-TEQ concentration in spring, summer, fall and winter were 37.4%, 73.0%,
297 42.7% and 9.9% in 2015; 35.5%, 78.7%, 48.5% and 11.0% in 2016; and 32.8%, 77.4%, 43.1%
298 and 11.3% in 2017.

299 In Bengbu, the seasonal average temperatures during 2015 were 15.9, 27.5, 16.9 and 5.0°C
300 in spring, summer, fall and winter, respectively; in 2016, the seasonal average temperatures in
301 spring, summer, fall and winter were 16.3, 27.3, 16.9 and 4.5°C, respectively; while in 2017, the
302 average temperatures of spring, summer, fall and winter were 16.5, 28.0, 16.2 and 4.9°C,
303 respectively. The fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ concentration in spring,
304 summer, fall and winter were 30.7%, 71.5%, 36.5% and 8.7% in 2015; 31.2%, 73.5%, 39.9% and
305 7.4% in 2016; and 28.6%, 72.7%, 37.3% and 7.2% in 2017. The above results show that the
306 fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ of the both cities were all lower than that in
307 the particle phase in spring, fall and winter, but were significantly higher than that in particle
308 phase in summer (Fig. 3 (A), 3 (B) and 3 (C)).

309 As for Wuhu, the three-year average fractions of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ in
310 spring, summer, fall and winter were 35.2%, 76.4%, 44.8% and 10.7%, respectively; the
311 three-year average temperatures in spring, summer, fall and winter were 17.3, 27.8, 18.5 and
312 6.5 °C, respectively. While in Bengbu, the three-year average fractions of gas phase total
313 PCDD/Fs-WHO₂₀₀₅-TEQ in spring, summer, fall and winter were 30.2%, 72.6%, 37.9% and 7.8%,
314 respectively; and the three-year average temperatures in spring, summer, fall and winter were 16.2,
315 27.6, 16.7 and 4.8 °C, respectively. The above results indicate that the fractions of gas phase total
316 PCDD/Fs-WHO₂₀₀₅-TEQ in summer were highest and those in winter were lowest, while the
317 values of spring and fall were both in the middle levels, but the former were slightly lower than
318 the latter. The fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ had a significant, positive
319 correlation with air temperature. As the temperature increased, a certain fraction of particle phase
320 PCDD/F evaporated into the gas phase, and thus the gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ
321 increased with an increasing temperature. In addition, the average temperature of Wuhu in spring,
322 summer, fall and winter were all slightly higher than those in Bengbu, while the corresponding
323 fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ were slightly higher than those in Bengbu
324 (Figs. 3 (A), 3 (B) and 3 (C)).

325 With regard to the gas-particle partitioning of PCDD/F-WHO₂₀₀₅-TEQ of each congener,
326 more gas phase PCDD/Fs-WHO₂₀₀₅-TEQ always occurred in summer, particularly for the low
327 molecular weight PCDD/Fs. For instance, the three-year averages for the fractions of
328 2,3,7,8-TeCDD and 2,3,7,8-TeCDF in the gas phase were 97.2% and 98.1% in summer, but were
329 39.2% and 49.9% in winter, respectively; those fractions of the middle molecular weight
330 PCDD/Fs for both 1, 2,3,6,7,8-HxCDD and 1, 2,3,6,7,8-HxCDF were 57.6% and 70.9% in

331 summer, but were 1.6% and 3.1% in winter, in the gas phase; while for the high molecular weight
332 PCDD/Fs of OCDD and OCDF, they were 5.1% and 7.4% in summer and 0.0% and 0.1% in
333 winter, respectively.

334 In Bengbu the situation was very similar to Wuhu, during 2015, 2016 and 2017, for the
335 average fractions of gas phase PCDD/Fs, the low molecular weight PCDD/Fs of 2,3,7,8-TeCDD
336 and 2,3,7,8-TeCDF were 96.4% and 97.6% in summer, but were 30.1% and 40.2% in winter,
337 respectively; for the middle molecular weight PCDD/Fs, such as 1, 2,3,6,7,8-HxCDD and 1,
338 2,3,6,7,8-HxCDF, were 51.2% and 65.3% in summer, but were 1.0% and 2.0% in winter,
339 respectively; however, the high molecular weight PCDD/Fs of OCDD and OCDF still had lower
340 fractions in the gas phase, and were 3.8% and 5.6% in summer and both 0.0% in winter,
341 respectively.

342 The results indicate that lower molecular weight PCDD/F congeners were primarily in the
343 gas phase, while the particle phase was usually associated with higher molecular weight PCDD/F
344 congeners (Wu *et al.*, 2009a; Lin *et al.*, 2010; Huang *et al.*, 2011a; Mi *et al.*, 2012; Suryani R. *et al.*,
345 *et al.*, 2015). The gas phase PCDD/Fs had higher fractions in summer than in winter, similar to in
346 previous studies (Xu *et al.*, 2009; Wang *et al.*, 2010; Huang *et al.*, 2011a; Lee *et al.*, 2016). This
347 may due to lower molecular weight PCDD/Fs usually having higher vapor pressure (Wang *et al.*,
348 2010; Huang *et al.*, 2011a). As the ambient temperature increased, the fraction of gas phase
349 PCDD/Fs also rose, while when the temperature decreased some of the gas PCDD/Fs were
350 exchanged and transferred into the particle phase. As a result, lower molecular weight PCDD/Fs
351 primarily existed in the gas phase, and the gas phase PCDD/Fs fractions increased with increasing
352 temperature.

353

354 **5. PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content**

355 The contents of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ of Wuhu and Bengbu in 2015,
356 2016 and 2017 are shown in Figs. 4 (A), 4 (B), and 4 (C).

357 During 2015, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were between 0.154
358 and 0.588 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.419 ng-WHO₂₀₀₅-TEQ g⁻¹ in Wuhu, were
359 between 0.104 and 0.633 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.435
360 ng-WHO₂₀₀₅-TEQ g⁻¹ in Bengbu. While in 2016, the level of PM_{2.5}-bound total
361 PCDD/Fs-WHO₂₀₀₅-TEQ in Wuhu ranged between 0.097 and 0.619 ng-WHO₂₀₀₅-TEQ g⁻¹ and
362 averaged 0.404 ng-WHO₂₀₀₅-TEQ g⁻¹, and the level of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ
363 in Bengbu ranged between 0.158 and 0.659 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.457
364 ng-WHO₂₀₀₅-TEQ g⁻¹. And in 2017, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were
365 between 0.141 and 0.878 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.498 ng-WHO₂₀₀₅-TEQ
366 g⁻¹ in Wuhu, were between 0.155 and 0.795 ng-WHO₂₀₀₅-TEQ g⁻¹ and with an average of 0.511
367 ng-WHO₂₀₀₅-TEQ g⁻¹ in Bengbu.

368 During 2015, the lowest three monthly PM_{2.5} concentrations were for July (32.0 µg m⁻³), June
369 (34.0 µg m⁻³) and August (38.0 µg m⁻³) in Wuhu, and July (47.0 µg m⁻³), August (48.0 µg m⁻³) and
370 June (49.0 µg m⁻³) in Bengbu; however, the lowest three monthly PM_{2.5}-bound total
371 PCDD/Fs-WHO₂₀₀₅-TEQ contents occurred in July, August and June in both Wuhu and Bengbu,
372 and the levels were 0.158, 0.154, 0.259 ng-WHO₂₀₀₅-TEQ g⁻¹ and 0.104, 0.197, 0.248
373 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. But in 2016, the three months with the lowest PM_{2.5}
374 concentrations were August (25.0 µg m⁻³), July (31.0 µg m⁻³) and September (36.0 µg m⁻³) in

375 Wuhu, accompanied by the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ
376 contents, which were 0.097, 0.099 and 0.206 ng-WHO₂₀₀₅-TEQ g⁻¹ in July, August and June,
377 respectively; and the three months with the lowest PM_{2.5} concentrations were August (33.0 µg m⁻³),
378 July (38.0 µg m⁻³) and October (39.0 µg m⁻³) in Bengbu, accompanied by the lowest three monthly
379 PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, which were 0.158, 0.159 and 0.218
380 ng-WHO₂₀₀₅-TEQ g⁻¹ in July, August and June, respectively. And in 2017, the lowest three
381 monthly PM_{2.5} concentrations were for August (27.0 µg m⁻³), July (29.0 µg m⁻³) and September
382 (35.0 µg m⁻³) and in Wuhu, and August (34.0 µg m⁻³), July (36.0 µg m⁻³) and September (38.0 µg
383 m⁻³) in Bengbu; however, the lowest three monthly PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ
384 contents occurred in July, August and June in both Wuhu and Bengbu, and the levels were 0.141,
385 0.141, 0.238 ng-WHO₂₀₀₅-TEQ g⁻¹ and 0.155, 0.161, 0.287 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. It
386 can be seen that in the ambient air the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content was not
387 closely correlated with the PM_{2.5} concentration, and similar results were reported in previous
388 studies (Xing *et al.* 2017).

389 As for the seasonal variation, for Wuhu, in 2015, the PM_{2.5}-bound total
390 PCDD/Fs-WHO₂₀₀₅-TEQ contents in spring, summer, fall and winter were 0.496, 0.190, 0.410 and
391 0.580 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; and in 2016 they were 0.530, 0.134, 0.389 and
392 0.561 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; while in 2017 they were 0.786, 0.173, 0.445 and 0.588
393 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. It was found that, in Wuhu, the PM_{2.5}-bound total
394 PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer were roughly only 68.8% in magnitude lower than
395 the average value of the other three seasons (spring, fall and winter). For Bengbu, in 2015, the
396 PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in spring, summer, fall and winter were

397 0.569, 0.183, 0.441 and 0.549 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; while in 2016, they were 0.578,
398 0.178, 0.496 and 0.575 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively; and in 2017, they were 0.745, 0.201,
399 0.509 and 0.591 ng-WHO₂₀₀₅-TEQ g⁻¹, respectively. The results indicated that in Bengbu the
400 PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in summer was approximately 66.7% in
401 magnitude lower than the average values of other three seasons (spring, fall and winter). This was
402 because that the ambient temperature in summer was much higher than the average values of other
403 three seasons (spring, fall and winter) and more of the PCDD/Fs bound to the particles were
404 evaporated to the gas phase, and so the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents
405 decreased.

406

407 6. Sensitivity Analysis

408 Sensitivity analysis can provide a better basis for confirming which environmental
409 parameters are important to the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents.
410 Several parameters, such as the ambient temperature, PM_{2.5} concentration, PM₁₀ concentration and
411 total PCDD/F mass concentration, could affect the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ
412 content. In this study, sensitivity analyses were carried out depending on the initial values of
413 ambient air temperature = 22.2°C, PM_{2.5} = 58.0 µg m⁻³, PM₁₀ = 87.0 µg m⁻³ and total-PCDD/F
414 mass concentration = 1.12 pg m⁻³. The parametric sensitivity for the atmospheric PM_{2.5}-bound total
415 PCDD/Fs-WHO₂₀₀₅-TEQ contents of Wuhu is shown in Fig. 5.

416 Where P: initial value of parameters;

417 ΔP: increase or reduction in parameters;

418 S: predicted value in each of the parameters at the initial value;

419 ΔS : response in each of the parameters.

420 The sensitivity analysis indicated that the total PCDD/F mass concentration was the most
421 sensitive parameter for atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents. When
422 $\Delta P/P$ was changed from 0% to +20%, +50%, $\Delta S/S$ responded from 0% to +42%, +106%. This
423 may be because PCDD/Fs are the root cause of total PCDD/Fs-WHO₂₀₀₅-TEQ, so the change in
424 PCDD/Fs mass concentration has a significant effect on PM_{2.5}-bound total
425 PCDD/Fs-WHO₂₀₀₅-TEQ contents.

426 From Fig. 5, it also can be seen that the PM₁₀ concentration is also an important sensitive
427 parameter for the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents. When $\Delta P/P$
428 was changed from 0% to +20%, +50%, $\Delta S/S$ responded from 0% to +29%, +72%. There was a
429 strong correlation between PM₁₀ and total PCDD/F mass concentrations, and the change in PM₁₀
430 concentration has a great impact on the PCDD/F mass concentration, and thus it can affect the
431 PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents in a significant manner.

432 The sensitivity analysis also demonstrated that an increase in air temperature has a significant,
433 negative effect on atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, when $\Delta P/P$
434 was changed from 0% to 50%, $\Delta S/S$ responded from 0% to -112%. But a decrease in air
435 temperature has less effect on the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ
436 contents, when $\Delta P/P$ was changed from 0% to -50%, $\Delta S/S$ responded from 0% to +73%. The
437 temperature affects the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents through
438 changing the gas-particle partitioning of PCDD/Fs, the high molecular weight PCDD/Fs have a
439 large contribution to the total PCDD/F mass concentration and primarily existed in the particle
440 phase. When the air temperature was increasing, the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ

441 contents decreased obviously as more of the particle-bound PCDD/Fs were evaporated to the gas
442 phase; when the temperature was not high, the PCDD/Fs were mostly existed in the particle phase,
443 when the temperature decreased, the rest of the gas phase PCDD/Fs changed into the particle
444 phase, and thus the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents increased. This is
445 consistent with the conclusion that the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in the
446 summer season was only approximately 34.1% in magnitude lower than the average of the other
447 three seasons (spring, fall and winter).

448 A decrease in the PM_{2.5} concentration had a negative or positive correlation with the
449 atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents, when $\Delta P/P$ was changed from 0%
450 to -50%, $\Delta S/S$ responded from 0% to -66%. The effect of increasing PM_{2.5} can also be divided into
451 two stages: when $\Delta P/P$ was changed from 0% to +30% and +70%, $\Delta S/S$ responded from 0% to
452 +23% and +33%, respectively, but when $\Delta P/P$ was changed from +70% to +100%, $\Delta S/S$ responded
453 from +33% to +25%. The PM_{2.5} concentration affects the PM_{2.5}-bound total
454 PCDD/Fs-WHO₂₀₀₅-TEQ contents mainly in terms of the particle-bound PCDD/Fs. Lower PM_{2.5}
455 concentrations were always accompanied by a better atmospheric stability, which is beneficial to
456 the dispersion of air pollutants, and thus PCDD/Fs also decreased obviously, and the PM_{2.5}-bound
457 total PCDD/Fs-WHO₂₀₀₅-TEQ content was the value of the total-PCDD/Fs-WHO₂₀₀₅-TEQ/PM_{2.5}
458 ratio, and the atmospheric PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents also decreased
459 slowly. However, when the PM_{2.5} concentrations were higher than 86 $\mu\text{g m}^{-3}$, because the
460 PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents were calculated by the ratio of the total
461 PCDD/Fs-TEQ concentration divided by that of PM_{2.5}, a higher PM_{2.5} concentration was always
462 accompanied by a low air temperature, and thus the PM_{2.5}/PM₁₀ ratio was high, which means the

463 rate of increase of the $PM_{2.5}$ concentrations was greater than that of PM_{10} , and also higher than
464 that of the total PCDD/F mass concentration, and the rate of increase of the $PM_{2.5}$ concentration
465 was greater than that of total PCDD/Fs-WHO₂₀₀₅-TEQ concentration, and therefore the
466 $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ contents decreased when the $PM_{2.5}$ concentration
467 continuous to rise.

468 The result of the sensitivity analysis suggested that atmospheric $PM_{2.5}$ -bound total
469 PCDD/Fs-WHO₂₀₀₅-TEQ contents were most sensitive to total PCDD/F mass concentration,
470 followed by PM_{10} concentration, and then the air temperature and $PM_{2.5}$ concentration.

471

472

473 CONCLUSION

474 The results of the current investigation of both $PM_{2.5}$ and PCDD/Fs in Wuhu and Bengbu are
475 summarized as follows:

476 1. The $PM_{2.5}$ concentration of the focal three years in Wuhu ranged between 25.0 and 108.0
477 $\mu g m^{-3}$, with an average of 53.0 $\mu g m^{-3}$; for Bengbu, the figures were 33.0-101.0 $\mu g m^{-3}$ and with
478 an average of 61.4 $\mu g m^{-3}$. The $PM_{2.5}$ level in Wuhu was lower than that in Bengbu. In general, the
479 time variation of the $PM_{2.5}$ concentration in both Wuhu and Bengbu fell slowly, but was still above
480 the WHO air quality regulated standard (10 $\mu g m^{-3}$).

481 2. As for the seasonal variations, the three-year average of $PM_{2.5}$ concentration for Wuhu in
482 summer (32.3 $\mu g m^{-3}$) was 58.2% in magnitude lower than that in winter (77.3 $\mu g m^{-3}$); and the
483 value for Bengbu in summer (41.9 $\mu g m^{-3}$) was 51.7% in magnitude lower than that in winter (86.8
484 $\mu g m^{-3}$). The high temperature in summer contributed to more violent vertical air current transport,

485 which can accelerate $PM_{2.5}$ dispersion, while stronger rainfall and wind speed also enhanced the
486 removal of $PM_{2.5}$. In winter, the low temperature hindered $PM_{2.5}$ dispersion, and polluted air from
487 northern cities caused the $PM_{2.5}$ concentration to increase.

488 3. The $PM_{2.5}/PM_{10}$ ratios of the three-year range in Wuhu were in the range of 0.42-0.89, with
489 an average of 0.63, and were 0.43-0.79 with an average of 0.65 in Bengbu. A higher $PM_{2.5}/PM_{10}$
490 ratio was always associated with a higher $PM_{2.5}$ concentration. This means that $PM_{2.5}$ is the major
491 portion of atmospheric particles.

492 4. The total PCDD/Fs-WHO₂₀₀₅-TEQ concentration of Wuhu in summer (0.028
493 pg-WHO₂₀₀₅-TEQ m^{-3}) was 48.1% in magnitude lower than that in winter (0.054
494 pg-WHO₂₀₀₅-TEQ m^{-3}); and the value for Bengbu in summer (0.035 pg-WHO₂₀₀₅-TEQ m^{-3}) was
495 44.4% in magnitude lower than that in winter (0.063 pg-WHO₂₀₀₅-TEQ m^{-3}). The values for the
496 total-PCDD/Fs-WHO₂₀₀₅-TEQ concentrations of these two cities were all higher than those in
497 previous studies of Taiwan (0.046 pg-WHO₂₀₀₅-TEQ m^{-3}).

498 5. In Wuhu, the three-year average fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ
499 concentration in summer (76.4%) was 2.53 times than that in the other seasons (30.2%); while in
500 Bengbu, the fraction in summer (72.6%) was 2.87 times than that in the other seasons (25.3%). As
501 such, the fraction of gas phase total PCDD/Fs-WHO₂₀₀₅-TEQ had a significant, positive
502 correlation with air temperature.

503 6. In Wuhu, the three-year average $PM_{2.5}$ -bound total PCDD/Fs-WHO₂₀₀₅-TEQ content in
504 summer (0.166 ng-WHO₂₀₀₅-TEQ g^{-1}) was approximately only 68.8% in magnitude lower than the
505 average value of the other three seasons (0.532 ng-WHO₂₀₀₅-TEQ g^{-1}); while in Bengbu, that of in
506 summer (0.187 ng-WHO₂₀₀₅-TEQ g^{-1}) was approximately 66.7% in magnitude lower than the

507 average value of the other seasons (0.561 ng-WHO₂₀₀₅-TEQ g⁻¹). Due to the higher temperature in
508 summer, more of the particle-bound PCDD/Fs evaporated to the gas phase, and the PM_{2.5}-bound
509 total PCDD/Fs-WHO₂₀₀₅-TEQ contents thus decreased.

510 7. The sensitivity analysis of PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ in Wuhu showed
511 that the PCDD/F concentration was the most positively correlated sensitive factor, when $\Delta P/P$
512 was changed from 0% to +20%, +50%, $\Delta S/S$ responded from 0% to +42%, +106%. The second
513 positively correlated sensitive factor was the PM₁₀ concentration, when $\Delta P/P$ was changed from 0%
514 to +20%, +50%, $\Delta S/S$ responded from 0% to +29%, +72%. This was followed by the atmospheric
515 temperature, and its effect was negatively correlated, when $\Delta P/P$ was changed from 0% to +50%,
516 $\Delta S/S$ responded from 0% to -112%; when $\Delta P/P$ was changed from 0% to -50%, $\Delta S/S$ responded
517 from 0% to +73%. The last sensitive parameter was the PM_{2.5} concentration, and the impact was
518 divided into two stages: when $\Delta P/P$ was changed from 0% to +30%, +70%, $\Delta S/S$ responded from
519 0% to +23%, +33%; but when $\Delta P/P$ was changed from +70% to +100%, $\Delta S/S$ responded from
520 +33% to +25%.

521 8. The results of this study can provide useful information in the search for more insights into
522 both atmospheric PM_{2.5} and PCDD/Fs.

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685 **Figure Captions**

686 Fig. 1 (A). Monthly Average Atmospheric PM_{2.5} Concentration in Wuhu and Bengbu during 2015

687 Fig. 1 (B). Monthly Average Atmospheric PM_{2.5} Concentration in Wuhu and Bengbu during 2016

688 Fig. 1 (C). Monthly Average Atmospheric PM_{2.5} Concentration in Wuhu and Bengbu during 2017

689 Fig. 2 (A). Monthly Average PM_{2.5}/PM₁₀ Ratio in Wuhu and Bengbu during 2015

690 Fig. 2 (B). Monthly Average PM_{2.5}/PM₁₀ Ratio in Wuhu and Bengbu during 2016

691 Fig. 2 (C). Monthly Average PM_{2.5}/PM₁₀ Ratio in Wuhu and Bengbu during 2017

692 Fig. 3 (A). Seasonal Variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the
693 ambient air in Wuhu and Bengbu during 2015

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695 ambient air in Wuhu and Bengbu during 2016

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697 ambient air in Wuhu and Bengbu during 2017

698 Fig. 4 (A). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during
699 2015

700 Fig. 4 (B). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during
701 2016

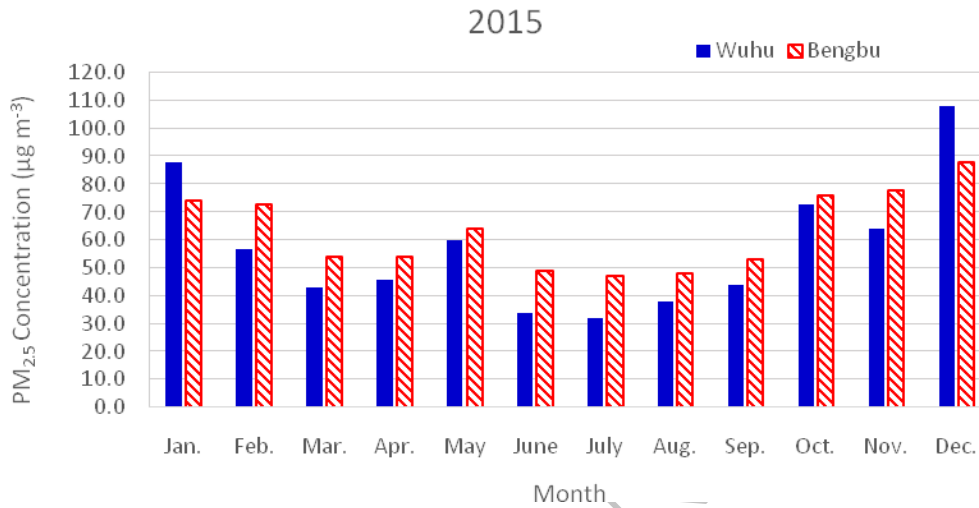
702 Fig. 4 (C). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during
703 2017

704 Fig. 5. Sensitivity Analysis for the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ Contents of Wuhu

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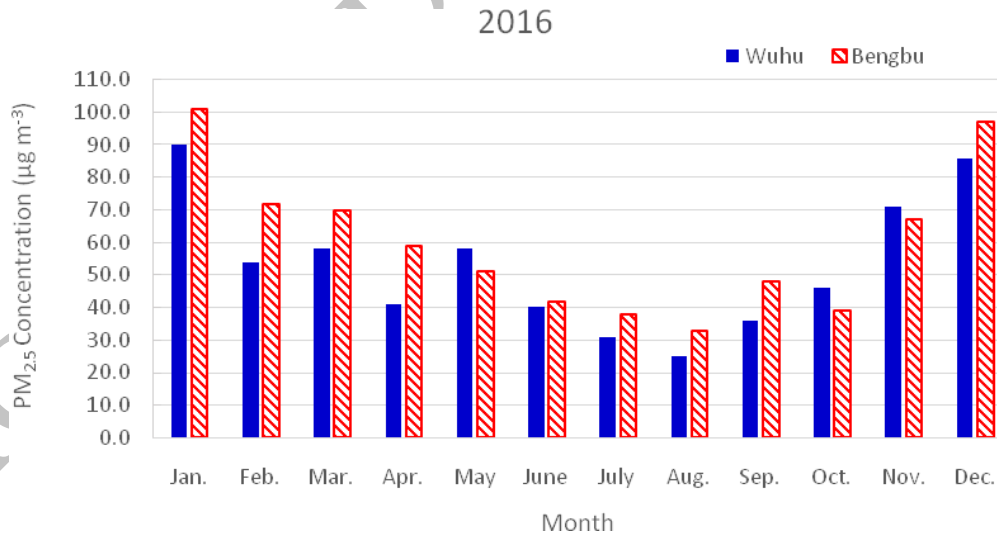
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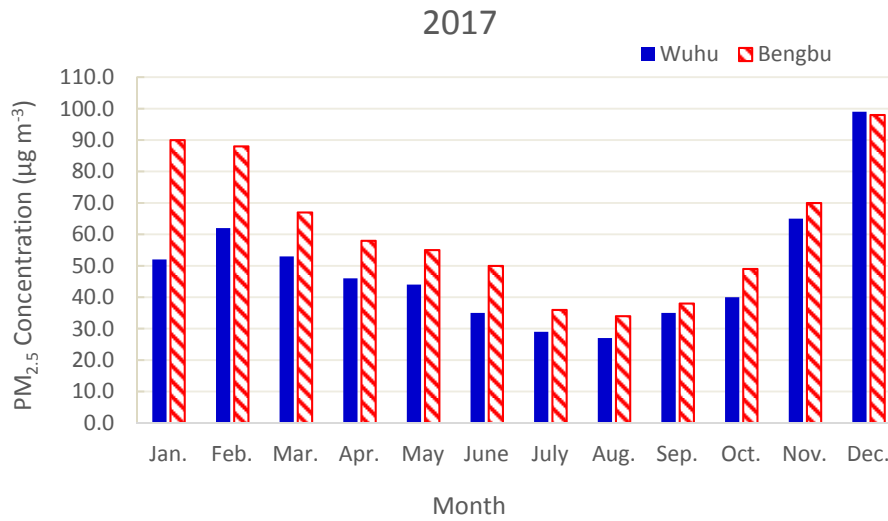
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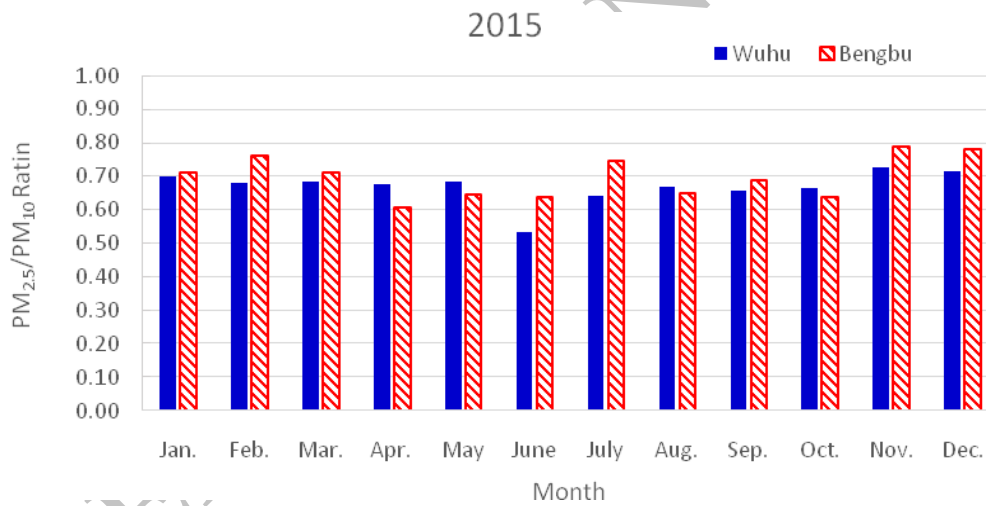
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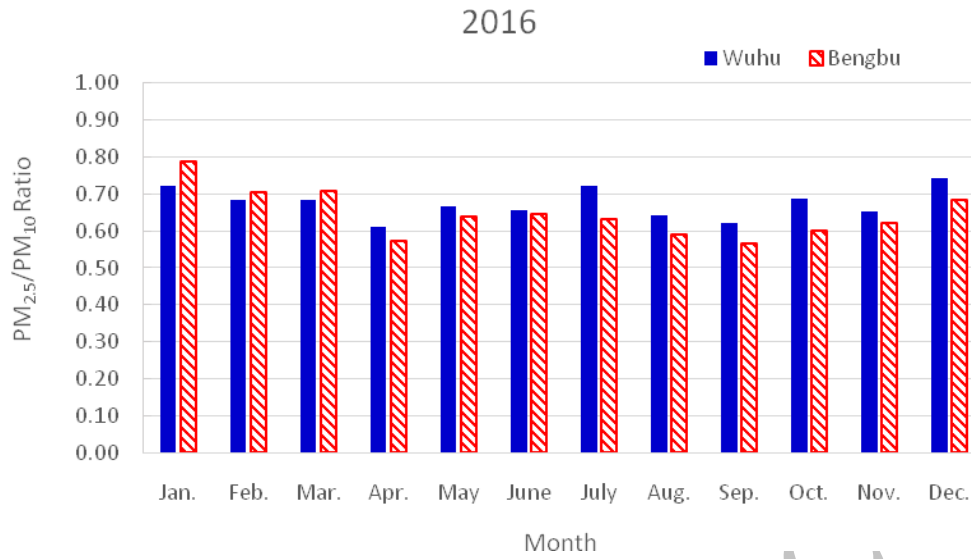
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718 Fig. 2 (A). Monthly Average PM_{2.5}/PM₁₀ Ratio in Wuhu and Bengbu during 2015

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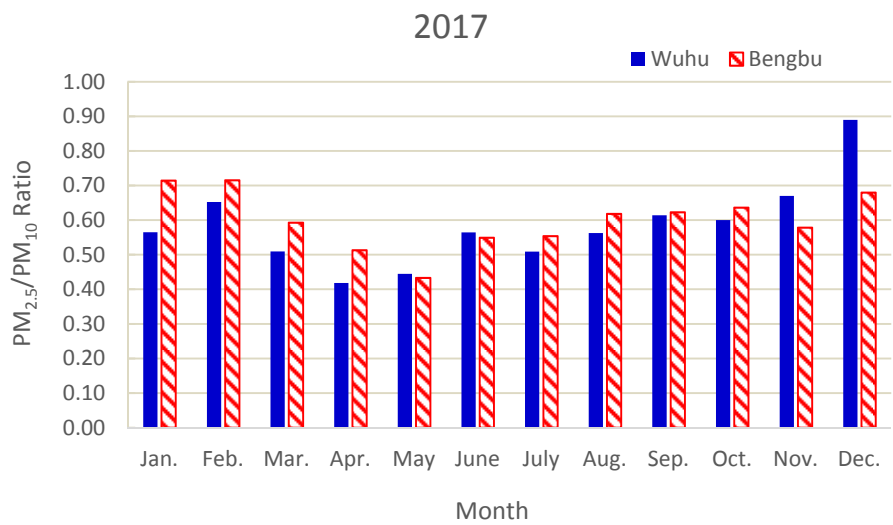


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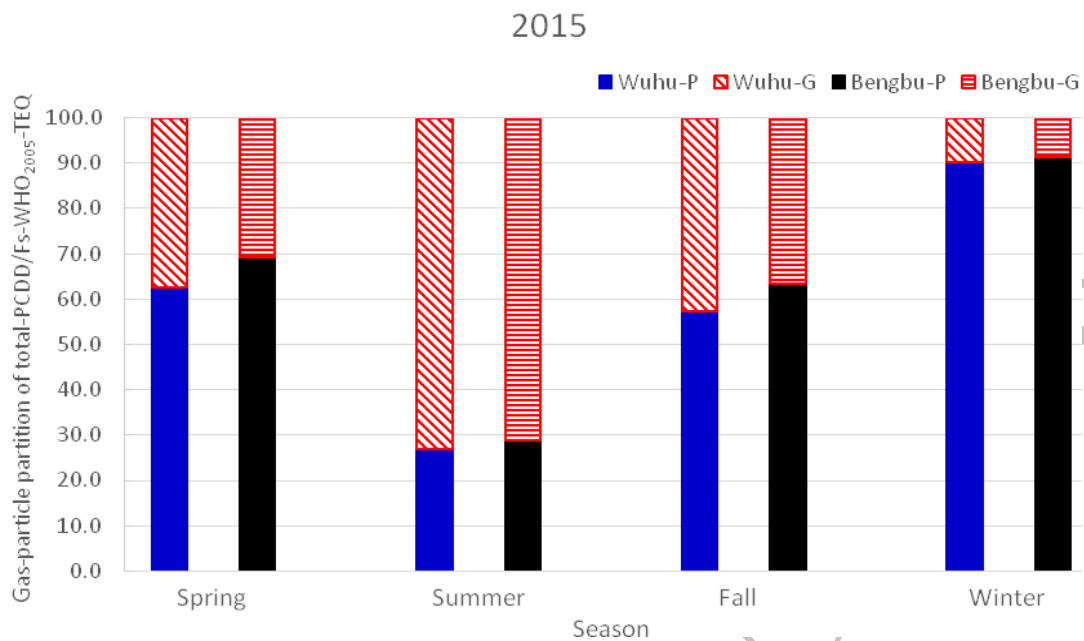


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Fig. 2 (C). Monthly Average PM_{2.5}/PM₁₀ Ratio in Wuhu and Bengbu during 2017

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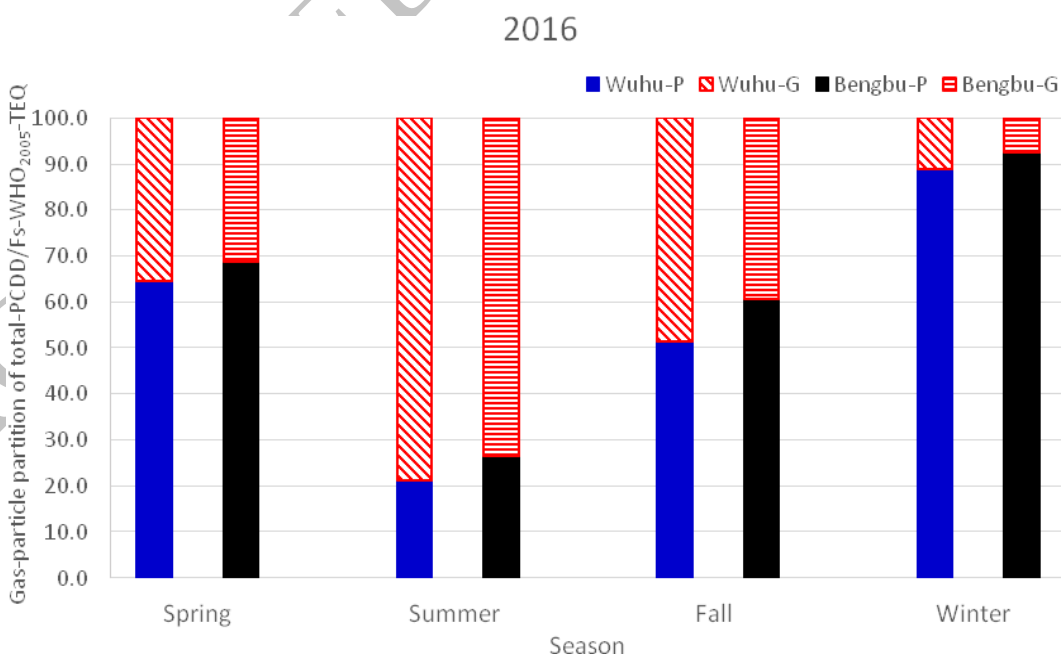
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728 Fig. 3 (A). Seasonal Variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the

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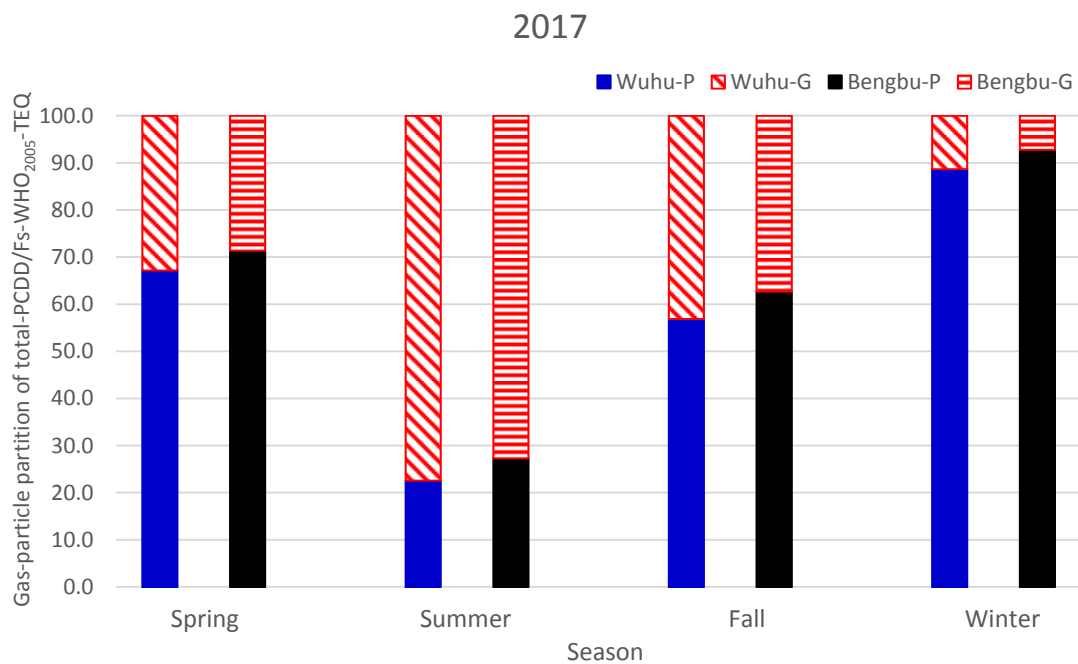
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733 Fig. 3 (B). Seasonal Variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the

734 ambient air in Wuhu and Bengbu during 2016

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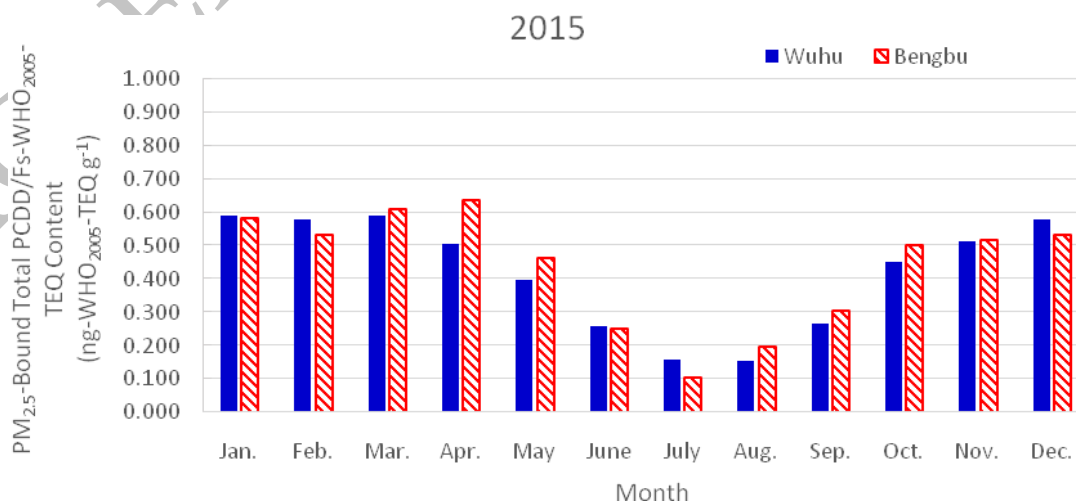


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737 Fig. 3 (C). Seasonal Variations of gas-particle partition of total-PCDD/Fs-WHO₂₀₀₅-TEQ in the

738 ambient air in Wuhu and Bengbu, respectively, during 2017

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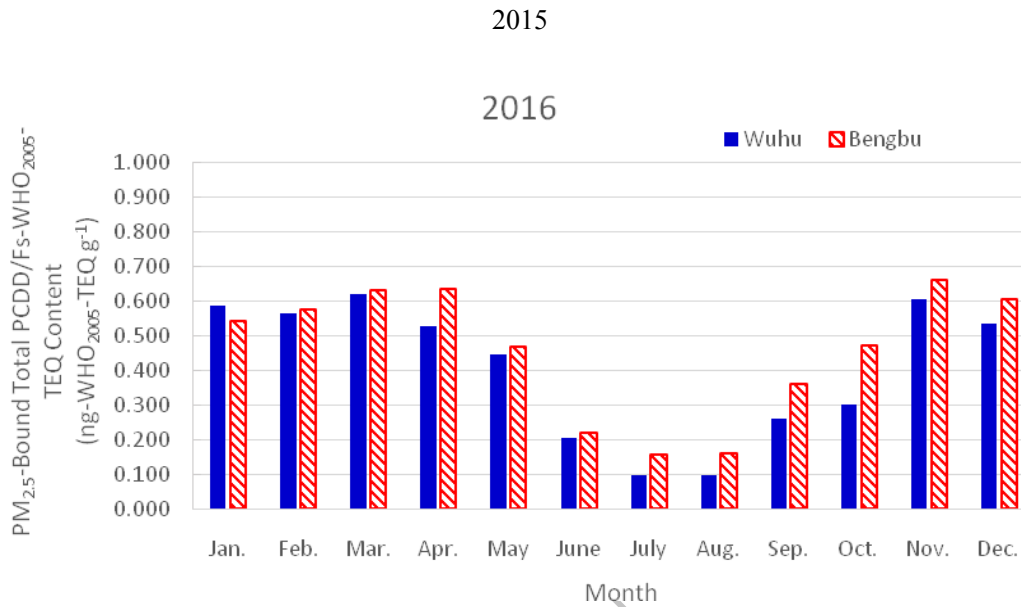


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742 Fig. 4 (A). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during

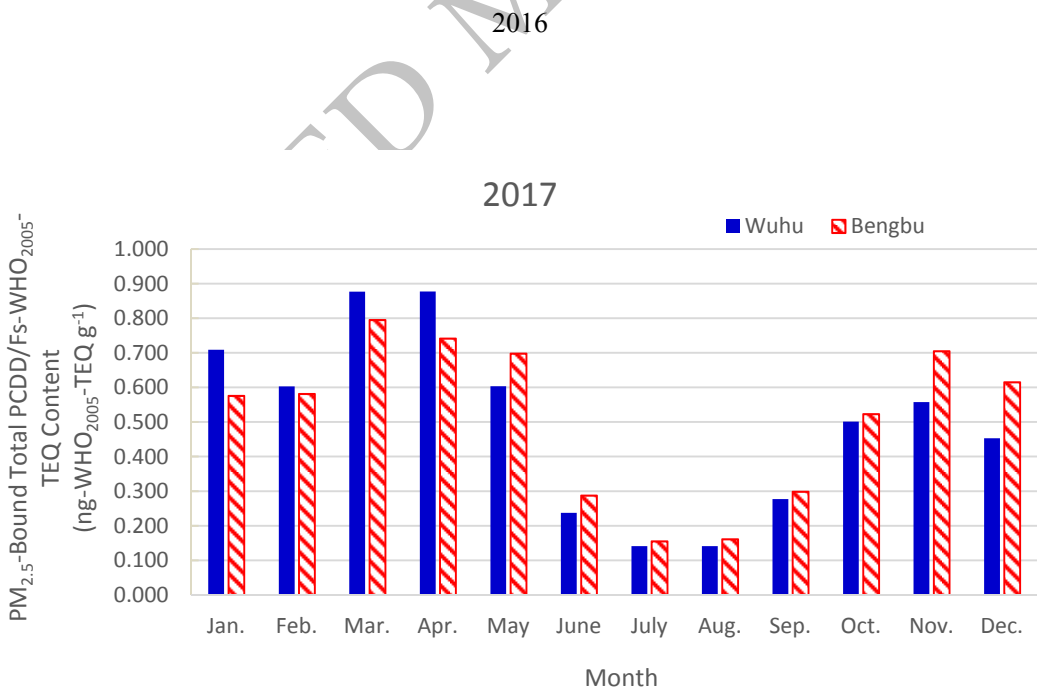
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745 Fig. 4 (B). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during

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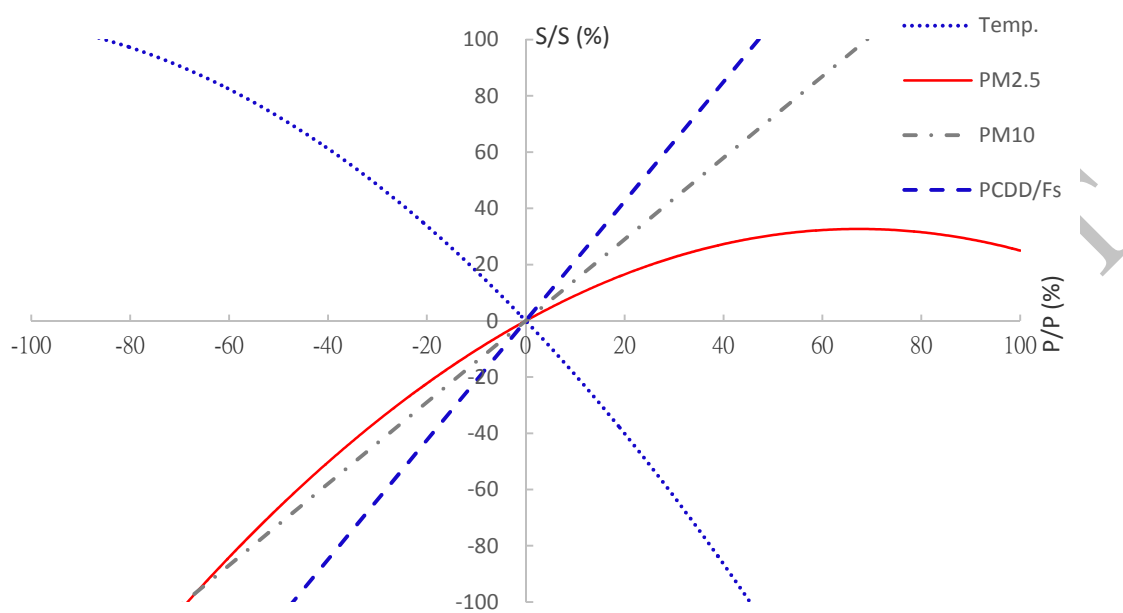
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749 Fig. 4 (C). PM_{2.5}-Bound Total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu and Bengbu during

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753 Fig.5. Sensitivity Analysis for the PM_{2.5}-bound total PCDD/Fs-WHO₂₀₀₅-TEQ Content of Wuhu

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