Developments in Environmental Science, Volume 7
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ISSN: 1474-8177/DOI:10.1016/S1474-8177(07)07006-4

Chapter 6

Sources and Occurrence of Persistent Organic Pollutants in the Pearl River Delta, South China

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Abstract

The Pearl River Delta is a fast-developing economic zone in South China. Relatively abundant monitoring data of POPs are available in the regional environment. DDT is still the most priority POP widely occurred at high concentrations in the air, wild fishes and human breast milk. Fast-increasing municipal waste incinerators, as well as electronic waste cycling activities in the region are important potential sources for PBDEs and PCDD/Fs. Atmosphere and aquatic biota are suggested to be the priority environmental media for monitoring POPs in the subtropical environment, and regional collaboration between mainland China, Hong Kong and Macau on the monitoring and inventory of POPs are of critical need in the Pearl River Delta.

6.1. Introduction: The Pearl River Delta

Low-latitude regions may be important source regions for persistent organic pollutants (POPs), and may also play an important role in the global atmospheric transport of these pollutants. In developed countries, the production and application of many important POPs, in particular those of organochlorine pesticides (OCPs), have been prohibited or well controlled/managed, and their distribution in the environment is relatively uniform (Jaward et al., 2004). However, in most developing countries, in particular those located in the low-latitude regions, there may still be new sources and current application of some POPs, as a result of exemption of the Stockholm Convention, illegal local use, as well as potential transfer of industries and electronic wastes from developed countries to developing countries (BAN, 2002). On the other hand, owing to their volatility, many POPs are capable of migrating in the atmosphere

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across a long distance, from warmer places to colder regions (Wania and Mackay, 1996). Therefore, the current situation of POPs contamination in the low-latitude regions is of interest in the context of their global sources, inventories and transport.

Located in the low-latitude region, below the Tropic of Cancer, the Pearl River Delta (PRD) is one of the most important metropolitan areas in China. The PRD has a land area of ca. 41,700 km^2, a population of over 40.2 millions. Large and important cities in the PRD include Guangzhou, Hong Kong, Shenzhen, Fushan, Macau, Zhuhai, Zhongshan, Dongguan etc. (Fig. 6.1). Administratively, the mainland part of the PRD is affiliated to Guangdong Province. Owing to the fast industrialization during the last 25 years, the PRD contributes more than 10% of the country's gross domestic product (GDP, excluding Hong Kong and Macau). The annual average air temperature and precipitation in the PRD are 21–22°C and 1690–1900 mm, respectively. The Pearl River is the third largest river in China, with an average discharge of 174 billion m^3/year and an associated suspended load of 37.3 million tones (Hong et al., 1999). Being the habitat of more than 320 fish species, the Pearl River estuary (PRE) has an area of 2110 km^2, forming one of the key ecosystems in south China costal area.

6.2. Concerned sources of POPs in the PRD

6.2.1. Organochlorine pesticides (OCPs)

The PRD had a long history of agricultural application of large amounts of technical DDTs and HCHs until their official ban implemented in 1983. In the PRD, it is estimated that OCPs usage was ~76,000–100,000 tons annually from 1972 to 1982 (Hua and Shan, 1996). The application rate of these chemicals has been averaged from 1.8 to 2.7 kg/metric acre in the agricultural zones around the Delta (Mai et al., 2003). The residues from the past usage in soil and sediment were believed to be the secondary sources for DDTs and HCHs in the contemporary environment, via evaporation, desorption and with land runoffs, enhanced by the intensifying human activities and land transform in the fast-developing region (Zhang et al., 2002).

An acaricide in wide application, dicofol, was suspected to be a new source of DDTs in China (Qiu et al., 2003). Dicofol is synthesized by using DDT as a key intermediate. The residue of DDTs is controlled to be $<0.1\%$ in many developed countries. However, due to the less advanced industrial technology, the concentration of DDT residue in dicofol in China is still high. According to a report, the average contents of

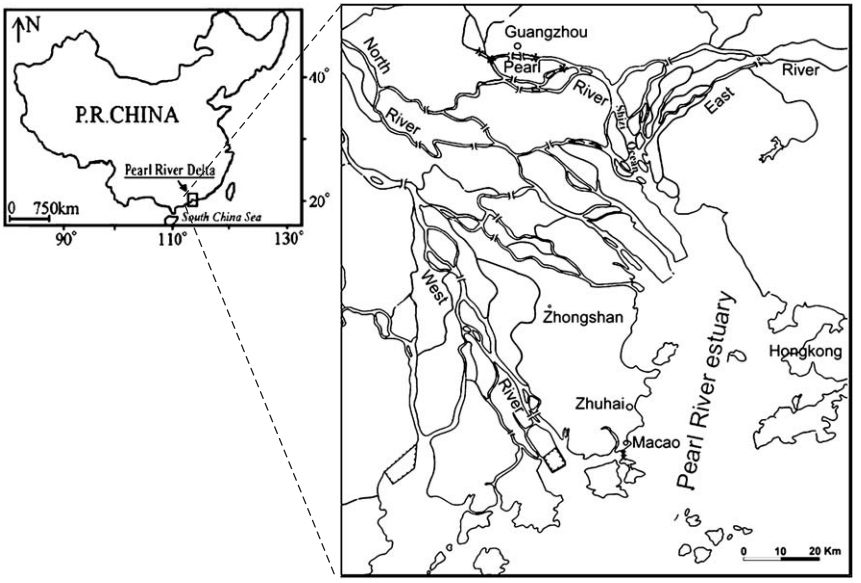

Figure 6.1. Map showing the Pearl River Delta, South China.

o,*p*′-DDT, *p*,*p*′-Cl-DDT, *o*,*p*′-DDE, and *p*,*p*′-DDT in some commercial available samples were 114, 69, 44, and 17 g kg^{-1} dicofol, respectively. On the basis of a production and distribution survey, the total input of DDTs to the environment from the dicofol use in China was estimated to be 8770 tons between 1988 and 2002 (Qiu et al., 2005). Dicofol is also applied in the PRD. A commercially available dicofol product on the market in Guangzhou contains ca. 5% of *p*,*p*′-DDE, 4.5% of *o*,*p*′-DDT and 5% of *p*,*p*′-DDT (Li Jun et al., 2003, unpublished data). In 2003, an average of 14.4 tons of dicofol (as pure product) were applied in the PRD (An et al., 2005), corresponding to the introduction of ca. 2 tons of DDTs (sum of *p*,*p*′-DDE, *o*,*p*′-DDT and *p*,*p*′-DDT) to the environment.

Another potentially important regional source of DDT in the PRD may be from antifouling paints used on fishing boats. The average annual production of DDT was 4519 tons during 2000–2003 in China, and there was no DDT import from other countries. Approximately 4% of total DDT production was used as the additive for the production of antifouling paint for fishing ships. It is estimated that ~150–300 tons of DDT per annum is emitted through antifouling paint usage in China, corresponding to roughly 5% by weight DDT content in the product. The coastal area of Guangdong province has ca. 60,000 fishing ships, which is ~1/5 of the total number in China. Roughly it can be estimated that ~30–60 tons of DDT may be introduced to the coastal environment of Guangdong, including the PRD. As most of this DDT is expected to be released directly to coastal waters, their potential ecological risk needs to be further evaluated.

The γ-isomer of HCH, as the active component in Lindane, is widely used as a pesticide in the mainland part of the PRD (Hong Kong banned Lindane in 1991; AFCD, 2001), both in agriculture and household. It has been reported that conversion of γ-HCH to α-HCH in the environment is possible (Walker et al., 1999), and this point will need to be considered when analyzing the potential source of HCH in the environment.

Chlordane and Mirex were widely used in the PRD for termite control. Chlordane was strictly banned by the local government in 2001, while Mirex is still in use in Guangdong due to the lack of less toxic substitutes. The estimated average annual application of Mirex in the PRD was ca. 44 tons in the PRD in 2000 (An et al., 2005).

6.2.2. Industrial chemicals: Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs)

PCBs can be contained in industrial products manufactured in the past, and/or formed as by-products during combustion processes. In the context of industrial PCBs, China issued a "Relevant Regulations for

Stopping the Production of PCBs'' in 1974, and issued a ''Circular on Preventing the Pollution of Hazardous Substances of PCBs'' in 1979. Approximately 10,000 tons of PCBs were manufactured, and of these, 9000 tons were trichlorobiphenyl applied in transformers and capacitors. The remaining 1000 tons were pentachlorobiphenyl mainly applied in paints, inks and lubricants as an additive. In Guangdong, most of the PCB-containing capacitors were replaced and sealed/stored for more than 20 years. They have been further treated safely in recent years gradually by the local government. Until 1999, the remaining untreated PCB-containing capacitors were estimated to be a few hundreds (Li, 1999).

PBDE is another group of industrial POPs. It is used as brominated flame retardants through the addition to or reaction with plastics and synthetic fibers. These fire retardants often make up a considerable proportion of product weight: plastic can contain up to 15% PBDEs, and polyurethane foam (PUF) up to 30% PBDEs (WHO, 1994; EU, 2003). The total global consumption of PBDEs in 2001 was 67,440 tons, among them deca-BDE accounted for more than 80% and octa-BDE more than 12%. The PRD has become an important manufacturing base in the world since the 1980s, for the production of electronics, appliances, personal computers and peripheral devices, textile and furniture. In particular, it is one of the three electronic manufacturing bases in China, accounted for more than 40% (ca. 100 billion US$ in 2002) of the total production of electronic products in the country. Therefore, the application amount of PBDEs in the PRD is estimated to be high. However, as the regional industry began to thrive largely in the 1990s, when the production of PBDEs was shifted from penta- and octa-BDE to deca-BDE, the major PBDE product applied is believed to be deca-BDE, as would be consistent with the use of PBDE products in Asia (50,710, 331, 3307 tons of deca-, penta-, and octa-BDE, respectively, in 2001). It should be noted that, in 2003, EU announced the new Regulation of Hazardous Substances (RoHS), which came into force on July 1, 2006. RoHS strictly regulates PBDE residues in electronic products exported to EU, and has consequently limited the PBDE application in the PRD, from where a large portion of electronic products are exported to EU. In addition, China will also implement its domestic RoHS, mainly targeting electronic products, in 2007, which is believed to further limit the industrial application of PBDEs.

6.2.3. Unintentional by-products: Polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans (PCDD/Fs), PCBs and PBDEs etc.

PCDD/Fs have various sources and are produced as unintentional by-products of many manufacturing and combustion processes involving the

use, production or disposal of chlorine or chlorine-derived chemicals, notably, polyvinyl chloride (PVC) polymers. Sources of significant environmental concern for PCDD/Fs include waste incineration, combustion of landfill gases, open burning, and many organochlorine production processes. Other sources include steel industry, ceramic industry, power generation etc. PCDD/Fs are also found as impurities in 2,4-D and pentachlorophenol (PCP). Hong Kong assembled its PCDD/Fs inventory in 2003. However, until now, there is no regional inventory of PCDD/Fs in the PRD as a whole.

Incineration is one of the major waste treatment options in the PRD under the huge burden of municipal solid waste (MSW). Small waste incinerators with capacity less than 300 ton/day have been strictly regulated since 2001, and most of them have been closed in recent years. However, until 2006, there were nine large MSW incinerator power plants located in Shenzhen, Guangzhou, Zhuhai, Fushan and Dongguan, treating 6640 tons of MSW daily, and there are other eight incinerators being built for an extra capacity of 6150 tons/day. It is planned that, before 2010, there will be a total of 27 large MSW incinerator power plants in the PRD, treating 23,490 tons of MSW, which account for 73.87% of the total MSW generated daily in the region. Most of the incinerator power plants adopted advanced technologies from European countries. It is estimated that 70% of the light industrial waste consists of PVC plastics, rubber, leather and cloth etc. (Fu et al., 2003). Although concentrations of PCDD/Fs in the emission of these large incinerators are still not available, it could be estimated that they may contribute significantly to the total environmental loadings of PCDD/Fs in the PRD.

Illegal electronic waste (e-waste) recycling is believed to be another important source for unintentional POP emissions, in particular for PBDEs, PCBs, PCDD/Fs (Soderstrom and Marklund, 2002; Wang et al., 2005; Cai and Jiang, 2006), and polycyclic aromatic hydrocarbons (PAHs; Yu et al., 2006a, b). The hazardous e-wastes include computers, electronic appliances and transformer carcasses. In the e-waste sites, recycling operations consist of toner sweeping, dismantling electronic equipment, selling computer monitor yokes to copper recovery corporations, plastic chipping and melting, burning wires to recover copper, melting circuit boards over open fires and using acid chemical strippers to recover gold and other metals (Cai and Jiang, 2006). It was estimated that more than 70% of the e-waste from North America is shipped/smuggled to Asia, most of them to the southeastern coastal China (BAN, 2002). In addition, it was estimated that more than 200 million sets of electronic appliances, mainly TV sets and electronic fans, were retired/disposed of in Guangdong. There are three large scale e-waste recycling towns in the province, among them two

are located in the PRD. Despite the potential importance of e-waste recycling as a source of PCDD/Fs, PCBs and PBDEs, the POP emission factors associated with the process of e-waste burning and recycling without sound management, as well as its contribution to POPs in the environment, are still largely unknown in the region.

6.3. Occurrence of POPs in the environment of the PRD

6.3.1. Organochlorine pesticides (OCPs)

6.3.1.1. Air

A screening of POPs in the atmosphere in East Asia using passive atmosphere samplers (PAS) showed that the PRD was among the regions with the highest concentrations of DDTs and HCB in Asia (Jaward et al., 2005). Nevertheless, there are only limited studies reporting levels, based on active high-volume samplers, of POPs in the air of the PRD. Cheng et al. (2000) analyzed OCPs in a few aerosol samples in this area, the results showed high concentrations of DDTs in Guangzhou (up to 109.38 pg m^{-3}). Louis and Sin (2003) collected air samples, using modified Anderson high-volume samplers, during the winter of 2000/2001 at the Tai Mo Shan weather station in Hong Kong. OCPs were found at relatively low concentrations of ~0.02–0.23 ng m^{-3}. Recently, a study was conducted to measure the atmospheric concentrations of OCPs in Guangzhou and Hong Kong fortnightly from December 2003 to December 2004. The results showed that the atmospheric DDT concentrations in the PRD were much high (Li et al., 2007) when compared with other places in the world (Table 6.1). The average concentrations of α- and γ-hexachlorocyclohexane (HCH), *trans*-chlordane (TC), *cis*-chlordane (CC), *p*,*p*′-DDT, *p*,*p*′-DDE, *p*,*p*′-DDD, *o*,*p*′-DDT, endosulfan-α and -β in the air of Guangzhou were 134, 539, 871, 1340, 627, 207, 93, 846, 246, and 60 pg m^{-3}, respectively; and in Hong Kong were 46, 51, 389, 380, 358, 56, 53, 191, 148, and 20 pg m^{-3}, respectively. In general, the concentrations of OCPs increased from winter to summer except for α-HCH, which showed no clear seasonal pattern.

6.3.1.2. Water

Significant amounts of OCPs were found in water of rivers, estuaries and seas, and generally, sewage water concentrations > river concentrations > estuary concentrations > sea concentrations. Levels of OCPs in sewage from Guangzhou had the highest concentrations of DDTs and HCHs

Table 6.1. Comparison of organochlorine pesticide concentrations (pg/m^3) in the atmosphere of Guangzhou and Hong Kong with reference data (Li et al., 2007)

	α-HCH	γ-HCH	TC	CC	*p,p′*-DDE	*p,p′*-DDD	*p,p′*-DDT	*o,p′*-DDT	End 1	Reference
Hong Kong	46±28 (5–108)	51±63 (5–261)	389±381 (7–1602)	380±358 (7–1496)	56±26 (8–151)	53±58 (3–235)	358±718 (6–2625)	191±188 (7–673)	148±212 (<LOD–840)	Li et al. (2007)
Guangzhou	134±70 (41–313)	539±797 (5–2639)	871±443 (127–1789)	1335±989 (129–4185)	207±151 (28–579)	93±85 (8–378)	627±740 (22–2836)	846±618 (86–2014)	246±291 (<LOD–1083)	Li et al. (2007)
South India	(530–27400)	(380–8170)			(400–5790)	(140–520)	(60–1020)	(0–150)		Rajendran et al. (1999)
Korea	169±250 (20.6–830)	50.5±86.6 (4.8–326)	5.0±9.4 (0.2–35.0)	3.7±7.1 (0.1–26.1)	34.6±51.3 (2.6–185)	6.2±18.3 (<5.6–67.0)	21.0±34.6 (<1.0–121)			Yeo et al. (2003)
Japan	103±67 (14–281)	40±22 (10–98)	70±89 (2–401)	61±76 (<1–329)	5.3±4.2 (<0.3–17.8)	<2 (<2–<2)	6±7 (<2–25)			Murayama et al. (2003)
Chicago	110±40	150±80	130±80	120±70		100±50	70±60			Bidleman et al. (1998)
Belize, Central America		63±22	34±39	32±42	458±127	35±45	556±356	145±45		Alegria et al. (2000)

(up to 1200 ng l^{-1}), and the total OCPs in river waters were from 130 to 1200 ng l^{-1} (Yang et al., 1997). The OCP concentrations in estuary water were significantly lower than those of river. For example, total OCP concentrations in estuary near Macao were from 25.2 to 67.8 ng l^{-1} (Luo et al., 2004). A survey of OCPs in surface sea waters in 2005 was carried out in the South China Sea (SCS). A total of nine 100 L surface seawater samples were collected, and relatively low concentrations of total OCPs with 1.3–7.4 ng l^{-1} were found (Zhang et al., 2007).

6.3.1.3. Sediment

The concentrations of total HCHs (t-HCHs) ranged from 0.08 to 1.38 ng g^{-1} with a mean value of 0.36 ng g^{-1}, which were comparable to those of Kyeonggi Bay, Korea (<0.15–1.2 ng g^{-1}) and Xiamen Harbor (0.14–1.12 ng g^{-1}), but were lower than those of the Yangtze River Estuary and its nearby coastal area (n.d.–30.4 ng g^{-1}). The levels of total DDTs (t-DDT; sum of *p*,*p*′-DDD, -DDE, and-DDT) ranged from 0.04 to 2.48 ng g^{-1} with an average of 0.87 ng g^{-1}. The mean concentration (1.54 ng g^{-1}) of t-DDTs within the PRE was slightly lower than the mean concentration of 2.84 ng g^{-1} obtained in the same area by Hong et al. (1999). Again, the DDT levels were lower than those reported for Kyeonggi Bay, Korea (0.048–32 ng g^{-1}), but were higher than the levels in the Yangtze River Estuary and its nearby coastal area (n.d.–0.57 ng g^{-1}). The concentrations of t-OCPs (sum of t-HCHs, t-DDTs, and heptachlor) exhibited positive correlations with the TOC contents ($r = 0.76$ with $p < 0.001$), suggesting that TOC was a significant factor controlling the contaminant distribution in sediments (Chen et al., 2006a, b, and references therein).

6.3.1.4. Soil

A survey of OCPs in surface soils was carried out recently in the PRD (Li et al, 2006). A total 74 soil samples containing crop, paddy and natural soils were collected. The concentrations and enantiomeric compositions of HCH, DDT, and chlordane were analyzed. Detailed information on the concentrations in the soils is shown in Table 6.2. The mean concentrations of total HCHs and DDTs in descending order were: crop soils > paddy soils > natural soils; whereas, only pesticides in the natural soils were significantly ($p < 0.001$) lower than crop soils and paddy soils. The spatial distribution of HCHs and DDTs was similar, with higher levels found at the center of the PRD. The DDT concentrations in crop (mean 54 ng g^{-1} dw) and paddy soils (mean 47 ng g^{-1} dw) in the PRD were similar to those reported in paddy soils in Dehradun of

Table 6.2. OCP concentrations in soils of different land uses in the Pearl River Delta (Li et al., 2006)

Subgroup	$ng\,g^{-1}$	α-HCH	β-HCH	γ-HCH	δ-HCH	*p*,*p*′-DDE	*p*,*p*′-DDD	*p*,*p*′-DDT	*o*,*p*′-DDT	TC	CC
Crop soils (37)	Mean	1.08	2.49	0.90	0.50	21.9	7.22	20.9	6.34	0.62	0.71
	Median	0.80	1.26	0.54	0.42	7.82	2.70	4.64	0.80	0.42	0.40
	Range	<DL–3.48	0.05–16.0	<DL–4.43	<DL–1.28	0.14–231	0.06–73.8	0.07–305	<DL–96.7	<DL–6.13	0.10–7.74
Paddy soils (14)	Mean	0.89	1.76	0.62	0.38	19.4	23.4	2.98	0.91	0.42	0.36
	Median	0.78	1.14	0.45	0.26	12.6	7.64	1.05	0.33	0.47	0.40
	Range	<DL–2.76	<DL–7.01	<DL–2.03	<DL–1.02	1.64–66.6	0.07–163	0.19–22.4	0.12–4.92	<DL–0.76	<DL–0.56
Natural soils (23)	Mean	0.25	0.27	0.11	0.20	0.82	0.53	1.40	0.46	1.53	1.61
	Median	0.17	0.11	0.04	0.14	0.71	0.48	1.12	0.41	0.45	0.38
	Range	<DL–1.46	<DL–1.11	<DL–0.75	<DL–0.70	<DL–2.22	0.05–1.40	0.09–4.20	0.05–1.22	<DL–13.7	<DL–15.5

India 13–238 ng g^{-1} (Babu et al., 2003), lower than those reported in soils in urban Guangzhou (ranged from 3.58 to 831 ng g^{-1}) (Chen et al., 2005), Tianjin (628.1–2840.5 ng g^{-1}) (Gong et al., 2002), Vietnam (averaged 110 ng g^{-1}) (Thao et al., 1993), the US Corn Belt (n.d.–11,800 ng g^{-1}) (Aigner et al., 1998), and Romania (9.0–1542.2 ng g^{-1}) (Covaci et al., 2001). Another independent survey of OCPs in soils conducted recently in Hong Kong (Zhang et al., 2006) found much lower concentrations of DDTs (0.52 ng g^{-1} dw) and HCHs (6.19 ng g^{-1} dw).

6.3.1.5. Biota

There have been limited reports on OCP levels in aquatic organisms in the PRD. OCP concentrations were reported in mussels from Hong Kong waters (Phillips, 1985; Fang et al., 2001), fishes from the Hong Kong market (Chan et al., 1999), fishes (Tilapia) from Hong Kong inland waters (Zhou et al.,1999) and from aquacultural fishponds in the PRD (Zhou and Wong, 2004). The concentrations in freshwater fishes were 0.01–7.8 ng g^{-1} lipid for HCHs, and 22.3–381 ng g^{-1} lipid for DDTs (Zhou and Wong, 2004). In a recent survey of OCPs in demersal fishes in the PRE and Daya Bay, 50 samples (34 fishes, 4 shrimps, 6 crabs) were collected and analyzed with GC-MS. The average concentrations of t-DDTs, t-HCH, and t-Chlordane in the fishes were 73.36 ± 102.37, 0.30 ± 0.18, 0.87 ± 0.35 ng g^{-1} ww in Daya Bay, and 123.68 ± 100.66, 0.41 ± 0.48, 12.23 ± 26.85 ng g^{-1} ww in the PRE, respectively. Concentrations of HCHs and chlordanes were comparable to those reported in other region, while DDTs levels were higher (Guo et al., 2007). The results suggested that DDT is still the priority pesticidal POP in the aquatic ecosystem of the PRD.

The mean concentration of total HCHs was 1164.7 ng g^{-1} ww, in the blubber of five live Indo-Pacific humpback dolphins in Hong Kong, 36 times greater than that of Zhuhai (32.8 ng g^{-1}). For total chlordanes, the Hong Kong samples (258.6 ng g^{-1}) had concentrations comparable to those of the Xiamen (249.5 ng g^{-1}) and Zhuhai (221.7 ng g^{-1}). However, the mean total DDT concentration in Hong Kong samples, 9110.5 ng g^{-1} ww, was lower than those of Xiamen and Zhuhai (Leung et al., 2005).

6.3.2. PCBs

6.3.2.1. Air

Limited reports are available on PCBs in the atmosphere of the PRD. The total PCB levels measured in a background site (Tai Mo Shan) in

Hong Kong ranged from 10 to 1910 (mean 480) pg m^{-3} (Louie and Sin, 2003). In 2005, air samples were collected at four urban sites in Guangzhou for the analysis of 64 PCB congeners. The study reported similar total PCB concentrations (sum of 64 congeners) of 363–1961 pg m^{-3} (Chen et al., 2006). This was similar to the range of 20–1700 pg m^{-3} in Europe as determined by PAS technology by Jaward et al. (2004), but much higher than the range in Asia (5–340 pg m^{-3}, by PAS, Jaward et al., 2005). The PCBs in the air of Guangzhou were dominated by congeners with 3–4 chlorine atoms (Chen et al., 2006).

6.3.2.2. Water

Nie et al. (2005) reported PCB concentrations from water samples collected from four major river outlets of the Pearl River. The concentrations of total PCBs in surface water ranged from 2.47 to 6.75 ng l^{-1}, with an average of 3.92 ng l^{-1}. There were no significant differences between various sampling sites. These levels were 100 times higher than the samples collected from the uncontaminated Atlantic Ocean (0.035–0.072 ng l^{-1}; Connel et al., 1999), close to the level obtained in Humber estuary, UK (1.0 ng l^{-1}; Zhou et al., 1996), but much lower than those observed in Daya Bay in China (313.6 ng l^{-1}; Zhou et al., 2001a, b) and in Minjiang River Estuary, Southeast China (214–1819 ng l^{-1}; Zhang et al., 2003). In terms of the pattern of congener composition in water, PCB-153, 180, 119, and 49 (with 6, 7, 5, and 4 chlorine atoms, respectively) were dominant.

6.3.2.3. Sediment

The levels of PCBs in the PRD sediments have been compared with those from other parts of the world in recent studies (Mai et al., 2002; Fu et al., 2003). The sediments in Zhujiang River (48.3–486 ng g^{-1}) and Macao Harbor (339 ng g^{-1}) were thought to be moderately to strongly contaminated by PCBs, when compared to the world-wide concentrations (0.2–400 ng g^{-1}) of near-shore surface sediments (Fowler, 1990). PCB levels in sediment from Shiziyang (10–30.3 ng g^{-1}) and Xijiang (11–13.6 ng g^{-1}) Rivers, and Lingding Bay (10.2–13.5 ng g^{-1}) were comparable to those of sediments from Canadian Mid-latitude and Arctic lake sediments (2.4–39 ng g^{-1}) (Muir et al., 1996). The DDT concentration (1629 ng g^{-1}) in sediment from Macao Inner Harbor is higher than those obtained in other heavily polluted locations, such as Rhone Prodelta in the north-west Mediterranean region (124–657 ng g^{-1}) (Tolosa et al., 1995). DDTs levels in sediments of Zhujiang (35.1–91.0 ng g^{-1}) and

Shiziyang (22.9–40.4 ng g^{-1}) rivers were in the high end of the world-wide concentration range, and comparable to those of moderately polluted locations, such as Ebro prodelta in the NW Mediterranean region (23–89 ng g^{-1}) (Tolosa et al., 1995). Nie et al. (2005) reported that the concentrations of total PCBs (36 congeners) in the sediments were 23.23, 17.82, 13.65, and 11.13 ng g^{-1} dw for Humen, Hengmen, Jiaomen, and Modaomen, respectively, and these concentrations were within the ranges of the above-mentioned studies (Mai et al., 2002; Fu et al., 2003). Another study conducted in Deep Bay of the PRE reported a total PCB (28 congeners) concentration range of 6.0–290 ng g^{-1} dw (Fung et al., 2005), which is also within the regional range (Mai et al., 2002; Fu et al., 2003).

6.3.2.4. Biota

The concentrations of PCBs in 71 fishes in the PRE were reported by Nie et al. (2005). The total PCB concentrations (38 congeners) ranged from 68.64 to 316.85 ng g^{-1}, lipid in fish. The concentration of PCBs in fish in this study were higher than that of shellfish ($<$6.78 ng g^{-1} dw) from Minjiang estuary (Chen et al., 2002), close to that of shrimp and fish from Mai Po Marshes in Hong Kong, and lower than or close to the concentration of PCBs in green-lipped mussels (*Perna viridis*, 82.8–615.1 ng g^{-1}, lipid) from the PRE (Fang et al., 2001). However, by comparison with other places in the world, the concentrations of PCBs in fish from the PRE were lower, well below the 2000 ng g^{-1} on a wet weight basis of maximum concentration of total PCBs in edible seafood allowable by the Food and Drug Administration. Notwithstanding, the PCB concentrations in some species exceeded the standard in meat, eggs, poultry and related products at 200 ng g^{-1} lipids set by the European Directive 1999/788/CE (Binelli and Provini, 2003). It was found that the feeding habits of fish were very significant in influencing the accumulation of polychlorinated biphenyls and their congeneric pattern. Carnivorous and benthic fishes such as eel (*Anguilla japonica*) and Chinese sea catfish (*Arius sinensis*) were found to have high concentrations of polychlorinated biphenyls (mainly of high chlorinated congeners), while herbivores such as shad (*Clupanodon punctatus*) and mullet (*Mugil cephalus*) exhibited an opposite trend. Congeners PCB-153, 138, 118, 87/81, 170, and 52 were found frequently in most fish samples. Another study investigated the PCB concentrations in 61 freshwater fish samples (Tilapia, common carp, big head, silver carp, and grass carp) from the PRD (Zhou et al., 2004), which reported total PCB concentration ranges of 60–480 ng g^{-1} lipid, similar to those in the estuarine fishes (Nie et al., 2005), and 16% of fish collected from the PRD exceeded the US EPA guideline concentration for PCBs. Concentrations

of total PCBs ranged from 5.3 to 40.8 μg g^{-1} ww (mean 19.6), in the blubber of five live Indo-Pacific humpback dolphins in Hong Kong, and 0.07 μg g^{-1} ww in a dolphin from Zhuhai (Leung et al., 2005).

6.3.3. PBDEs

6.3.3.1. Air

There have been two noteworthy studies of PBDE measurements in air of the PRD. One using high volume air samplers in the urban city of Guangzhou in June of 2004 (Chen et al., 2006) and the other using PUF passive air samplers in three locations (Dinghushan, Guangzhou and Hong Kong) of the PRD from Sept. to Nov. 2004 (Jaward et al., 2005). The concentrations of Σ_{10}PBDEs in air samples (particle plus gas phases) collected from Guangzhou ranged from 88.8 pg m^{-3} at an urban site to ~3672 pg m^{-3} near an industrial area, while BDE 209 ranged from 263.8 pg m^{-3} to ~4200 pg m^{-3}. The arithmetic mean atmospheric concentrations of ΣPBDEs (sum of all target PBDE congeners except for BDE 209) in samples from the urban and city background sites in this study were comparable to or slightly higher than those from other places around the world. The arithmetic mean atmospheric concentrations of BDE 209, however, were higher than those in North America and Europe, and similar to the values from Japan. Congeneric compositions were dominated by BDE 209 in all (> 70%) but an industrial site, with an average abundance of 48% for BDE 209. The PBDE congeneric profiles in air samples indicated that deca- and penta-BDE were the main source mixtures consistent with the results from the sediments. In a study of air across Asia, the PBDE concentrations (sum of BDE 17, 28, 32, 47, 49, 75, 99, 100) in PUF samplers from three locations of the PRD region ranged from 3.97 to 23.8 pg/sample, and these were generally lower than those in developed countries (Jaward et al., 2005).

6.3.3.2. Water

Very few data is available for PBDEs in water of the PRD. A recent study reported PBDE concentrations in five paired subsurface layer (1 m) and micro-surface layer water samples in Hong Kong. The total PBDE (eight congeners) concentrations ranged from 11.3 to 62.3 pg^{-1} in the dissolved phase and from 26.2 to 32.5 pg^{-1} in the suspended particulate matter. BDE 28, 47, and 100 were the most abundant congeners, with no BDE 209 detected. This was different from those reported in San Francisco

Bay in the US, where BDE 47, 99, and 209 were found to be the most abundant congeners (Oros et al., 2005).

6.3.3.3. Sediment

There have been several studies of PBDE concentrations in sediments of the PRD (Zheng et al., 2004; Liu et al., 2005a, b; Mai et al., 2005a, b). A total of 66 river and estuarine surface sediment samples from the PRD region have been analyzed for PBDEs in a recent study (Mai et al., 2005). The Σ_{10}PBDEs (BDE 28, 47, 66, 100, 99, 85, 154, 153, 138, 183) and BDE 209 concentrations in surface sediments ranged from 0.04 to 94.7 ng g^{-1} dw with an average of 9.9 ng g^{-1} dw and from 0.4 to 7340.8 ng g^{-1} dw with an average of 465 ng g^{-1} dw, respectively. In general, the concentrations of BDE 209 were one to two orders of magnitude higher than those of ΣPBDEs. The mean concentrations of ΣPBDEs decreased in the order of Dongjiang River > Zhujiang River > Macao coast > the PRE > the SCS > Xijiang River for ΣPBDEs, while those of BDE 209 decreased in the order of Dongjiang River > Zhujiang River > Macao coast > the PRE > Xijiang River > the SCS. The Σ_{10}PBDEs concentrations in most surface sediments are well within in the range of the world riverine and coastal sediments (< 50 ng g^{-1}), while the BDE 209 concentrations in Zhujiang and Dongjiang rivers were at the high end of the worldwide figures for BDE 209 in sediments. Congeneric profiles were dominated by BDE 209 (72.6–99.7%), consistent with deca-BDE as the main source mixture, followed by the congeners from the penta-BDE mixture (BDE 99: 4.34%, DE 47: 2.87%, BDE 153: 0.83%, BDE 100: 0.60%, and BDE 154: 0.58%) and some octa-BDEs (BDE 183: 0.52% and BDE 138: 0.30%).

6.3.3.4. Biota

There is limited PBDE data in literature on the levels in organisms from the PRD. Small cetacean samples collected from Hong Kong waters during 1995–2001 had PBDE concentrations ranging from 230 to 6000 ng g^{-1} lw (Ramu et al., 2005), which were apparently higher when compared to northern fur seals from the Pacific coast of Japan (Kajiwara et al., 2004). The levels observed in cetaceans from Hong Kong were comparable to harbor seals from San Francisco Bay (She et al., 2000) and harbor porpoises from British Columbia (Ikonomou et al., 2002). Congeneric profiles in cetaceans showed that BDE 47, 99, and 100 made up 90% on an average of the total PBDE load. Another study has analyzed PBDEs in green mussel samples collected from nine locations along

Hong Kong marine waters (Liu et al., 2005). The Σ_{15}PBDEs (BDE 3, 15, 28, 47, 60, 85, 99, 100, 138, 153, 154, 183, 197, 207, 209) in Hong Kong mussels ranged from 27.0 to 83.7 ng g^{-1} dw.

A recent study compared PBDE concentrations in demersal organisms from the PRE and Daya Bay (Guo et al., 2007). The PBDE concentrations in fishes from the PRE (21–170 ng g^{-1} lipid, 0.2–4.2 ng g^{-1} ww) were generally within the reported ranges in literature, while those from Daya Bay were at the low end of the worldwide figures. Relatively higher total PBDE concentrations were observed in tank gorby (*Glossogobius giuris*, 170), robust tonguefish (*Cynoglossus robustus*, 140), and thread herring (*Clupanodon thrissa*, 100 ng g^{-1} lipid) from the estuary. Among the 15 PBDEs analyzed in this study, BDE 47 was the most abundant congener, accounting for 39 and 50% of total PBDEs in fishes from the PRE and from Daya Bay, respectively. BDE 100 accounted for 10% and 14% in fishes from the estuary and Daya Bay, respectively. Despite the large quantity of BDE 209 used in Asia/China (Ueno et al., 2004) as well as its high concentrations in some of the sediments from the PRD (Mai et al., 2005), BDE 209 comprised only $<3\%$ of the total PBDEs in the fishes. This may imply that BDE 209 has a low bioaccumulation potential to fishes (Sellström et al., 1998; Boon et al., 2002).

6.3.4. PCDD/Fs

Published data on PCDD/Fs in the environment of the PRD is still limited. A short-term sampling scheme was conducted to determine the levels of particle-bound polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in Guangzhou atmosphere and evaluate the impact of PCDD/Fs to inhabitants. Samples were collected from four different districts of Guangzhou. The results showed that industrial activities had more influences on levels of PCDD/Fs in the sampled districts. The mean PCDD/Fs concentrations (i.e., mean I-TEQ values) of Huadu district, Liwan district, Tianhe district and Huangpu district were 3815–26,530 fg m^{-3} (104.6–769.3 fgI-TEQ m^{-3}). Emission source analysis indicated that small diffuse sources were relatively important (Yu et al., 2006).

Atmospheric deposition of PCDD/Fs was also investigated at four locations in Guangzhou. The annual deposition fluxes of tetra- to octa-CDD/Fs (total PCDD/Fs) were found to range from 170 to 3000 (mean 1500) pg m^{-2} day, and the fluxes of total 2,3,7,8-substituted PCDD/F congeners ranged from 2.1 to 41 (mean 20) WHO-TEQ m^{-2} day. The average deposition fluxes of total 2,3,7,8-substituted PCDD/F congeners in rainy season were found to be 27–37 pg WHO-TEQ m^{-2} day, and the PCDD/F deposition fluxes were obviously higher in rainy season than in

dry season. Results from regression analysis showed that number of rainy days, the amount of wet precipitation, PCDD/F concentrations in particles and organic carbon content played important roles in accounting for the variation of PCDD/F deposition fluxes. It was found that the profiles of PCDD/F homologues or congeners in the samples were the same either spatially or temporally, indicating that the PCDD/F emission sources were similar to one another. It was suggested that vehicle exhaust might be an important source for PCDD/Fs in Guangzhou, and that atmospheric deposition possibly tended to remove lower chlorinated DD/Fs from air and was one of sources for PCDD/Fs in soils (Ren et al., 2007).

An investigation of PCDD/Fs in soils across the PRD was conducted by the same research group. A total of 61 samples including 58 soils and 3 sediments were analyzed for PCDD/PCDFs with the isotope dilution method using a high resolution gas chromatograph/high resolution mass spectrometer (HRGC/HRMS). The results showed that the total concentrations of PCDD/PCDFs were 85.5–14,525 pg g^{-1} dw, and the concentrations of 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin (TCDD) equivalents (I-TEQ) were 0.09–19.7 pg g^{-1} dw. According to the German and the USA guidelines for I-TEQ concentrations, 14 in the 58 samples contained TEQs between 5 and 40 pg g^{-1} dw, and may be a cause for concern. The relative higher total concentrations and I-TEQ of PCDD/DFs in selected samples indicated the presence of potential point-source pollution. The homologue profile of PCDD/DFs and the spatial distribution suggested that the sources of PCDD/DFs were mixed in soils, but the uses of HCH and PCP were possibly the main sources in the samples that contained high concentration of PCDD/DFs. PCDD/DFs were also detected in soils from three mountain tops, suggesting wide dispersal of PCDD/DFs in this region via dry and wet deposition (Zhang, 2006).

6.4. Human exposure to POPs

POPs in human breast milk may directly reflect human exposure to POPs in the environment. A survey was conducted to examine *p*,*p*′-DDT, *p*,*p*′-DDE, β-HCH, and PCB concentrations in human breast milk, one of the most reliable bioaccumulation indicators. Milk samples (115 from Hong Kong and 54 from Guangzhou), in the lactation period from 3 to 5 weeks were analyzed. The results demonstrated that the mean levels of *p*,*p*′-DDT (Hong Kong: 0.39; Guangzhou: 0.70 μg g^{-1} of fat), *p*,*p*′-DDE (2.48; 2.85), and β-HCH (0.95; 1.11) were 2–15-fold higher when compared with studies conducted elsewhere (i.e., United Kingdom, Germany,

Sweden, Spain, and Canada). In contrast the concentration of PCBs (0.035; 0.031) was ~10 times lower. When compared to a similar study conducted 10 years ago in Hong Kong (*p*,*p*′-DDT: 2.17 μg g^{-1} of fat, *p*,*p*′-DDE: 11.67, β-HCH: 15.96, and PCB: 0.64), a considerable reduction in the levels of these contaminants was observed (Wong et al., 2002).

A recent study investigated PBDEs in 21 paired maternal and fetal blood, and 27 breast milk, with most of the samples collected in Guangzhou (Bi et al., 2006). The concentrations of total PBDEs ranged from 1.5 to 17 ng g^{-1} in the samples, and were within the range reported in European samples for a similar population, but lower than the human tissue levels in North America. BDE 47 and 153 were the dominant PBDE congeners in all samples and accounted for 60% of the total PBDEs.

6.5. Regional cycling and time trends of POPs

Natural archives of POPs, such as sediment cores, serve as an important source to retrieve the past POP contamination. Contrary to the decreasing trends observed in studies in many developed countries, OCPs, PCBs, and PAHs all displayed surface/sub-surface layer enrichment along the sediment cores in the PRD. The DDT and HCH concentrations increased sharply in the sediment layers deposited in the early 1990s, which correspond to the sharp cultivated land loss as a result of fast urbanization in the region, implying that soil runoff enhanced by the large scale land transformation may have mobilized these pesticides from the soil to the water environment (Zhang et al., 1999, 2002). The PCB concentrations in the sediment cores also displayed an up-profile increasing trend in relation to the regional GDP and cultivated land area loss during the same period when fast development of regional economy took place (Mai et al., 2003). The up-profile increase in PAH concentrations was also concurrent with the development of power generation, vehicle numbers in the PRD (Liu et al., 2005). Though most POPs trapped in the sediment may be immobilized, vertical migration of lighter PCBs with 3–4 chlorine atoms was observed in two cores (Mai et al., 2003). The POP pollution history reconstructed by sediment cores reflected that the intensive anthropogenic activities, typically during the last two decades in the PRD, which may have a strong impact on the sources and biogeochemical cycling of POPs in the region.

6.6. Conclusion remarks

Compared to other regions in China, the environmental occurrence and fate of POPs in the PRD have been more extensively characterized. In

view of the potential negative impact on human health, DDTs in the air and wild fishes in the region occur in very high concentrations, and should be taken as the first priority POP in the context of current source reduction. In particular, the DDT-containing dicofol application and antifouling paints for fishing ships should be paid a great attention. Secondly, PCDD/Fs were found at high levels in the urban environment. It is suggested that monitoring and assessment measurements should be taken urgently to enable a sound management of the MSW incinerators, the number of which is sharply increasing in the region. Thirdly, for PCBs and PBDEs, although they were generally within the concentration levels around the world and less important at a regional scale, distinctively high concentrations were found in several e-waste recycling sites. This calls for a better management and best applicable technology to be applied in the e-waste recycling industry, as well as characterization and remediation of relevant potential contaminated sites. Fourthly, a sound management/control/reduction of POPs at a regional scale cannot be made without a good understanding of the regional inventories of POPs, on which the knowledge and information is still very limited in the PRD. A regional collaboration between Guangdong province and Hong Kong is of great importance.

It is worth pointing out that, although high concentrations of DDT and PCDD/Fs were evident in the atmosphere, their concentrations in other abiotic compartments, i.e., soil, sediment and water, were generally comparable to other regions around the world. This is likely to be resulted from the high temperature in the tropical-subtropical region, and may generally be applicable to other low-latitude regions. It is thus suggested that atmosphere and aquatic organisms might be the priority environmental media for regional monitoring of POPs in the PRD, to elucidate their regional sources and future time trends.

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