Review article

Hexachlorobenzene sources, levels and human exposure in the environment of China

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ABSTRACT

This article summarizes the published scientific data on sources, levels and human exposure of hexachlorobenzene (HCB) in China. Potential sources of unintended HCB emission were assessed by production information, emission factors and environmental policies. HCB was observed in various environmental compartments in China. HCB levels increased from South China to North China in most of environmental compartments (air, soil and mussel). Some hotspots were identified near the factories producing and using HCB. In terms of spatial distribution, HCB concentrations in air and shellfish showed much variation, which indicated some primary emission sources in China. HCB levels in air and human milk in China were relatively higher than those in other countries, but HCB levels in other compartments were similar to those in Europe and other countries in Eastern Asia. In the limited studies on temporal trends of HCB levels in China, HCB concentrations in air, sediment, fish and human milk did not show a consistent downward trend. Although HCB levels in food and human milk does not pose a health risk in China at present, long-term exposure to HCB should not be overlooked.

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1. Introduction

Hexachlorobenzene (HCB) is one of the twelve persistent organic pollutants (POPs) listed under the Stockholm Convention, and has characteristics of persistence, bioaccumulation, toxicity, and long-range environment transport. HCB is extremely stable, globally distributed, and to be a most persistent POP (Meijer et al., 2003a).

HCB in the environmental compartments is not only from its industrial and agricultural application, but also produced as trace contaminant in several pesticides and as an unintended byproduct during the industrial manufacture and combustion processes (Bailey, 2001). HCB was first introduced in 1933 as a fungicide. Agricultural use of HCB dominated its emissions during the 1950s and 1960s (Courtney, 1979). Although HCB production and use has been ceased before 2000 in most countries, HCB is still being released to the environment as an unintended byproduct in chemical processes, incomplete combustion and an impurity in pesticides. Current emissions are estimated to be 70%–95% lower than that in 1970 because of the banning of HCB for agricultural use (Barber et al., 2005).

HCB has been observed all over the world in air, soil, water, sediment, biota and human tissues. The atmospheric concentrations are now fairly uniform, varying by less than an order of magnitude at background sites. However, highest HCB air concentrations were still found in historical source regions, indicating that there are some ongoing primary or secondary emissions (Jaward et al., 2005). HCB concentrations in background soils were strongly influenced by source region and soil organic matter content (Meijer et al., 2003a; Valle et al., 2005). HCB levels in biota and human were affected by regional and local historical HCB emissions. Levels of HCB in the environment have dropped substantially in the past 20 years as shown by many studies (Braune, 2007; Bustnes et al., 2007; Kannan et al., 1992), which shows the consistent decrease of HCB emissions and HCB is being degraded in the environment. HCB has been reported in tissues of polar bears and penguins from the Arctic and the Antarctic, there are no consistent downward trend of HCB in certain polar locations (Corsolini et al., 2002; Kannan et al., 2005).

Although emission and distribution of HCB have been well documented at global scale, information about that in China is limited. There are many studies on HCB concentrations in various environmental compartments in China, but only a few focused on the countrywide distribution of HCB. Moreover, most of the studies on HCB levels were scattered in the articles about organochlorine pesticides (OCPs) and only listed the data of concentrations with few discussions about the relations between the sources and levels of HCB. Furthermore, some results were partial because of insufficient information about HCB emissions and distribution in China, which may lead to misunderstanding of HCB pollution status in China. Therefore, the objectives of this study are to assess all potential sources of HCB in China, to discuss spatial distribution and temporal trends of HCB levels, to find relations between the sources and levels of HCB, to indicate human exposure to HCB, and to give recommendations for further studies through the overview of published scientific data.

2. HCB sources in China

2.1. Direct production and use of HCB in China

HCB has never been used directly as a pesticide in China, but has been used to produce pentachlorophenol (PCP) and pentachlorophenol-Na (PCP-Na), reagent and fireworks (Wei et al., 2007b). Hexachlorocyclohexane (HCH) was used to produce lindane and HCB in China (HCH is thermally dehydrohalogenated to trichlorobenzene and then chlorinated to produce HCB which is treated with NaOH to produce PCP and PCP-Na). China began to produce HCB in 1958, and in total there were 6 HCB production enterprises. Due to control over HCH production and usage after 1983, the production of HCB

has been reduced drastically. After 1983, there is only one enterprise in Tianjin continuing with HCB production in the whole country. Cumulative production of HCB after 1988 was 79,278 tons, of which 78,323 tons were used to produce Na-PCP and PCP, accounting for 98.8% of the total production. The rest was mainly used to produce fireworks. Data on HCB production in China from 1988 to 2003 are listed in Fig. 1. Production was reduced year by year after 2000 and completely stopped in 2004. As a part of China's National Implementation Plan (NIP) for Stockholm Convention, the government prohibited the production and use of HCB in 2009 (SEPA, 2007).

2.2. Unintended HCB emissions in China

There are no unintended HCB emission inventories in China at present, and the literature about unintended HCB emissions from potential sources is limited. In this section, we try to collect and summarize the available information on unintended HCB sources in China and assess emissions and trends of all potential HCB sources.

2.2.1. Pesticides with HCB impurity

HCB was reported to be a trace contaminant in several pesticides containing chlorine (Benazon, 1999; Pacyna et al., 2003). In the 1990 HCB inventories of the US, the following pesticides contained HCB as a contaminant: atrazine, simazine, picloram, pentachloronitrobenzene (PCNB), chlorothalonil, dimethyl tetrachloroterephthalate (DCPA), lindane, technical HCH, PCP and PCP-Na (USEPA, 1998). In China, production and use of technical HCH and lindane were prohibited in 1983 and 2000 respectively (Wang et al., 2005a). PCP was produced until 1995 (Fig. 2) and registered as wood preserving agent by the Ministry of Agriculture of China. PCP-Na was produced and used as wood preserving agent and molluscicide from 1958 to 2003 (Fig. 2). PCNB is not allowed for crops. Other pesticides containing HCB are produced and used widely in China, of which only chlorothalonil has been given National Standard for HCB impurity with permissible value of 0.04% (GB-9551-1999). HCB concentration in picloram in China was 1.15 mg/kg (Lu, 2007), much lower than that (50 mg/kg) reported in the US 1990 emission inventories. HCB concentration in chlorothalonil was 42.2 mg/kg in China (Wang et al., 2005b), similar to the data (40 mg/kg) reported in US 1990 emission inventories. Pesticide application may be an important source of HCB in the environment because of widespread use of pesticides and the lack of strict regulations on HCB impurity in pesticides in China.

Fig. 1. HCB production from 1988 to 2003 in China^a (SEPA, 2007). ^aNo data about HCB production available before 1988.

Fig. 2. PCP and PCP-Na production in China^{a, b} (SEPA, 2007). ^aNo data about PCP production available before 1983. ^bNo data about PCP-Na production available before 1988.

2.2.2. Manufacturing emissions

Many studies revealed that HCB production and use in some chemical plants caused elevated HCB levels in the surrounding environment (Barber et al., 2005). There were also some studies on HCB pollution of chemical plants in China (Table 1). High levels of HCB were detected in sediment and soil in the Ya-Er lake area in central China, where a factory primarily produced PCP and PCP-Na through HCB (Wu et al., 1997). High levels of HCB were also detected in the sediments from Nanpaiwu River which received the wastewater from a HCB and PCP-Na factory in Tianjin (Hu et al., 2005; Ding et al., 2007). Very high HCB levels in water, soil, sediment and vegetation were observed near a chemical plant using HCB as a chemical material in Hubei Province (Wang et al., 2007c). Concentrations of HCB in soil were significantly higher in one site near a former organochlorine pesticides (DDT and HCH) factory (894 ng/g dry wt.) than those in other locations (0.0531–12.7 ng/g dry wt.) in Anhui Province (Shi et al., 2007a). Moreover, soils were seriously contaminated by HCB in two factories producing PCP and PCP-Na through HCB (SEPA, 2007). These studies indicated that a large amount of HCB emission could be caused by historical wastewater emission and air deposition from these chemical factories. However, there are no detailed data of HCB emissions from the chemical factories. Therefore, it is essential to identify HCB residue levels and potential risk in environmental compartments close to these factories. Furthermore, volatilization of “old” HCB from these contaminated soils need to be studied.

The manufacture of chlorine using carbon anodes is known to produce small amounts of HCB (Bailey, 2001). There is no study about

HCB emission from chlorine production using carbon anodes in China. Carbon anodes were eliminated in China according to the regulation issued by the National Development and Reform Commission. It is not a source of HCB in China. HCB was not detected in perchloroethylene with a detection limit of 2 ppb and found at less than 2 ppb in trichloroethylene (IPCS, 1997). Although these chlorinated solvents were produced and used in China, a small amount of HCB could be emitted from their production because of their low emission factors.

2.2.3. Metals industry and combustion processes

Hexachloroethane in aluminum oxide can produce small amount of chloro-organics, including HCB (Westberg et al., 1997). Hexachloroethane is still used in molten aluminum process in China, but the amount decreases gradually because of the toxicity of hexachloroethane. Secondary copper recovery was popular in China (the production of recovered copper was 744,026 ton in 2005) (NMII, 2004), which can also lead to HCB emissions. No literature is available about HCB emission from metals industry in China.

In many studies HCB was reported as a product of incomplete combustion where chlorine is present. These combustion processes includes all kinds of waste incineration (hazardous waste, municipal waste, medical waste, and sewage sludge), cement production, iron ore sintering, coal combustion, and biomass burning. Except for sewage sludge incineration (not used in China), other combustion processes may be potential sources of HCB in China. No HCB emission data from combustion processes is available in China. There is no direct regulation on HCB emissions in combustion processes, but some indirect regulations and actions can contribute to reduction of HCB emission. There are complete controlling standards of regular air pollutants and particulate from waste incinerations in China. Moreover, China has planned to phase out the outdated ironstone sinter and electric arc furnace steel-making techniques. In addition, China is making efforts to reduce emissions of PCDD/Fs from combustion processes by improving combustion condition (SEPA, 2007), which is also expected to reduce HCB emissions.

3. HCB distribution in the environment

Compared to the studies on DDT and HCH residue levels, there are much fewer studies on HCB levels in China. We try to integrate these regional studies to provide a general profile of the spatial and temporal distribution of HCB residue levels in China.

3.1. Air

The studies on HCB levels in air in China were conducted in different regions during the period 2000–2007. HCB was detected in all the sampling sites, which implies its long-range transport potential and persistence. Jaward et al. (2005) studied the spatial distribution of HCB concentrations across Asia by passive sampling survey using PUF discs in 2004, which revealed that air HCB levels in China showed much variation, increasing gradually from southwest to northeast (Table 2). It is different from the uniform distribution found in South Korea (14–95 pg/m³), Japan (26–136 pg/m³) (Jaward et al., 2005) and Europe (1.4–8.9 pg/m³) (Jaward et al., 2004), which indicated that there were some primary emission sources of HCB in China. Although HCB production was completely stopped in 2004, HCB emissions from waste dumpsites and contaminated soils in the former HCB factories in Tianjin and Fuzhou may be responsible for the high air HCB levels in Tianjin, Beijing, and Fuzhou. Air HCB levels in four provinces (Anhui, Hubei, Jiangsu and Hunan province) along the Yangtze River also showed much variation, and there were high HCB levels in limnological regions or the areas around some factories producing pesticides (Shao et al., 2007a). Air HCB concentration (126 pg/m³) in Qingdao was similar to the mean HCB concentration (near 100 pg/m³) in China (Lammel et al., 2007). The HCB concentrations in air in the Pearl River Delta were uniform with high concentration/low concentration ratios

Table 1
Elevated HCB levels in surrounding environment of chemical plants.^a

Location	Sample type	Range (average)	Sampling year	Reference
Ya-Er lake	Sediment	3.15×10^7 – 5.71×10^7	1991–1994	Wu et al. (1997)
	Soil	3.54×10^7 – 3.77×10^7		
Nanpaiwu River	Sediment	799–141000	2002	Hu et al. (2005)
Hubei province	Sediment	17.5–1320(240)	2003	Ding et al. (2007)
	Water	2.54–125	2005	Wang et al. (2007c)
	Sediment	251000–2280000		
	Soil	59.8–1960		
Anhui province	Vegetation	52.9–687		
	Soil	894	2004	Shi et al. (2007b)

^a HCB in vegetation, sediment and soil is measured with ng/g dry weight, while in water is ng/mL.

Table 2
HCB levels in air in China.

Area	Range/average (pg/m ³)	Sampling year	Sampling method	Reference
Hong Kong	46–232 (137)	Winter of 2000/2001	Active PUF	Louie and Sin (2003)
Anhui province	36.6–669	2005.5–2005.10	Active PUF	Shao et al. (2007a)
Hubei province	65–236			
Jiangsu province	41.1–183			
Hunan province	21.4–81			
Beijing Tongzhou district	222	Spring, 2004	Active PUF	Shao et al. (2007b)
	85.3	Autumn, 2004		
Beijing Daxing district	330	Spring, 2004		
	52.7	Autumn, 2004		
Throughout China	10–460	2004.9–2004.11	Passive PUF	Jaward et al. (2005)
Pearl River Delta	27–216 (81)	2005.8–10	Passive PUF	Wang et al. (2007b)
	108–567 (257)	2005.12–2006.2		
The Mt. Everest region	4.8–12.6 (8.9)	2002.5–2002.6	Active PUF	Li et al. (2006)
Qingdao	142	2003.6	Active PUF	Lammel et al. (2007)

of 1.7–4.8, which implied the lack of significant primary sources in this area (Wang et al., 2007b). The atmospheric HCB levels in the Mt. Everest region (a remote area far from local sources) were very low (Li et al., 2006), which could represent the background levels of HCB in China. HCB levels in air in colder seasons (Winter and Spring) were much higher than those in warmer seasons (Summer and Autumn) in China, which was different from the seasonal variation of other OCP (DDT and Chlordane) levels (Shao et al., 2007b; Wang et al., 2007b). This result also differed from the studies in other countries that HCB levels were higher in warmer season (Ma et al., 2003; Murayama et al., 2003). It proved that temperature did not strongly affect HCB levels in air in China. Some HCB emission sources in colder seasons might be responsible for the higher air HCB levels.

There is only one study on temporal trends of HCB in air in China. HCB levels (46–232 pg/m³) in Hong Kong in winter of 2000–2001 (Louie and Sin, 2003) were similar to the levels (108–189 pg/m³) in winter of 2005–2006 (Wang et al., 2007b). Because there were no other studies, it is difficult to estimate the temporal trends of air HCB levels in China.

Compared to other countries, air HCB levels in China were high. Even in the remote area (the Mt. Everest region), air HCB levels were also higher than the levels in Europe (1.4–8.9 pg/m³) (Jaward et al., 2004). HCB levels in air were highest in China among the countries (Japan, South Korea, and Singapore) across Asia in 2004. It was similar to air HCB levels (40–340 pg/m³) in Great lakes in 1990/1991 (Hoff et al., 1996) and Baikal region (70–170 pg/m³) in 1992 (Iwata et al., 1995). As a whole, HCB air levels in China in the 2000s were similar to those in Europe and USA in the 1990s. This time lag can be mainly attributed to the later production and prohibition of HCB in China.

3.2. Soil

The studies on soil HCB residues focused on eastern China (Table 3). The highest levels of HCB in soils were reported at the places where HCB was used or produced in large amounts (Table 1). HCB was not detected in soil samples of Mt. Qomolangma area with the elevation from 4700 m to 5620 m (Wang et al., 2007d), much lower than HCB levels (0.13–0.80 ng/g dry wt.) in European Pyrenees mountain soils (Grimalt et al., 2004). There was no study about spatial distribution of soil HCB levels throughout China. According to some regional studies in China, HCB levels in soils increased gradually from south to north China. In Hong Kong, HCB levels were detected from 5 samples out of 46 samples and lower than those in other cities in China (Zhang et al., 2006). In other studies (Table 3), HCB could be detected in most of soil samples. According to three studies in different sampling locations in Beijing, HCB levels in soils of Beijing were distributed in the order: chemical industry zone (1.85 ± 1.27 ng/g dry wt.) > urban (1.58 ± 0.68 ng/g dry wt.) > average Beijing (0.849 ng/g dry wt.) (Li et al., 2005; Shi et al., 2007b; Zhou et al., 2007). It indicated that industrial emissions of HCB were the most important sources in this region. In agricultural soils, HCB was also found. HCB levels in arable lands of Pearl River Delta in South China (Wan and Jia, 2005) were much lower than those in Taihu lake region (East China) (Gao et al., 2005; Wang et al., 2007a) and Yangling (Northeast China) (Wang et al., 2006). High HCB levels in agricultural soils in Taihu lake region and Yangling could be attributed to the use of PCP-Na and other pesticides containing HCB impurity. In comparison, HCB levels in China were lower than those in Katowice (6.4 ± 9.6 ng/g dry wt.), Poland (Falandyisz et al., 2001), higher than those in Australia (<0.2 ng/g dry wt.) (Cavanagh et al., 1999), while close to global background soil HCB levels (ranging from 0.010 to 5.20 ng/g dry wt.

Table 3
HCB levels in soil in China.^{a,b}

Area	Land use	Average/range (ng/g dry weight)	Sampling year	Reference
Hong Kong	Woodland, grassland, arable land, wetland and reclamation land	0.007–0.31	2000.12	Zhang et al. (2006)
Pearl River Delta	Arable land	ND – 2.12 (0.306)	2003.11	Wan and Jia (2005)
Taihu lake region	Tree land	(5.13)	2004	Wang et al. (2007a)
	Paddy land	(3.76)		
	Fallow land	(1.86)		
Taihu lake region	Agricultural soils	0.9–6.2 (2.45)		Gao et al. (2005)
Lake Tai, Hangzhou Bay and Shanghai city region	Surface soils	0.08–3.2	2000–2001	Nakata et al. (2005a)
Anhui province	–	0.0531–12.7 (0.502)	2004.5–2005.6	Shi et al. (2007a)
Yangling, Shaanxi province	Agricultural soils with wheat-corn rotation	(2.50)	2000.6	Wang et al. (2006)
Beijing urban city	Urban soils	0.46–3.17 (1.58)	2003	Li et al. (2005)
Chemical industry zone in Beijing	Industrial soils	ND – 4.12 (1.85)	2003.11	Zhou et al. (2007)
Beijing	–	ND – 6.12 (0.849)	2004.5–2004.7	Shi et al. (2007a)
Qomolangma area	–	ND	2005.4	Wang et al. (2007d)

^a ND: below the detection limit.

^b All data base on dry weight.

Table 4
HCB concentrations in water (ng/L) in China.

Location	Average/range (ng/L)	Sampling year	Reference
Tonghui River, Beijing	2–660 (145)	2003.11	Zhou et al. (2005)
Guanting reservoir, Beijing	1.43–27.3 (12.4)	2003.11–2004.7,8	Xue et al. (2005)
Shallow surface ground water in Taihu basin	0.7–19 (3)	–	Li et al. (2007a)
Pearl River Estuary	<1.0–13.6 (2.6)	2005.4–2005.11	Wei et al. (2007a)
The Nanjing section of Yangtze river	0.52–0.62	1998.5	Jiang et al. (2000)
Yellow River Estuary	1260	2004.8	Jiang et al. (2006)
Huaihe River	4700–12200	1998.4–5	Wang et al. (2002)

with a mean of 0.680 ng/g dry wt.) (Meijer et al., 2003b). There is no available information on temporal distribution of soil HCB levels in China.

3.3. Water and sediment

There have been a number of reports about HCB concentrations in water and sediment in China. Most studies focused on rivers, while a few on lake, reservoir, and sea. High HCB levels in water and sediment are supposed to come from point sources producing and using HCB (Table 1). In the major water systems in China, significant amounts of HCB can be detected (Table 4). HCB levels in the rivers (Huaihe River, Yellow River, Tonghui River) in north China (Wang et al., 2006; Wang et al., 2002; Zhou et al., 2005) were several magnitudes higher than the levels in the rivers (Pearl River and Yangtze River) in South China (Jiang et al., 2000; Wei et al., 2007a). HCB concentrations in water from Beijing Guanting reservoir were generally lower than those in rivers (Xue et al., 2005). HCB concentrations were also detected out in surface ground water in Taihu basin (Li et al., 2007a) (Table 5).

As a river receiving wastewater from many industries, the sediments in the Tonghui River had the highest HCB residues in China (Zhou et al., 2005). In major rivers in China, HCB concentrations in the sediments from the Haihe River were higher than those in other rivers, which indicated the effects of industrial activities (Ding et al., 2006; Jiang et al., 2000; Sun et al., 2007; Yang et al., 2004). HCB concentrations in sediments in the Beijing Guanting reservoir were generally lower than those in rivers (Xue et al., 2005). HCB concentrations in Taihu Lake were higher than those in most of rivers because of industrial discharge to the Lake (Yuan et al., 2003). HCB concentrations in the coastal sediments in the Pearl River Delta were much higher than those in the Pearl River estuary (Fung et al., 2005; Wei et al., 2007a).

Table 5
HCB concentrations in sediments (ng/g) in China.^a

Location	Average/range (ng/g)	Sampling year	Reference
Guanting reservoir, Beijing	0.0552–0.494 (0.146)	2003.11–2004.7,8	Xue et al. (2005)
Tonghui River, Beijing	(135)	2003.11	Zhou et al. (2005)
Haihe River, Tianjin	6.8–16.7 (11.9)	–	Ding et al. (2006)
Middle and lower reaches of the Yellow River	0.03–4.58	2005.11	Sun et al. (2007)
Nanjing section of Yangtze river	1.2–6.6 (4.1)	1998.5	Jiang et al. (2000)
Taihu Lake	0.06–9.69 (2.16)	Spring and Summer, 2000	Yuan et al. (2003)
Huangpu River	3.2	2001.9	Nakata et al. (2005a)
Wuhan section of Yangtze river	0.94–4.39 (1.86)	2001.10	Yang et al. (2004)
Pearl River Estuary	0.26–1.82 (0.69)	2005.4	Wei et al. (2007a)
Coastal sediments in the Pearl River Delta	1.1–55	2003.03	Fung et al. (2005)

^a All data base on dry weight.

Temporal trends of HCB levels were studied in dated sediment cores in Naisihu Lake in Shandong province and coastal areas in Hong Kong (Li et al., 2007b; Wei et al., 2008). HCB concentrations in Naisihu Lake increased significantly from 0.80 ng/g dry wt. to 2.27 ng/g dry wt. over the period 1960–2000. In Hong Kong, HCB in coastal sediments was first detected in the 1940s, and two peak-time periods were observed in the 1960s and 1990s, which didn't show a decreasing trend either. The results were different from decreasing trends of HCB concentrations in the sediments in other countries, because HCB was produced from 1958 to 2003 in China while its production and use was banned in most of the other countries before the 1980s.

3.4. Vegetation

Vegetation can serve as a biomonitor to trace the levels of atmospheric contamination of HCB. HCB concentrations in pine needles were determined in six regions of China in 2001 (Xu et al., 2004b). There was significant positive correlation between the GDP per capita and the concentration of HCB in pine needles. The high levels of HCB were found at relatively high-income areas in East China. HCB levels in pine needles at the chemical industrial area (5.3 ng/g dry wt.) were higher than those at the residential area near chemical industries (2.9 ng/g dry wt.) and residential area (1.1 ng/g dry wt.) in Beijing in 2002 (Xu et al., 2004a), which indicated that HCB in Beijing was mainly from chemical industries. Another study in 2003 showed that HCB could be found in 100% of the leaf samples from the plants in Beijing's southeast chemical industrial zone (Zhou et al., 2007). Around the sewage ditch from a chemical plant using HCB as industrial material in Hubei Province, the roots of vegetation were seriously contaminated by HCB (52.9–687 ng/g dry wt.). Lower concentrations (ND – 18.4 ng/g dry wt.) in vegetations further from the chemical plant showed the indicative of vegetation to HCB emissions (Wang et al., 2007c). In remote area of Qomolangma in China, HCB levels (0.0156–1.25 ng/g dry wt.) in grasses were detected. The significant correlation between HCB levels and the altitude suggests that cold condensation was occurring in elevated areas (Wang et al., 2007d). In remote area of southeast Tibet, HCB could also be detected in conifer needles (Yang et al., 2008). HCB levels in the needles in China are similar to those (1.4–30 ng/g dry wt.) in Europe (Calamari et al., 1994; Holoubek et al., 2007), but are generally higher than those in Africa and Canada (Davidson et al., 2004; Tremolada et al., 1993) (Table 6).

3.5. Biota

3.5.1. Bird

HCB concentrations in eggs of *Egretta garzetta* in Poyang Lake (3.3 ng/g dry wt.) and Taihu lake (6.3 ng/g dry wt.) were investigated, and the contamination levels did not affect the development and breeding success of *E. garzetta* (Ruan et al., 2003). HCB concentrations in *Acrocephalus aedon* around Taihu Lake in 2000 were 32 ± 23 ng/g

Table 6
HCB concentrations in vegetation in China.^{a,b}

Location	Range/average (ng/g)	Vegetation types	Sampling time	Reference
18 areas in China	ND–11.7 (4.2)	Needles	2001	Xu et al. (2004b)
Beijing City	1.1–5.3 (2.3)	Needles	2002.3–4	Xu et al. (2004a)
Chemical industrial zone in Beijing	0.132–2.14 (1.44)	Leaves of some redwood and grass	2003.11	Zhou et al. (2007)
Qomolangma area (4700–5600 m)	0.0156–1.25	Grasses Jidou and Rouzi	2005.4	Wang et al. (2007d)
Southeast Tibetan Plateau	0.69–4.3	Needles	2006.5	Yang et al. (2008)

^a ND: below the detection limit.^b All data based on dry weight.

lipid wt. (Nakata et al., 2005a). No other studies on HCB levels in birds were conducted in China.

3.5.2. Fish and shellfish

HCB levels in fish and shellfish in China were studied widely. Concentrations of HCB in fishes from Taihu Lake were relatively consistent, ranging from 20 ng/g lipid wt. in spotted steed to 37 ng/g lipid wt. in topmouth culter. HCB levels in shrimp and farm-raised carp around Taihu lake were relatively low (11–14 ng/g lipid wt.) (Nakata et al., 2005a). Concentrations of OCPs in different tissues of Chinese Sturgeon were investigated (Wan et al., 2007). HCB was detected in all the samples, and HCB concentrations (224–2600 ng/g lipid wt.) in Chinese Sturgeon muscles were comparable to those in sturgeons collected from the Caspian Sea (Kajiwara et al., 2003).

The studies on HCB in halobios focused on mussels. In the study by Monirith et al., HCB concentrations were generally uniform in China excluding extremely high HCB levels (540 ng/g lipid wt.) in samples collected from Jiao Zhou Bay, Shandong province. If the sites with extreme high concentration are excluded, HCB levels of mussels in China were similar to those in South Korea and Japan, but higher than those in Vietnam and Philippines (Monirith et al., 2003). In the study by Ramu et al., the distribution of HCB in mussels was comparable with the previous study, but the maximum level of HCB in mussel was 12 ng/g lipid wt. HCB concentrations in China were lower than those in South Korea and similar to those in Japan (Ramu et al., 2007). In two studies, HCB levels in mussels increased from south to north in China. The chemical factories producing HCB in Tianjin and Fuzhou may contribute to the unusually high HCB concentrations in mussels from Bohai Sea and Fuzhou. HCB concentrations in bivalves collected from Pearl River Estuary were comparatively low, except for HCB levels in bivalves from one site (26.8 and 14.9 ng/g dry wt.) (Wei et al., 2006). Almost all of the shrimp samples had extremely low levels

of HCB (<0.5 ng/g dry wt.), except for the samples from the site (1.14 ng/g dry wt.). In marine fishes in Hangzhou Bay, HCB levels ranged from the detection limit in halfsmooth tonguefish to 61 ng/g lipid wt. in trident goby. No significant difference in HCB concentrations was observed between fishes from Taihu Lake and Hangzhou Bay (Nakata et al., 2005a). HCB levels in fish in China were higher than those (1.67–4.53 ng/g lipid wt.) in eastern and southern Asia (India, Thailand, Vietnam, Indonesia, Papua New Guinea, and the Solomon Islands), but were lower than those in Australia and Europe (124 ng/g lipid wt.) (Kannan et al., 1995; Falandysz et al., 1994). In the study using skipjack tuna as a bioindicator of global PCBs and OCPs monitoring, HCB levels in the liver of skipjack tuna in the East China Sea (<0.16–7.0 ng/g lipid wt.) and South China Sea (2.2 ng/g lipid wt.) were medium in global environment (Ueno et al., 2003). HCB was detected in all the blubber of finless porpoises collected from South China Sea in 1990 (140–230 ng/g lipid wt.) and 2000/2001 (87–250 ng/g lipid wt.), concentrations of HCB in finless porpoises did not change significantly in the last decade (Ramu et al., 2006). There are no other studies about temporal trend of HCB in fish and shellfish (Table 7).

3.6. Sewage sludge

HCB levels were detected in sewage sludge from wastewater treatment plants in two studies. According to the study (Cai et al., 2007), samples were collected from 1998 to 1999 from 9 cities in China. HCB was detected in all samples with the levels from 9 to 3170 ng/g dry wt. (mean: 584 ng/g dry wt.). In another study by (Wang et al., 2007e), 31 sewage sludge samples in total were collected from 26 cities in 2005 in China. The concentrations of HCB ranged from 7.5 ng/g to 319 ng/g dry wt., with an average of 145 ng/g dry wt. HCB levels in sewage sludge were significantly higher than those in the river sediments (Table 5) in China, which indicated that wastewater was also a pollution source of HCB.

4. Human exposure

4.1. HCB residues in food and human exposure

There were some studies on HCB residue levels in various food groups in China. HCB levels in butter collected from 23 countries were determined, with particularly high levels recorded in the sample (6.15 ng/g lipid wt.) from China (Kalantzi et al., 2001). The dietary intakes of HCB by the general population were evaluated in some studies after 2000 (Table 8). These studies focused on eastern China. In all the studies, the estimated daily intakes of HCB were much lower than Canadian tolerable daily intake (270 ng/kg body weight/day), indicating that the intakes would not pose a health risk in China at the present (Nakata et al., 2002; Wei et al., 2006; Yang et al., 2006;

Table 7
HCB concentrations in fish and shellfish in China.

Location	Range/average (ng/g lipid wt.)	Sample types	Sampling time	Reference
Taihu Lake	11–37	Carp, catfish, spotted steed topmouth culter and shrimp	2000.8	Nakata et al. (2005a)
Han Zhou Bay	<0.01–61 (16)	11 species of fishes and shrimps	2001.9	
Along the Coast of China	<1–12	Mussels (<i>Mytilus edulis</i> and <i>Perna viridis</i>)	2003–2005	Ramu et al. (2007)
Hong Kong	<1			
Along the Coast of China	<0.5–540 (56)	<i>Mytilus edulis</i> and <i>Perna viridis</i>	1999–2001	Monirith et al. (2003)
Hong Kong	<0.50–<1.5 (<1.0)		1998–1999	
East China Sea	<0.16–7.0 (2.1)	Skipjack tuna liver	1997	Ueno et al. (2003)
South China sea	2.2		2001	
Yangtze River	224–2600	Chinese sturgeon muscle	2003–2005	Wan et al. (2007)
Dong shan	140–230	Blubber of finless porpoises	1990	Ramu et al. (2006)
Hong Kong	87–250		2000–2001	

Table 8Food residue levels and estimated dietary intake (EDI) of HCB in China.^{a,b,c}

Location	Food group	Range/average (ng/g)	HCB EDI (ng/kg bw/day)	Sampling time	Reference
Pearl River Delta	Mussel and oyster	0.30–26.8 ^a	1.4×10^{-3} –8.0	2003.3	Wei et al. (2006)
Local supermarkets of Dalian, Tianjin Shanghai	Shrimp	<0.05–1.14 ^a			
Pingqiao, Zhejiang province	Fish and shellfish	0.01–2.28 ^b (0.38)	0.19	2002.4	Yang et al. (2006)
	Rice	44.7–83.5 ^a (median: 52.1)	0.3	2003.10–2005.6	Zhao et al. (2007)
	Hen egg	0.03–536 ^a (median: 135)			
	Crucian carp	0.03–11.8 ^a (median: 1.70)			
Luqiao, Zhejiang province	Rice	29.8–78.1 ^a (median: 70.6)	0.3		Zhao et al. (2007)
	Hen egg	0.15–396 ^a (median: 107)			
	Crucian carp	0.03–2.71 ^a (median: 0.39)			
Shanghai and its vicinity (Yixing)	Seafoods	<0.01–0.24 ^b	46/60 = 0.77	2001.8–2002.4	Nakata et al. (2002)
	Meat	<0.01–0.37 ^b			
	Cereals	<0.01 ^b			
	Fruits	<0.01–0.04 ^b			
	Vegetables	<0.01–0.75 ^b			
	Tea leaves	0.79–3.1 ^b			
	Others (mud snail, crawfish milk sunflower seed egg pulse oil)	<0.01–1.0 ^b			
Local market in Zhoushan City	Fishes	0.07–2.0 ^b	Not available	2003.10	Jiang et al. (2005)

^a Data base on dry weight.^b Data base on wet weight.^c Canadian HCB Health guidelines (270 ng/kg body weight/day).

Zhao et al., 2007). They were also lower than the daily intakes of HCB in India (1989), Thailand (1988), Vietnam (1990), and Japan (1992) (Kannan et al., 1997). However, the cancer risks associated with fish consumption based on 95th centile concentrations of HCB were greater than unity, suggesting that daily exposure to HCB due to fish consumption had a lifetime cancer risk of greater than one in one million in Zhoushan city (Jiang et al., 2005). It proves that HCB levels in foods need to be taken into consideration of potential health risk in China, although the risks are low at the present.

4.2. HCB residues in human milk and tissue

The studies on HCB concentrations in human tissues in China mainly focused on human adipose tissue and human milk (Table 9). HCB levels in human adipose tissues in Guizhou province (Nakata et al., 2005b) and Shanghai city (Nakata et al., 2002) were comparable to those in Japan (Minh et al., 2000) and Mexico (Waliszewski et al., 1999) and slightly higher than those in the United States (Archibeque-Engle et al., 1997) and Korea (Kang et al., 1997). Most of human milk samples were collected from eastern China during the period 1987–2005. HCB could be detected in all the studies, but HCB levels in various areas ranged widely (Kunisue et al., 2004; Zhao et al., 2007). Human milk from some cities such as Changsha, Shijiazhuang had high HCB concentrations (Cao et al., 2000; Sun et al., 2005). HCB levels in China were relatively higher compared to those in the US and other Asian countries (Tanabe and Kunisue, 2007; Johnson-Restrepo et al., 2007). It is concerned that estimated daily intake (0.15 ng/g body wt./day) of HCB by infants from breast milk was close to tolerable daily intake (0.27 ng/g body wt./day). Temporal trends of HCB in human milk have been reported in China. HCB levels in milk decreased obviously in Beijing from 1988 to 2002 and in Changchun city from 1987 to 1998 (Li et al., 2000; Yu et al., 2005a), while increased by 60% in Changsha from 1988 to 1998 (Cao et al., 2000). The differences in temporal changes of HCB may be related to HCB emissions of point sources in China.

5. Conclusion

In China, the characteristics of HCB production and usage were different from other countries. HCB was produced from 1958 to 2003 in China, although it was banned in most of countries before the 1980s. Large amount of HCB was produced in China. Most of HCB was used as an intermediate for producing PCP and PCP-Na. There was no direct use

Table 9

HCB residue levels in human milk and tissue in China.

Location	Type of human tissue	Range/median (mg/kg lipid wt.)	Sampling year	Reference
Beijing	Human milk	Median: 0.02–0.07	1988–2002	Yu et al. (2005b)
Changsha	Human milk	0.01–0.24 (0.06)	1988	Cao et al. (2005b)
Changchun	Human milk	0.06–0.81 (0.19)	1988	Cao et al. (2000)
		0.22	1987	Li et al. (2000)
Dalian	Human milk	0.0595 (0.06)	1998	Kunisue et al. (2000)
		0.03–0.22 (mean 0.081 ± 0.046)	2002.11–12	Kunisue et al. (2004)
Shenyang		0.025–0.12 (mean 0.056 ± 0.03)		
Shijiazhuang city	Human milk	0.102	2002.11	Sun et al. (2005)
Tangshan countryside		(G mean: 0.0978)		
		0.0876	2003.08	
Luqiao, Zhejiang province	Human milk	(G mean: 0.0846)		
Pingqiao, Zhejiang Province		0.01–262 ng/g (38.5)	2003.10–2005.06	Zhao et al. (2007)
Guizhou Province, southern China	Human adipose tissues	23.6–65.8 ng/g (48.2)		
Shanghai	Human adipose tissues	7.6–100 ng/g (mean: 40 ± 19)	2002	Nakata et al. (2005b)
		28–127 ng/g (mean: 64 ± 37)	2001	Nakata et al. (2002)

of HCB in agriculture in China. Moreover, most PCP-Na produced through HCB was used as a molluscicide in limnological regions of south China. Some chemical plants producing and using HCB caused serious HCB pollution. There are some potential unintended HCB emission sources but little information of detailed emission from these sources in China. Pesticides application may be an important HCB emission source because of their high emission factors and vast use. Other sources cannot be omitted due to the uncertain factors associated with such emissions. Because of the prohibition of HCB, PCP-Na and PCP, technical innovation and implementation of stricter environmental regulations, HCB emission is expected to reduce in China.

HCB is widely distributed in various environmental compartments in China. HCB can be detected even in remote areas. HCB levels increased from South China to North China in most of environmental compartments (air, soil, and mussel). Except for high HCB levels in air and human milk, HCB levels in other environmental compartments in China are similar to those in Europe and eastern Asia from regional perspective. Moreover, HCB levels in air and shellfish tend to much variation, which are different from the uniform distribution in Europe and eastern Asia, indicating that there are some primary emission sources of HCB in China. High HCB concentrations in air, soil and vegetation are found near industrial areas, especially the plants producing and using HCB.

In general, HCB in food and human milk is not expected to pose a health risk in China at the present, although HCB levels in human milk and some foods were higher than those in most of other countries. However, the increasing trend of HCB levels in human milk in some areas needs to be taken into consideration. In addition, long-term exposure to HCB should not be overlooked.

In the limited studies, the temporal trends of HCB levels in the environment in different areas were various. HCB levels in some areas were decreased obviously, but increased strikingly in other areas. In addition, air HCB levels in the colder season were much higher than the warmer season. Some sources of HCB emissions in colder season may be responsible for the elevated levels.

Further studies need to be performed on HCB emission inventories in China, HCB levels in central China and western China, and temporal trends of HCB levels in China. Direct regulations and policies for control over HCB emissions should be developed based on the results of the studies on HCB.

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