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Chapter 4

Polychlorinated Dibenzo-*p*-Dioxins, Dibenzofurans, and Biphenyls, and Polybrominated Diphenyl Ethers in China☆

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Abstract

China was one of the first proponents of the Stockholm Convention, but only recently have we started to prepare the national inventory of PCDD/Fs and PCBs, which are among POPs specified by the convention. PBDEs have been extensively used as flame retardants in various products, and recently, the environmental problems associated with these compounds have become great concern. Data about the pollution status of these pollutants were extremely scarce in China due to the lack of regulations and monitoring capacity. In this chapter, sources of PCDD/Fs, PCBs, and PBDEs and their levels in the environmental media in China are summarized, based mainly on available scientific literature. The challenges for management of these compounds are also discussed.

4.1. Introduction

As polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) are compounds with similar structures and monitoring methods, they are discussed together in this chapter. The structures of PCDDs, PCDFs, PCBs, and PBDEs are shown in Fig. 4.1.

PCDDs and PCDFs (PCDD/Fs) are often just called "dioxins". The number of chlorine atoms in each molecule can vary from 1 to 8, and there are 75 and 135 congeners for PCDDs and PCDFs, respectively. The congeners with chlorine substitutions in the 2, 3, 7, and 8 positions are thought to have dioxin-like toxicity. PCDD/Fs are formed as unwanted

☆Taiwan, Hong Kong and Pearl River Delta are not included this chapter.

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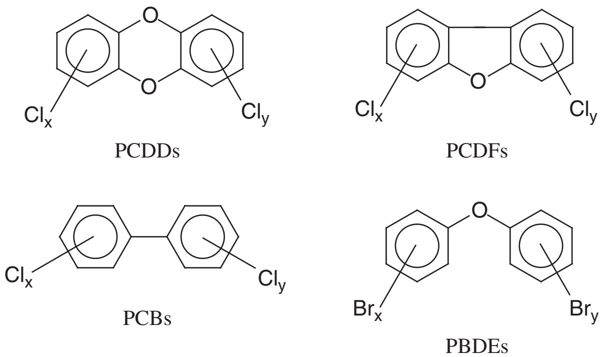

Figure 4.1. Chemical structures of PCDD/Fs, PCBs, and PBDEs.

by-products in many combustion and industrial processes. They have also been formed from natural processes, such as forest fires and volcanoes, but these formations are in general of less importance compared to the anthropogenic emissions. The major sources of environmental contamination with PCDD/Fs can be cataloged as: waste incineration, chemical industry, and pulp and paper industry.

There are 209 PCB congeners, differentiated by the numbers and positions of the substituted chlorine atoms. The 12 PCB congeners with 4 or more chlorines with just 1 or no substitution in the ortho position are thought to have dioxin-like toxicity and are often called "dioxin-like PCBs". PCBs have been used commercially since 1929 as dielectric and heat exchange fluids, as well as in a variety of other applications. The total amount of PCBs produced worldwide has been estimated to be 1.5 million metric tons (t). The majority of the PCBs in the environment are thought to be the result of leaks from PCB-containing electrical capacitors and transformers. Waste incineration is also a potential air emission source of PCBs.

PBDEs have been called one of the "emerging pollutants". They are extensively used as flame retardants (FRs) in various polymers and especially in electronic equipment such as computers and television sets. Similar to PCBs, there are 209 congeners of PBDEs, and the nomenclature system is also based on the same IUPAC scheme used for PCBs. But unlike PCBs, for which large-scale production has been banned for many years, PBDEs are still widely used and their transport and transformation in the environment are still poorly understood. The global demand for PBDEs has increased rapidly since the 1970s. In 1992, the global production of PBDEs was ~40,000 t, but in 1999 this had increased to approximately 70,000 t (Renner, 2000).

PCDD/Fs, PCBs, and PBDEs are usually classified as groups of persistent organic pollutants (POPs). They have been found in almost all compartments of the global ecosystem in at least trace amounts. They can be transported over long distances in the atmosphere, resulting in widespread distribution across the Earth, including regions where they have never been used. PCDD/Fs and PCBs were specified as POPs in the Stockholm Convention. China was one of the first proponents of the convention and ratified it on June 25, 2004. However, due to a lack of monitoring capacity and regulation, these pollutants have not yet been regularly monitored in China. Although PBDEs are categorized as emerging POPs, few scientific papers have reported their occurrence in China. Therefore, only very limited knowledge on the pollution of PCDD/Fs, PCBs, and PBDEs in China is available.

4.2. Sources of PCDD/Fs in China

According to a number of pilot surveys of dioxins carried out by some Chinese institutions, dioxins have a broad distribution in China. Based on the available data, the major sources of PCDD/Fs in China are discussed below.

4.2.1. Solid waste incineration

With the explosive development of the economy and urbanization in China, more and more municipal solid wastes (MSWs) are generated. In 2000, 150 million tons of MSW were generated in China, and the total MSW dumped was 6 billion tons (Zhao, 2004). The MSW is mainly disposed of by three processes in China: landfill, MSW incineration (MSWI), and compost. Whereas solid waste incinerators (SWI), especially MSW incinerators, have proven to be the most significant PCDD/F generators in industrialized countries, SWI are presently not considered as the biggest source in China because only a small portion of MSW are currently being disposed of by incineration. However, numerous MSW incinerators are under construction in various cities to save limited land space. The emission of dioxins will increase rapidly from the boom of MSWI in China.

The MSWI furnaces in China can be divided into four major types: stack furnaces, fluid-bed furnaces, rotary kiln furnaces, and gasification. Incinerators with the largest capacity are mostly stack furnaces. Monitoring data on dioxins emitted from MSWI in China are extremely scarce because there is no regular monitoring requirement. A project funded by

United Nation Environmental Program (UNEP) to investigate PCDD/Fs in some Chinese industries and processes is presently being carried out, but the results are not yet available.

A preliminary investigation on dioxin emission from MSWI in China has been campaigned by Tian and Ouyang (2003). Flue gas of 15 different types of MSW incinerators was monitored. About half of the data exceeded the national standard for dioxin emissions limit (1 ng TEQ m^{-3}; National Standard of the People's Republic of China, 2001) and the highest was at 100 ng TEQ m^{-3} level. However, this report did not provide precise concentrations. Total dioxin emissions to air from MSWI was estimated to be 72 ng TEQ annually in China based on the monitoring data of Tian and Ouyang (2003). Stack furnaces (with bag filters for dust removal) are the major type of MSW incinerators in China but not the major contributor to dioxin emissions. Fluid-bed furnaces accounted for ~60% of the dioxin emissions from MSWI.

Dioxins in fly ash of stack MSW incinerators from three cities were investigated by Jin et al. (2003b) and the concentrations were 7.53, 1.52, and 0.44 ng TEQ g^{-1}, respectively. The concentration of dioxins in flue gas will decrease if active carbon powder is used before the bag house equipment, but the concentration of dioxins in the fly ash will increase accordingly.

4.2.2. Chemical industry

PCDD/Fs can be formed during the manufacture of chlorophenols, chlorobenzenes, and chlorobiphenyls, and the production and use of them could be important sources. Some researchers believe that the manufacture of halogenated organic chemicals is the largest contributor of PCDD/Fs in China (Huang et al., 2001). While some specified sources have been closed in developed countries, the release of PCDD/Fs from chemical manufacturing is still a problem in China due to the use of dated processes or technologies, despite their limited use. These chemical manufactures are described below.

4.2.2.1. Chloralkali industry

Rapp et al. (1991) found that the total concentration of PCDD/Fs in a Swedish graphite electrolytical sludge was 650 ng g^{-1}. The use of graphite electrodes has been banned in developed countries because of the associated high pollution levels. The chloralkali industry is the fundamental raw material industry in China. Among the 500 chloralkali manufactures all over the world, China has 230 of them and ranks second in the world behind America. In these plants, it is common to use graphite electrode as the anode and ferric net covered with asbestos as the cathode. This production technique may be a significant source of PCDD/Fs in China. Graphite electrode sludge collected from a huge chloralkali plant in central China was analyzed, and the concentrations of total PCDD/Fs and I-TEQ were 379 ng g^{-1} and 21.7 ng g^{-1}, respectively (Xu et al., 2000). The congener pattern in the electrode sludge corresponded very well to the typical "chloralkali pattern", and it indicated the same formation mechanism in the manufacturing process. The annual dioxin emissions from graphite electrode sludge in China were estimated to be 5.4 kg I-TEQ based on the 254,000 t sludge generated annually. Some of the sludge can enter the environment through effluent or runoff and seriously pollute the surroundings.

4.2.2.2. Chlorophenols

Chlorophenols have been widely used for a variety of pesticidal applications. Chlorophenols and their sodium salts have been primarily used for wood preservation and as chemical intermediates in the manufacture of other pesticides or chemicals. In China, sodium pentachlorophenate (Na-PCP) was especially used to eradicate snails, *Oncomelania hupensis*, in the schistosomiasis area in China. Chinese commercial PCP or Na-PCP contained appreciable amounts of PCDD/Fs, and their concentrations in PCP and Na-PCP were 142 and 92 ng I-TEQ g^{-1}, respectively (Bao et al., 1995).

4.2.2.3. Hexachlorocyclohexanes (HCHs)

γ-HCH or lindane is used as an insecticide on fruit and vegetable crops. Global technical HCH and lindane usage was estimated to be as high as 6,000,000 t in 1998 (Willett et al., 1998). Because of the environmental and biological persistence of HCHs, their use has been regulated. China banned technical HCH in 1983, but the use of lindane is not completely prohibited in China because there are exemptions that HCH can still be used as pesticides for forests and to control plagues of pests. The annual production of HCH was $\sim$4000 t before 1983, and the amount decreased to $\sim$500 t in recent years. In China, the most common method to produce trichlorobenzene (TCB) is pyrolysis of HCH wastes. Solid wastes from the production of TCB contained extremely high level of PCDD/Fs (Bao et al., 1994). The average concentrations of total PCDD/Fs and TEQ were 129 mg g^{-1} and 768 μg g^{-1}, respectively. The TEQ generated from this source were estimated to be 90 kg annually based on the quantity of HCH production. Although HCH has been banned for many years, large quantities exist in the environment and pose potential threats to human and environmental health.

4.2.2.4. PCBs

PCDD/Fs can be found as contaminants in commercial PCB products and may contribute more to the toxicity of the mixture due to their much higher TEFs than PCBs. PCBs were once produced in China until they were banned in 1974. TEQs of PCDD/Fs in two Chinese commercial PCBs were 217 and 417 ng g^{-1}, respectively (Li and Jiang, 1995). Many transformers containing PCBs as dielectric fluids were discarded, but they were not registered properly. In some metal recovery sites, leaks and spills from illegal disposal of PCBs in the 1980s during the dismantling of transformers have seriously polluted the environment.

4.2.2.5. Chloranil and dye

While chloranil is an important raw material in the dye industry, it is considered to be a source of PCDD/Fs (Christmann et al., 1989). Levels of OCDD/F between 1 μg g^{-1} and 100 μg g^{-1} were detected in chloranils and C. I. Violet 23 dye, which was synthesized from chloranil (Zhang et al., 2000a). Chlorophenol proved to be the real source of PCDD/Fs in the chloranils because technical PCP or trichlorophenol (TCP) was used as raw material. After substituting PCP and TCP with hydroquinone as an intermediate, the PCDD/Fs in chloranils were dramatically reduced (Zhang et al., 2002a).

4.2.3. Chlorine bleaching pulp

Traces of PCDD/Fs can be generated as by-products during the bleaching of pulp with chlorine. Emission from the pulp-bleaching process was characterized by US EPA as the third largest source of dioxins in the United States. Non-wood plant fibers such as cereal, rice, and reed are extensively used as raw material for paper production in China. Much higher concentrations of PCDD/Fs than from wood pulping in European or American pulp mills were reported from Chinese pulp and paper mills that bleach non-wood fibers. The typical bleaching sequence in China includes three stages: C (chlorine), E (alkaline extraction), and H (hypochlorite). Stage C has been identified as the major PCDD/F formation process. The PCDD/Fs in different stages were investigated in a paper mill using non-wood plant fibers as raw material in southern China (Zhang et al., 2000b). The concentration of PCDD/Fs in discharge water from stage C was 253 pg L^{-1}, which is much higher than that from other stages. The level of PCDD/Fs in the bleaching pulps ranged from 34 to 44 pg TEQ g^{-1} dw (Zheng et al., 1997a). The PCDD/F concentration in the wastewaters from a pulp mill in China using a bleaching sequence C-E-H was 316 pg I-TEQ L^{-1} whereas 2,3,7,8-TCDD was 230 pg L^{-1} and 2,3,7,8-TCDF was 122 pg L^{-1} (Zheng et al. 2001).

4.2.4. Iron and steel industry

Several operations for the production of iron and steel, including sinter production, coke production, and electric arc furnaces, have been identified as potential emission sources of PCDD/Fs. China is the largest producer of steel in the world. According to 2005 statistical data, the iron and steel production of China was 349 million tons. It is reasonable to assume that the iron and steel industry could be a major source of PCDD/F emission to air in China, but data are not available for an assessment of the emissions from this source.

4.2.5. Other sources

In addition to those sources discussed above, there are various other sources that could release considerable amount of dioxins. For instance, power/energy generation, non-ferrous metal processes, medical waste incineration, transportation, and cement kilns are all potentially significant dioxin sources, according to the experiences of other countries. However, there are no related data reported in China.

4.3. Sources of PCBs in China

PCBs were manufactured in China between 1965 and 1974, and the production was estimated to be 10,000 t. This is a minor fraction of the 1.5 million tons of total production worldwide. The major components in the Chinese PCB technical mixtures were tri- to penta-chlorinated congeners (named as PCB3 and PCB5, respectively). PCB3 was mainly used as an additive in paint, and PCB5 was mainly used as dielectric fluid in transformers. Some transformers containing PCBs as dielectric fluids were imported mainly for the steel industry and power plants, but the exact quantity is not known. Leaks and spills of PCBs could be a major source of releases.

In the 1980s, the activity of dismantling electric equipment to reclaim copper and other metals occurred in some places in China. Many of the transformers containing PCBs had been dismantled manually, and the environment was seriously polluted by PCBs because of illegal disposal.

According to the Chinese disposal regulations, wastes that contain PCBs are categorized as dangerous wastes and must be incinerated to destroy the PCBs. Jiang et al. (1999) analyzed fly ash collected from a PCB incinerator and found that the residual PCBs in fly ash were 0.022 ng TEQ g^{-1}. Compared to the 2–3 μg TEQ g^{-1} in technical PCBs, PCBs were destroyed effectively. The destruction and removal efficiency (DRE) for PCB incineration of the incinerator had achieved the requirements of design ($>$99.9999%) (Jin et al., 1997), although considerable levels of PCDD/Fs could still be detected. Most of the existing PCB wastes are sealed up in chemical waste landfills or caves for safekeeping. Many of these were not registered because of a lack of management. Leakage of some stockpiled PCBs has been found (Shao, 2001).

MSWI has been identified as an important existing potential PCBs emission source, but currently no information on stack gas concentrations of PCBs in MSWI has been reported in China. Dioxin-like PCBs are not categorized as dioxins in China so there are no regulations for PCBs in stack gas from MSWI and other combustion processes.

4.4. Sources of PBDEs in China

The release of PBDEs to the environment can occur during initial synthesis, incorporation into products, and the use of such products. In China and mainly in the eastern part of the country, large quantities of deca-BDE have been produced. In 2001, the production of deca-BDE was ~13,500 t per annum and the consumption has increased rapidly in recent years (Xia et al., 2005). In addition, three of the largest brominated FR manufacturers in the world (i.e., Great Lakes Chemical, Indianapolis, IN; Albemarle Chemical, Richmond, VA; and Dead Sea Chemical, Beer-Sheva, Israel) all have distributors in China.

The recycling of electronic items is another important source of PBDEs released into the environment. PBDEs make up from 5 to 30% of the weight of plastics and ~6 to 12 × 108 kg of PBDEs were released into environment via electronic wastes (E-wastes) from 1997 to 2004 (Martin et al., 2004). In China, much E-waste has long been imported for recycling, such as the world's obsolete computers and electronic components.

The current volume of E-waste disposed of in China is unknown. Martin et al. (2004) estimated that up to 261,000 t of PBDEs were imported into Guangdong province in 2002 in scrap electronic devices. The Chinese government is unable to provide details of the E-waste disposal practices but now has its own ban on such imports of E-waste.

In February 2006, China passed the Law on Pollution Control of Electronic and Information Products, which among other things, forbids producing, selling and importing of electronic and information products containing PBDEs. The legislation will come into force on July 31, 2007. Like PCBs, the levels of PBDEs in the environment will decline in China and around the world due to our awareness and the regulations.

4.5. Levels of PCDD/Fs, PCBs, and PBDEs in China

This section briefly reviews the existing status of PCDD/Fs, PCBs, and PBDEs in China. The amount of available data is inadequate to precisely evaluate the pollution situation. PCDD/Fs, PCBs, and PBDEs are not regular monitoring targets because the regulation limits for them in water, air, and soil are not yet established in China. Most of the data are collected from public scientific works and are not systematic. Some seriously polluted areas where investigation has been extensively campaigned are described separately in the next section.

4.5.1. Contamination levels of PCDD/Fs, PCBs, and PBDEs inenvironmental media in China

4.5.1.1. Aquatic environment

Aquatic media, including sediment and water, were more extensively investigated than other environmental media for PCDD/Fs, PCBs, and PBDEs in China. But the data are difficult to compare because of the inconsistency in the analytical procedures. In many cases, the data were reported without providing the results of quality control, making it impossible to evaluate the data quality. Most of the results reported were performed with GC-ECD and congener-specific data were rarely reported. Table 4.1 summarizes the level of PCDD/Fs, PCBs, and PBDEs in water and sediment collected around China from both freshwater and marine environments. Most of the investigations were carried out in eastern China. The research focused on sediments, and the targets were mostly PCBs. Chen et al. (1999) investigated PCBs in sediments of 11 rivers in eastern China and reported that the total concentration of PCBs (14 congeners) ranged

Table 4.1. The PCDD/Fs, PCBs, or PBDEs levels in aquatic environmental media in some locations of China (unit: ng g^{-1} dw for sediment samples and ng L^{-1} for water samples if not specified)

Location	Media	Time	Pollutants	Level	Reference
Huanghe River	Sediment	2004	PCBs^{-}	n.d.–5.98	Sun et al., 2005
Huaihe River	Sediment	–	PCBs^{57}	6.34, 8.24	Wang et al., 2001
Jiulongjiang River	Water	1999	PCBs^{12}	0.36–150	Zhang et al., 2000c
	Porewater	1999	PCBs^{12}	209–3869	
Liaohe River	Sediment	1998	PCBs^{4}	0.9–42.9[b]	Zhang et al., 2000d
	Water	1998	PCBs^{4}	n.d.	
Minjiang River	Sediment	1999	PCBs^{12}	15.13–57.93	Zhang et al., 2002b
	Water	1999	PCBs^{12}	0.20–2.47 μg L^{-1}	
	Porewater	1999	PCBs^{12}	3.19–10.86 μg L^{-1}	
Second Songhuajian g River	Sediment	–	PCBs^{-}	0.6–337	Liu et al., 1998
Yangtze River	Sediment	2000	PCBs^{-}	18.12	Chen et al., 2003
Yongding River	Sediment	1999	PCBs^{26}	0.81, 9.72	Ma et al., 2001
Guanting Reservoir	Sediment	1999	PCBs^{26}	4.23, 5.63	Ma et al., 2001
Baiyangdian Lake	Sediment	1992	PCBs^{-}	31.1–510.9	Zhu et al., 1995
Taihu Lake	Sediment	2004	PCDD/Fs	0.12–1.32	Zhang and Jiang, 2005
			PCBs^{209}	0.89–29.75	
Dalian Bay	Sediment	1999	PCBs^{9a}	0.040–3.230 (2.141)	Liu et al., 2001
Qingdao Bay	Sediment	1996	PCBs^{-}	1.021–153.132 (19.104)	Li et al., 1998
		1997/1999	PCBs^{50}	0.65–32.9	Yang et al., 2003b
		1997/1999	PBDEs^{21}	0.12–5.5	Yang et al., 2003c
Jinzhou Bay	Sediment	1996	PCBs^{-}	0.598–32.563 (5.863)	Li et al., 1998
Xiamen Bay	Sediment	1998	PCBs^{12}	n.d.–0.32	Zhang et al., 2000e
	Water	1998	PCBs^{12}	0.08–1.69	Zhang et al., 2000f
Xiamen coastal area	Sediment	–	PBDEs^{12}	0.10–2.06	Ou, 2006
	Sediment	2001	PCBs^{7}	0.19–18.95 (2.70)	Yang et al., 2003a
Yangtze River Delta	Sediment	2002	PBDEs^{12}	n.d.–0.55	Chen et al., 2006b
	Sediment	2002	BDE 209	0.16–94.6	Chen et al., 2006b

[a]Number of congeners of PCBs or PBDEs determined; "–" means unknown.
[b]Expressed as min-max (mean) (if available).

from 10.5 to 25.5 ng g^{-1}. In general, the level of PCBs in the sediments of Chinese waters was relatively low compared with those reported in industrialized countries. Considering that the levels in eastern China tend to be higher than those in western China, we can conclude that the pollution of PCBs in the Chinese aquatic environment is generally low.

4.5.1.2. Soil

Although soil is believed to act as a significant repository for POPs, we have almost no idea about the pollution situation of PCDD/Fs, PCBs, and PBDEs in soils of China, because only a few surveys have been conducted in some specified areas. A preliminary study showed that the level of PCDD/Fs in soils in the Beijing area ranged from 12 to 260 pg g^{-1} (Li et al., 2004). PCB concentrations in soils of industrial plants and rural areas of Shanghai were reported to be 0.05–587 ng g^{-1} (Xie et al., 2005). A chloralkali plant was suspected to be an emission source of PCBs. Soil in the former Cheoy Lee Shipyard at Penny's Bay, Hong Kong was contaminated with dioxins, petroleum hydrocarbons, and metals. Approximately 100,000 m^3 of soil were decontaminated before the land was used for infrastructure development.

4.5.1.3. Air

Several atmospheric aerosol samples were collected in Shanghai and Dalian and PCDD/Fs in total suspended particles were reported (Yang et al., 2004). The mean concentrations of total PCDD/Fs were 55.5 and 19.2 pg m^{-3}, and the mean I-TEQs were 0.928 and 0.334 pg m^{-3}, respectively. The predominant congeners were the lower chlorinated congeners. The pollution level is comparable to the general trend of urban industrial sites (0.1–0.4 pg TEQ m^{-3}, Lohmann and Jones, 2000).

Jaward et al. (2005) conducted a large-scale passive air sampling survey for PCBs, OCPs, and PBDEs across Asia in 2004. Thirty-two samplers were successfully deployed in 13 rural and 19 urban sites. Air concentrations in China ranged from 7.0 to 117 pg m^{-3} for PCBs (sum of seven congeners) and from ND to 340 pg m^{-3} for PBDEs (sum of eight PBDEs). They concluded that the levels of PCBs were elevated in some Chinese locations, and PBDE level were generally low in the region.

4.5.1.4. Sewage sludge

In addition to PBDE manufacturing and specific industries, wastewater treatment including storm water runoff and sewage sludge is considered to be a major source of environmental PBDEs. The concentrations of PBDEs in sewage sludge from wastewater treatment plants give an indication of the general exposure to and uses of these compounds. In the early 1990s, European researchers identified high levels of PBDEs (i.e., for BDE-47 and -99, at concentrations between 100 and 190 ng g^{-1} dw) in sludge samples (Darnerud et al., 2001). In the United States, a survey of sewage sludge biosolids from 26 publicly owned treatment works (POTWs) in 9 states revealed that the PBDE concentrations exceeded those found in Europe 10–100 times (Hale et al., 2001, 2003). In China, data on PBDEs in sewage sludge are scare. PBDEs were recently detected in sewage sludge collected from three different provinces of China (Wang et al., 2005). The total PBDE (19 congeners) concentrations were 27.2, 8.0, and 5.2 ng g^{-1} dw, respectively. The results showed that PBDE levels in the sewage sludge in China were substantially lower than those in the sludge from Europe and North America. Furthermore, data analysis indicated that the concentration of PBDE congeners was not correlated with facility location, size of the population served, or processing capacity.

4.5.1.5. Biota

Most of the available data on PCDD/Fs, PCBs, or PBDEs in biota were obtained using fish or mussel samples. Table 4.2 summarizes the results reported in China. PCBs in marine biota were found to be substantially higher than that from freshwater, and compared with that in biota from north or east, the levels of PCBs from the south of China were significantly higher. In addition to fish and mussels, pine needles have been used as passive samplers to determine regional contamination of POPs in China (Zhu et al., 1999; Chen et al., 2006a). The PCDD/F and PCB levels in these pine needles are low or comparable with other regions that were not impacted by evident sources (Chen et al., 2006a).

4.5.2. Food

An investigation about PCDD/Fs and PCBs in Chinese food is being conducted by the Chinese Center for Disease Control and Prevention to evaluate the dietary exposure to dioxins of Chinese people, but no data are available at this time.

4.5.3. Human blood and milk

Jin et al. (2003a) investigated the dioxin concentrations in the breast milk of 79 first-time mothers in Shengyang and Dalian, Liaoning Province, by

Table 4.2. The PCDD/Fs, PCBs, and PBDEs levels in biota samples in some locations of China

Location	Biota	Time	Pollutants	Level	Reference
Huaihe River	Silver carp	–	PCBs^{57}	4.65 ng g^{-1} ww	Wang et al., 2001
Minjiang River		1995/1996		n.d.–6.78 ng g^{-1} dw	Chen et al., 2001
Fujian	Pine needle	1998	PCBs	16–32 (19) ng g^{-1} ww	Zhu et al., 1999
Shanghai	Pine needle	1997	PCBs	54–62 (61) ng g^{-1} ww	Zhu et al., 1999
Zhejiang	Crucian carp	–	PCBs^{6}	150–1757 (528.2) pg g^{-1} ww	Tie et al., 2005
	Silver carp	–	PCBs^{6}	48–524 (176.0) pg g^{-1} ww	
	Pine needle	1998	PCBs	48–63 (55) ng g^{-1} ww	Zhu et al., 1999
Dalian	Pine needle	1997	PCDD/Fs	127 $\pm$ 40 ng kg^{-1} dw	Chen et al., 2006a
Dalian	Pine needle	1997	PCBs	4389 $\pm$ 1575 ng kg^{-1} dw	Chen et al., 2006a
Bohai Sea	Mollusks	–	PCDD/Fs	0.9–15, 317 pg g^{-1} lipid	Zhao et al., 2005
			PCBs^{209}	66.1–583.6 pg g^{-1} lipid	
East China Sea	Skipjack tuna	1996–2001	PBDEs^{11}	21 ng g^{-1} lipid	Uneo et al., 2004
Fujian coastal area	Shellfish	2002	PCBs	n.d. (<0.8 ng g^{-1} ww)	Xue et al., 2004
North of Yangtze River mouth	Shellfish	1990	PCBs	n.d.–22.9 ng g^{-1} ww	Liu et al., 1996
Qingdao Bay	Mussel	1997–1999	PCBs^{50}	8.4, 4.4 ng g^{-1} ww	Yang et al., 2003b
South China Sea	Finless porpoise	1990, 2000/2001	PCBs^{62}	1400–28,000 ng g^{-1} lipid	Ramu et al., 2006
			PBDEs^{10}	84–980 ng g^{-1} lipid	
	Skipjack tuna	1996–2001	PBDEs^{11}	23, 34 ng g^{-1} lipid	Uneo et al., 2004
Xiamen Island	Shellfish	1995	PCBs	n.d.–234 ng g^{-1} dw	Chen et al., 2001
Yangtze River estuary	Mussel/crab/fish	2002	PCBs^{36}	43.7–1260.4 (342.5) ng g^{-1} ww	Liu et al., 2004
		1997	PCBs	104–128 (115) ng g^{-1} ww	

CALUX bioassay. The mean concentrations of 15.84 and 7.21 pg TEQ g^{-1} fat were obtained for the two cities, respectively. They concluded that the dioxin level in Chinese breast milk was at the global average level, although it was higher than the lowest limit of the Tolerable Daily Intake (TDI) proposed by WHO. Pollution levels of dioxins and related compounds in pooled samples of human breast milk collected from primiparae in Dalian and Shenyang have been determined (Kunisue et al., 2004). The levels of PCDD/Fs and coplanar PCBs were in the range of 3.5–5.7 and 2.5–3.2 pg TEQ g^{-1} lipid wt., respectively. Concentrations of dioxins and PCBs in humans in China are in general lower than those in many other countries.

Bi and Xu (2000) reported the residue levels of PBDEs in human serum and breast milk samples from southern China. The concentrations of seven congeners ranged from 1.5 to 17 ng g^{-1} lipid. The levels were within the range reported in European samples but lower than that reported in North American samples.

4.5.4. Some severely polluted areas in China

4.5.4.1. The Ya-Er Lake

Ya-Er Lake is located in the east part of Wuhan, Hubei province, China. It is a shallow, eutrophic lake along the middle-lower reach of the Yangtze River. The water covers 20 km^2, and there are 270 km^2 of farmland with a population of 300,000 people around the lake. Ya-Er Lake was once famous for fishing before a large chemical factory was built on the bank in 1962. From 1962 to 1987, the lake was seriously polluted with PCDD/Fs and other pollutants, such as hexachlorocyclohexane (HCH), chlorinated benzenes (CBs), chlorinated phenols (CPs) and its sodium salts, and mercury, by direct discharge of effluent from the factory. The macrophytes vanished completely and the fishing was ruined. Nowadays, the wastewater is discharged by other means, which has resulted in the reappearance of the lake biota. In view of the historical input to lake sediments, PCDD/Fs were found to originate from the production of chlorinated chemicals in the 1980s, but more recently these compound mainly originate from graphite electrode sludge (Xu, et al., 1999). In the highest polluted sublake, the concentration of total PCDD/Fs in the surface sediment was 13.8 ng g^{-1} and I-TEQ was 0.42 ng g^{-1}. To investigate the transfer and bioaccumulation of PCDD/Fs in the foodweb, samples from different aquatic animals, aquatic plants, duck eggs, bird eggs, mother's milk, and human hair from the Ya-Er Lake area were collected and analyzed. The results showed that the aquatic organisms were seriously contaminated by PCDD/Fs. These compounds have bioaccumulated in biota of various trophic levels and biomagnified along the food chain. The concentrations of PCDD/Fs in fish muscle tissue ranged from 34.4 to 134 ng I-TEQ kg^{-1} lipid. Fish was the primary protein intake source in the diet of residents in the area. The high accumulation of PCDD/Fs in the foodweb has been seriously threatening the health of local residents in the Ya-Er Lake area.

4.5.4.2. The Dongting Lake

Dongting Lake, located in southern China, is the second largest freshwater lake in China. The total area of the lake is ~3,000 km^2, and it is regarded as very important due to its multiple functions: flood prevention, water supply, fishing, sightseeing, and shipping. However, a severe blood parasite, *Schistosomiasis japonica*, prevailed in this region for a long time. Technical Na-PCP has been sprayed since the 1960s to kill *O. hupensis* the snail hosts of the parasite, and Na-PCP has polluted most of the area (Zhang et al., 2001). The region was also seriously polluted by PCDD/Fs, which are contaminants in Na-PCP technical products (Zheng, et al., 1997b). The highest concentration of PCDD/Fs in sediments was 891 pg TEQ g^{-1}, and OCDD was the major contributor with a concentration of 151 ng g^{-1}. Now the molluscacide has been mostly replaced because of the awareness of the harm it causes (Liu et al., 2005). Fortunately, PCDD/Fs in human breast milk samples collected from the region were low, with the exception of several samples (Zheng et al., 2003).

4.5.4.3. Tianjin

Tianjin, located in northen China, is adjacent to the Bohai Sea and is one of the largest industrial cities in China. The Haihe River, the largest water system in north China, flows through Tianjin city and empties into the Bohai Sea. With the rapid economic growth, industrialization, and urbanization, accompanied by inadequate infrastructure investment and management capacity, water shortage has become a bottleneck to the further development of the economy and agriculture production in this region. Meanwhile, water pollution is becoming extremely serious. The Dagu Drainage River, situated in the south of the Haihe River, is a very important channel for the discharge of various domestic and industrial wastewater to reduce water pollution in the Haihe River. Along the Dagu Drainage River, there are several large-scale chemical factories including the largest DDT and PCP producer in China. Endemic chloracne among the workers in a plant here had been noted since 1974. The prevalence of chloracne was 73.4% in total and 95.2% in a TCB tank area where PCDD/Fs levels were thousands of ppm (Cheng et al., 1993). In addition, there are hundreds of small-scale chemical factories in this area, including pulp and paper, printing and dyeing, leather, medicine, etc. Hu et al. (2005) reported the occurrence of trace organic contaminants in Bohai Bay and its adjacent Nanpaiwu River (another name for the Dagu Drainage River). The maximum concentration of PCDD/Fs in sediment samples from the river was 8714 ng g^{-1} dw and 22 ng TEQ g^{-1} dw and the maximum concentration of co-PCBs of the sediment samples was 315 ng g^{-1} dw and 0.88 ng TEQ g^{-1} dw. Another investigation (Zhang, 2004) of this area showed that the maximum levels of dioxins in the sediment of Dagu Drainage River were 557 ng g^{-1} dw and 893 pg TEQ g^{-1} dw, and the concentrations of PCBs were 154 ng g^{-1} dw and 21 pg TEQ g^{-1} dw. The pollution level increased significantly after the river passed through a big chemical industry area. The congener profile of dioxins of both investigations indicated that the most significant source was the production of pentachlorophenol (PCP) and PCP-Na in this area. However, the source of PCBs in this area is still not clearly understood.

4.5.4.4. Taizhou

Taizhou, situated in the coastal central section of Zhejiang Province, is one of the most active cities in the Yangtze River Delta. The main industries of Taizhou include plastic moulds, automobile, chemical medicine, and electric appliances. In the 1980s, dismantling operations of scrap electric equipment to reclaim metals emerged and boomed in some villages of Taizhou. At present, Taizhou is one of the largest dismantling centers in China (Fig. 4.2). In 1989, serious pollution of PCBs in soil and sediment was found because of the scrap transformer dismantling operation in this region (Chu et al., 1995). The concentrations of PCBs in a farmland soil and a sediment sample were 788 and 691 ng g^{-1}, respectively. PCDD/Fs were detected in two farmland soils in this area in 2005, and the mean total PCDD/Fs and TEQs were 2.6 ng g^{-1} and 21 pg g^{-1}, respectively (Luo et al., 2005). Because the scrap transformers containing PCBs as dielectric fluids are rarely available, open burning of wires and other parts with PVC insulation to recover metals such as copper and steel has been considered the major source of PCDD/Fs in the region in recent years.

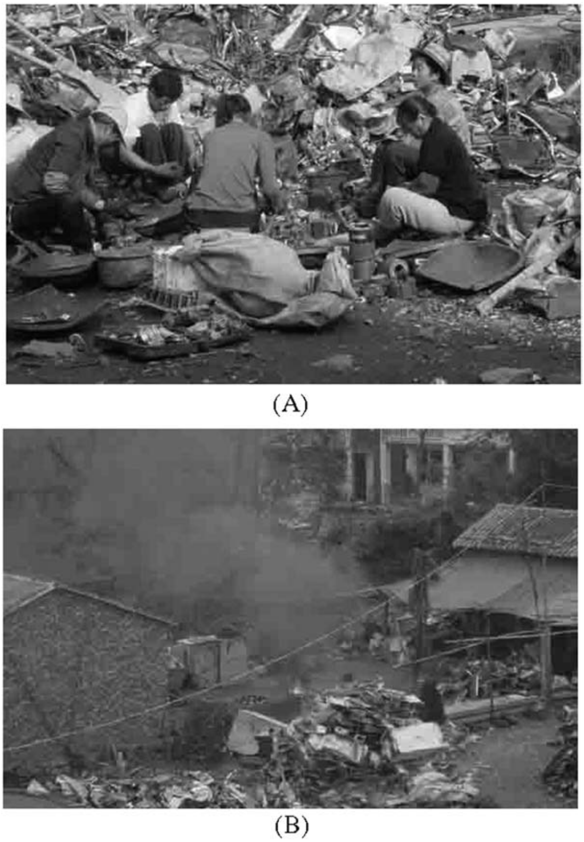

Figure 4.2. (A) Workers are dismantling electric appliances in Taizhou. In 1980s, dismantling the scrap transformers containing PCBs as dielectric fluids has severely polluted the environment. (B) Open backyard burning of wires and other parts to recycle copper or other metals is very typical although it is illegal. The activity has been considered the major source of PCDD/Fs in the region in recent years.

4.5.4.5. Guiyu

The town of Guiyu is located in Guangdong province, China, and very famous for its E-waste recycling. Before the 1990s, it was only a cluster of small villages. It became a booming recycling center for E-waste arriving from various regions of the world since 1995 (Fig. 4.3). Today, millions of tones of E-waste are imported to this area for recycling metals, plastics, and other useful materials. The potentially hazardous recycling practices included the manual and unprotected removal of printer cartridge toner,

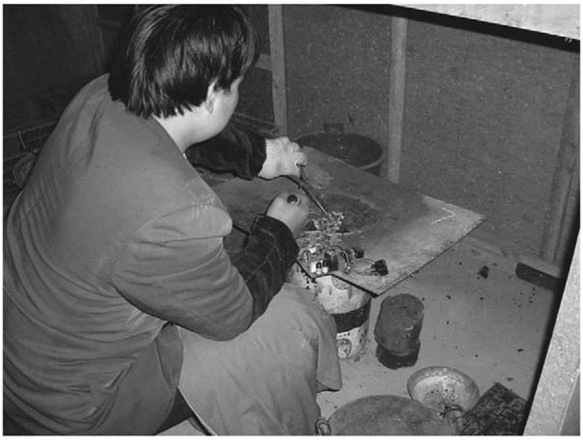

Figure 4.3. A worker is de-soldering circuit boards over a grill. The E-waste recycling practice causes severe environmental pollution of PBDEs and other contaminants in Guiyu.

the open incineration of wires to recover copper, etc. Aside from contributing to the prospering the local economy and improving the living standards of the inhabitants, these operations have also brought serious environmental problems to this area. Nowadays, the government has realized the severity and is preparing legislation on electronic waste to limit the import of E-waste. However, it might need a long time and large amounts of economic investments to remediate the severe environmental impacts caused by the pollution. More and more research has also been focused on this area. Information on pollution status in this area is being collected, such as PBDE congener profiles. The reported data of PBDEs from one recent published paper (Wang et al., 2006) showed that the concentrations are in the range of 0.26–824 ng g^{-1} dw. The isomer profiles of PBDEs were similar to various technical formulations of FR products.

4.6. Challenges of PCDD/Fs, PCBs, and PBDEs management in China

Implementation of the Stockholm Convention will be pivotal in the fight against POPs. Implementing the Convention involves a number of important departments and covers economic, ecological, and social fields. It is thus a very difficult and challenging task. Compared to other specified POPs, PCDD/Fs, and PCBs are more difficult to control because they can be formed and released unintentionally. The main challenges for China are summarized below.

1. Analysis and monitoring capacity is still low, particularly for dioxins. The current number of laboratories qualified for PCDD/Fs, dioxin-like PCBs and PBDEs analyses are not sufficient for the implementation of the Stockholm Convention. There are 13 laboratories equipped with HRGC/HRMS, and more are under construction for dioxin analysis in China. However, these laboratories belong to different blocks of the political infrastructure and were built for different purposes. Effective coordination among existing laboratories capable of dioxin analysis will help with building and strengthening the monitoring network in order to meet the need to implement the Stockholm Convention. In addition, currently available equipment in many laboratories and the proficiency of the personnel are also in need of updating.
2. Regulatory standards are urgently needed for waste discharges, food safety, etc. For example, there are no regulations for dioxins in food and numerous products or processes. There is a regulatory limit for dioxins in stack gas from MSWI, but there is no regulated monitoring. The heavy industry and chemical industry with possible dioxin emission have grown rapidly in recent decades. Some processes differ from those used in other countries, thus the standards for emissions from other countries should not be directly applied.
3. Relevant policy and financial support plans need to be improved to meet the requirement of the implementation of the Stockholm Convention. Public awareness plays a very important role in the fight against POPs, but in China it suffers from a lack of sufficient funding for public environmental education, and for research results dissemination and technology transfer.
4. There is a lack of effective reduction technologies. Fundamental research on dioxins emission control needs to be effectively promoted. The best available techniques (BAT) and best environmental practices (BEP) for the destruction of the existing PCB wastes need to be implemented. More environmentally benign alternatives to PBDEs are to be sought to meet the demand for FRs.

Solutions to problems caused by POPs are closely linked to China's environmental protection, social and economic development. Updating management policy and monitoring capability is likely the first step that needs to be taken at the present stage.

4.7. Conclusions

This chapter reviews the sources, environmental levels, and current situation of PCDD/Fs, PCBs, and PBDEs in China. Although there are diverse sources and serious contamination in some places, the current status of pollution throughout the country is still not clear. China has not been able to conduct nationwide inventory surveys on these chemical pollutants of high concern. A national or regional inventory has proven to be a crucial step toward the reduction and elimination of dioxins and PCBs. As one of the components of the Stockholm Convention, updating management policy and monitoring capability, and applying BAT and BEP are urgent tasks for the Chinese government and research institutions.

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