Pollutants in Hong Kong Soils: Organochlorine Pesticides and Polychlorinated Biphenyls

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Pollutant in Hong Kong soils series:

1.

Chung, M. K., Hu, R., Cheung, K. C. & Wong, M. H. Pollutants in Hong Kong soils: Polycyclic aromatic hydrocarbons. Chemosphere 67, 464–473 (2007).

http://doi.org/10.1016/j.chemosphere.2006.09.062

2.

Chung, M. K., Cheung, K. C. & Wong, M. H. Pollutants in Hong Kong Soils: As, Cd, Cr, Cu, Hg, Pb and Zn

https://doi.org/10.1101/2020.02.16.951558

3.

Chung, M. K., Hu, R., Cheung, K. C. & Wong, M. H. Pollutants in Hong Kong Soils: Organochlorine Pesticides and Polychlorinated Biphenyls

https://doi.org/10.1101/2020.02.16.951541

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47   Abstract

48   Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were

49   investigated in 138 soil samples collected in Hong Kong. Results showed that within

50   the selected OCPs, only DDT and its metabolites (DDD and DDE) were frequently

51   detected. Of 138 soil samples, 18% were non-detected for any DDT residues, while

52   25% were contaminated sporadically with DDT only (non-detected to 1090 µg kg^{-1})

53   and 57% contained various combinations of DDT, DDD and DDE (2.03 to 1118 µg kg^{-1}

54   ). In contrast, total PCBs ($\sum$66 congeners) contamination was distributed more evenly

55   (0.22 to 154 µg kg^{-1}) than DDTs, but serious contamination was found in industrial

56   areas and soils collected near highways. Concentrations of 7 indicator PCBs ranged

57   between non-detected to 34.5 µg kg^{-1}. The ratios of DDT/metabolites were typically

58   greater than 1, thus suggesting recent application of DDT. Medium-range deposition

59   from industrial areas within as well as away from the city is also suspected to be the

60   origins of DDTs and PCBs found in Hong Kong soils. The concentrations of DDTs

61   and PCBs in all soil samples did not exceed the recommended values in soil quality

62   guidelines except 3 samples for DDT from locations far away from densely populated

63   districts such as Tuen Mun and Tin Shui Wai. Therefore, DDTs and PCBs were not of

64   significant concern in terms of their impacts on public health and environment.

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69   *Keywords: Persistent Toxic Substances (PTS), South China, Persistent Organic*

70   *Pollutants (POPs), Urban Soils.*

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1. Introduction

Polychlorinated biphenyls (PCBs) and a number of organochlroine pesticides (OCPs) such as dichlorodiphenyltrichloroethane (*p,p'*-DDT) and hexachlorocyclohexanes (HCHs) are candidates of designated 12 Stockholm Convention persistent organic pollutants (POPs). These anthropogenic chemicals, unlike other nature contaminants that can be degraded by physiochemical or biological means, are much more persistent in the environment. Their potential carcinogenicities and the possibility of resulting more toxic metabolites have spotlight attention of the general public in recent years (Connell *et al.*, 2003; Megharaj *et al.*, 1999).

PCBs were first commercially produced in 1929 (Mai *et al.*, 2005) and cumulative production has been estimated as much as 1.2 million tonnes around the world (Harrad *et al.*, 1994). The primary uses of PCBs included congener-mixture in dielectric fluids, flame retardants, and industrial lubricant fluids in transformers and capacitors (Schuhmacher *et al.*, 2004). They are organochlorine chemicals that have 209 congeners. Among these congeners, 12 are especially dangerous for their dioxin-like (DL) activities (van den Berg *et al.*, 1998). Their residues have been found in the environment and have been either banned or restricted on production or usage in many countries since 1970s (Harrad *et al.*, 1994).

DDT was first synthesized in 1874 and the discovery of DDT's insecticidal activity by Paul Müller in 1939 subsequently led to his award of the Nobel Prize (Carson *et al.*, 1962). After World War II, it was applied on agricultural crops world wide and stimulated the synthesis and development of other organochlorine pesticides. However, later evidences showed its detrimental effects on non-target organisms (Carson *et al.*, 1962; Christen, 1999), leading to the restriction on its production and usage in many countries. Nevertheless, several tropical and subtropical countries are being exempted for using DDT for public health purpose to control the spread of malaria (World Wildlife Fund, 2004).

In Asia, Japan and Korea have banned the use of DDT in the 1970s (Phillips and Tanabe, 1989), whereas DDT production was prohibited in China in 1983 (Wolfe *et al.*, 1984). PCBs have been banned or regulated in China in early 1980s (Mai *et al.*, 2005). Nevertheless being officially regulated for more than 20 years, PCBs, DDTs

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111 and other OCPs such as HCH were still being detected in various environmental
112 compartments throughout China (Ding *et al.*, 2005; Fung *et al.*, 2004; Mai *et al.*, 2005;
113 Wu *et al.*, 1999; Yuan *et al.*, 2001), but overall the PCB levels were relatively low and
114 serious contaminations were highly restricted in area such as storage locations of PCBs
115 (Xing *et al.*, 2005). In Hong Kong, the production and use of DDT have been banned
116 since 1988, however, there is no legislation to ban the production of PCBs as no record
117 of such industrial activity by the Hong Kong government. Instead, historical
118 equipments with PCBs such as transformers and capacitors are being phased out
119 voluntarily by their owners (Hong Kong Environmental Protection Department, 2002).
120

121 In the Pear River Delta (PRD) where Hong Kong is located, Lingding Yang was
122 reported to be one of the hotspot sites polluted with OCPs in the Pearl River Estuarine
123 (Fang, 2004), which might transport pollutants to coastal areas of Hong Kong.
124 Contamination of PCBs and DDTs in mussels farmed in Hong Kong was reported as
125 early as 1990s (Phillips, 1989). DDT levels in human milk collected from both
126 Guangzhou and Hong Kong were generally higher when compared with 19 countries
127 (Wong *et al.*, 2005). Recent studies indicated that several freshwater fishes cultivated
128 in fish ponds around the PRD and purchased from local markets all contained DDTs
129 (Cheung *et al.*, 2006), and in particular the Mandarin fish (*Siniperca kneri*) are of actual
130 concern, which could contain DDTs up to 4.3 times higher (62 µg kg$_{-1}$ wet wt.) than
131 the recommended value set by USEPA (14.4 µg kg$_{-1}$ DDTs, wet wt.) (Kong *et al.*,
132 2005).
133

134 The major objectives of this paper are to study the contemporary levels of edaphic
135 OCPs and PCBs and assess their potential risks to general public in Hong Kong. It is
136 hoped that the data would serve as a valuable reference for redeveloping of some areas
137 which were affected by various industries in the past, as well as fulfilling part of the
138 obligation imposed by the Stockholm Convention on POPs and the duty of monitoring
139 POPs in the environment. In addition to these, potential sources of OCPs and PCBs
140 were also discussed.
141

142 **2. Materials and Methods**
143 *2.1 Sampling, Preparation and Analysis*

5

144 Ten land use categories had been designated to reveal the pollution impacts from
145 various human activities. These included urban park, greening area, country park, rural
146 area, restored landfill, agricultural farmland, orchard farm, crematorium, industrial area
147 and nearby highway. There were totally 138 composite soil samples collected in early
148 2003 at various locations in Hong Kong. Surface soils (0-5 cm depth) were taken by
149 using a stainless steel soil core with the uppest organic vegetative materials were
150 removed in advance. Samples were air-dried and sieved through a 2-mm mesh.

151

152 Extraction of OCPs and PCBs from soil samples were performed according to the US
153 EPA Standard Method 3540C (U.S. Environmental Protection Agency, 1996a).
154 Briefly, 10 g of soil sample was transferred into a soxhlet apparatus and extracted by
155 80 ml acetone (pesticide grade, Tedia) and dichloromethane (DCM) (pesticide grade,
156 Tedia) mixture (1:1, v:v) for 18 h. Florisil columns were used to minimize interferences
157 to target compounds.

158

159 Fifteen OCPs were analyzed: α-HCH, β-HCH, δ-HCH, heptachlor, aldrin, heptachlor
160 expoxide, endosulfan I, endosulfan II, dieldrin, p,p'-DDE, endrin, p,p'-DDD, endrin
161 aldehyde, endosulfan sulphate and p,p'-DDT. For PCBs, 66 of the congeners were
162 quantified. Using the IUPAC nomenclature, they are 1, 2, 3 (total mono-PCBs), 4, 6,
163 8, 9, 15 (total di-PCBs), 16, 18, 19, 20, 22, 25, 27, 28, 29, 34 (total tri-PCBs), 40, 42,
164 44, 47, 52, 56, 66, 67, 69, 71, 74 (total tetra-PCBs), 82, 87, 92, 93, 99, 101, 105, 110,
165 118, 119 (total penta-PCBs), 128, 134, 136, 138, 144, 146, 147, 151, 153, 157, 158
166 (total hexa-PCBs), 173, 174, 177, 179, 180, 187, 190, 191 (total hepta-PCBs), 194, 195,
167 199, 203 (total octa-PCBs), 206, 207, 208 (total nona-PCBs) and 209 (total deca-PCBs).
168 ∑PCBs is defined as the sum of the concentration of 66 congeners. Standards for OCPs
169 and PCBs congeners mixtures were purchased from ChemService Inc. and AccuStand
170 Inc. respectively.

171

172 The analytical methods were based on using GC-MS (Gas Chromatrography-Mass
173 Spectrometry) instrument (U.S. Environmental Protection Agency, 1996b). In short,
174 OCPs and PCBs were quantitatively analyzed by a Hewlett Packard (HP) 6890 GC
175 system equipped with an HP 5973 mass selective detector (MS) and a 30m × 0.25mm
176 × 0.25µm DB-5 capillary column (J & W Scientific) with helium as the carrier.

177

178   Limit of detection (LOD) for OCPs was: 2 µg kg-1 for *p,p'*-DDD and *p,p'*-DDE, 20 µg

179   kg-1 for DDT and 10 µg kg-1 for the rest of OCPs. LOD for PCBs varied, and a universal

180   safe value for mono to penta PCBs was set to 0.5 µg kg-1 while 1 µg kg-1 for the rest of

181   the PCBs.

182

183   *2.2 Quality Control*

184   Laboratory analytical blank and Certified Reference Material (CRM) CRM105-100

185   (Resource Technology) and HS-2 (Institute for Marine Biosciences) were included in

186   every 2 batches (14 samples) of soxhlet extraction to assess the recoveries and

187   performance for measurement of OCPs and PCBs respectively. None of the analytical

188   blanks were found to have detectable contamination of the monitoring OCPs and PCBs.

189   Average individual OCPs recoveries were: $88 \pm 3\%$ (*p,p'*-DDT), $92 \pm 6\%$ (*p,p'*-DDD),

190    $86 \pm 2\%$ (*p,p'*-DDE), $67 \pm 4\%$ (dieldrin), $64 \pm 7\%$ (endosulfan I), $72 \pm 6\%$ (endosulfan

191   II), $59 \pm 9\%$ (endrin). While average individual PCBs recoveries were: $77 \pm 3\%$

192   (IUPAC 101)**,** $89 \pm 5\%$ (IUPAC 138)**,** $87 \pm 5\%$ (IUPAC 151)**,** $103 \pm 9\%$ (IUPAC 153)**,**

193    $76 \pm 12\%$ (IUPAC 180)**,** $82 \pm 3\%$ (IUPAC 194)**,** $86 \pm 10\%$ (IUPAC 199)**,** $78 \pm 9\%$

194   (IUPAC 209). Mean recovery for OCPs and PCBs were > 75% and >84% respectively

195   and all samples were not corrected.

196

197   *2.3 Data Analyses*

198   Statistical analyses were calculated using Statistica (version 6.0, StaSoft). Kriging

199   maps were developed using SADA (version 4.0, University of Tennessee) and

200   graphical plots of data were produced by either Statistica or Sigmaplot (version 8.0,

201   Systat). Not detected values were substituted with half of LOD only for descriptive

202   statistics.

203

204   **3. Results and Discussion**

205   *3.1 Spatial values*

206   In general, most OCPs were not detected except DDTs, and their mean concentrations

207   are listed in Table 1. Two locations were found to have endosulfan sulfate at

208   concentration about 12 µg kg-1 while heptachlor epoxide was found in 1 soil sample.

209   This finding is in HK is in line with the observation that DDTs were the mostly

210   predomonant OCPs in tropical Asia (Kannan *et al.*, 1995). Uneven distribution of

211   DDTs was observed, with 18% of soil samples showing no detectable DDTs. Areas

212 near highways had higher concentrations of DDT when compared to other land use

213 categories (median 77.8 µg kg-1). DDT concentrations decreased according to the

214 following pattern: areas near highways > urban park > crematorium > orchard farm >

215 rural area > agricultural farmland > greening area > industrial area > country parks >

216 restored landfills. The variation in concentrations was greatest for DDT, ranged from

217 not detected to 1000 µg kg-1, whereas not detected to around 200 µg kg-1 for DDD and

218 DDE.

219

220 To assess the spatial distribution of individual DDTs in Hong Kong, kriging maps

221 (Figure 1) were constructed by interpolating the data from various locations (ordinary

222 kriging). A single hotspot for DDT, DDD and DDE contamination was observed in the

223 southern tip of Tsing Yi island. Another hotspot for DDT compounds was found in the

224 roadside located in Lung Kwu Tan, where no DDD and DDE were detected. The half

225 life of DDTs ranges from a few months to 30 years, or even up to centuries (Aigner *et*

226 *al.*, 1998; Dimond and Owen, 1996). The traffic in the aforementioned locations was

227 mainly contributed by trucks powered by diesel engine, which may release high levels

228 of toxic pollutants such as PAHs and lead, creating an adverse condition which is

229 unfavorable to biodegradation, and thus slow down the biodegradation of DDTs in the

230 vicinity.

231

232 In Hong Kong, relatively high levels of DDTs (2.87 mg kg-1 fat) and β-HCH (0.95 mg

233 kg-1 fat) were found in human milk (Wong *et al.*, 2002), whereas the mean

234 concentration of DDTs in whale and dolphin found in local water was 46 mg kg-1 wet

235 wt. (Minh *et al.*, 1999). The present results indicated that the mean edaphic DDT

236 concentrations was 72 µg kg-1, while levels of DDTs in river sediments and tilapia of

237 the inland water systems in Hong Kong were 2.8 to 8.6 and 28 to 40 µg kg-1 dry wt.

238 respectively (Zhou *et al.*, 1999), and average DDT levels in surface sediment of Mai

239 Po marsh was 8.15 µg kg-1 dry wt (Zheng *et al.*, 2000). It can be seen that there are up

240 to 1000 folds of difference in DDT concentrations between the backgrounds and living

241 organisms, and this implied that DDTs are being accumulated into organisms and

242 finally reaching human beings through various pathways including ingestion of

243 contaminated food and soil particles, and inhalation of dust. Being a large agricultural

244 country, China has produced and used a large quantity of DDTs not until early 1980s

245 when DDT was banned for agriculture application (Wong *et al.*, 2005). The level of

8

246   DDTs in China is highly varied, from maximum values below 100 µg kg-1 in crop soils

247   of PRD (5 to 80 µg kg-1) (Fu *et al.*, 2003), to below 1000 µg kg-1 in Tianjin (0.35 to

248   963 µg kg-1) (Gong *et al.*, 2004) and above 1000 µg kg-1 in outskirts of Beijing (0.77 to

249   2178 µg kg-1) (Zhu *et al.*, 2005). Large difference between the minimum and maximum

250   values is also observed in the current study (Table 1).

251

252   The concentrations of total PCBs, dioxin like (DL) and non-dioxin like (NDL) PCBs in

253   soil samples are shown in Table 1. Concentrations of total PCBs could be grouped into

254   3 categories: The most contaminated which included industrial and areas near highway

255   (median concentration around 8 µg kg-1); followed by urban park and greening area (5

256   µg kg-1) and the rest of the land uses which contained the lowest concentration (3 to 4

257   µg kg-1). The PCB toxicity is expressed as toxic equivalent quantity (TEQ) and is

258   calculated according to toxic equivalency factor (TEF) stated by World Health

259   Organization (WHO). It represents the total toxicity of a mixture of related substances

260   that equal to their combined toxic effects. Median PCB WHO-TEQs (calculated from

261   3 DL PCBs) in all the land use categories were below 0.06 ng kg-1. Zhao *et al.* (2006)

262   reported that the PCB WHO-TEQs (9 DL PCBs) of soils collected from a polluted and

263   abandoned farmland in southern China was 57 ng kg-1, whereas the PCB WHO-TEQs

264   equaled to 5.13 ng kg-1, which is much higher than the corresponding data of the present

265   study. Among the 3 DL-PCBs, PCB-157 was contributing greater than 40% in all land

266   use categories, followed by PCB-118 and PCB-105. For the sum of 6 and 7 NDL-

267   PCBs, PCB-52 was the most dominant congener (usually greater than 35% of total

268   NDL-PCBs), while PCB-101 was the lowest among the indicator PCBs. It was reported

269   that $\sum 13$ PCBs ranged from 0.051 to 22 µg kg-1 in soils of United Kingdom and

270   Norway (Meijer *et al.*, 2002), while concentration of $\sum 7$ PCBs ranged from 0.09 to 150

271   µg kg-1 (Seine River basin) in France (Motelay-Massei *et al.*, 2004). In addition, $\sum 6$

272   PCBs in Moscow soils were found between 2 to 34 µg kg-1 (Wilcke *et al.*, 2006). PCBs

273   in soils from Shenyang, China ranged from 6.4 to 15.2 µg kg-1, but the polluted areas

274   in southeast coast of China where PCBs related activities such as dismantling of PCB-

275   containing transformers reached 788 µg kg-1 (Xing *et al.*, 2005). The PCB

276   concentrations in Hong Kong soils were within the lower range of the levels reported,

277   because of a lack of PCB related industry.

278

279   *3.2 Sources*

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280   Correlation matrixes for DDTs in different land use categories are presented in Table

281   2. In most cases DDD were significantly correlated to DDE (p<0.05). Since DDT

282   could be degraded to DDE and then DDD in the environment, or directly to DDD

283   (Agency for Toxic Substances and Disease Registry, 2002), the positive correlation of

284   DDD and DDE suggested that DDE to DDD was not an important pathway for DDT

285   degradation. The results also showed that DDT had a significant correlation with its

286   metabolites in agricultural farmlands, industrial areas and restored landfills, thus a slow

287   but continuous input of DDT to these areas is suspected, as increase in metabolites

288   should be accompany with a decrease in parent compound.

289

290   Various DDT ratios could be used to assess any recent inputs of DDT in our

291   environment (Kong *et al.*, 2005). These include DDD/DDT, DDE/DDT and

292   DDT/(DDE+DDD) ratios. Table 3 presents the various calculated ratios for DDTs in

293   each soil land use category. Metabolite(s) (DDD, DDE, DDE+DDD) to parent

294   compound (DDT) ratios were less than 0.15 on average. In contrast,

295   DDT/(DDE+DDD) ratios were much higher, particular in the vicinity of highways.

296   Two hot spots (in Tsing Yi island, 796 µg kg$_{-1}$ and Lung Kwu Tan, 1090 µg kg$_{-1}$)

297   identified in the kriged map (Figure 1) were both located adjacent to industrial

298   activities, e.g. power station/oil depot/chemical plants/machinery factories in Tsing Yi

299   island, and steel mill/power station/cement plant in Lung Kwu Tan. Out of the 138 soil

300   samples collected, 18% of them did not show detectable level of neither DDT nor its

301   metabolites. In addition, DDT without metabolites was detected in 25% of the 138

302   samples. These results suggested 2 possibilities: 1) there was still a small-scale,

303   localized non-mobile DDT input or 2) the highly heterogeneous soil micro-

304   environment could either promote degradation of DDT or it could inhibit the

305   degradation of organic compounds, depending on the sampling locations. Photo-

306   oxidation and volatilization of DDT are possible mechanisms for the loss of DDT

307   (Hussain *et al.*, 1994), but the principal pathway for the loss of DDT in soils is mainly

308   through microbial actions (Mohn and Tiedje, 1992).

309

310   The current official use of DDT is for the control of disease vectors in indoor house

311   spraying as specified by the World Health Organization (WHO) (World Wildlife Fund,

312   2005). Apart from DDT, *p,p'*-DDD was also used as an insecticide but it is not as

313   common as DDT (Agency for Toxic Substances and Disease Registry, 2002).

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314

315   In Hong Kong, DDT is not a registered pesticide and has been banned for use since the

316   beginning of 1988. Currently, it can be traded only under permit in Hong Kong (UNEP

317   Chemicals, 2002). Between 1979 to 1982, about 5,023 to 5,996 kg of DDT pesticide

318   was imported annually (Morton, 1990) and the existence of residual DDT and its

319   metabolites in soils are therefore expected.

320

321   OCPs are routinely found in the atmosphere (Cortes *et al.*, 1998). A recent study

322   observed that OCPs like α-HCH, hexachlorobenzene, DDT, DDE, heptachlor, and

323   endosulfan I were detected in local atmospheric compartment at relatively low

324   concentrations 0.02–0.23 ng m$_{-3}$ (Louie and Sin, 2003). It was proved that the northeast

325   monsoon wind can bring air pollutants from China to Hong Kong (Lee and Hills, 2003),

326   and it is reasonable to deduce that pollutants such as DDTs bound in fine particles could

327   also be a potential source in Hong Kong soils via atmospheric deposition of dust

328   particles from the PRD.

329

330   Dicofol is an organochlorine acaricide (a chemical that kills mites) which is highly toxic

331   to aquatic life and can cause egg-shell thinning in some bird species. As Dicofol is

332   manufactured from technical DDT in China, it also contributes to the sources of fresh

333   DDT in the surrounding (Qiu *et al.*, 2005). Though DDT is also released to the

334   environment by pyrogenic degradation of animal fat in food during cooking in PRD,

335   but its contribution to the edaphic environment is expected to be comparatively low

336   (Cheng *et al.*, 2000). In addition, some OCPs (such as DDT and HCH) may still be

337   illegally used for agricultural purpose in the PRD (Wong and Poon, 2003).

338

339   PCB homologues profile for corresponding land use categories is plotted in Figure 2.

340   PCB patterns were not characterized in terms of lower or higher chlorinated biphenyl,

341   but by individual homologues. Di-PCBs was the most dominant homologue, together

342   with tetra- and octa-PCBs, they accounted for more than 70% of all the investigated

343   PCBs. Aroclor is a commercial mixture of PCBs that has been used intensively.

344   Homologue composition of Arochlor such as Arochlor 1016, 1242, 1254 and 1260

345   showed domination of tri- to hexa-PCBs (U.S. Environmental Protection Agency,

346   2005). However, the present results revealed that the major homologue was di-PCBs

347   and thus rule out the possibility of recent Arochlor contamination. Interestingly, the

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348   contribution of octa-PCB accounted for more than 40% of total PCBs found in

349   industrial areas. The greater distribution of heavier homologues is possibly due to the

350   preferential atmospheric deposition in the vicinity of the source (Meijer *et al.*, 2002).

351   It has been reported that combustion process such as municipal solid waste incineration,

352   automobile exhaust are potential sources of PCBs (Granier and Chevreuil, 1991; Mai

353   *et al.*, 2005) and the industrial activities, with mobile trucks could be the major

354   contributor to PCBs in industrial areas.

355

356   The general domination of di-PCB contaminations could be originated from the short-

357   range atmospheric transport from industrial sites (Motelay-Massei *et al.*, 2004), and

358   medium-range regional atmospheric deposition is also suspected (Wilcke *et al.*, 2006).

359   Volatization and re-deposition during dredging of contaminated sediments is also a

360   possible source of PCBs in soils (Vorhees *et al.*, 1999). However, a study indicated

361   that total PCBs ($\sum$112 congeners) in the sediments of PRD was in the range of 26 to 32

362   μg kg-1, with tri- to hexa-PCBs accounted for more than 80% of total PCBs (Mai *et al.*,

363   2005), thus this volatization and re-deposition pathway from sediment is unlikely to

364   play a crucial role in PCB contaminations in soils.

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367   *3.3 Cleanup guidelines*

368   The guideline values for DDTs and PCBs from various countries are listed in Table 4.

369   In general, concentrations of DDT obtained in the present study were below the soil

370   quality limits imposed by Netherlands (Ministry of Housing Spatial Planning and

371   Environment, 2000), Denmark (Danish Environmental Protection Agency, 2002),

372   Canada (Environment Canada, 2003) and China (State Environmental Protection

373   Administration of China, 1995) except samples from southern Tsing Yi island (796 μg

374   kg-1) and Lung Kwu Tan (1090 μg kg-1). For total DDTs (DDT + DDE + DDD), only

375   one sample from Lung Kwu Tan (1090 μg kg-1) was slightly above of the guideline

376   values from Denmark (1000 μg kg-1) while the rest of 137 samples were below the

377   safety values from Netherlands, Denmark and China. In contrast to the sporadic

378   occurrence of hazardous samples concerning DDTs, none of the samples was found to

379   contain PCBs at alarming concentration when benchmarking with the soil quality

380   guidelines from the aforementioned countries. In a word, the potential risks to human

381   and ecosystem imposed by the investigated OC chemicals are minimum.

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382

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4. Conclusions

385   In the present study of DDT and PCB concentration in Hong Kong soils, sporadic

386   contamination of DDTs and it's metabolites were found at a range of 0.22 to 154 µg

387   kg-1 and the concentration was found to be highest in industrial areas. There were

388   widespread but low levels of PCB contamination (0.22 to 154 µg kg-1), while the

389   highest level was observed in industrial areas. PCB-157 was the most active DL-PCBs

390   among 3 (PCB -105, 118, and 157) that have been investigated and contributed more

391   than 40% of WHO-TEQ in all land use categories, while PCB-152 was the most

392   dominant congener among the 6 or 7 indicator PCBs. The current levels of DDT and

393   PCB in Hong Kong were within the guideline values of the soil quality guidelines

394   adopted in countries such as Netherlands and Denmark. However, 3 out of 138 samples

395   exceeded DDT(s) concentrations but their potential hazards were minimal as the sites

396   were located in remote areas. It is advised that a thorough site investigation on DDTs

397   should be made prior to any residential development in these locations.

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411   **List of Figures:**

412   Figure 1. Kriged maps DDT (DDT, DDD, DDE and total DDTs) and PCB (3-PCBs

413   WHO-TEQ congeners, 6 indicator PCBs, 7 indicator PCBs and sum of 66 PCB

414   congeners) levels (µg/kg) in surface soils of Hong Kong. The 3-PCBs WHO-TEQ is

415   in ng kg$_{-1}$ scale.

416

417   Figure 2. Homologue profiles of PCBs in corresponding land uses in Hong Kong.

418   Total PCBs is sum of 66 selected congeners.

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Table 1. Mean concentrations of DDTs and PCBs of soils in Hong Kong. Units are in µg kg-1 except for the WHO-TEQ PCBs.

Classified soil categories	Sample no.	DDT M*	DDT Medi*	DDT Range	DDD M*	DDD Medi*	DDD Range	DDE M*	DDE Medi*	DDE Range	Total DDTs M*	Total DDTs Medi*	Total DDTs Range
							Concentration in soils (µg/kg)						
Urban park	39	39.7	34.2	[N.D. - 160]	4.4	4.4	[N.D. - 27.9]	2.5	2.3	[N.D. - 8.3]	2.5	2.3	[N.D. - 165]
Greening area	14	24.4	14.3	[N.D. - 162]	3.8	4.5	[N.D. - 6.4]	1.6	2.1	[N.D. - 4.1]	1.6	2.1	[N.D. - 169]
Country park	9	18.1	2.0	[N.D. - 51]	0.8	0.2	[N.D. - 3.5]	0.2	0.2	[N.D. - 1.7]	0.2	0.2	[N.D. – 51]
Rural area	19	35.9	19.4	[N.D. - 252]	1.3	0.2	[N.D. - 4.4]	0.7	0.2	[N.D. - 2.4]	0.7	0.2	[N.D. - 253]
Restored landfill	11	4.0	2.0	[N.D. - 31.5]	2.5	0.2	[N.D. - 7.7]	1.3	0.2	[N.D. - 3.8]	1.3	0.2	[N.D. – 43]
Agricultural farmland	9	42.0	19.0	[7.8 - 125]	3.5	3.6	[N.D. - 7.5]	3.8	2.1	[0.75 - 15.7]	3.8	2.1	[8.71 - 149]
Orchard farm	5	35.0	19.8	[16.3 - 67.2]	6.3	3.9	[N.D. - 19.4]	21.9	1.9	[N.D. - 103]	21.9	1.9	[20.2 - 190]
Crematorium	10	22.0	21.1	[N.D. - 54.1]	1.9	2.4	[N.D. - 3.6]	1.0	1.2	[N.D. - 2.1]	1.0	1.2	[4.03 – 55]
Industrial area	18	257.0	12.7	[N.D. - 1090]	25.5	2.6	[N.D. - 215]	12.8	0.2	[N.D. - 108]	12.8	0.2	[N.D. - 1118]
Nearby Highway	4	103.0	77.8	[22.6 - 232]	1.4	0.2	[N.D. - 5.4]	0.8	0.3	[N.D. - 2.6]	0.8	0.3	[23 - 241]

Classified soil categories	Sample no.	PCB 105 (DL) M*	PCB 105 (DL) Medi*	PCB 105 (DL) Range	PCB 118 (DL) M*	PCB 118 (DL) Medi*	PCB 118 (DL) Range	PCB-157 (DL) M*	PCB-157 (DL) Medi*	PCB-157 (DL) Range	Total 3 DL PCBs M*	Total 3 DL PCBs Medi*	Total 3 DL PCBs Range	ng PCB WHO-TEQ kg-1 M*	Medi*	Range
							Concentration in soils (µg/kg)							Concentration (ng/kg)		
Urban park	39	0.11	N.D.	[N.D. - 0.68]	0.18	0.1	[N.D. - 1.05]	0.05	N.D.	[N.D. - 0.4]	0.34	0.1	[N.D. - 2]	0.05	0.02	[N.D. - 0.36]
Greening area	14	0.15	0.1	[N.D. - 0.8]	0.2	0.1	[N.D. - 1.24]	0.04	N.D.	[N.D. - 0.16]	0.39	0.2	[N.D. - 2.12]	0.05	0.03	[N.D. - 0.24]
Country park	9	N.D.	N.D.	[N.D. - N.D.]	N.D.	N.D.	[N.D. - N.D.]	N.D	N.D.	[N.D. - N.D.]	N.D.	N.D.	[N.D. - N.D.]	N.D.	N.D.	[N.D. - N.D.]
Rural area	19	0.04	N.D.	[N.D. - 0.12]	0.07	0.1	[N.D. - 0.3]	0.02	N.D.	[N.D. - 0.11]	0.13	0.1	[N.D. - 0.48]	0.02	0.02	[N.D. - 0.09]
Restored landfill	11	0.04	N.D.	[N.D. - 0.12]	0.09	0.1	[N.D. - 0.24]	0.01	N.D.	[N.D. - 0.06]	0.14	0.1	[N.D. - 0.42]	0.02	0.01	[N.D. - 0.07]
Agricultural farmland	9	0.05	N.D.	[N.D. - 0.2]	0.06	N.D.	[N.D. - 0.25]	0.02	N.D.	[N.D. - 0.06]	0.13	N.D.	[N.D. - 0.47]	0.02	N.D.	[N.D. - 0.07]
Orchard farm	5	0.05	N.D.	[N.D. - 0.12]	0.04	N.D.	[N.D. - 0.12]	N.D	N.D.	[N.D. - N.D.]	0.1	0.1	[N.D. - 0.25]	0.01	0.01	[N.D. - 0.03]
Crematorium	10	0.03	N.D.	[N.D. - 0.11]	0.05	0.0	[N.D. - 0.11]	N.D	N.D.	[N.D. - N.D.]	0.09	0.1	[N.D. - 0.22]	0.01	0.01	[N.D. - 0.03]
Industrial area	18	0.04	N.D.	[N.D. - 0.22]	0.07	N.D.	[N.D. - 0.43]	0.01	N.D.	[N.D. - 0.1]	0.13	N.D.	[N.D. - 0.66]	0.02	N.D.	[N.D. - 0.08]
Nearby Highway	4	0.16	0.1	[N.D. - 0.44]	0.51	0.3	[N.D. - 1.53]	0.16	0.1	[N.D. - 0.51]	0.83	0.4	[N.D. - 2.48]	0.15	0.06	[N.D. - 0.45]

Values in square brackets represent range of concentration. *M=Mean, Medi=Median. †: Not included for total 6 NDL PCBs. ‡:Calculated from sum of PCB 105, 118 and 157.

Table 1 (con't)

Classified soil categories	Sample no.	PCB-28 (NDL)			PCB-52 (NDL)			PCB-101 (NDL)			†PCB-118 (NDL)			PCB-138 (NDL)		
		M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range
							Concentration in soils (µg/kg)									
Urban park	39	0.33	0.2	[0.09 - 5.19]	0.5	0.5	[N.D. - 0.98]	0.16	0.1	[N.D. - 2.45]	0.18	0.1	[N.D. - 1.05]	1.61	0.3	[N.D. - 13.89]
Greening area	14	0.36	0.2	[N.D. - 2.49]	0.66	0.4	[0.09 - 2.8]	0.14	0.1	[N.D. - 0.91]	0.2	0.1	[N.D. - 1.24]	0.39	0.3	[N.D. - 1.52]
Country park	9	0.13	0.1	[0.07 - 0.39]	0.41	0.4	[0.24 - 0.76]	N.D.	N.D.	[N.D. - N.D.]	N.D.	N.D.	[N.D. - N.D.]	0.04	N.D.	[N.D. - 0.12]
Rural area	19	0.23	0.2	[N.D. - 1.42]	0.48	0.5	[N.D. - 1.2]	0.06	0.1	[N.D. - 0.24]	0.07	0.1	[N.D. - 0.3]	0.32	0.3	[N.D. - 1.03]
Restored landfill	11	0.19	0.2	[0.1 - 0.3]	0.57	0.6	[0.26 - 0.89]	0.07	N.D.	[N.D. - 0.29]	0.09	0.1	[N.D. - 0.24]	0.29	0.1	[N.D. - 1.11]
Agricultural farmland	9	0.14	0.1	[0.07 - 0.32]	0.44	0.4	[0.28 - 0.69]	0.04	N.D.	[N.D. - 0.15]	0.06	N.D.	[N.D. - 0.25]	0.18	0.1	[N.D. - 0.62]
Orchard farm	5	0.13	0.2	[0.07 - 0.17]	0.37	0.4	[0.32 - 0.41]	0.04	N.D.	[N.D. - 0.08]	0.04	N.D.	[N.D. - 0.12]	0.15	0.1	[N.D. - 0.34]
Crematorium	10	0.16	0.1	[0.06 - 0.51]	0.49	0.4	[0.31 - 0.73]	0.05	N.D.	[N.D. - 0.19]	0.05	0.0	[N.D. - 0.11]	0.28	0.2	[0.13 - 0.88]
Industrial area	18	0.23	0.1	[N.D. - 1.86]	0.38	0.4	[N.D. - 1.03]	0.05	N.D.	[N.D. - 0.36]	0.07	N.D.	[N.D. - 0.43]	0.16	0.1	[N.D. - 0.64]
Nearby Highway	4	0.5	0.2	[0.09 - 1.58]	2.34	0.7	[0.42 - 7.47]	0.36	0.2	[N.D. - 1.02]	0.51	0.3	[N.D. - 1.53]	1.03	1.0	[0.11 - 1.91]

Classified soil categories	Sample no.	PCB-153 (NDL)			PCB-180 (NDL)			Total 6 NDL PCBs			Total 7 NDL PCBs			Total 66 PCB congeners		
		M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range	M*	Medi*	Range
							Concentration in soils (µg/kg)									
Urban park	39	0.94	N.D.	[8.28 - 0.22]	1.21	0.2	[N.D. - 9.52]	4.76	1.4	[0.22 - 33.77]	4.94	1.5	[0.23 - 34.58]	6.44	5.0	[3.21 - 25.42]
Greening area	14	0.27	N.D.	[1.04 - 0.22]	0.22	0.2	[N.D. - 0.49]	2.04	1.3	[0.15 - 9.15]	2.24	1.5	[0.16 - 10.39]	5.87	5.2	[0.93 - 14.73]
Country park	9	0.09	N.D.	[0.35 - N.D.]	0.04	N.D.	[N.D. - 0.11]	0.73	0.7	[0.38 - 1.26]	0.74	0.7	[0.39 - 1.27]	4.01	3.7	[2.79 - 6.31]
Rural area	19	0.24	N.D.	[1.04 - 0.2]	0.31	0.2	[N.D. - 1.41]	1.65	1.4	[0.39 - 4.35]	1.72	1.5	[0.43 - 4.65]	4.67	3.9	[2.13 - 13.52]
Restored landfill	11	0.27	N.D.	[0.83 - 0.25]	0.21	0.1	[N.D. - 0.84]	1.61	1.3	[0.73 - 4.06]	1.7	1.4	[0.74 - 4.29]	4.49	4.4	[2.84 - 5.92]
Agricultural farmland	9	0.24	N.D.	[0.81 - 0.08]	0.1	N.D.	[N.D. - 0.51]	1.13	0.9	[0.46 - 2.71]	1.19	0.9	[0.47 - 2.97]	3.84	3.7	[2.39 - 5.12]
Orchard farm	5	0.09	N.D.	[0.24 - N.D.]	0.09	0.1	[N.D. - 0.23]	0.86	0.6	[0.53 - 1.38]	0.91	0.6	[0.54 - 1.5]	5.09	4.0	[3.79 - 9.84]
Crematorium	10	0.25	0.1	[0.83 - 0.17]	0.22	0.2	[0.06 - 0.74]	1.45	1.4	[0.87 - 3.01]	1.5	1.5	[0.88 - 3.12]	3.48	3.2	[2.18 - 5.64]
Industrial area	18	0.1	N.D.	[0.39 - N.D.]	0.09	0.0	[N.D. - 0.32]	1.01	0.8	[N.D. - 4.16]	1.07	0.8	[N.D. - 4.29]	33.13	7.1	[0.22 - 154.11]
Nearby Highway	4	0.63	0.1	[1.11 - 0.67]	0.44	0.4	[0.15 - 0.8]	5.3	3.1	[1.17 - 13.89]	5.81	3.3	[1.18 - 15.43]	24.78	8.7	[7.34 - 74.49]

Values in square brackets represent range of concentration. *M=Mean, Medi=Median. †: Not included for total 6 NDL PCBs. ‡:Calculated from sum of PCB 105, 118 and 157.

Table 2. Correlation matrix of DDT concentrations for each individual land use in Hong Kong.

Urban park				Agricultural farmland		
	DDT	DDD	DDE		DDT	DDD	DDE

Greening area:

	DDT	DDD	DDE
DDT		0.001	0.30
DDD	0.27		0.43*
DDE	0.27	0.63*	

Orchard farm (Agricultural farmland):

	DDT	DDD	DDE
DDT		0.63	0.70*
DDD	0.82		0.74*
DDE	0.77	0.98**	

Country park — Rural area:

	DDT	DDD	DDE
DDT		0.35	-0.01
DDD	0.08		0.66
DDE	0.08	0.98*	

Crematorium — Industrial area:

	DDT	DDD	DDE
DDT		-0.31	-0.24
DDD	0.48*		0.48
DDE	0.48*	1.00**	

Restored landfill — Nearby highway:

	DDT	DDD	DDE
DDT		0.71*	0.68*
DDD	0.89		1.00*
DDE	0.85	0.99*	

Pearson correlation coefficients were shown. Values with * indicated that significant correlations were found at $p=0.05$, and values with ** indicated that significant correlations were found at $p=0.01$.

Table 3. Various ratios for DDT and its metabolites calculated for each land use in Hong Kong soil.

Land uses	DDD / DDT	DDE / DDT	DDT / (DDD+DDE)
Urban park	0.17 ± 0.30 (a)	0.08 ± 0.08 (a)	6.33 ± 5.96 (a)
Greening area	0.15 ± 0.10 (a)	0.09 ± 0.08 (a)	4.35 ± 6.54 (a)
Country park	0.07 ± 0.10 (a)	0.02 ± 0.05 (a)	8.38 ± 7.14 (a)
Rural area	0.06 ± 0.08 (a)	0.03 ± 0.04 (a)	9.45 ± 7.67 (a)
Restored landfill	0.38 ± 0.19 (a)	0.20 ± 0.11 (ab)	0.80 ± 1.21 (a)
Agricultural farmland	0.12 ± 0.11 (a)	0.12 ± 0.07 (a)	6.92 ± 5.07 (a)
Orchard farm	0.16 ± 0.11 (a)	0.36 ± 0.66 (b)	3.56 ± 2.76 (a)
Crematorium	0.10 ± 0.10 (a)	0.05 ± 0.05 (a)	4.79 ± 4.00 (a)
Industrial area	0.20 ± 0.19 (a)	0.08 ± 0.09 (a)	4.84 ± 5.92 (a)
Nearby highway	0.01 ± 0.01 (a)	0.01 ± 0.01 (a)	58.73 ± 42.14 (b)

Values in parenthesis represent grouping by one way ANOVA analysis (Tukey HSD, $p<0.05$).

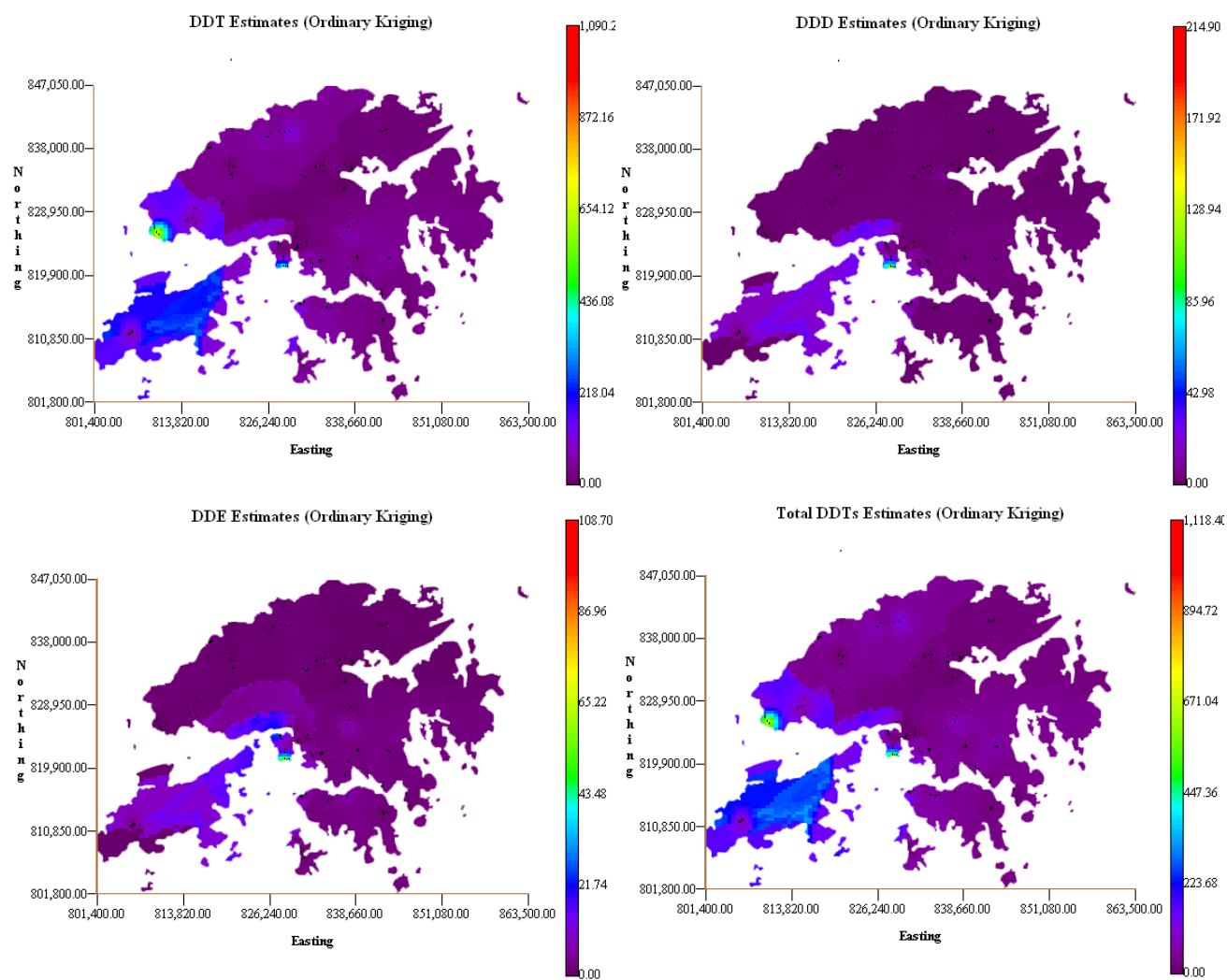**Table 4. Soil quality guidelines and their criteria for DDTs and PCBs in various countries.**

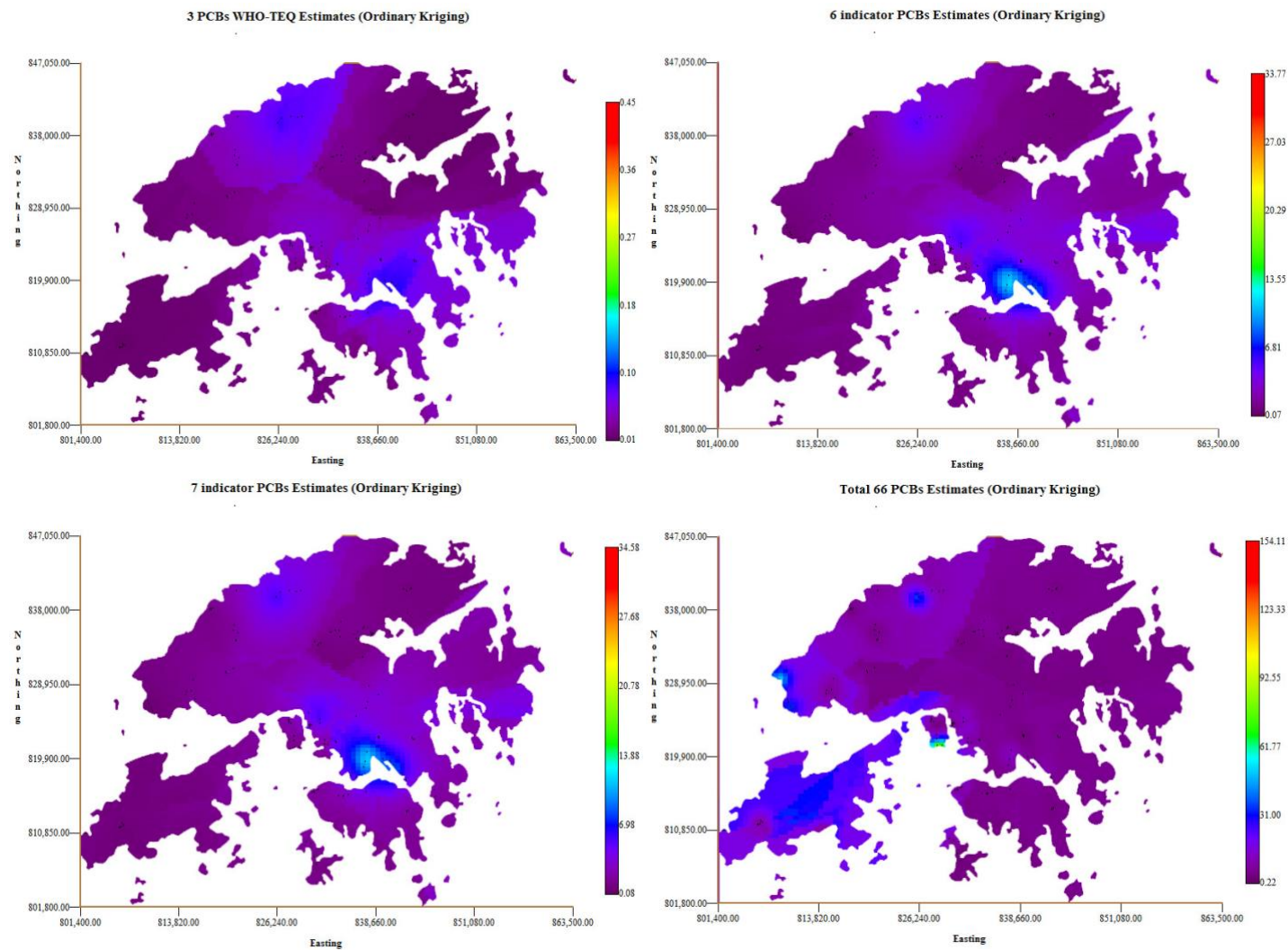| Country of implementation	Quality guidelines	DDT	DDD	DDE	Total DDTs	PCBs
		Concentration in soils (μg/kg)				
Netherlands	Dutch target value	N.A.	N.A.	N.A.	10*	20‡
	Dutch intervention value	N.A.	N.A.	N.A.	4000*	1000‡
Denmark	Danish soil quality criteria	N.A.	N.A.	N.A.	1000#	N.A.
Canada	Canadian environmental quality guidelines	700†	N.A.	N.A.	N.A.	(500) 1300†
China	Environmental quality standard	N.A.	N.A.	N.A.	N.A.	N.A.

* Sum of DDT, DDD and DDE.

Quality criteria for soil in areas with very sensitive land use.

† Vales in parenthesis represent criteria for agricultural land uses; values without parenthesis represent criteria for residential/parkland uses.
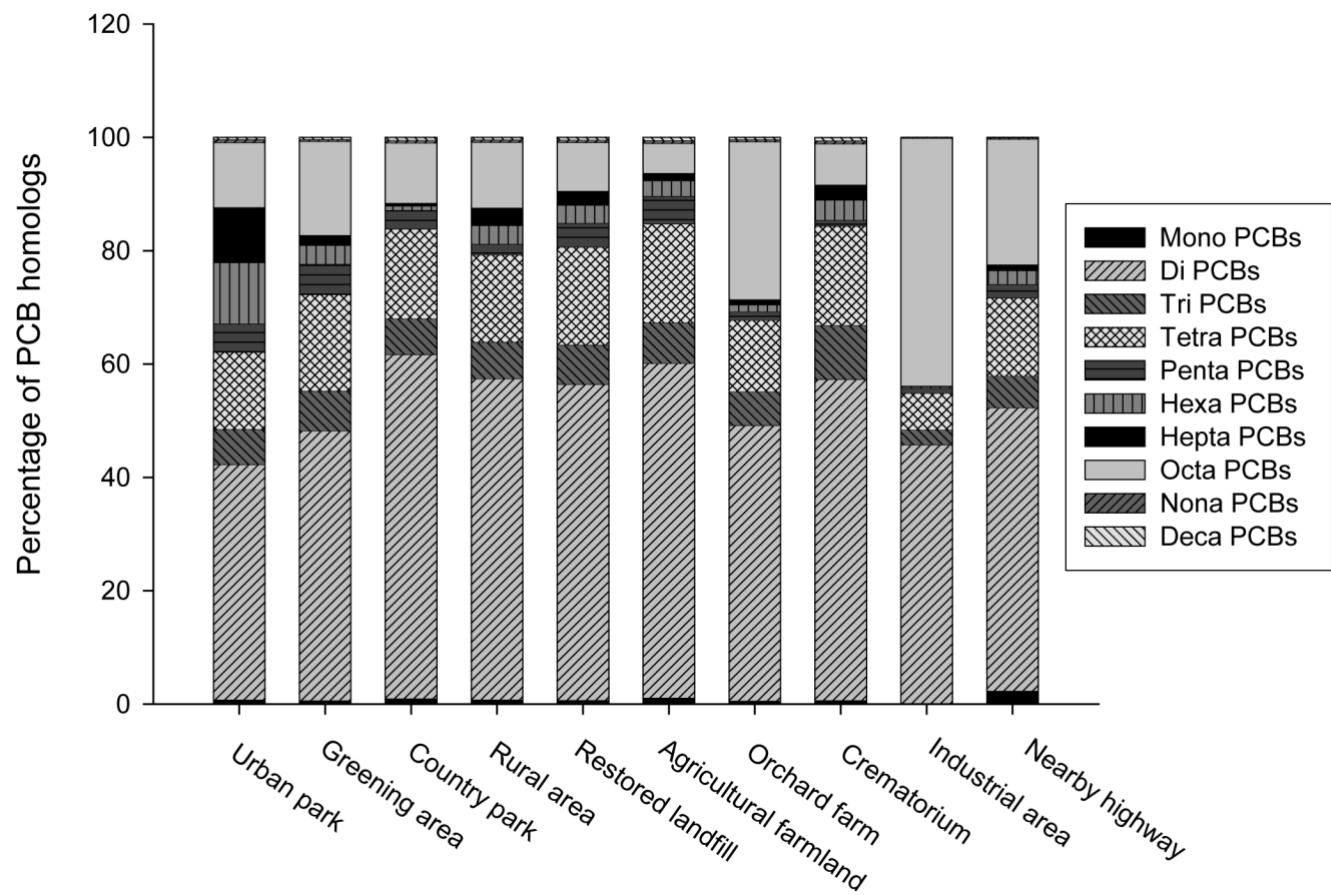
‡ Intervention value means sum of 7 PCB congeners: PCB-28, -52, -101, -118, -138, -153 and -180. Target value refers to total PCBs excluding PCB-118.

Figure 1. Kriged maps DDT (DDT, DDD, DDE and total DDTs) and PCB (3-PCBs WHO-TEQ congeners, 6 indicator PCBs, 7 indicator PCBs and sum of 66 PCB congeners) levels (µg/kg) in surface soils of Hong Kong. The 3-PCBs WHO-TEQ is in ng kg$_{-1}$ scale.

3 PCBs WHO-TEQ Estimates (Ordinary Kriging)

6 indicator PCBs Estimates (Ordinary Kriging)

7 indicator PCBs Estimates (Ordinary Kriging)

Total 66 PCBs Estimates (Ordinary Kriging)

55   Figure 1 (Con't)

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Figure 2. Homologue profiles of PCBs in corresponding land uses in Hong Kong. Total PCBs is sum of 66 selected congeners

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