

1 **Organic Contamination and Remediation in the Agricultural Soils**  
2 **of China: A Critical Review**

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17  
18 **Abstract**

19 Soil pollution is a global problem in both developed and developing countries. Countries with  
20 rapidly developing economies such as China are faced with significant soil pollution  
21 problems due to accelerated industrialization and urbanization over the last decades. This  
22 paper provides an overview of published scientific data on soil pollution across China with  
23 particular focus on organic contamination in agricultural soils. Based on the related

24 peer-reviewed papers published since 2000 (n = 203), we evaluated the priority organic  
25 contaminants across China, revealed their spatial and temporal distributions at the national  
26 scale, identified their possible sources and fates in soil, assessed their potential environmental  
27 risks, and presented the challenges in current remediation technologies regarding the  
28 combined organic pollution of agricultural soils. The primary pollutants in Northeast China  
29 were polycyclic aromatic hydrocarbons (PAHs) due to intensive fossil fuel combustion. The  
30 concentrations of organochlorine pesticides (OCPs) and phthalic acid esters (PAEs) were  
31 higher in North and Central China owing to concentrated agricultural activities. The levels of  
32 polychlorinated biphenyls (PCBs) were higher in East and South China primarily because of  
33 past industrial operations and improper electronic waste processing. The co-existence of  
34 organic contaminants was severe in the Yangtze River Delta, Pearl River Delta, and  
35 Beijing-Tianjin-Hebei Region, which are the most populated and industrialized regions in  
36 China. Integrated biological-chemical remediation technologies, such as surfactant-enhanced  
37 bioremediation, have potential uses in the remediation of soil contaminated by multiple  
38 contaminants. This critical review highlighted several future research directions including  
39 combined pollution, interfacial interactions, food safety, bioavailability, ecological effects,  
40 and integrated remediation methods for combined organic pollution in soil.

41 **Keywords:** Organochlorine pesticides; Polycyclic aromatic hydrocarbons; Polychlorinated  
42 biphenyls; Phthalate esters; Agricultural soil; Combined organic contamination

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## 63 **1. Introduction**

64       The contamination of agricultural soil has been observed around the world due to the  
65 long-term application of pesticides, fertilizer, plastic film, wastewater irrigation, sewage  
66 application and other activities. Unlike contamination in industrial sites, agricultural soil  
67 pollution and its adverse impacts tend to be chronic. The cumulative loading of pollutants in

68 agricultural soil may pose significant risks to the ecological functions of soils, plant growth,  
69 and eventually, human health. For example, nitrobenzene inhibited the growth of soybean  
70 seedlings and caused genotoxicity in soybean root tip cells (Guo et al., 2010). Polycyclic  
71 aromatic hydrocarbons (PAHs) were found to change the abundance of functional genes in  
72 soils (Han et al., 2014). Exposure to PAHs damaged both enzymatic and non-enzymatic  
73 antioxidant defenses and harmed human health (Garcon et al., 2001). Agricultural soils were  
74 often contaminated in large areas by combined and non-point source pollution, which are  
75 difficult to address through normal remediation operations.

76 The global occurrence and adverse effects of organic pollution have caused increasing  
77 public concern. Many organic contaminants (OCs), such as organochlorine pesticides (OCPs),  
78 polychlorinated biphenyls (PCBs), phthalate esters (PAEs), and PAHs, are characterized by  
79 high toxicity, persistence, and bioaccumulation in the environment (Pies et al., 2007; Sun et  
80 al., 2016c). Vast areas of farmland soils are an important reservoir for OCs (Zhang et al.,  
81 2013c; Zhong and Zhu, 2013), which can in turn serve as a secondary emission source to air  
82 and water (Bidleman and Leone, 2004; Tao et al., 2008; Cabrerizo et al., 2011). Many OCs in  
83 agricultural soils are difficult to degrade biologically under normal environmental conditions.  
84 The residues of OCs in agricultural soils can enter food chains and eventually present a  
85 potential risk to human health through trophic transfers (Fantke and Jolliet, 2015; Liu et al.,  
86 2016c).

87 As one of the major agricultural countries, China has extensive arable land areas (2.03  
88 billion acres) with various soil types (e.g., Anthrosols and Histosols) (IUSS Working Group  
89 WRB, 2014) and a wide variety of soil contaminants. Thus, the national-scale summary in

90 this review can serve as a representative example for addressing the problem of global  
91 agricultural soil pollution. Recently, the State Council of China published the Action Plan on  
92 Prevention and Control of Soil Pollution (The Central Government of the People's Republic  
93 of China, 2016). Understanding the spatial distribution and temporal trend of OCs in  
94 agricultural soils across China is necessary for devising pragmatic and enforceable legislation.  
95 The fate, source, and interfacial behavior of coexisting organic chemicals in soil should also  
96 be studied for efficient pollution control and soil remediation. The revealed spatiotemporal  
97 variations based on the published data can help to explain the combined pollution situation in  
98 agricultural soils and the effect of pollution regulation in China since 2000. There has been a  
99 large volume of literature on organic pollution in the agricultural soils of China. However,  
100 these surveys of varying scale (e.g., site-specific, city-wide, or regional-scale) have been  
101 scattered, and there is no clear understanding of the status of organic contamination in  
102 agriculture throughout China, given that the types of OCs vary across the vast farmland of the  
103 country. Most relevant studies have focused on a single class of OCs in a relatively small  
104 sampling area during a very short sampling period. A critical review is greatly needed to  
105 summarize the concentration levels and characteristics of combined organic pollution in  
106 agricultural soils at a national scale.

107       The primary pollutants in the densely populated regions of China have not been assessed.  
108 The priority contaminants that should be controlled would vary across different regions due  
109 to the disparity in industrial operations, agricultural practices, and economic development  
110 levels. For example, in the Yangtze River Delta, Pearl River Delta, and Beijing-Tianjin-Hebei  
111 Region, the combined contamination of pollutants may be more severe due to intensive

112 industrial and urbanization activities. Some regions accommodate numerous electronic waste  
113 recycling sites or coal industry plants, whereas some regions are made up of large and  
114 intensive agricultural farmland. Reviewing the up-to-date literature can reveal the geographic  
115 distribution of OCs and inform the development of management plans for combined pollution.  
116 The source, fate, and risk of coexisting OCs in agricultural soils should be evaluated to reveal  
117 their different source-sink mechanisms and devise an effective control strategy. A low-cost  
118 and effective approach is essential for the remediation of extensive areas of polluted  
119 agricultural soils in addition to industry-impacted soil. The challenges involved in developing  
120 proper remediation technologies for soil contaminated with multiple organic contaminants  
121 need to be better understood and well addressed.

122 The present paper provides an extensive and critical review of studies on agricultural  
123 soils in China, particularly in addressing the recent literature regarding organic pollution. A  
124 total of 203 papers about organic contaminants in Chinese agricultural soils have been  
125 published since 2000, of which a total of 155 papers focused on the four classes of OCs  
126 (OCPs, PAHs, PCBs, and PAEs), implying that they are of significant concerns within the  
127 current research (Fig. S1). The objectives were to (a) reveal the concentration levels, regional  
128 characteristics, spatial distribution, and temporal variation of principal organic contaminants  
129 in the agricultural soils of China; (b) assess the development and challenges involved in  
130 remediation technologies for combined organic pollution in agricultural soils; and (c) identify  
131 and suggest possible future directions for research in this field.

## 132 **2. Characteristics of organic contaminants in agricultural soils**

### 133 *2.1. OCPs*

134 OCPs were extensively used in agriculture for pest control (Li et al., 2001; Liu et al.,  
135 2015b). Although their usage was banned for decades, OCPs remain ubiquitous in the  
136 environment worldwide even in remote regions (He et al., 2013; Huang et al., 2014c). As a  
137 large agricultural country, China has been the largest consumer of pesticides in the world.  
138 Abundant OCPs were reportedly produced during the 1950s-1980s, e.g., a total of 4.5 million  
139 tons of HCHs and 0.44 million tons of DDTs (Cao et al., 2007).

140 More than 20 OCPs have been detected in Chinese agricultural soils (Fig. S2). DDT,  
141 HCB, heptachlor, Aldrin, and dieldrin were listed in the Stockholm Convention on Persistent  
142 Organic Pollutants (POPs) by the United Nations Environment Program (UNEP) (World  
143 Wide Fund, 2005). HCHs, endosulfan, and methoxychlor have been proposed as potential  
144 POP candidates (World Wide Fund, 2005). Furthermore, HCH is considered to be a primary  
145 pollutant by the USEPA (UNEP, 2003). The total concentrations of OCPs ranged from < LOD  
146 (limit of detection) to 3,520 ng/g, with a mean of  $58.9 \pm 51.5$  ng/g ( $n = 40$ ) (Table 1). The  
147 concentrations of OCPs in Central China were higher than other regions (Kruskal-Wallis,  
148  $0.289 < p < 0.796$ ), possibly due to intensive agricultural activities (Fig. 1 and Fig. 2). The  
149 amount of historical pesticide usage (including major OCPs) in farmland was the largest in  
150 this region, especially Henan Province (Zhao, 2013). Among the reported OCPs, *p,p'*-DDE  
151 exhibited the highest concentration (mean  $\pm$  standard deviation:  $14.6 \pm 20.7$  ng/g,  $n = 66$ ),  
152 followed by *p,p'*-DDT (mean:  $12.7 \pm 15.8$  ng/g,  $n = 66$ ) and chlordane (mean:  $8.36 \pm 26.7$   
153 ng/g,  $n = 15$ ). Generally, the total concentrations of DDTs and HCHs ranged from < LOD to  
154 3,515 ng/g (mean:  $41.6 \pm 57.2$  ng/g,  $n = 68$ ), and from < LOD to 760 ng/g (mean:  $11.4 \pm 18.2$   
155 ng/g,  $n = 64$ ), respectively (Table 1), which was comparable to those reported in Romania and

156 Germany (Covaci et al., 2001; Manz et al., 2001). According to the Chinese Environmental  
157 Quality Standard for Soil (GB-15618-1995) (1995), the average concentrations of DDTs and  
158 HCHs in all the reports were within in the Class II Limit (Safe). However, the concentrations  
159 of DDTs and HCHs in some samples exceeded this safety standard. For example, a relatively  
160 high level of DDTs was detected in samples collected from Jiangsu, Tianjin, Hohhot, and  
161 Jiangxi (Table 1). To date, the environmental quality standard concerns only DDTs and HCHs.  
162 Several studies revealed that some OCPs, such as chlordane and methoxychlor, also exhibited  
163 high concentrations in agricultural soils, indicating that the standard should be updated for  
164 better soil protection.

165 Soil becomes the major sink and reservoir of OCPs (Tao et al., 2008). However, OCPs  
166 can also be emitted into the atmosphere and be recycled continuously between the  
167 atmosphere and the terrestrial environment. The air-soil exchange analysis of OCPs could  
168 help to predict the distribution patterns of OCPs in a certain region (Liu et al., 2016b). It was  
169 reported that the re-emission of OCPs from soil is a dynamic process that may be affected by  
170 soil properties. Previous studies suggested that DDTs were much easier to be volatilized from  
171 soil to air in Zhejiang as compared to southern Ontario, Canada (Kurt-Karakus et al., 2006;  
172 Zhang et al., 2012a), indicating that DDTs have higher volatility in soils with higher  
173 temperatures and lower organic contents. Comparing evaporation of OCPs from soil to air  
174 showed that *p,p'*-DDE,  $\gamma$ -HCH, and  $\delta$ -HCH have a higher tendency to evaporate from soil to  
175 air when compared with *p,p'*-DDD and *p,p'*-DDT (Yu et al., 2013).

176 To assure the quality of agricultural products, particularly in polluted areas, it is  
177 necessary to study the bioavailability of OCs that can help evaluating plant uptake and



178 selecting appropriate crops. It is known that the organic pollutants in soils could enter plant  
179 via root uptake (Fantke and Jolliet, 2015). Tao et al. (2005) proposed that the accumulation of  
180 OCPs was lower in tuberous plants as compared to the plants of fibrous root system. The  
181 accumulation of OCPs in plants is closely related to the properties of the chemical concerned  
182 and soil medium, for instance, high concentration of SOM in soil could inhibit the  
183 accumulation of OCPs in plants (Wu and Zhu 2016). Recently, it was reported that the  
184 accumulation of OCPs in plants could be influenced by nanoparticles (De La Torre-Roche et  
185 al., 2012).

186 Temporal trends in OCPs in agricultural soils during the last 16 years were analyzed (Fig.  
187 3). Except for one study (Wang et al., 2016a), the concentrations of OCPs in recent studies  
188 are lower than those that were identified decades ago, indicating that the residues of OCPs  
189 decreased significantly after the ban on agricultural uses. Considering that some samples still  
190 exhibited high concentrations of OCPs, there may be some new inputs of OCPs, such as  
191 dicofol and lindane (Sun et al., 2016c). The spatial analysis results show that the average  
192 levels of OCPs in the soils of North and Central China were higher (Fig. 2). The areas, such  
193 as Henan and Hebei, in which the agricultural crop sown area is 14,425 and 8,739 hectares,  
194 respectively, are also important production bases for foodstuffs, cotton, and edible oil crops  
195 (Zhou et al., 2013a; National Bureau of Statistics of China, 2017). Large quantities of OCPs  
196 have been used intensively to increase agricultural production for several decades (Wang et  
197 al., 1999).

198 The concentrations of OCPs were also closely associated with the land use types. The  
199 concentrations of HCHs and DDTs in vegetable soils were reportedly higher than those in

200 paddy soils collected from the Pearl River Delta (Li et al., 2006), which was probably  
201 because the anaerobic-aerobic rotation conditions in paddy fields were more favorable for  
202 OCP degradation (Hao et al., 2008). However, according to some other studies, the total  
203 concentrations of OCPs in soils decreased in the order paddy field > upland field, because  
204 there were greater amounts of OCPs applied during the rice growth periods (An et al., 2005;  
205 Gao et al., 2013). Taking the technical HCH as an example, it was estimated that  
206 approximately half of this insecticide was used in rice paddies (Li et al., 2001).

## 207 2.2. PAHs

208 PAHs tend to be retained in soils due to their high hydrophobicity (Terashima et al.,  
209 2003). PAHs are formed primarily during the incomplete combustion or pyrolysis of organic  
210 matter. Sixteen PAHs were identified in a priority control list by the USEPA. Seven PAHs,  
211 namely, BaA, Chr, BbF, BkF, BaP, DahA, InP and BghiP, are confirmed to be carcinogenic  
212 (Harvey, 1991). Some studies have indicated that China suffers serious PAH contamination  
213 from the combustion of fossil fuel and biomass. The total emission of the 16 priority PAHs  
214 was 25,300 t in 2003 for China, contributing to over 20% of the global total PAH emissions  
215 (Xu et al., 2006; Zhang and Tao, 2009).

216 PAHs have been detected in a large number of agricultural soil samples. The total PAH  
217 concentrations ranged from < LOD to 27,580 ng/g, with a mean of  $772 \pm 895$  ng/g ( $n = 40$ )  
218 (Table 2). The concentration of carcinogenic PAHs ranged from < LOD to 7,940 ng/g, with  
219 an average of  $464 \pm 889$  ng/g ( $n = 17$ ). In Northeast China, the concentrations of PAHs were  
220 higher than other regions (Kruskal-Wallis,  $0.180 < p < 0.938$ ) due to extensive coal combustion  
221 and petroleum industrial operations (Fig. 1 and Fig. 2). Among the carcinogenic PAHs, Chr

222 and BbF were the most abundant compounds in the soil (Fig. S3). There is currently no  
223 recommendation or guideline for regulating the occurrence of PAHs in agricultural soils in  
224 China. A classification of soil contamination based on PAHs was proposed as follows, in a  
225 previous study: non-contaminated soil (< 200 ng/g); weakly contaminated soil (200-600 ng/g);  
226 contaminated soil (600-1000 ng/g) and heavily contaminated soil (> 1000 ng/g)  
227 (MaliszewskaKordybach, 1996). According to this classification, the concentrations of PAHs  
228 exceeded the limits of heavily contaminated soil in the soils of Nanjing (Ge et al., 2006;  
229 Wang et al., 2015a), Shanghai (Sun et al., 2008), Taizhou (Tang et al., 2010), the Pearl River  
230 Delta (Cai et al., 2007), Beijing (Ma et al., 2005; Zhou et al., 2013b), Changchun (Chen et al.,  
231 2016), and Xi'an (Zhou et al., 2012) (Table 2).

232 The study of air-soil exchange of PAHs showed that LMW PAHs were more easily to be  
233 volatilized from the soil, whereas the HMW PAHs exhibited a net deposition from the  
234 atmosphere (Liu et al., 2011). Previous study demonstrated that the soil moisture could  
235 accelerate the volatilization of PAHs from soil (Wang et al., 2015c). HMW PAHs were more  
236 easily to be adsorbed to soils due to higher hydrophobicity, in other words, the volatilization  
237 tendency of HMW PAHs was much lower than that of LMW PAHs (Wang et al., 2015c; Cetin  
238 et al., 2017; Dumanoglu et al., 2017).

239 Accumulation of PAHs in plants was associated with the properties of PAHs and the  
240 species of plants. It was proposed that HMW PAHs have greater mobility from soils to plants.  
241 The accumulation of PAHs was greater in vegetables than corn, exemplifying the  
242 physiological difference between different plants (Chen et al., 2016). It should be noted that  
243 the direct contact of plant roots with water might facilitate the uptake of PAHs from soils to

244 plants (Wang et al., 2015d). The cropping pattern of plants could be a major factor that  
245 influences the accumulation of PAHs in plants. Furthermore, a sudden decrease in PAH  
246 concentrations was observed in the vegetative organs (roots, stems, and leaves) of rice during  
247 the heading stage, which may be due to the organic nutrient-associated transport of PAHs  
248 from the vegetative organs to the rice seeds. Factors, such as the properties of PAHs, plant  
249 species, tillage method, and growth time, should be considered comprehensively with respect  
250 to food safety and phytoremediation concerns.

251       The temporal trends in PAHs in the farmland topsoil are shown in Fig. 3. During the last  
252 16 years, the concentration of PAHs showed no significant increasing or decreasing trends.  
253 However, the total PAH concentrations in Tianjin in 2012 were reportedly higher than those  
254 in 2008 (Chen et al., 2015). Further studies should be conducted to measure the temporal  
255 trends of PAHs in the city or in certain regions. The PAH levels in Northeast and Northwest  
256 China were higher than those in the other regions (Fig. 2). The higher PAH concentrations in  
257 the soils of Northeast China are potentially the result of frequent coal combustion for  
258 home-heating in the winter, especially in rural areas. Shaanxi and Xinjiang of Northwest  
259 China are also important coal and petroleum production areas in China. The production of  
260 coal and petroleum could cause significant PAH pollution in the environment. In addition,  
261 due to the rapid growth of industrial production, population, and traffic density, cities in East  
262 China are also faced with serious PAH pollution.

263       Several studies have compared the concentrations of PAHs in different types of fields.  
264 The total concentrations of PAHs in vegetable fields were higher than they were in paddy  
265 fields (Yang et al., 2007a; Cao et al., 2013). Multi-cropping practices, such as cover cropping,

266 double cropping, and integrated crop-livestock systems, have been employed in the vegetable  
267 fields to meet the growing demand for agricultural commodities. Thus, more agricultural  
268 materials, such as organic fertilizer and pesticide, were applied to the vegetable fields, and  
269 they led to PAH pollution (Yang et al., 2007a). However, the deep plowing activity in paddy  
270 fields may also dilute the LMW PAHs in surface soil, but facilitate their transfer to the  
271 subsoil (Wang et al., 2012b).

### 272 2.3. PCBs

273 PCBs are halogenated aromatic compounds, and they were recognized as POPs by the  
274 Stockholm Convention in 2001 (Zhang et al., 2008). Some PCB congeners, such as PCB-77,  
275 81, 105, 114, 118, 12, 126, 156, 157, 167, 169, and 189, were considered as dioxin-like PCBs  
276 due to their higher toxicities (Sun et al., 2016b). Despite being banned in many countries,  
277 PCBs still represent an important class of pollutants that present high risks to humans and  
278 ecosystems (Su et al., 2012; Sun et al., 2016b).

279 Soils are among the most important environmental sinks for PCBs. However, in  
280 comparison with the studies on OCPs and PAHs, the surveys on PCBs in agricultural soils  
281 were fewer (Fig. S1). The total concentrations of PCBs in agricultural soil were between <  
282 LOD and 202 ng/g, with an average of  $9.31 \pm 15.4$  ng/g ( $n = 21$ ) (Table 3 and Fig. S4). The  
283 total concentrations of dioxin-like PCBs ranged from < LOD to 78.9 ng/g, with a mean of  
284  $4.64 \pm 4.41$  ng/g ( $n = 6$ ). Higher PCB levels were found in South (Kruskal-Wallis,  
285  $0.180 < p < 0.655$ ) and East China (Kruskal-Wallis,  $0.088 < p < 0.827$ ), as compared to other  
286 regions (Fig. 1 and Fig. 2). The average non-cancer risk of PCBs was relatively high (Hazard  
287 Index = 44.0 for children, and 24.7 for adults) in agricultural soils from the Yangtze River

288 Delta (Sun et al. 2016b). High cancer risks from PCBs in children and adults were also found  
289 in that region. The high risks were primarily from the dioxin-like PCBs, such as PCB-126,  
290 PCB-81, and PCB-169, which should be considered for new legislation and policy relating to  
291 soil pollution protection in the future.

292 Li et al. (2010) demonstrated that soil was more supersaturated when compared to air  
293 since the PCB concentrations in the air declined relatively faster, whereas the concentrations  
294 in the soils needed longer to respond. Generally, the soil composition and environment  
295 parameters, including the organic carbon, particle size composition, soil porosity, relative  
296 humidity, and temperature, could affect the air-soil exchange of PCBs. A comparison of the  
297 coupled soil and air concentration in China has been conducted by Zhang et al (2008), in  
298 which the low-chlorinated PCBs were more easily volatilized to air, while the  
299 high-chlorinated PCBs were more strongly bound to soil particles. Soil could be an important  
300 contributor of the low-chlorinated PCBs (very volatile 2-Cl and in some cases 3-Cl PCBs)  
301 that were observed in the air. For high-chlorinated PCBs, the soil was likely to be the sink for  
302 a long time (Li et al., 2010).

303 Accumulation of low-chlorinated PCBs was higher in plant tissue as compared to the  
304 high-chlorinated PCBs (Li et al., 2015b). The accumulation of PCBs in plant was also plant  
305 species dependent: higher accumulation of PCBs was found in wild plants, followed by the  
306 vegetable (*Raphanus sativus* L., *Brassica rapa* L., etc.) and rice stalk (Wang et al. 2011b).  
307 The accumulation of PCBs in plant was positively correlated with the growing time (Li et al.,  
308 2015b).

309 Higher concentrations of PCBs were observed in the electronic waste-dismantling areas

310 in China (especially for Guiyu and Taizhou). It has been estimated that globally,  
311 approximately 40 million tons of e-waste are generated every year, with an increasing rate of  
312 4% per year (Wong et al., 2007). Most of the world's e-waste is exported from developed  
313 countries to developing countries such as China (Fu et al., 2012a). Discarded PCB-containing  
314 equipment has become an important source of PCBs in the environment (Ogunseitan et al.,  
315 2009; Sepulveda et al., 2010; Wang et al., 2013a), and it may explain the elevated PCB  
316 concentrations in the soils of Guangdong and Zhejiang (Cai et al., 2008).

317 The concentrations of PCBs varied with the land-use types, in the order paddy fields >  
318 upland fields > uncultivated fields (Zhang et al., 2007; Wang et al., 2010b; Zhang et al.,  
319 2013d; Sun et al., 2016b). This difference is probably related to the soil conditions and  
320 planting patterns. The water in the paddy fields reduces the volatilization of PCBs to the air,  
321 and it prevents the aerobic degradation of PCBs. However, the concentrations of PCB in the  
322 upland field soils of Guangdong province were nearly four times higher than those in the  
323 paddy field soils (Wang et al., 2011b). The probable reason is that the deep plowing activity  
324 of paddy fields and the irrigation water diluted the PCBs in the topsoil and transferred them  
325 to the deep soil layer.

#### 326 2.4. PAEs

327 PAEs are a class of chemicals that are widely used as plasticizers in polyvinyl acetates,  
328 polyvinyl chloride, and polyurethanes (Wang et al., 2010a). Due to their potentially  
329 hazardous impacts on human health and the environment, both the USEPA and the China  
330 State Environmental Protection Administration have classified six PAEs as priority pollutants.

331 During the last decade, PAEs were frequently identified in farmland from different

332 regions of China (Cai et al., 2005; Yang et al., 2007b; Zhang et al., 2015; Sun et al., 2016c).  
333 PAEs exhibited the highest levels among the studied POPs with an average concentration of  
334  $3,738 \pm 5,840$  ng/g ( $n = 21$ ) in Chinese soils, covered from  $< \text{LOD}$  to  $157,620$  ng/g (Table 4).  
335 The highest concentration was observed in the farmlands of Hubei (Wu et al., 2015) (Fig. 1).  
336 The high concentration of PAEs might be due to the widespread application of plastic  
337 materials, such as agricultural plastic film. The extensive use of fertilizer and pesticides could  
338 also cause elevated PAE levels. In the polluted area, DEHP presented the highest  
339 concentrations (mean:  $3,011 \pm 5,964$  ng/g,  $n = 14$ ), followed by DBP (mean:  $1,131 \pm 2,039$   
340 ng/g,  $n = 7$ ) (Fig. S5).

341 The studies on the air-soil exchange of PAEs indicated that the polluted soils could be a  
342 secondary source of atmospheric contamination of PAEs. Previous studies suggested that one  
343 of the dominant losses of PAEs from the soils was attributable to soil-air volatilization  
344 (Cheng et al., 2015). DMP was more readily volatilized to air as compared with other five  
345 EPA-PAEs, and the volatilization from soil to air decreased with the increase of  $\log K_{ow}$  of  
346 PAEs.

347 The studies of the uptake, translocation, and accumulation of PAHs in plant showed that  
348 the accumulation of MnBP in plants was higher as compared with DnBP, DEHP, and MEHP  
349 (Sun et al., 2015). The concentrations of PAEs differed in various plants (Ji et al., 2014; Sun  
350 et al., 2015). More studies on the accumulation and tissue distribution of PAEs in plants  
351 should be conducted in future.

352 No significant temporal trend in PAEs was observed in China during the last 16 years  
353 (Fig. 3). However, the concentrations of PAEs in South China showed an increasing trend



354 from 2005 to the time of this study. The production and utilization of agricultural plastic films  
355 have reportedly increased in recent years, which could be causing the increasing pollution in  
356 South China. In general, the concentration of PAEs in South China was much higher than  
357 those in other regions (Fig. 2 and Table 4). Some cities in South China, such as Guangzhou,  
358 were large metropolitan areas and intensive agricultural areas for a long-time period. Along  
359 with the fast urbanization and industrialization of the last few decades, the environmental  
360 quality of these urban areas in South China has severely deteriorated.

361 The cultivation methods of farmland might affect the residual levels and fates of PAEs in  
362 the soils. The concentrations of PAEs were lower in non-cultivated fields when compared  
363 with the cultivated fields (Xu et al., 2008). This finding was because the cultivation methods  
364 and farm management could introduce PAEs into the fields. Regarding the cultivated fields,  
365 the concentrations of PAEs were higher in paddy fields than those in the upland field (Wang  
366 et al., 2013b). Plastic agricultural films, municipal biosolids, agricultural chemicals, and  
367 wastewater irrigation have been identified as the main sources for PAEs contamination in  
368 soils (He et al., 2015). Surface coverage by irrigation water could partially result in different  
369 PAE levels between paddy and upland fields. In paddy fields, the surface soils were  
370 frequently covered with water, which may considerably reduce the volatilization of PAEs  
371 (Wang et al., 2013b). Moreover, the water coverage may greatly reduce the contact that PAEs  
372 have with sunlight irradiation and air exposure, thus lowering the abiotic degradation of the  
373 PAEs (Wang et al., 2013b). By contrast, PAEs were more easily degraded by microbes under  
374 aerobic conditions, and thus a higher biodegradation of PAEs was found in upland soils (Liu  
375 et al., 2010). However, it is also reported that orchard and vegetable fields showed higher

376 PAEs concentration than paddy field since orchard and vegetable fields require more  
377 intensive application of fertilizers and pesticides, and more plastic packing (Liu et al. 2010).

### 378 **3. Remediation of soil organic contamination**

379 Various organic pollutants remain ubiquitous in the agricultural soils of China. These  
380 pollutants will not only pose adverse effect to soil properties, but they can also enter food  
381 chains and eventually pose risks to human health. It is thus important to reduce and eliminate  
382 organic pollutants and lower the contamination that is already present in the environment.  
383 Many remedial attempts, including physical, chemical, and biological treatments, have been  
384 made to reach an environmentally sound and cost-effective remediation of contaminated  
385 lands. A summary table of all the remediation technologies for soil organic pollution  
386 according to different categories is provided in Table S1 (Supporting Information).

387 Previous studies have demonstrated that physical remediation (such as thermal treatment  
388 and vapor extraction technology) and chemical remediation (such as oxidation and reduction  
389 technology, and soil washing technology) could be effectively used to remedy organic  
390 pollutant-contaminated soils (Gan et al., 2009; Falciglia et al., 2016; Jia et al., 2016).  
391 However, the use of physical or chemical remediation technologies may severely  
392 compromise the biological and chemical quality of soils (Lim et al., 2016). Problems such as  
393 high equipment and treatment costs and damage to soil quality have not been resolved, which  
394 limits the large-scale application of physical or chemical methods in the remediation of  
395 extensive areas of agricultural soil that is contaminated by persistent organic pollutants. To  
396 date, physical and chemical remediation methods are mainly used for industry-impacted soil.

#### 397 *3.1. Bioremediation and phytoremediation*

398 The area of pollution in farmland is usually much larger than that of contaminated  
399 industrial fields. The contamination degree of agricultural soil is comparably lower than that  
400 of the soil in industrial zones. The remediation of agricultural soil requires the protection of  
401 soil fertility and ecological function. Compared with physical and chemical remediation,  
402 bioremediation and phytoremediation have greater potential applications for the remediation  
403 of agricultural soil because they have the advantages of low cost, easy *in situ* operation, and  
404 no secondary pollution. Bioremediation/phytoremediation in agricultural soils have been  
405 applied both in China and abroad (Gerhardt et al., 2009; Teng et al., 2010; Ma et al., 2012).

406 Bioremediation, primarily microbial degradation, is a natural way of eliminating  
407 pollutants by breaking down polluted substrates for nutrients. Soil microorganisms, either in  
408 the presence of oxygen (aerobic biodegradation) or without oxygen (anaerobic  
409 biodegradation), have been found to degrade and mineralize OCs. Bioremediation of OCs has  
410 been successfully conducted in agricultural fields under natural conditions (Labana et al.,  
411 2005; Odukkathil and Vasudevan 2016; Ayotamuno et al., 2006). Small-scale field studies  
412 showed that *p*-Nitrophenol could be completely depleted in the presence of *Arthrobacter*  
413 *protophormiae* AKJ100 (Labana et al., 2005). Efficiency of bioremediation could be  
414 optimized by agitation, aeration, and addition of nutrients. For instance, heterotrophic  
415 bacteria have been used in remediation of crude-oil polluted agricultural soil. Results showed  
416 that between 75 and 200 g of fertilizer per 0.16 m<sup>2</sup> lead to the best biodegradation  
417 (degradation efficiency covered from 93% to 95%) (Ayotamuno et al., 2006). Highly  
418 chlorinated PCBs could be the substrate for aerobic bacteria (Gomes et al., 2013), and they  
419 can be dechlorinated to form less chlorinated congeners under anaerobic conditions

420 (Furukawa and Fujihara, 2008). The significant anaerobic biodegradation of LMW PAHs  
421 such as fluorine, phenanthrene, and pyrene has been observed in paddy soil with nitrate being  
422 used as an electron acceptor (Ambrosoli et al., 2005). The microbial degradation of PAEs in  
423 soils is more effective under aerobic than anaerobic conditions (Yuan et al., 2002). The  
424 biodegradation efficiency of fluxapyroxad in soils was investigated, and the results showed  
425 that higher oxygen levels caused a higher potential for fluxapyroxad degradation (Li et al.,  
426 2015c). Currently, researchers are proposing that coupling anaerobic and aerobic conditions  
427 could increase the efficiency of remediation strategies (Master et al., 2002; Meade and  
428 D'Angelo, 2005). Sequential anaerobic then aerobic composting was suggested for the  
429 biodegradation of PCBs (Long et al., 2015).

430 Plants have been reported to mineralize a wide range of complex organic pollutants into  
431 non-toxic constituents such as chlorine, carbon dioxide, and nitrate (Vidali, 2001).  
432 Phytoremediation is an environmentally friendly, low cost, and sustainable method for  
433 remediation. The accumulation of OCPs in plant tissue has been studied (Gonzalez et al.,  
434 2005). The concentration of OCPs in plant tissues might be 4- to 45-fold higher than those in  
435 soils. The grasses of the *Poaceae* and the legumes of the *Fabaceae* were considered as  
436 excellent candidates for removing PAHs from soils. Moderately hydrophobic chemicals are  
437 most likely to be bioavailable to rooted and vascular plants. Some hydrophilic OCs could also  
438 be taken up by plants through hydrogen bonding with transpiration water or the partition  
439 effect (Chiou et al., 2001; Dietz and Schnoor, 2001). The OCs in the plants might then be  
440 translocated to the stem and leaf tissues, which were subsequently degraded (Liu and Schnoor,  
441 2008; Zhai et al., 2013). Phytoextraction and phytovolatilization are the two approaches used

442 to remove and detoxify OCs in agricultural soils. Phytoextraction removes pollutants from  
443 soil by concentrating them in the harvestable plant part. Previous studies have demonstrated  
444 that zucchini (*Cucurbita pepo*) is a good accumulator of OCs from agricultural soil (Wang et  
445 al., 2004; White et al., 2006). For phytovolatilization, some OCs could be converted into  
446 volatile forms within the plants and subsequently volatilized. It has been reported that  
447 compounds with low octanol-air partitioning coefficients ( $\log K_{oa} < 5$ ) are more prone to  
448 being phytovolatilized (Limmer and Burken, 2016). Field experiments showed that the plant  
449 could facilitate the mobilization and/or degradation of OCs in soils and the *Fimbristylis*  
450 *littoralis* could be useful for the bioremediation of crude oil polluted agricultural soils (up to  
451 92% of total PAHs were removed after 90 days) (White, 2001; Nwaichi et al., 2015).

### 452 3.2. Integrated remediation technologies

453 Each remediation technology discussed in the preceding section has its own problems.  
454 Therefore, some integrated remediation technologies were developed.  
455 Plant-microbe-associated bioremediation has been used for agricultural soil remediation.  
456 Synergistic interactions between plants and microbial communities in the rhizosphere are  
457 effective for the degradation of recalcitrant OCs (Vergani et al., 2017). Root exudates (amino  
458 acids, flavonones, sugars, enzymes, phenolic compounds and other organic materials) could  
459 enhance the bioavailability of OCs and microbial activities in the immediate vicinity of the  
460 roots (Javorska et al., 2009). To eliminate PAHs with increasing ring numbers, root  
461 exudate-enhanced degradation becomes more important (Sun et al., 2010). Rhizosphere  
462 microbes can occur naturally, or they can be activated by introducing specific microbes into  
463 the rhizosphere (Gerhardt et al., 2009). Chaudhry et al. (2005) suggested that microbial

464 strains capable of breaking down OCs were widely present in rhizosphere soils.

465 Bioremediation alone was usually unable to remove persistent and highly toxic pollutants  
466 quickly from agricultural soil (Huang et al., 2017). The use of a biological treatment as a  
467 secondary step after chemical remediation has been performed (Kulik et al., 2006). It was  
468 ascertained that a combined Fenton-like treatment and biological remediation was more  
469 efficient at PAH removal than either one alone. Surfactant-enhanced bioremediation (SEBR)  
470 is a promising technology for improving the bioavailability and removal efficiency of OCs in  
471 agricultural soil (Wang et al., 2016d). Several studies have shown that surfactant can not only  
472 increase the partition of OCs to microbial cells, but it can also facilitate the transmembrane  
473 transportation of OCs into the cells and hence accelerate intracellular biodegradation (Zhang  
474 and Zhu, 2012; Li and Zhu, 2014; Li et al., 2014). Different types of surfactants imposed  
475 various impacts on the biodegradation of PAHs via distinct mechanisms such as disrupting  
476 bacterial membranes and the modification of cell surface hydrophobicity (Zhang et al.,  
477 2013a). Recently, the ring-hydroxylating dioxygenase (RHDase) and the  
478 1-hydroxyl-2-naphthoate dioxygenase genes (1H2Nase), which play a key role in the  
479 decomposition of hydrophobic aromatic compounds, were found to be induced in the  
480 presence of surfactants (Li et al., 2015a).

481 Surfactants could be applied to increase the solubility and bioavailability of OCs and thus  
482 assist in the mobilization of OCs into the soil solution (Chirakkara et al., 2016). A recent  
483 study revealed that surfactants could enhance the bioavailability of PAHs and the efficiency  
484 of plant-microbe-associated bioremediation (Ni et al., 2014). Another study found that  
485 surfactant can enhance the degradation of DDTs by microorganisms in agricultural soil

486 (Wang et al., 2016e). Overall, surfactant-enhanced bioremediation could be a promising  
487 technology for addressing combined organic pollution in agricultural soil, and it should be  
488 developed in future studies.

#### 489 **4. Future research needs**

##### 490 *4.1. Combined pollution and interfacial interactions*

491 Because of the rapid growth of industrial processes, traffic density, agricultural activities,  
492 and the population, a wide range of chemicals such as POPs, heavy metals, antibiotics, and  
493 nanomaterials may coexist in the soil environment at elevated concentrations. From the  
494 reviewed literature, it is clearly illustrated that the research highlights have gradually  
495 transferred from single pollution to combined pollution in recent years. Emerging pollutants  
496 enter the soil ecosystem and possibly interact with conventional pollutants that remain from  
497 the historical contamination (Zhu et al., 2005; Li et al., 2013a). These potential interactions  
498 between the coexisting chemicals and soil constituents can influence their behavior and fates  
499 in soils (Liang et al., 2015). There were recent studies on combined pollution in a simulated  
500 aqueous environment (Lei et al., 2016) and an atmospheric environment (Pandey et al., 2013;  
501 Ercan and Dinçer, 2015; Hassanvand et al., 2015). Future studies are needed to reveal the  
502 adsorption, transportation, and transformation of multi-class pollutants in soil environment  
503 and at the air-water-soil interfaces. It is important to clarify the interactions associated with  
504 physical and chemical processes for the pollution control and remediation of contaminated  
505 agricultural soils to accomplish environmental sustainability under a rapidly changing climate.  
506 Climate changes (temperature and precipitation, etc.) and more often occurrence of extreme  
507 weather may affect the soil quality and the air-soil exchange of OCs (Dumanoglu et al., 2017),

508 which should be considered in the future.

509 A number of factors may influence the interfacial interactions. The characteristics of  
510 contaminants (i.e., their hydrophobicity, polarity, and electron density), and the composition  
511 of natural and amended soil media (i.e., minerals, humus, biochar, and nanoparticles) could  
512 be the key contributing factors. It is important to reveal the molecular-level mechanisms of  
513 the pollutant fate and interactions in future studies. For example, electrospray ionization  
514 coupled with Fourier-transform ion cyclotron resonance mass spectrometry (ESI-FT-ICR-MS)  
515 have been proved helpful in the characterization of interfacial behavior (Lv et al., 2016). To  
516 characterize the interactions in soil interfaces, new analytical methods should be developed.

#### 517 *4.2. Ecological effects on soil microbial communities*

518 Soil microbes are crucial to biogeochemical cycles on Earth and play a key role in soil  
519 ecosystems. Numerous studies have found that environmental pollution may affect the  
520 metabolic activities of soil microbes and consequently change the soil microbial community  
521 composition and diversity (Liu et al., 2015a; Sun et al., 2016b). Pollutants in soils could  
522 decrease the diversity of microorganisms but enrich the tolerant species via the environmental  
523 filtering process, which may in turn affect the overall ecosystem functions and natural  
524 balance of soil microorganisms. The consequence of soil microbial responses to  
525 environmental changes is still the key question that is in urgent need of an answer.

526 For example, antibiotics along with antibiotic resistance genes (ARGs) have been  
527 extensively investigated through a metagenomic study recently (Martinez, 2009; Xu et al.,  
528 2016). The large amounts of antibiotics applied to agricultural production systems can exert  
529 selection pressure on environmental microorganisms, contributing to proliferating antibiotic



530 resistance in microorganisms. The ARGs were further disseminated among bacteria through  
531 horizontal gene transfer mechanisms via various mobile genetic elements (Fang et al., 2015).  
532 However, methods for analyzing the genetic diversity in soil microbes were rarely employed  
533 in the study of soil pollutants, such as PAHs, OCPs, PCBs, and PAEs. There were very few  
534 field studies focused on the effects of combined pollution on the indigenous microbiota. The  
535 responses of soil microbiota community to the combined pollution and its ecological  
536 consequences should be explored in the future.

537       However, soil microorganisms also play an important role in the remediation of polluted  
538 soil through active involvement in the degradation and transformation of different organic  
539 compounds. Understanding the effects of OC pollution on soil ecosystems is helpful for  
540 developing the selection and application of microbes for remediation purposes.  
541 High-throughput sequencing has enabled the in-depth exploration of microbial biodiversity in  
542 the polluted soil environment. The rapidly developing metagenomic analysis provides  
543 advanced technology for exploring the ecological consequences caused by combined organic  
544 pollution of soil. The predictions for indigenous bacteria in terms of their ability to degrade  
545 various pollutants on the dynamics of agricultural production are necessary for systematic  
546 ecological evaluations.

#### 547 *4.3. Bioavailability, food safety, and risk assessments*

548       The uptake and accumulation of OCs in crops is closely related to their bioavailability  
549 (White, 2002). Although a great number of studies on the occurrences of OCs in Chinese  
550 agricultural soils have been published, few data are available on the bioavailability of OCs in  
551 soil. Thus, one of the major issues is to investigate the *in vivo* bioavailability of OCs in a

552 wide range of edible crops and poultry animals, to provide important information on risk  
553 evaluations of OCs in soil and agricultural products. To predict bioavailability accurately, the  
554 distribution of OCs and the influencing factors should be studied. The available concentration  
555 in the soil integrated with the plant metabolism coefficient and the foliar uptake factor should  
556 be considered (Sun et al., 2016a). The regulation principle regarding the bioavailability of  
557 OCs based on the behaviors at multimedia soil-water-air-plant-cell interfaces have not been  
558 revealed yet.

559 Food safety is closely related to the bioavailability and accumulation of OCs in  
560 agricultural produce, which is a priority for future research. The availability of arable land is  
561 limited in many countries such as China. In response to rapid population growth and  
562 increasing food demand, inappropriate strategies including excessive application of pesticides  
563 and fertilizers have been adopted to achieve high production. These agricultural practices can  
564 lead to the accumulation of pollutants in vegetable and crops. Root-accumulated DDTs in  
565 vegetables were reported from 7.1 to 152 ng/g (cabbage, spinach, etc.) in a study (Tao et al.,  
566 2005). Total concentrations of PAHs in the corn along main roadside ranged from 220 to 627  
567 ng/g (Chen et al., 2016). Organic pollutants at these concentrations may present serious  
568 health risk for human consumption. PAEs were found to show broad inhibition on the activity  
569 of enzymes UDP-glucuronosyltransferases (UGTs) (Cao et al., 2017). Hydroxylated-PCBs  
570 are considered to disrupt the thyroid hormone metabolism (Meerts et al., 2002). Therefore, to  
571 protect the diet safety of local residents, it is essential to understand the accumulation status  
572 and potential food safety risk in soil-vegetable system.

573 The potential risks of OCs to human health and the ecological system are less understood.

574 Due to the widespread distribution of OCs, individuals are unavoidably exposed to OCs via  
575 dietary and non-dietary (e.g., ingestion, dermal contact, and inhalation) routes. Soil is the  
576 major sink and reservoir of pollutants in the environment. Soil-air partitioning makes soil a  
577 secondary source of pollutants to the atmosphere (Sweetman et al., 2005; Cabrerizo et al.,  
578 2011). The leaching of pollutants from soil is one of the most important threats to the  
579 groundwater quality (Hantush et al., 2000; Calderon-Preciado et al., 2011). In addition, global  
580 climate change can affect the pollution situation of the environment (Nadal et al., 2015;  
581 Tambo et al., 2016). It is necessary to thoroughly evaluate the risk from soil pollutants and  
582 establish a systematic strategy for the control of the whole process. Valid and realistic risk  
583 assessments of OCs to both human health and ecological receptors should be considered in  
584 future studies to provide key information for risk management and remediation decisions.

#### 585 *4.4. Novel control and remediation method*

586 Control and remediation methods for soil pollution should be developed according to the  
587 land use types and pollution levels. Compared to industrial sites in China, farmlands are  
588 characterized by large areas of low-level but combined pollution. The integrated techniques  
589 of chemical treatment and bioremediation methods have many advantages. For agricultural  
590 soils with low pollution, mitigation methods may be developed to enhance the sorption of  
591 pollutants to soil components, for reducing their bioavailable concentrations as well as  
592 decreasing the uptake and accumulation in plants. The addition of biochar or surfactant has  
593 been demonstrated to be useful for fixing pollutants in soil (Zhou and Zhu, 2008; Kusmierz et  
594 al., 2016). However, the biochemistry processes and mechanisms in the crop rhizosphere  
595 have not been clearly evaluated. New mitigation methods should aim to produce safe food in

596 relatively lightly polluted soil.

597 The selection of suitable crops is also important for the sustainable utilization of  
598 agricultural soil. Collaborations between plants and soil microorganisms can accelerate the  
599 degradation of OCs, although the mechanism still must be elucidated (Jin et al., 2013; Ni et  
600 al., 2014). For agricultural soils with heavy pollution, remediation methods should be  
601 developed to remove OCs from the soil and assure the environmental safety of farmlands.  
602 The addition of certain chemicals such as biosurfactants can promote the elution of OCs from  
603 soil particles (Song et al., 2008). The development of nanotechnology has attracted  
604 significant attention. Nanomaterials have great application potentials for the remediation of  
605 polluted soil due to their superior reactivity (Zhan and Jiang, 2015; Zhu et al., 2015). The use  
606 of nanomaterials for enhancing plant-microbe-associated bioremediation technology has a  
607 bright future. The recently developed electrokinetic-biological remediation of  
608 pesticide-contaminated soil has an advantage in that it enables *in situ* pollutant elimination  
609 (Barba et al. 2017). Monitored natural attenuation has attracted increasing attention from the  
610 perspective of economic management strategies. The prudent and controlled use of naturally  
611 occurring pollutant degradation and retardation processes is applicable for a large area of  
612 complex contaminated agricultural soils (Rugner et al., 2008). In addition, to ensure  
613 productivity, the control and remediation technology for agricultural soil in China should  
614 consider a field-scale application in vast areas of farmland as well as different regions of  
615 China.

## 616 **5. Conclusions**

617 As widely detected environmental pollutants, OCPs, PAHs, PCBs, and PAEs are the

618 primary organic contaminants in the agricultural soils of China. The average concentrations  
619 of PAEs, PAHs, OCPs, and PCBs in agricultural soils of China were  $3,724 \pm 5,844$  ng/g,  $772$   
620  $\pm 895$  ng/g,  $58.9 \pm 51.5$  ng/g, and  $9.31 \pm 15.4$  ng/g, respectively. These organic contaminants  
621 present distinct spatial patterns across the nation. Therefore, management guidelines for  
622 organic pollution should be adapted to local conditions. It is necessary to conduct regular  
623 surveys to monitor the evolutionary trends. Integrated remediation technologies, such as  
624 surfactant-enhanced bioremediation, should be further refined for the cost-effective  
625 remediation of large areas of contaminated farmlands. The combined pollution, interfacial  
626 interactions, ecological effects, bioavailability, food safety, risk assessment, and integrated  
627 pollution control and remediation methods for agricultural soil pollution are major research  
628 topics in the future. More effective and imperative activities are needed for the management  
629 and control of organic contaminants in the agricultural soils in developing countries.

630

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637

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1345 **Figure legend:**

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1347 **Fig. 1:** National distributions of OCPs, PAHs, PCBs, and PAEs in agricultural soils of China.

1348 The average concentrations were calculated from the data in literatures of the same province.

1349 The name of province/city where only one paper was published was given in the map.

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1351 **Fig. 2:** Concentrations of organic pollutants in agricultural soils of different regions. The

1352 primary regions include East China (e.g., Shandong, Anhui, Jiangsu, Shanghai, and Zhejiang),

1353 South China (e.g., Fujian, Guangdong, Hainan, and Guangxi), North China (e.g., Inner

1354 Mongolia, Shanxi, Hebei, Beijing, and Tianjin), Central China (e.g., Jiangxi, Hunan, Hubei,

1355 and Henan), Northeast China (e.g., Heilongjiang, Jilin, and Liaoning), Southwest China (e.g.,

1356 Guizhou, Yunnan, Chongqing, Sichuan, and Tibet), and Northwest China (e.g., Xinjiang,

1357 Qinghai, Gansu, Ningxia, and Shaanxi). The bars represent mean values. The error lines

1358 represent the maximum.

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1360 **Fig. 3:** Temporal trends of organic pollutants in the agricultural soils of China.

Table 1 Concentrations of OCPs (ng/g) in agricultural soil of China

Location	Total concentrations of DDTs			Total concentrations of HCHs			Total concentrations of OCPs			References
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	
East China										
Zhejiang/Shanghai/Jiangsu	0.13	3515	56.2	0.37	30.3	2.46	1.0	3520	59.3	(Sun et al., 2016c)
Shandong	3.88	79.55	26.51	0.41	9.67	4.01	5.07	87.77	32.58	(Geng et al., 2006)
Anhui	—	—	17.61	—	—	28.64	—	—	48.58	(Wang et al., 2011a)
Shanghai	0.14	77.89	15.80	1.14	22.43	4.52	3.11	117.47	26.25	(Lv et al., 2011)
Zhejiang	0.08	5.39	1.22	0.05	5.90	2.20	ND	41.3	—	(Feng et al., 2012)
Jiangsu	3.31	43.81	11.13	0.81	9.43	3.6	—	—	—	(Ge et al., 2006)
Jiangsu	6.3	1050.7	64.1	2.7	130.6	13.6	10.3	1059.6	77.6	(An et al., 2005)
Shandong	0.46	17.6	5.84	ND	7.11	1.49	—	—	—	(Li et al., 2016)
Jiangsu	ND	600	88.8	ND	99	3.23	—	—	—	(Hu et al., 2014)
Shandong	—	—	19.8	—	—	21.8	7.17	171	44	(Zhu et al., 2014)
Jiangsu	ND	184.02	23.33	ND	42.59	8.26	4.80	219.10	32.2	(Gao et al., 2013)
Zhejiang	4.0	529	82	—	—	—	4.0	529	82	(Zhang et al., 2012a)
Zhejiang	—	—	—	0.028	2.80	1.23	—	—	—	(Zhu et al., 2017)
Jiangsu	ND	600	88.8	ND	99	3.2	—	—	—	(Hu et al., 2011)
Shanghai	0.44	247.45	21.41	ND	10.38	2.41	3.16	265.24	—	(Jiang et al., 2009)
Jiangsu	0.46	484.24	28.87	0.28	17.93	3.23	—	—	—	(Zhang et al., 2009)
Jiangsu	4.2	678.6	99.81	—	—	—	4.2	678.6	99.81	(Yang et al., 2008)
Zhejiang/Shanghai/Jiangsu	0.57	302.73	36.49	—	—	—	0.57	302.73	36.49	(Li et al., 2008b)
Jiangsu	19	92.6	50.2	10.1	41.5	28.5	—	—	86.4	(Wang et al., 2007a)
Anhui	ND	211	23.7	1.51	14	4.69	3.63	227	29.7	(Meng et al., 2013)
South China										
Fujian	6.51	469.5	51.17	0.41	9.61	2.02	7.36	473.09	53.19	(Lin et al., 2013)
Fujian	0.01	107.99	7.48	1.05	25.07	3.98	—	—	—	(Zhang et al., 2011b)
Fujian	0.22	384.75	32.86	1.39	247.4	22.75	2.7	632.15	55.61	(Huang et al., 2014b)
Fujian	0.1	240.38	14.94	0.78	90.71	5.03	2.56	465.99	38	(Qu et al., 2013)
Guangdong	0	152.7	26.9	0	104.4	15.3	4.6	1021.5	113.4	(Hao et al., 2007a)
Hainan	0	12.38	1.25	0	2.23	0.16	ND	17.37	2.30	(Wu et al., 2014)
Guangdong	1.82	60.3	8.57	0	1.74	0.53	2.71	62.4	11.9	(Yue et al., 2012)
Fujian	0.51	241.3	29.61	0.45	151.21	14.51	3.66	658.42	78.83	(Qu et al., 2015)
Guangdong	ND	110	8.8	ND	62	3.4	—	—	—	(Yu et al., 2013)
Fujian	10.54	1.53	53.26	0.41	7.90	2.125	—	—	—	(Yang et al., 2013a)
Fujian	0.94	700.99	71.17	0.38	39.52	9.51	—	—	—	(Zhang et al., 2012b)
Fujian	0.64	78.07	3.86	0.72	30.16	9.79	—	—	—	(Yang et al., 2012)
Fujian	0.91	27.89	6.725	0.96	7.47	3.18	4.62	38.2	12.58	(Zhang et al., 2011c)
Guangdong	1.93	199	60.8	0.35	281	22	—	—	—	(Ni et al., 2011)
Guangdong	—	—	82.1	—	—	4.42	—	—	86.5	(Ma et al., 2008)
Guangdong	0.27	414	—	ND	24.1	—	—	—	—	(Li et al., 2006)
North China										
Tianjin	—	—	16.085	—	—	93.61	—	—	140.07	(Zhang et al., 2010)
Beijing	18.04	101.33	64.44	11.64	29.80	15.77	—	—	—	(Ma et al., 2003)
Beijing	ND	64.91	6.46	ND	14.97	0.73	—	—	—	(Wang et al., 2012a)
Tianjin	ND	616.98	—	ND	92.74	—	ND	690.9	62.78	(Lv et al., 2010)
Beijing	ND	116.74	6.64	ND	5.56	0.67	—	—	—	(Hu et al., 2010)
Tianjin	—	—	157.5	—	—	52.8	—	—	—	(Tao et al., 2005)
Beijing	ND	57.9	9.82	ND	7.33	0.69	—	—	—	(Tieyu et al., 2005)
Beijing	7.21	2910	381.3	2	760.3	32	—	—	—	(Shi et al., 2005)
Tianjin	0.7	972.2	56.0	—	—	—	—	—	—	(Gong et al., 2004)
Hohhot	ND	994.39	137.22	4.84	281.44	52.39	10.69	1384.12	184.87	(Zhang et al.,

2013b)

Central China										
Jiangxi	0	43.04	7.11	0.37	7.52	1.66	—	—	—	(Xie and Ding, 2013)
Henan	—	—	135.3	—	—	89.0	—	—	—	(Wang et al., 2009c)
Hubei	8	570	68	4.9	27	11	21	590	88	(Liu et al., 2016a)
Hunan	0.33	2421	111.2	—	—	—	—	—	115.3	(Li et al., 2008a)
Henan	ND	206.1	54.9	2.9	56.4	20.3	85.0	1392.1	193	(Wang et al., 2016a)
Jiangxi	ND	1690	16.35	0	178	1.46	—	—	—	(Teng et al., 2015)
Hubei	0.31	5.36	1.80	0.23	3.77	1.27	1.26	22.15	6.49	(Liu et al., 2015b)
Hubei	ND	1198	151.56	ND	100.58	15.39	11.43	1253.3	196.59	(Zhou et al., 2013a)
Jiangxi	0.14	6.981	2.138	—	—	—	0.14	6.981	2.138	(He et al., 2013)
Northeast China										
Liaoning	1.6	21.55	6.67	0.7	2.61	1.42	—	—	—	(Teng et al., 2013)
Jilin	1.45	81.75	—	1.17	27.83	—	1.45	83.88	—	(Liu 2013)
Jilin	3.16	48.35	13.19	4.37	44.77	11.17	12.10	72.51	29.69	(Zhang et al., 2016b)
Jilin	0.94	107.81	16.59	0.89	98.29	11.62	2.44	177.06	31.95	(Zhang 2016a)
Liaoning	ND	40.25	—	ND	42.79	—	ND	51.32	6.86	(Shi et al., 2011)
Heilongjiang	0.72	28.2	5.430	0.136	51.8	7.12	—	—	—	(Wang et al., 2009b)
Heilongjiang	0.072	28.222	5.84	0.136	51.762	9.3	0.28	81.34	17.5	(Ma 2007)
Southwest China										
Sichuan	—	—	1.73	—	—	1.08	—	—	—	(Xing et al., 2010)
Guizhou	—	—	14.39	—	—	5.595	—	—	—	(Wei et al., 2007)
Yunnan	—	—	—	2.88	17.62	7.448	—	—	—	(He et al., 2010)
Chongqing	4.31	213.5	41.76	0.55	26.54	4.05	7.29	222.42	46.15	(Fu et al., 2012b)
Sichuan	—	—	53.89	—	—	5.01	20.18	104.33	61.46	(Pan et al., 2011)
Tibetan	ND	41.6	1.36	ND	8.36	0.349	—	—	—	(Wang et al., 2016c)
Sichuan	0.29	5.72	1.755	0.43	10.6	1.93	0.97	17.6	4.915	(Gai et al., 2014)
Northwest China										
Xinjiang	ND	40.03	18.51	ND	30.86	14.37	16.4	84.86	41.89	(Chen et al., 2014)
Lanzhou	0.141	120	16.9	0.0822	4.49	0.865	—	—	—	(Mao et al., 2013)
Gansu/Xinjiang/Qinghai/Ningxia	0.1	120.49	12.52	0.17	9.38	1.45	0.9	133.44	—	(Huang et al., 2014c)
Shaanxi	ND	80.9	7.65	0.01	86.16	2.22	0.33	130.34	11.05	(Wei et al., 2015)
Shaanxi	—	—	5.75	—	—	0.45	—	—	6.2	(Lu and Liu, 2015)

ND: Not detected

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Table 2 Concentrations of PAHs (ng/g) in agricultural soil of China

Location	Total concentrations of Car-PAHs			Total concentrations of 16-PAHs			Total concentrations of PAHs			References
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	
East China										
Shandong	—	—	—	84.2	1076	289	120	1486	415	(Feng et al., 2013)
Anhui	—	—	—	—	—	—	58.2	437.8	216.8	(Gao et al., 2012)
Jiangsu	—	—	—	—	—	—	312.2	27580.9	4292.4	(Ge et al., 2006)
Shanghai	—	—	—	—	—	—	203.8	6753.9	1172.7	(Sun et al., 2008)
Jiangsu	—	—	—	—	—	—	45.6	2286.8	627.15	(Ding et al., 2007)
Zhejiang	4.9	355.2	75.1	22.6	757.6	179.1	22.6	757.6	179.1	(Hu et al., 2015)
Jiangsu	16.7	2330	747	—	—	—	21.5	3350	1060	(Wang et al., 2015a)
Shandong	—	—	—	—	—	—	27	753	118	(Yuan et al., 2014)
Shanghai	10.6	3040	402	18.8	6320	807	25.8	7380	976	(Wang et al., 2015b)
Shanghai	47.3	1159.5	428.6	92.2	2062.7	665.8	140.7	2370.8	756.8	(Jiang et al., 2011b)
Zhejiang	70.5	1254.1	282.8	—	—	—	262.6	3420.2	1118.2	(Tang et al., 2010)
South China										
Guangdong	ND	3125	124.8	—	—	—	3.3	4079	244.2	(Yang et al., 2007a)
Fujian	13.12	394.5	141.48	—	—	—	70.7	1667.83	480.28	(Sun et al., 2016d)
Guangdong	—	—	—	—	—	—	44.8	3206	582	(Yu et al., 2006)
Guangdong	ND	2400	585	—	—	—	160	3700	1480	(Cai et al., 2007)
Guangdong	—	—	—	—	—	—	22.1	1256.9	318.2	(Hao et al., 2007b)
Guangdong	—	—	—	—	—	—	58	3077	315.4	(Ma et al., 2008)
Guangdong	—	—	—	—	—	—	—	—	514.4	(Huang et al., 2014a)
Guangdong	ND	2773	147.5	—	—	—	ND	4079	316.4	(Li et al., 2007)
Guangxi	—	—	—	—	—	—	—	—	76.5	(Shi et al., 2015)
Hongkong	—	—	—	—	—	—	—	—	31.1	(Zhang et al., 2006)
Guangdong	309	473	—	—	—	—	562	934	—	(Wang et al., 2015d)
North China										
Beijing	—	—	—	—	—	—	1200	3350	1970	(Zhou et al., 2013b)
Beijing	—	—	—	—	—	—	48.4	381.6	140	(Li et al., 2013b)
Tianjin	—	—	—	—	—	—	20.1	4074.7	509.9	(Shi et al., 2012)
Shanxi	—	—	—	—	—	—	78	325	210	(Zhang et al., 2014)
Beijing	—	—	—	—	—	—	37.5	1245.9	219.2	(Peng et al., 2016)
Beijing	—	—	485	—	—	—	16	3884	1347	(Ma et al., 2005)
Beijing/Tianjin	8.6	658.6	128.8	—	—	—	31.6	1475	336.4	(Wang et al., 2010c)
Shanxi	ND	607	56	—	—	—	ND	782	202	(Zhao et al., 2014)
Tianjin	—	—	—	—	—	—	22.86	14722.14	957.825	(Chen et al., 2015)
Central China										
Jiangxi	ND	961	30.1	—	—	—	1.86	3810	195	(Teng et al., 2015)
Northeast										
Liaoning	—	—	—	—	—	—	—	—	223	(Wang et al., 2007b)
Liaoning	3	236	33.7	—	—	—	50	3309	390	(Cao et al., 2013)
Jilin	—	—	—	—	—	—	1572.4	4390.2	2954.9	(Chen et al., 2016)
Southwest										
Chongqing	72.08	1800.81	309.9	—	—	—	277.4	3301	752.6	(Lan et al., 2014)
Northwest										
Shannxi	92	7940	3816	—	—	—	125	9057	2727	(Zhou et al., 2012)
Ningxia	—	—	—	—	—	—	17.2	1199.3	190.6	(Li et al., 2014b)
Shannxi	—	—	—	—	—	—	48.4	440.01	141.34	(Han et al., 2015)
Xinjiang	22	288	102	—	—	—	331	2752	1742	(Chen et al., 2013)
Total China	—	—	—	—	—	—	9.9	5910	377	(Ma et al., 2015)

Car-PAHs: carcinogenic PAHs

16-PAHs: 16 kinds of PAHs which were on a priority control list by the USEPA

ND: Not detected

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Table 3 Concentrations of PCBs (ng/g) in agricultural soil of China

Location	Total concentrations of DL-PCBs			Total concentrations of PCBs			References
	Min	Max	Mean	Min	Max	Mean	
East China							
Zhejiang/Jiangsu/Shanghai	0.41	65.8	9.37	0.41	73.5	15.2	(Zhang et al., 2011a)
Jiangsu	ND	3.791	0.352	0.016	3.791	1.071	(Wang et al., 2010b)
Jiangsu	—	—	—	1.18	29	12.74	(Hu et al., 2009)
Shandong	—	—	—	3.06	14.88	8.04	(Geng et al., 2006)
Zhejiang/Jiangsu/Shanghai	0	78.9	7.5	ND	130	20.2	(Sun et al., 2016b)
Jiangsu	—	—	—	ND	32.83	4.13	(Zhang et al., 2007)
Shandong	—	—	—	ND	87	2.6	(Xie et al., 2012)
South China							
Guangdong	—	—	—	18	130	66	(Wang et al., 2011b)
Guangdong	—	—	—	0.04	0.79	—	(Li et al., 2015b)
Guangdong	—	—	—	ND	32.79	0.42	(Jiang et al., 2011a)
Guangdong	0.15	61.2	9	0.3	202	18.4	(Zhang et al., 2013d)
North China							
Beijing	—	—	—	0.39	13	3.1	(Liu et al., 2006)
Tianjin	0.03	4.36	0.87	0.36	16.88	4.02	(Li et al., 2011)
Central China							
Jiangxi	—	—	—	ND	93	6.75	(Teng et al., 2015)
Northeast China							
Liaoning	0.36	1.36	0.77	0.36	1.36	0.77	(Lang et al., 2014)
Southwest China							
Guizhou	—	—	—	8.9	55.9	29.8	(Wei et al., 2007)
Tibet	—	—	—	0.0019	0.0132	0.0051	(Wang et al., 2016b)
Tibet	—	—	—	0.0471	0.4226	0.1856	(Wang et al., 2009a)
Tibet	—	—	—	0.0086	0.0391	0.0162	(Tian et al., 2014)
Sichuan	—	—	—	0.22	2.31	1.005	(Gai et al., 2014)
Northwest China							
Shannxi	—	—	—	—	—	0.67	(Lu and Liu, 2015)
Total China	—	—	—	0.138	1.14	0.424	(Ren et al., 2007)

DL-PCBs: dioxin-like PCBs

ND: Not detected

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Table 4 Concentrations of PAEs (ng/g) in agricultural soil of China

Location	Total concentrations of six PAEs			Total concentrations of PAEs			References
	Min	Max	Mean	Min	Max	Mean	
East China							
Shandong	716	16007	2219.5	794	19504	2385	(Yang et al., 2013b)
Zhejiang/Shanghai/Jiangsu	68	9330	—	167	9370	782	(Sun et al., 2016c)
Shandong/Zhejiang/Shanghai/Anhui	—	—	—	1340	7140	2282	(Hu et al., 2003)
South China							
Guangdong	3000	45670	21030	3000	45670	21030	(Cai et al., 2005)
Guangdong	ND	25900	670	ND	25990	670	(Yang et al., 2007b)
Guangdong	—	—	1984	—	—	1984	(Li et al., 2015d)
Guangdong	—	—	—	195	33600	2570.4	(Zeng et al., 2008)
Guangxi/Fujian/Guangdong	—	—	—	2930	6970	4353	(Hu et al., 2003)
North China							
Hebei	191	457	294	191	457	294	(Zhang et al., 2015)
Tianjin	50	10400	754	50	10400	754	(Kong et al., 2012)
Neimenggu/Beijing/Tianjin/Shanxi	—	—	—	17600	3780	2800	(Hu et al., 2003)
Beijing	ND	1360	310	20	2900	630	(Cheng et al., 2015)
Central China							
Hubei	5630	157620	19330	5630	157620	19330	(Wu et al., 2015)
Jiangxi	ND	22800	530	ND	22800	530	(Teng et al., 2015)
Henan/Hubei/Hunan	—	—	—	890	2250	1430	(Hu et al., 2003)
Northeast China							
Heilongjiang	—	—	—	—	—	109.2	(Wang et al., 2013b)
Heilongjiang/Liaoning	—	—	—	4410	10030	6713	(Hu et al., 2003)
Southwest China							
Sichuan/Yunnan/Guizhou	—	—	—	1850	2960	2220	(Hu et al., 2003)
Northwest China							
Gansu	—	—	—	—	—	2810	(Hu et al., 2003)
Total China	—	—	—	75	6369	1088	(Niu et al., 2014)

Six PAEs: PAEs which were classified as environmental priority pollutants by USEPA

ND: Not detected