A spatial temporal assessment of pollution from PCBs in China

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Abstract

This research represents an assessment of the current state of pollution from polychlorinated biphenyls (PCBs) in China. Various environmental media including sediment, water, organism and soil were analyzed and the spatial character of PCB pollution in each environmental medium was determined. On a national basis, PCB levels in all environmental media were relatively low, with little evidence of major contamination in China's main regions and rivers. However, there were some locations with high PCB concentrations. Sediments in Pearl River (83.1 ng/g) and its estuary (58.9 ng/g), Dalian Bay (58.1 ng/g) and Songhua River (36.8 ng/g) had relatively high level of PCBs. There were also some areas, which were mainly the industrial pollution sites or PCB equipment storage locations, remained seriously polluted with the highest PCB residue level of 150000 ng/g. In addition, Minjiang Estuary (985.2 ng/g) and Taihu Lake (631 ng/g) had high levels of water pollution, while Pearl Estuary (635.7 ng/g) and Jiaozhou Bay (273.3 ng/g) had relatively high PCB levels in organisms. PCB pollution in soil was limited to a few special pollution areas with the highest PCB level of 4.5448×10^6 ng/g. Point source pollution was the common pattern of contamination, influenced primarily by local geographic, economic and historical factors. Analysis of PCB concentrations from the 1980s to 1990s shows an increasing trend, possibly due to the improper disposal of and leakage from PCB containers, chemical transfers, and the general rise of industrial pollution.

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1. Introduction

Polychlorinated biphenyls (PCBs), as persistent organic pollutants (POPs), have been extensively studied during the last 30 years in view of their extensive pro-

duction and usage, their long-range transport capability within the environment, and their bioaccumulation, persistence, and impact on both ecosystems and human health. Because of increasing concern for the risks posed by their use, they were listed along with eleven other compounds as serious threats to health and the environment by the Stockholm Convention (May 22, 2001).

Prior to 1974, PCBs generally entered the air, soil, water and sediment during their manufacture and use (Wong and Poon, 2003). In China, approximately

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10000 tons of PCBs were produced from 1965 to 1974 (production of PCBs was banned in 1974), with 9000 tons as trichlorobiphenyl and 1000 tons as pentachlorobiphenyl. Trichlorobiphenyl was used primarily in power capacitors applied in electricity production, distribution and transmission, while pentachlorobiphenyl was used mainly as a paint additive (China SEPA, 2003). Following the ban on the production and use of PCBs, most of the outdated PCB-containing equipment (equipment filled with PCBs as dielectric fluid) was removed from use and stored. Unfortunately, some of that equipment has recently been found leaking into the surrounding environment and as such, continues to pose a threat to health and the environment. Other sources of selective PCB pollution stem from the illegal dismantling and/or disposal of PCB-containing equipment, the unwanted by-products from the combustion of chlorine-containing waste through unprotected incineration systems and certain other chemical processes involving organochlorines.

While implementation of PCB controls became an important task in China following ratification of the Stockholm Convention, there was only limited information at the national level regarding the current status of PCB pollution and emission sources. The number of studies about PCBs in China has increased recently in light of a series of global initiatives on POPs and an increasing concern about the risks POPs pose to human health and the environment. Among this growing body

of existing research, several have focused on monitoring PCBs within the environment. For instance, Chen et al. (1999), Wu et al. (1999) and Yuan et al. (2001b) investigated PCB levels in sediments from Chinese river/estuary systems, and PCB contaminations in various water organisms such as mussels was surveyed by Monirith et al. (2003) and Fung et al. (2004). While these studies have advanced our understanding of the nature of the problem posed by PCBs, a significant gap still exists in our knowledge of the extent and spatial character of the PCB situation in China. This study aims to assess the temporal and spatial distributions of PCB levels presented in existing data as a preliminary assessment of the extent and distribution of PCBs in China.

2. Assessment of PCB pollution in the environment

Most of the research about environmental contamination from PCBs in China has focused on the eastern part of the country, especially estuarial areas such as those of the Pearl and Yangtze rivers, Bohai Bay on the east coast in north China, and along other major rivers such as Minjiang, Jiulongjiang, Yangtze, and Songhua. The spatial distribution of sampling sites in China along with the relevant quantities (percentages) of PCBs deployed in the different areas is shown in Figs. 1 and 2 (China SEPA, 2003). 45.2% of all PCBs used in China were used in the east, with an average gross application

Fig. 1. Sampling locations of PCBs in sediment, organism and water in China. A: Heilongjiang River; B: Nenjiang River; C: Songhua River; D: Yalujiang Estuary; E: Dalian Bay; F: Bohai Sea; G: Yellow River; H: Jiaozhou Bay; I: Huaihe River; J: Lusi Coast; K: Hanshui River; L: Yangtze River; M: Taihu Lake; N: Yangtze Estuary; O: Dongting Lake; P: Ningbo Coast; Q: Zhoushan Coast; R: Minjiang River; S: Jiulongjiang River; T: Pearl River; U: Daya Bay; V: Najiabawa Peak, Tibet.

Fig. 2. Sampling locations of PCBs in sediment, organism and water around the Bohai Sea area.

density of 2.86 kg/km². After that, 35.7% were used in the middle section of the country with an average gross application density of 1.29 kg/km², while 19.1% were used in west China with an average gross density of 0.36 kg/km². This is not surprising since the vast majority of China's industrial development has occurred in the east and east central portions of the country, especially along major rivers and coastal areas where industrial

concentration is highest and manufactured goods could be easily shipped to their final destination.

2.1. PCBs in sediments

Average PCB concentrations in sediments collected from the literature are presented in Fig. 3. If different workers investigated the same area, the average level was selected or calculated for presentation in Fig. 3.

Because there is not an environmental standard for the concentration of PCBs in sediments in China, we used a criterion presented by Long et al. (1995) for appraisal purposes. Long et al. (1995) developed two guidelines for use as comparative values, an effects range-low (ERL) standard and an effects range-median (ERM) standard, for assessing a number of trace metals and organic compounds including PCBs in the environment. The ERL and ERM values were intended to define chemical concentration ranges that are rarely, occasionally, or frequently associated with adverse biological effects including altered benthic communities (depressed species richness or total abundance), histopathological disorders in demersal fish, EC₅₀ or LC₅₀ concentrations and toxicity predicted by models. The ERL and ERM guideline values for PCBs adopted here were set at 22.7 ng/g and 180 ng/g, respectively.

Fig. 3. Average PCB levels in sediments at various sites in China (ng/g dry wt.). Heilongjiang River (Chen et al., 1999), Nenjiang River (Chen et al., 1999), Songhua River (Li et al., 1989; Liu et al., 1998; Chen et al., 1999), Daliaohe River (Wu et al., 1999), Luanhe River (Wu et al., 1999), Haihe River (Wu et al., 1999), Yongding River (Ma et al., 2001), Tonghuihe River (Zhang et al., 2004), Yellow River (Chen et al., 1999), Huaihe River (Chen et al., 1999; Wang et al., 2001), Yangtze River (Chen et al., 1999; Jiang et al., 2000), Qiantangjiang River (Chen et al., 1999; Wu et al., 1999), Huangpujiang River (Wu et al., 1999), Hanshui River (Chen et al., 1999), Minjiang River (Chen et al., 1999), Pearl River (Chen et al., 1999; Kang et al., 2000; Nie et al., 2001; Mai et al., 2002), Beijiang River (Chen et al., 1999), Yellow Estuary (Wu et al., 1999), Yangtze Estuary (Wu et al., 1999; Yang et al., 2002; Chen et al., 2003; Liu et al., 2003; Yang et al., 2003), Site 1 (Chu et al., 1995b), Minjiang Estuary (Yuan et al., 2001a; Zhang et al., 2002, 2003), Jiulongjiang Estuary (Hong et al., 1995; Chen et al., 1996; Wu et al., 1999; Zhou et al., 2000; Yuan et al., 2001a,b), Pearl River Estuary (Hong et al., 1995, 1999; Kang et al., 2000; Yuan et al., 2001b), Baoding (China SEPA, 2003), Dalian Bay (Li et al., 1998; Liu et al., 2001), Jinzhou Bay (Li et al., 1998), Daya Bay (Zhou et al., 2001), Ya-Er Lake (Wu et al., 1997), Dongting Lake (Chen et al., 1999), Site 2 (China SEPA, 2003), Site 3 (China SEPA, 2003).

Among the 31 sites identified in Fig. 3, there were some heavily polluted areas. Site 1 (a PCB pollution area on the southeast coast), an industrial drainage canal used by paper mills in Baoding, Ya-Er Lake near Wuhan which had been polluted by industrial emission 20 years before, Site 2 (a river near a PCB equipment storage location) and Site 3 (another river passing adjacent to a PCB equipment storage facility) had constituents frequently posing biological impairment with concentrations much greater than ERM. Six sites (Nenjiang and Songhua rivers in northeast China, Dalian Bay, and Minjiang River, Pearl River and Pearl Estuary in south China) posed possible adverse biological effects, with average concentrations (25 ng/g, 36.8 ng/g, 58.1 ng/g, 25 ng/g, 83.1 ng/g, and 58.9 ng/g) greater than the ERL limits recommended by Long et al. (1995). Although these locations are important industrial and agricultural areas, none of the samples suggested a serious biological threat, with PCB concentrations less than the ERM standard. Samples taken from Heilongjiang River (northeast China), Yellow and Huaihe rivers, Minjiang Estuary, and some branches of Yangtze River (Qiantangjiang, Huangpujiang, Hanshui rivers and Dongting Lake) in middle China had average PCB concentrations near the ERL level, suggesting that necessary concern should be given to these locations. Other areas such as the Luanhe, Yongding and Tonghuihe rivers near Beijing, the Daliaohe and Haihe rivers and Jinzhou Bay near Bohai Sea and Yellow River Estuary had relatively low levels of contamination.

Regionally, the distribution of PCB pollution tends to be related to drainage area, industrial patterns, and pollution emissions during the two decades since their production and use was banned. PCB sampling locations and concentrations in sediments along Pearl River are presented in Figs. 4 and 5. PCB levels from Fangcun (485.45 ng/g) to Huangpu at Guangzhou City were probably higher than those in the rural sector (Shiziyang River and Lingding Bay) because of greater levels of industrial emission and municipal sewage as potential sources. Concentrations in sediments in old industrial parks (Zhichang, Yuancun and Huangpu of Guangzhou sector) were also higher than those at the new industrial park or in adjacent residential areas. Along the Guangzhou branch of the Pearl and Shiziyang rivers and Lingding Bay, PCB contamination decreased as the distance to the source increased. Pollution in the estuary was very low (1.58 ng/g) possibly owing to the influx of large amounts of water from other branches of the river in this area. Also heavily polluted were harbor areas such as Macao and Victoria Harbor 3, as the result of both industrial sources (e.g., a landfill near the Macao's inner Harbor) and hydrological conditions (e.g., a typhoon shelter at Victoria Harbor 3). Fu et al. (2003) also reported that there were two different PCB pollution sources in the Pearl River/Estuary according to the

Fig. 4. Sampling locations of PCBs in sediments in Pearl River Estuary.

Fig. 5. Average PCB levels in sediments in Pearl River/Estuary (ng/g dry wt.). Fangcun, Xijiang River, Shiziyang River, Lingding Bay, Macao Harbor (Kang et al., 2000); Yagang, Xicun, Zhichang, Donghaoyong, Yuancun, Huangpu (Nie et al., 2001); Estuary (Hong et al., 1999; Yuan et al., 2001b); Victoria Harbor 1, 2, 3 (Hong et al., 1995).

PCB distribution profile, one from Guangzhou and another from Hong Kong. PCB pollution in the Pearl River Delta sediments seemed to be the result of trace discharges into storm waters, sewage, industrial waste, agricultural runoff, and some atmosphere deposition.

2.2. PCB pollution in water

The available surveys about PCBs in water were very deficient and the limited data on PCB concentrations are given in Table 1. According to the USEPA guidelines, the concentration of PCBs should be less than 14 ng/l

Table 1
Concentrations of PCBs in water from various sites in China (ng/l)

Location	Concentration range	Average concentration	Reference
Songhua River	0.003–0.088	0.013	Li et al. (1989)
Tonghui River	31.58–344.9	105.5	Zhang et al. (2004)
Jiaozhou Bay	9.01–12.63	10.82	Chu et al. (1995a)
Huaihe River	0.24–0.28	0.26	Wang et al. (1999)
Yangtze River	0.29–2.00	1.24	Chen et al. (1996); Sun et al. (2002)
Taihu Lake		631	Wang et al. (2003)
Minjiang Estuary	203.9–2473	985.2	Zhang et al. (2003)
Jiulongjiang Estuary	0.1–1505	177.9	Zhang et al. (2000); Zhou et al. (2000)
Pearl Estuary	1.13–3.11	2.3	Nie et al. (2002)
Daya Bay	91.1–1355.3	313.6	Zhou et al. (2001)
Dong Lake, Wuhan		2.7	Xi et al. (1998)
Nanjiabawa Peak, Tibet		0.0093	Sun et al. (1986)

for surface water to be considered no hazard to aquatic or human health (USEPA, 2002). Table 1 identifies those locations with low-levels of PCBs in water. At Nanjiabawa Peak in Tibet, for example, where there are no industrial emissions, the concentration level was only 0.0093 ng/g. Although the contamination level was extremely low in this area, the trace amounts present suggest the possibility that atmospheric transfer may be a working pathway for PCBs to enter the area (Sun et al., 1986). High upper limit concentrations of PCBs were found in the Minjiang and Jiulongjiang estuaries (2473 ng/g and 1505 ng/g, respectively), Daya Bay

(1355.3 ng/g) and Tonghui River (344.9 ng/g). Taihu Lake, an important industrial and agricultural area in middle China, also had a high PCB average level at 631 ng/l (Fig. 1 and Table 1).

2.3. PCB pollution in organisms

PCB concentrations in organism are presented in Table 2. For the levels of same areas investigated by different workers, the average levels were calculated and then listed. Some dry weight values were multiplied by 10 and presented as wet weight according to an

Table 2
Concentrations of PCBs in organisms from various sites in China (ng/g wet wt.)

Location	Concentration range	Average concentration	Reference
Yalujiang Estuary		3.8	Liu et al. (1996)
Daliaohe Estuary		0.4	Liu et al. (1996)
Bohai Bay		5.4	Liu et al. (1996)
Jinzhou Bay		1.4	Liu et al. (1996)
Dalian Bay	4.6–120	44.3	Liu et al. (1996)
			Yu et al. (2002)
			Fung et al. (2004)
Jiaozhou Bay	6.6–540	273.3	Liu et al. (1996)
			Fung et al. (2004)
Lusi Coast		3.2	Liu et al. (1996)
Yangtze Estuary		60.69	Fung et al. (2004)
Zhoushan Coast		47.65	Fung et al. (2004)
Ningbo Coast		41.13	Fung et al. (2004)
Minjiang Estuary	2.82–69	30.6	Chen et al. (2001)
			Fung et al. (2004)
Jiulongjiang Estuary	37.1–270	120.7	Chen et al. (2001)
			Fung et al. (2004)
Pearl Estuary	25–2480	635.7	Fang et al. (2001)
			Monirith et al. (2003)
			Sun et al. (2003)
Pearl River	130.1–288.5	181.9	Nie et al. (2001)
Songhua River ^a	18.6–161.7	61.3	Li et al. (1989)
Huaihe River ^a	4.65–7.27	5.96	Wang et al. (2001)

^a The samples in those locations were fish, while samples in other locations were mussels.

experiential relationship suggested by Sun et al. (2003). The Environmental Chemical Contaminants Control Guidance set up by the United States Food and Drug Administration (USFDA) provided a federal tolerance of PCBs (2000 ng/g wet wt.), which was used as a criterion to assess the PCB contamination status in organisms including fish and fisheries products such as fresh or saltwater finfish, crustaceans, other forms of aquatic animal life and all mollusks (USFDA, 2001).

Most areas had relatively low concentrations (less than USFDA standard of 2000 ng/g) of PCBs in mussels or fish. Areas in the north (Yalujiang Estuary and some locations around the Bohai Sea namely Daliaohe Estuary, Bohai Bay, Jinzhou Bay, and Dalian Bay) were slightly less contaminated than the sites in the south, especially the Jiulongjiang and Pearl estuaries and Jiaozhou Bay where industrial and wastewater throughput is relatively large.

Based on evidence collected from crustacean tissue samples, some areas along the southeast coast had higher levels (691 ng/g) than found just 2 years prior (22.6 ng/g) or at a nearby island (0.88 ng/g, Chu et al., 1995a), indicating that PCB contamination from point sources remains an issue in the region.

2.4. PCB pollution in soil

Few of the studies surveyed PCB contamination in the soil, and when undertaken, the focus was on already heavily polluted areas. In China, most of the PCB-containing equipment was stored at special sites when they were taken out of use. Because of poor management and storage conditions, however, PCBs from some of these special storage locations have leaked into the surrounding media especially the soil. China SEPA (2003) reported that directly polluted soils in some sealed storage locations reached as high as 4.5448×10^6 ng/g, with levels in surrounding lands and farmlands having as much as 2930 ng/g.

Along China's southeast coast where some residents were illegally trading in or dismantling PCB-containing equipment, PCB concentrations have been found as high as 788 ng/g dry wt. (Chu et al., 1995b). In the Wentai Area, for example, the arbitrary disposal of used electric appliances resulted in a major PCB pollution accident in 1989, with PCB levels reaching 1101.4 ng/g in an adjacent rice field in 1993. Following the ban on such activity, pollution levels decreased to 568.15–920.08 ng/g in 1997 and by 1999 were down to 2.01–7.34 ng/g (Bi et al., 2002).

Agricultural soils sampled in suburban Beijing reported concentration levels of only 0.18 ng/g (Chu et al., 1995b) and soils sampled from Shenyang, a major industrial city in north China's Liaoning Province, had PCB levels of 6.4–15.2 ng/g (Jing et al., 1992). Compared to the allowable level of 60 ng/g for PCBs in ambient soil

approved by the former Union of Soviet Socialist Republics (USSR) Ministry of Health in 1991, both areas had relatively slight PCB contamination levels on average (Bobovnikova et al., 1993; Bi et al., 2002). While the broad average pollution situation seems to be in control, the threat posed by seriously polluted point sources remains real and should not be overlooked in assessing the risks identified by current data.

3. Temporal PCB pollution in China

Monitoring of PCBs in the environment has not been extensive, with inadequate funding and little policy support adding to the difficulties in China. In addition, few of the places were investigated more than once. Data collected over time for some of the main rivers and estuaries are presented in Fig. 6. The results indicate that concentrations of PCBs in most areas of China increased during the 1980s and 1990s, with the exception of Yangtze River and Dalian Bay. This variation in environmental PCB levels is in contrast to results presented in most foreign studies where concentrations of PCBs have been decreasing since the 1970s (Giesy and Snyder, 1998; Muir and Norstrom, 2000; Marvin et al., 2004).

There are two possible reasons behind this difference. In China, the production of PCB capacitors was greatest between 1965 and 1974 and China also imported large quantities of PCB transformers in the 1970s and 1980s accompanied by other large size/scale purchases of integrated mechanical and electrical equipment (China SEPA, 2003). If the calculated service life of 15–20 years for such equipment is taken into consideration, they would be scheduled for removal from service during

Fig. 6. Temporal trend of PCB levels in sediments in some main rivers of China. Jiulongjiang Estuary (Hong et al., 1995; Chen et al., 1996; Wu et al., 1999; Yuan et al., 2001b), Minjiang Estuary (Yuan et al., 2001a,b; Zhang et al., 2002, 2003), Yellow River (Chen et al., 1999; Wu et al., 1999), Qiantangjiang River (Chen et al., 1999; Wu et al., 1999), Songhua River (Li et al., 1989; Liu et al., 1998; Chen et al., 1999), Pearl River (Chen et al., 1999; Kang et al., 2000; Nie et al., 2001), Yangtze River (Chen et al., 1999; Jiang et al., 2000) and Dalian Bay (Li et al., 1998; Liu et al., 2001).

the 1980s and 1990s. Further, because of excessive lags in management, lack of knowledge about the negative effects of PCBs, and poorly developed institutional support, most of the PCB-containing equipment was disposed improperly such as through uncontrolled combustion or storage under bad conditions, which caused the gradual emission of PCBs into environment in 1980–1990s. In addition, the Chinese economy was developing rapidly during this same period, and dramatic increases in pollution emissions accompanied the booming economy. As a by-product, PCBs could have entered the surrounding environment through multiple pathways. Given the trend observed during the 1980s and 1990s, it is likely that some areas in China will continue to experience increases in PCB levels in the future if controlling measures are not carried out.

4. Conclusion

Based on very general environmental criteria PCB concentrations in environmental media in China were relatively low on a national scale. That may be partly because PCBs were not produced in China with any great abundance relatively speaking, and they were used for a relatively short time. Based on existing information, there was little contrast in PCB contamination among the main regions, rivers, and estuaries investigated. On a regional scale, however, there were some significantly polluted areas and historic use sites causing serious local pollution with possible long-term affects. PCB residues in the sediments of some rivers and estuaries in China's northeast and southern regions were more seriously polluted (Pearl River/Estuary system, Songhua River, Ya-Er Lake and Dalian Bay) than other areas. Waters in the Minjiang Estuary and Taihu Lake in southeast China were extremely polluted, and Jiaozhou Bay and Pearl Estuarine had relatively high PCB level in organisms. Soils polluted by PCBs were found near special point sources reflecting problems associated with the improper dismantling and storage of old PCB-containing equipment. Local geographic, economic and historical factors also had a noticeable effect on the incidence and diffusion of pollution resulting from PCBs. In addition to the rise of pollution emissions from rapid economic development, PCB concentrations in most areas of China increased during the 1980s and 1990s due to improper disposal as well as leakage from PCB-containing equipment.

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