Persistent Organic Pollutants (POPs) in Sea food of China- A review

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Abstract: The coastal zone of China has been undergoing rapid economic growth in the past few years. Large quantities of persistent organic pesticides have been either used in agriculture and public health or released from manufacturing plants in this region. Weak environmental management framework has over the years permitted improper waste disposal use and disposal of pesticides have caused serious environmental problems. This paper attempts to review the state of sea food contamination and evaluate its risk to consumers in China using recent research data. The daily intake of Organochlorine pesticides (OCPs) and Polychlorinated biphenyles (PCBs) ingested by people living in coastal cities in China through fish and shellfish was also estimated. Risk assessment against various standards showed that seafood products have been contaminated by Dichlorodiphenyltrichloroethane (DDTs) and may pose health threat to local residents and the consumers due to the fact that China has been a major producer and consumer of DDTs in the past, and also uses DDT for vector control, resulting in higher background levels of DDTs in different ecological compartments. It is recommended to impose a tighter control on the use of DDT for vector control as well as for agricultural application, conduct regular monitoring of DDT concentrations in different ecological compartments. However, most research in coastal area is limited to a few kinds of POP compounds. [Journal of American Science 2009; 5(5):164-174]. (ISSN: 1545-1003).

Key words: POPs, Risk Assessment, Coastal Zone

1. Introduction

Organochlorinated substances are listed as Persistent Organic Pesticides (POPs) by the United Nations Environment Program (UNEP) in the 1995 Stockholm Convention. Twelve POPs were identified of which, nine are pesticides (aldrin, endrin, dieldrin, heptachlor, chlordane, mirex, toxaphene, Dichlorodiphenyltrichloroethane (DDT) and hexa chlorobenzene (HCB) (Wei et al. 2007)**.** The other substances are industrial chemical products or byproducts including Hexachlorocyclohexane (HCH), Polychlorinated biphenyles (PCBs) and Polychlorinated dibenzo-p-dioxins and furans. Due to their persistence, bioaccumulation, and adverse effects on wildlife and human, production and use of these chemicals were banned in the early 1970s in developed countries (Loganathan and Kannan, 1994).

Large amounts of Organo Chlorine Pesticides (OCPs) were used in past decades to obtain high yield to sustain overpopulation in China. Even after the ban of technical HCH and DDT in 1983, 3200 t of lindane (almost pure γ-HCH) was still in use between 1991 and 2000, and DDT production also continues due to export demand and dicofol production (Zhang et al. 2002; Qiu et al. 2004; Tao et al. 2005).During past few years, a number of surveys and studies on OCPs and PCBs in various environment phases has been conducted in China (Wu et al. 1999; Zhou et al. 2000; Zhou et al. 2001; Bi et al. 2002 ; Monirith et al. 2003). The foodstuffs, especially meat, fish and dairy products are important routes of exposure to organic contaminants for human (Yang et al. 2006; Harrison et al. 1996; Dougherty et al. 2000).

The rapid socio-economic development during the past two decades in China, especially the coastal area has given rise to severe economic, environmental and health problems (Wong et al. 2005). According to Wong et al. (2005) and National Implementation Plan (2007), the most pesticide application areas belong to eastern and southern area of China. Many investigation reports have been documented that these contaminants might be transported widely through aquatic environment. Fisheries and Aquaculture activities have been successfully done along the coastal zone of P.R.China. China has been the world's largest producer and exporter of fishery products since

2002(Guo et al. 2007). Fish and shellfish are important food for supplying essential trace elements and certain vitamins; moreover, the polyunsaturated *n*-3 fatty acids in fatty fish species are biologically important and have been associated with a decreased risk for cardiovascular disease (Svensson et al. 1995; Kromhout et al. 1985). This is more important in China, where human dietary habits are changing and widening with the ongoing rapid economic development and changing facets in life-styles. While consumption of grains and vegetables decreased from 1989 to 1997, consumption of meat, fish and dairy products increased during this period. In 1997, consumption of meat, fish and dairy products accounted for about 21% of foodstuffs consumed by Chinese and fish consumption was an important portion of them (Du et al. 2004). Given the importance of China's seafood products to the human health, information regarding the state of OCPs and PCBs contamination in seafood products is needed in order to evaluate risk of exposure to the contaminants on consumers. Therefore, this paper attempts to review the production, exploitation of pesticidal POPs and PCBs in China and the distribution and the fate of them in marine matrix based on recent research findings in coastal zone in China with emphasis on potential human health risk related to sea food consumption.

2. Production and Exploitation of POPs in China

Being one of the largest agricultural production countries, China has been a major producer and consumer of organochlorine pesticides, until their ban on production and agricultural use were enforced (Yang et al. 2004). In China ten pesticidal POPs were recorded. Seven of them were DDT, HCH, toxaphene, hexachlorbenzene (HCB), chlordane, heptachlor and mirex and they were produced at an industrial scale , and the other three (aldrin, dieldrin and endrin) were produced, only at a pilot plant level, or in research phase during 1950s to 1980s.

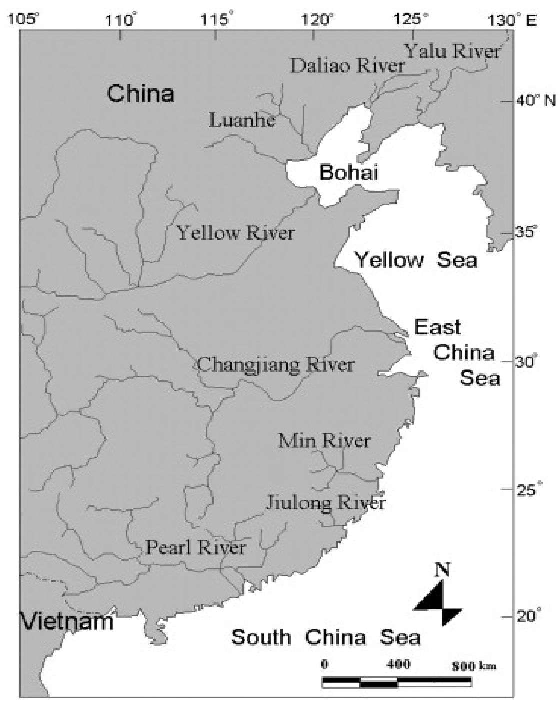

Figure 1. The map showing the coastal zone in China

2.1 Dichlorodiphenyl trichloroethane (DDTs)

The Organochlorine Pesticide DDT was one of the first synthetic chemicals to be produced in large quantities in China for the purpose of agricultural and disease vector control and dispersed widely in the environment (Li et al., 2005). It was first produced and mainly used in agriculture in 1951 (Wong et al. 2002) for controlling army worm, ball worm, pink ball worm, apple tortrix moth, greenish brown hawk moth on wheat, maize, cotton, orchard, soybean and sorghum (Cai et al. 1992).

The total production of commercial DDT was more than 430 kilo tons (Wong et al. 2002) until its agricultural use was banned in 1983 (Cheng, 1990). Before the ban, China was the third largest consumer of DDT for agricultural purposes (Figure 2) after US and Former Soviet Union and fifth consumer country for overall usage of the chemical (Figure 3). The importation of DDT has also been banned in China since 1994 (Wei et al. 2007). Since 1995, the output of technical grade DDT in China has been maintained at the level of 5,000 - 6,000 tons/year, and the output in 2004 was 3,945 tons (NIP, 2007).However, there remain only two enterprises producing technical grade DDT and one enterprise producing DDT preparations.

Small amount of DDT currently produce in China under the exemption of Stockholm Convention is to use as the intermediate in dicofol production, to export for disease vector control in the tropical regions where malaria breaks out heavily such as Southeast Asia and Africa, and to use in antifouling paint.

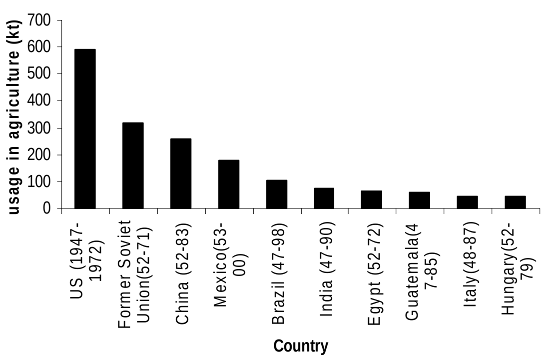

Figure 2. The top 10 countries with historical highest DDT use in agriculture (Source: Li, 2003a, 2003b)

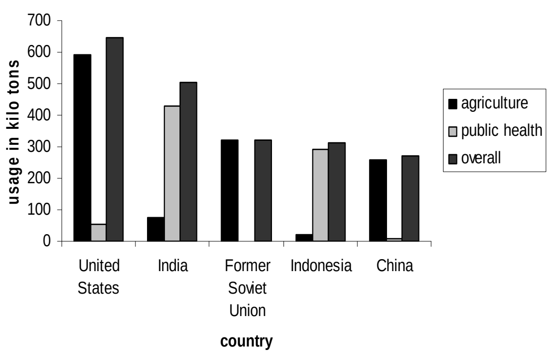

Figure 3. Top 5 countries for overall DDT use (Source: Li et al. 2004)

2.2 Hexachlorohexane (HCHs)

The Organochlorine Pesticide HCH has two formulations as technical hexachlorocyclohexane (HCH) and lindane. Technical-grade HCH consists principally of five isomers, α-HCH (60–70%), β-HCH (5–12%), γ- HCH (10–15%), δ-HCH (6–10%) and ε -HCH (3–4%) (Walker et al.1999).

China has consumed the highest amount of technical HCH (Figure 4), accounting for almost half of the total global usage, followed by India to kill pests on rice, wheat, maize, cotton, soybean, sorghum, orchards and some vegetables. Among them, more than half of HCH was used in rice paddies, 25% on wheat, and 10% on each of soybean/sorghum and maize (Cai et al. 1992). China started to produce and use technical HCH in 1952 and its use was banned in 1983. The total amount of technical HCH produced in China was 4.5 million tons by 1983 (Li et al. 1998b). Although technical HCH is no longer used, applications of lindane continue in many countries including China. The total lindane usage between 1970 and 1993 was 720 kilo tons (Voldner and Li, 1995).

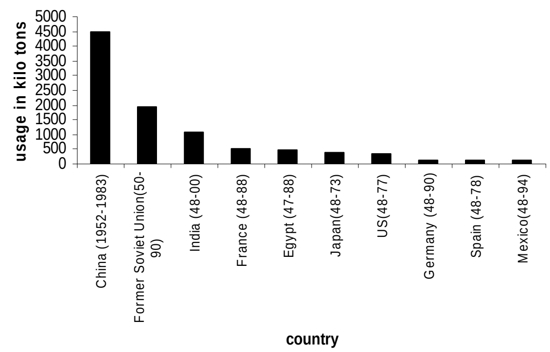

Figure 4. Top 10 countries with highest technical HCH use (Li et al., 1999a, 2004a)

2.3 Chlordane and Mirex

Chlordane and Mirex were used in China due to a lack of highly efficacious and low cost termiticites. China started to produce and use Chlordane in 1950s and the highest production could observe in 1999 accounting 520.6 tons (NIP, 2007). The production of mirex started in 1960s and the highest production could observe in 2000 accounting 31 tons (NIP, 2007).

From 1995 to 2003, about 5000 tons of chlordane and 140 tons of mirex were produced, especially, within 5 years from 1997 to 2001, 2300 tons t of chlordane and 14 tons of mirex was used in China (WB, 2005). Most of these pesticides may have been used in the south and south east China, as most of termites severely affected areas belong to these areas.

2.4 Hexachlorbenzene (HCB)

China began to produce HCB in 1958 and in total there were six enterprises. These enterprises used HCHs to produce HCBs and lindane. Due to control of HCH production, the production of HCB was reduced drastically. In 1990, the maximum HCB output of the whole country was 7365 tons. Production was reduced year by year after 2000 and completely stopped in 2004 (NIP, 2007).HCB has not been used as a direct pesticide in China, but as an intermediate for the production of other chlorinated substances such as Na-PCP which used to be employed for schistosomiasis prevention and control.

2.5 Toxaphene, heptachlor and other OCPs

Toxaphene was broadly used for cotton pests, fruit trees pests and maize borer prevention (Wei et al. 2007). The production was started in 1970. The highest production was in 1973 accounting 3740 tons (NIP, 2007).

Heptachlor was mainly used for soil pests, controlling termites in railway crossties and vector pests' prevention. The production was started in 1967 and it was stopped in 1982 (UNEP, 2002). The maximum amount used (20 tons) identified between 1967 and 1978. However, these two chemicals have not been produced or consumed because they were banned in 1982.No production carried out for aldrin, dieldrin and endrin at large scale in China, and the research or trial production has been stopped (NIP, 2007).

2.6 Polychlorinated biphenyles (PCB)

Polychlorinated biphenyls (PCBs) are amongst the more dangerous environmental contaminants due to their persistence, bioaccumulative properties and toxicity. PCBs have been used worldwide as plasticizers, hydraulic and dielectric fluids, fire retardants and paint additives (Kennisch, 1996). The production of PCBs oils began in 1965 in China (NIP, 2007). During the 1950's and the 1980's, China used to import PCBs-containing electrical equipment from other countries without being informed, most of which were specific transformers and capacitors for large facilities. (Jiang et al. 1997) reported that approximately 8000 tons of PCBs were produced under trade name 1 PCB and 2 PCB during period of 1960s–1970s. Most of them were used as dielectric fluids in electrical appliances and a small portion was used as additives in paints. Even today, large proportion of the original amounts of PCBs, still remain in old transformers and capacitors (Jiang et al. 1997). In January 1974, the Chinese government promulgated the decree on stopping production of capacitors with PCBs, as well as the decree on restricting import of electrical equipment containing PCBs.

3. Sources of OCPs and PCBs in Coastal Zone in China

The coastal zone of China comprises an area of more than three million square kilometers covering, four major Seas; Bohai, Yellow, East China and South China Sea (Figure 1). The area possesses an 18000-km coastline stretching across tropical, subtropical and temperate zones (Cao et al. 2007). The coastal zone is an interface between the land and sea, which comprises of a continuum of coastal land, intertidal area, aquatic systems including the network of rivers and estuaries, islands, transitional and intertidal areas, salt marshes, wetlands, and beaches. Fifty seven per cent of China total population comprise in East, Central and South China (NIP, 2007). Rapid industrialization, urbanization, and conversion of massive agricultural lands to commercial use have accelerated environmental deterioration in coastal region (Guo et al. 2007). Large amount of electrical waste are being carried from many other places to coastal areas in China (Zhao et al. 2007). Since China has been a major producer of seafood, coastal area consists of considerable amount of boats and they are being used antifouling chemicals. Hence, the major sources of OCPs and PCBs in the coastal zone in China are riverine exports of agricultural chemicals from coastal catchments, high shipping activities, heavy manufacturing effluent discharge, and municipal and industrial sewage disposal practices of low standard, used electrical items and atmospheric deposition.

Once discharged into the ocean, these chemicals disperse into three phases, namely water, sediment and biota (Pandit et al. 2002). These contaminants then accumulate in the sediment-dwelling organisms which may be transferred to higher trophic levels through the food chain (Lee et al. 2001).

4 Contamination levels of sea food in coastal zone

4.1 Increasing trend of DDTs towards Pearl River Delta, South China Sea

Many research activities have been carried out in China to investigate the OCPs in marine organisms (Table1). The results showed that the fish/shell fish species in Chinese coastal zone is contaminated by OCPs. DDTs were at the top level. A recent report launched in Asian countries revealed that the contents of DDTs in the marine benthos along the coastal areas of China were at the top level (Monirith et al. 2003). The highest DDTs could observe in the South China Sea. Results from (Monirith et al. 2003; Chen et al. 1996, 2000, 2002; Fang et al. 2001; Phillips, 1985; Tanabe et al. 1987) also showed that South China is the most contaminated coastal sea among the major seas in China. There are possible reasons behind this observation. The coastal area of Guangdong province in South China has 60,000 fishing ships, which is above 1/5 of the total number in China. It can be estimated that about 30–60 tons of DDT may be introduced to the coastal environment of Guangdong, including the Pearl River Delta. According to Fu et al. 2003, the Pearl River is believed to carry a considerable load of chlorinated pesticides, up to 863 tones per annum, which is the highest amongst China's rivers. High ratio of DDT/(DDD+DDE) in sediment (Hong et al. 1995; Zhang et al. 2001 ; Mai et al. 2002), as well as water (Zhou et al. 2001; Luo et al. 2004) samples indicated the relatively recent releases of DDT. Previous studies from (Zhang et al. 2002; Luo et al. 2004; Zhou, 2004; Chen et al. 2006) also showed that there were new inputs of DDTs in the PRD. However, comparatively lower level of DDTs found in marine organisms collected from East coast. But the survey of the National Bureau of Coastal Zone Protection during 1980–1987 showed that the organochlorines flux just carried by Yangtze River (the longest river in China) was 239.3 tons per year, which accounted for 19.8% of the total flux by Chinese river catchments into the marine coastal sites. Bohai Sea, Northern China collects pollutants from major rivers namely, Yalu, Daliao, Luanhi and Yellow (Figure 1) and showed comparatively intermediate levels of DDTs among the recent researches. Wu et al. (1999) noted high concentrations of DDTs in the river sediments from Northern China where a factory with high manufacturing capacity of DDT is located.

Table 1. The concentrations of OCPs and PCBs in marine organisms in ng/g (wet weight basis, dry weight basis in parenthesis; Wet weight = 0.16 * Dry weight (Ramesh et al. 1990)) in China.

Coastal region	DDTs	HCHs	CHLs	HCBs	PCBs	Reference
east Xiamen Island, South Eastern China	(75.2 - 2143)	(0.18 – 345)			(n.d. – 234)	Chen et al. (2002)
Minjing Estuary, South Eastern China	(21.5–2396)	(n.d.-5.07)			(n.d.-6.78)	Chen et al. (2002)
South coast, China	(150–200)					Klumpp et al. (2002)
South China Sea	65.70	< 1.5				Guo et al. (2007)
PRD	(4.1 - 7840)	(10.8)				Guo et al. (2008)
East coast, China	(14.4–640)	(0.17–9.91)	(0.13–1.86)		(1.34–13)	Fung et al. (2004)
Bohai Sea	29.40	1.27				Yang et al. (2004)
Dalian, Tianjin and Shanghai	28.9	0.92	0.47	0.38		Yang et al (2006)
northeast coast	(54.8–2680)	(1.42–25.5)	(n.d.–2.28)		(3.27–25.4)	Jin et al., 2008

4.2 Reasons for the different levels of other OCPs and PCBs

The results indicate that the HCHs concentrations were below than concentrations of DDTs. Historically, the usages of technical HCHs were much more (Figure 4) than those of DDTs in China. The discrepancy between the usages of HCHs and DDTs and their accumulative levels in seafood products may be due to the difference in physicochemical and biochemical properties between HCHs and DDTs, where in HCHs have higher biodegradability and lower lipophilicity compared to DDTs (Guo et al. 2007). The PCB levels in some areas showed high levels while other places had low levels. The physicochemical properties of PCBs vary widely and depend on the number and position of chlorine atoms in biphenyl rings. Vapor pressure, water solubility and biodegradability decrease with increasing number of chlorine atoms. In contrast, lipophilicity and adsorption capacity show a reverse trend (Loganathan, 1994). Therefore, high levels of PCBs in body tissues explained by accumulation of low chlorinated PCB congeners. PCBs were banned in 1983, yet a large proportion still remains in use at present in older transformers and capacitors. The high assimilative and self-purification capabilities of the estuary against anthropogenic activities and pollution impact via large runoff discharge during wet season and enormous sediment loads might be the major abating factors for high PCBs. However, (Jiang et al. 1997) reported that in the previous decades, only about 8000 tons of PCBs were produced in China. The chlordanes were also observed in marine organisms but the level was low. It was reported that technical chlordane is still being used in China against termites (Xu et al. 2004; Nakata et al. 2005), and trans-chlordane, cis-chlordane, trans-nonachlor are dominant constituents in technical chlordane (Kawano et al. 1988; Kawano et al. 1992 ; Xu et al. 2004).

4.3 Comparison the OCPs and PCBs in Chinese seafood with different countries

Table 2 summarizes the OCPs and PCBs analyzed in fish/shell fish species in different countries. P.R.China shows the highest DDT value among the sea foods in different countries. The reason behind this statement is high back ground levels in history. A substantial number of studies have focused on the contamination by POP pesticides in different ecological compartments in China, with pesticide residues remaining highly abundant in soils and crops (Wong et al. 2005). These contaminants ultimately reach to coastal environment through water bodies. In contrast, studies on the levels of POPs in the global environment show that emission sources of a number of POPs (including DDT) in the last 20 years have shifted from industrialized countries of Northern Hemisphere to less developing countries in tropical and sub-tropical regions including India and China (Wong et al. 2005). This may be due to the late production ban otherwise DDT is still being used in agriculture and for the control of disease such as malaria, typhus and cholera (Iwata et al. 1994 ; Loganathan and Kannan, 1994). But in tropical environment, these POPs compounds biodegrade and volatilize soon due to high temperatures. Therefore, the levels become reduced (Table 2). Hong Kong also shows comparatively high DDT and those values are implying that South China Sea is receiving DDT residues. Li et al. (2006) reported that higher concentrations of HCHs and DDTs in water, sediment, fish, and human breast milk in Hong Kong where no HCHs and DDTs were presently in use. This may due to the dispersion of OCPs in marine matrices and in turn to the uptake of OCPs via aquatic food chain.

The highest PCBs, HCHs, CHLs and HCBs concentrations in Table 2 are in the samples collected from Russia, India, Japan and Malaysia respectively. In the former USSR, technical PCBs mixtures have been used and produced as a dielectric fluid in the manufacture of power capacitors and transformers (Ivanov and Sundell, 1992). Therefore high PCBs level could explain by the presence of local PCB sources. In history, China was the top in use of HCHs. But in the present, the reason for the high level of HCHs in the marine organisms of India may be the usage of certain amount of technical HCHs for public health purposes and on certain food crops after its ban for agriculture in 1983 (Li et al. 1998). The chlordanes had been used largely for termite control until in 1986 in Japan (Loganathan et al. 1993) and high levels in marine organisms implied that they may be still discharged into the marine environment. HCB is not only used as a fungicide, but also generated as a byproduct during the production of agrochemicals and industrial chemicals (Monirth et al. 2003). Furthermore the HCB has been released to the environment by waste incineration (Van-Birgelen, 1998). Those reasons could account to the high levels of HCBs in Malaysia. However, the low residual levels of these pesticides and their low frequency of detection in Chinese coastal environment may be attributed to their relatively low residual levels in coastal environment and in seafood products in China or their relatively low potential for bioaccumulation in the species under consideration.

Table 2. The concentrations of OCPs and PCBs in marine organisms in $ng^{-1}g$ (wet weight basis, dry weight basis in parenthesis; Wet weight = 0.16 * Dry weight (Ramesh et al. 1990)) in different coastal regions in the world.

Country/coastal region	DDTs	HCHs	CHLs	HCBs	PCBs	References
China	240	0.80	3.0	1.3	2.5	Monirith et al. (2003)
Perth (western Australia)	0.2					Sericano et al. (1993)
Taiwan (China)	(0–121)	(0–7)				Ling and Teng (1997)

The marine coastal (USA)	(0.51–27.9)					O'Connor and Beliaeff (1998)
The coast of north Vietnam	(12.0 - 23.3)				(5.1- 25.3)	Nhan et al. (1999)
Taiwan-						Cheng Han et al. (2000)
Machu	(340)					
Kim-man	(337)					
Coastal water of Thailand	0.05–5.7		0.22–12.0			Boonyatumanond et al. (2002)
Korea entire coast	3.13	0.98	0.62	0.04	3.90	Kim et al. (2002)
Japan	3.5	0.32	6.0	0.08	30	Monirith et al. (2003)
India	4.2	2.0	0.6	0.02	3.8	Monirith et al. (2003)
Black sea coast	<0.12-14			0.364		Ozcok et al. (2007)
Egypt	98.1-629.8					Khalid et al. (2004)

5. Health Risk Assessment

Human epidemiological surveys have been proven that the exposure to chlorinated pesticides make adverse effects on human health (Ribas-Fito et al. 2003 ; Longnecker et al. 2001; Cooper et al. 2004). Table 3 summarizes the acceptable daily intakes of these pesticides issued by some authorities. The results revealed that the DDTs are the predominant contaminant. Most of the studies showed higher levels of DDTs than the Chinese government's first level criterion (10 ng/g) for marine biological quality (GB-18421-2001) (Chen et al. 2002) but conformed to the first level criterion (20 ng/g and 50 ng/g) for HCHs and PCBs respectively. DDT is highly persistent in the environment with a half life of 2–15 years (USEPA, 1989). In human fatty tissue, the half life of DDT has been reported to be 7–8 years (NACEC, 2001). To assess the human health risk, the estimated daily intakes (EDI) were calculated using the average values per each pollutant and the value for the consumption of fish and seafood in China (30.5 g/person/day) in 1997 (Du et al. 2004). The results (Table 4) reveal that the estimated daily intakes (EDI) for DDTs in South China Sea exceed the maximum admissible DDTs concentration established by the European Union which is more feasible value from the farmer's point of view. Hence, the sea food in South China could consider overloaded with DDTs. However, the exposure levels depend on the lipid content of the fish and on the amount of seafood consumed.

Table 3. The acceptable limit of certain POPs pesticides for humans (ng/g wet weight)

Authority	DDTs	HCHs	PCBs	Reference
Food and Agricultural Organization, World Health Organization	20	8		FAO/WHO (1993)
European Union	50	10		Binelli and Provini, 2003

Table 4. Estimated daily intakes (EDI) of PCBs, DDTs and HCHs through sea food by human (average body wt. 60 kg) in China. EDI (ng /kg /day) = [daily fish consumption (g /day)] × [mean OCP concentration (ng/g wet wt.)]/ [human body weight (kg)].

Pollutant	Average concentration (ng/g wet wt.)	EDI (ng/kg body wt./day)	ADI (FAO/WHO) (ng/kg body wt./day)
PCBs	China : 2.5	1.27	
DDTs	South China: 65.70	33.39	20
	Bohai Sea : 29.40	14.94	
HCHs	South China : < 1.5	0.76	8
	Bohai Sea : 1.27	0.64	

The results confirm that the concentrations of HCHs and PCBs in sea foods are far below to all the maximum admissible limits establish by different authorities. (Nakata et al. 2002) reported that the levels of HCHs in foodstuffs (including aquatic products) dramatically declined during the last 30 years. Moreover, low residual levels of HCHs suggest that HCHs are no longer an environmentally significant organic contaminant in seafood products from China.

Conclusion

Sea foods in Chinese coastal zone are contaminated with OCPs. Risk assessment against various standards clearly showed that seafood products in South China Sea are contaminated by DDTs and may pose health threat to local residents and the consumers if they totally rely on sea foods as their prime animal proteins. A national wide survey is needed to investigate the intake of OCPs, due to dietary differences, with a strong focus on the more sensitive populations, e.g., coastal residents who consume a large amount of fish.

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References

[1]Bi X, Chu S, Meng Q, Xu X. Movement and retention of polychlorinated biphenyls in a paddy field of WenTai area in China, Agriculture**.** Ecosystems and Environment 2002; 89: 241–252.

[2]Binelli A, Provini A. POPs in edible clams from different Italian and European markets and possible human health risk. Marine Pollution Bulletin 2003; 46:879–886.

[3]Boonyatumanond R, Jaksakul A, Puncharoen P, Tabucanon M S. Monitoring of organochlorine pesticides residues in green mussels (*Perna viridis*) from the coastal area of Thailand. Environmental Pollution 2002; 119: 245–252.

[4]Cai D J, Sun L J, Ke J L, Tang G C. Technical report RP032: pesticide usage in China, Ontario, Environment Canada, Canada. 1992.

[5]Cao W, Wong M H. Current status of coastal zone issues and management in China: A review. Environment International 2007; 33(7): 985-992.

[6]Chen L G, Mai B X, Bi X H, Ran Y, Luo X J, Chen S J, Sheng G Y, Fu J M, Zeng E Y. Concentration levels, compositional profiles, and gas-particle partitioning of polybrominated diphenyl ethers in the atmosphere of an urban city in South China. Environmental Science and Technology 2006; 40: 709–714.

[7]Chen W, Zhang L, Xu , Wang X, Hong L, Hong H. Residue levels of HCHs, DDTs and PCBs in shellfish from coastal areas of east Xiamen Island and Minjiang Estuary, China. Marine Pollution Bulletin**.** 2002; 45(1-12): 385-390.

[8]Chen W, Hong H, Zhang L, Xu L, Wang X, Hong, L. Residue levels and distribution features of organochlorine pollutants in the surface sediments of the sea areas between Minjiang Estuary and Mazu. Marine Science Bulletin. 2000; 19: 53–58 (in Chinese).

[9]Chen W, Zhang L, Xu L, Wang X, Hong H. Concentrations and distributions of HCHs, DDTs and PCBs in surface sediments of sea areas between Xiamen and Jinmen. Journal of Xaimen University (Natural Science). 1996; 35: 936–940 (in Chinese).

[10]Cheng Han, B, Lih Jeng W, Chang Hung T, Chien Ling Y, Jer Shieh M, Chu Chien L. Estimation of metal and organochlorine pesticide exposures and potential health threat by consumption of oysters in Taiwan. Environmental Pollution. 2000; 109(1): 147-156.

[11]Cheng, X. The pesticides production of China going to a period of higher efficient and more secure. Pesticides (in Chinese). 1990; 29 (1): 15–19.

[12]Cooper G S, Martin S A, Longnecker M P, Sandler D P, Germolec D R. Associations between plasma DDE levels and immunologic measures in African-American farmers in North Carolina. Environmental Health Perspectives. 2004; 112: 1080–1084.

[13]Dougherty C P, Holtz S H, Reinert J C , Panyacosit L, Axelrad D A, Woodruff T J. Dietary exposures to food contaminants across the United States. Environmental Research. 2000; 84:170–185.

[14]Du S, Zai F, Yang X. Report on nutritional and physical status of Chinese population. Databank of nutritional and physical status of Chinese population. 2004. Available from: http://www.in fh.ac.cn/database.htm.

[15]Fang Z Q, Cheung R Y H, Wong M H. Concentrations and distribution of organochlorinated pesticides and PCBs in green-lipped mussels, *Perna viridis* collected from the Pearl River estuarine zone. Acta Scientiae Circumstantiae 2001; 21**(**1): 113–116 (in Chinese).

[16]FAO/WHO. Food Standards Programme vol. 9 (4). Codex Alimentarius Commission, 1993;149–158.

[17]Fu J M, Mai B X, Sheng G Y, Zhang G, Wang X M, Peng P A, Xiao X M, Ran R, Cheng F Z, Peng X Z, Wang Z S, Tang U W. Persistent organic pollutants in environment of the Pearl River Delta, China: An overview. Chemosphere. 2003 ; 52:1411–1422.

[18]Fung C N, Lam J C W, Zheng G J, Connel D W, Monirith I, Tanabe S, Richardson B J, Lam P K S. Mussel based monitoring of trace metal and organic contaminants along the east coast of China using *Perna viridis* and *Mytillus edulis.* Environmental Pollution. 2004; 127: 203-216.

[19]Guo Y, Meng X Z, Tang H L, Zeng E Y. Tissue distribution of organochlorine pesticides in fish collected from the Pearl River Delta, China: Implications for fishery input source and bioaccumulation. Environmental Pollution. 2008; 155(1): 150-156.

[20]Guo J Y, Zeng E Y, Wu F C, Meng X Z, Mai B X, Luo X J. Organochorine Pesticides in Sea Food Products from Southern China and Health Risk Assessment. Environmental Toxicology and Chemistry. 2007; 26 (6): 1109–1115.

[21]Harrison R M, Smith D J T, Luhana L. Source apportionment of atmospheric polycyclic aromatic hydrocarbons collected from an urban location in Birmingham, U.K. Environmental Science and Technology1996; 30: 825–832.

[22]Hong H, Xu L, Zhang L, Chem J C, Wong Y S, Wan T S M. Environmental fate and chemistry of organic pollutants in the sediment of Xiamen and Victoria Harbours. Marine Pollution Bulletin.1995; 31: 229–236.

[23]Ivanov V, Sundell E N. Characterization of polychlorinated biphenyl isomers in Sovol and trichlorobiphenyl formulations by high resolution gas chromatography with electron capture detection and high resolution gas chromatography mass spectrometry technique. Environmental Science and Technology 1992; 26: 2012-2017.

[24]Iwata H, Tanabe S, Sakai N, Nichimura A, Tatsukawa R. Geographical distribution of persistent organochlorines in air, water and sediments from Asia and Oceania and their implications for global redistribution from lower latitudes. Environmental Pollution. 1994; 85: 15–33.

[25]Jiang K, Li L, Chen Y, Jin J. Determination of PCDD/Fs and dioxin-like PCBs in Chinese commercial PCBs and emissions from a testing PCB incinerator, Chemosphere.1997 ; 34: 941–950.

[26]Jin Y, Hong S H, Li D, Shim W J, Lee S S. Distribution of persistent organic pollutants in bivalves from the northeast coast of China Marine Pollution Bulletin. 2008; 57(6-12): 775-781.

[27]Kawano M, Inoue T, Wada T, Hidaka H, Tatsukawa R. Bioaccumulation and residue patterns of chlordane compounds in marine animals: Invertebrates, fish, mammals, and seabirds. Environmental Science and Technology**.** 1988; 22: 792–797.

[28]Kawano M, Ramesh A, Thao V D, Tatsukawa R, Subramanian A N. Persistent organochorine insecticide residues in some paddy, upland and urban soils of India. International Journal of Environmental Analytical Chemistry. 1992; 48: 163–174.

[29]Kennisch M J. Practical handbook of estuarine and marine pollution. Boca Raton, NY: CRC Press, Marine Science Series. 1996.

[30]Khalid A, Nemr A E, Said T O, El-Sikaily A, ABD-Alla A M A. Polychlorinated biphenyls and chlorinated pesticides in mussels from the Egyptian Red Sea Coast. Chemosphere. 2004; 54: 1407-1412.

[31]Kim S K, Oh J R, Shim W J, Lee D H, Yim U H, Hong S H, Shin Y B, Lee D S. Geographical distribution and accumulation features of organochlorine residues in bivalves from coastal areas of South Korea. Marine Pollution Bulletin. 2002; 45(1-12): 268-279.

[32]Klumpp D W, Hong H, Humphrey C, Wang X, Codi, S. Toxic contaminants and their biological effects in coastal waters of Xiamen, China. I. Organic pollutants in mussel and fish tissues. Marine Pollution Bulletin. 2002; 44: 752–760.

[33]Kromhout D, Bosschieter E B, Lezenne Coulander C. The inverse relation between fish consumption and 20-year mortality from coronary heart disease. New England Journal of Medicine. 1985; 312: 1205–1209.

[34]Lee K T, Tanabe S, Koh CH. Distribution of organochlorine pesticides in sediments from Kyeonggi Bay and nearby areas, Korea. Environmental Pollution. 2001; 114: 207–213.

[35]Li J, Zhang G., Qi S, Li X, Peng, X. Concentrations, enantiomeric compositions, and sources of HCH, DDT, and chlordane in soils from the Pearl River Delta, South China. Science of the Total Environment. 2006; 372:215–224.

[36]Li YF, Macdonald R W. Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Science of the Total Environment. 2005; 342(1-3): 87-106.

[37]Li YF, Li D C. Global emission inventories for selected organochlorine pesticides. Internal report, meteorogical service of Canada, Environment Canada, Toronto, Canada. 2004.

[38]Li YF, Zhulidov JAV, Roberts R D, Korotova LG. Hexachlorocyclohexane use in the Former Soviet Union. Archives of Environmental Contamination and Toxicology**.** 2004a.48: 10–15.

[39] Li YF, Bidleman TF. Correlation between global emissions of hexachlorocyclohexane and its concentrations in the Arctic Air. Journal of Environmental Informatics. 2003a; 1: 52–57.

[40]Li YF Bidleman TF. Usage and emissions of organochlorine pesticides, Canadian Arctic contaminants assessment report II, Indian and Northern Affairs Canada, Ottawa, 2003b:49–70 Chap. A.2.

[41]Li YF. Global technical hexachlorocylohexane usage and its contamination consequences in environment: from 1948 to 1997. Science of the Total Environment 1999a; 232**:** 123–160**.**

[42]Li YF, Cai DJ, Singh A. Technical hexachlorocyclohexane use trends in China and their impact on the environment. Archives of Environmental Contamination and Toxicology1998b; 5: 688–697.

[43]Li YF, Cai DJ, Singh A. Technical hexachlorocyclohexane use trends in China and their impact on the environment. Archives of Environment Contamination Toxicology1998; 35**:** 688–697.

[44]Ling YC, Teng HC. Supercritical fluid extraction and cleanup of organochlorine pesticides and polychlorinated biphenyls in mussel. Journal of Chromatography. 1997; 90: 153–160.

[45]Loganathan BG, Kannan K. Global Organochlorine Contamination Trends: An Overview. Ambio1994; 23 (3): 187–191.

[46]Loganathan BG, Tnabe S, Hidaka Y, Kawano M, Hidaka H, Tatsukawa R. Temporal trends of persistent organochlorine residues in human adipose tissue from Japan, 1928-1985. Environmental Pollution1993; 81: 31-39.

[47]Longnecker MP, Klebanoff MA, Zhou H, Brock JW. Association between maternal serum concentration of the

DDT metabolite DDE and preterm and small-for-gestational-age babies at birth. Lancet2001; 358: 110–114.

[48]Luo XJ, Mai BX, Yang QS, Fu JM, Sheng GY, Wang ZS. Polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides in water columns from the Pearl River and the Macao harbor in the Pearl River Delta in South China. Marine Pollution Bulletin2004; 48:1102–1115.

[49]Mai BX, Fu JM, Sheng GY, Kang YH, Lin Z, Zhang G, Min YS, Zeng EY. Chlorinated and polycyclic aromatic hydrocarbons in riverine and estuarine sediments from Pearl River Delta, China. Environmental Pollution2002; 117: 457–474.

[50]Monirith I, Ueno D, Takahashi S, Nakata H, Sudaryanto A, Subramanian A, Karuppiah S, Ismail A, Muchtar M, Zheng JJ, Richardson B, Prudente M, Due Hue N, Tana TVS, Tkalin A, Tanabe S. Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries. Marine Pollution Bulletin2003; 6 (3): 281–300.

[51]NACEC.Actions for eliminating DDT in Mexico and Central America. Q & A: NACEC, NARAP on DDT. North America Commission for Environmental Cooperation, 2001. (6/4/01)

[52]Nakata H, Hirakawa Y, Kawazoe M, Nakabo T, Arizono K, Abe S, Kitano T, Shimada H, Watanabe I, Li W, Ding X. Concentrations and compositions of organochlorine contaminants in sediments, soils, crustaceans, fishes and birds collected from Lake Tai, Hangzhou Bay and Shanghai city region, China. Environmental Pollution2005; 133 (3): 415–429.

[53]Nakata HM, Kawazoe K, Arizono S, Abe T, Kitano H, Shimada W, Li XD. Organochlorine pesticides and polychlorinated biphenyl residues in foodstuffs and human tissues from China: Status of contamination, historical trend, and human dietary exposure Archives of Environmental Contamination and Toxicology2002; 3: 473–480.

[54]Nhan D D, Am N M, Carvalhoc FP, Villeneuve JP, Cattini C. Organochlorine pesticides and PCBs along the coast of North Vietnam. The Science of the Total Environment1999; 237/238: 363-371.

[55]NIP; The People's Republic of China National Implementation Plan for the Stockholm Convention on Persistent Organic Pollutants. 2007.

[56]O'ConnorTP, Beliaeff B. Recent Trends in Coastal Environmental Quality (Result from the Mussel atch Project, NOAA Silver Spring, MD, 40) 1998.

[57]Ozkoc B, Bakan G Ariman S. Distribution and bioaccumulation of organochlorine pesticides along the Black Sea coast. Environmental Geochemistry and Health2007; 9(1):59-68.

[58]Pandit G G, Sahu S K, Sadasivan S. Distribution of HCH and DDT in the coastal marine environment of Mumbai, India. Journal of Environmental Monitoring2002; 4: 431–434.

[59]Phillips DJH. Organochlorine and trace metals in green-lipped mussels *perna viridis* from HongKong waters: a test of indicator ability. Marine Ecology Progress Series1985; 21:252–258.

[60]Qiu X, Zhu T, Li J, Pan H, Li Q, Miao G, Gong J. Organochlorine pesticides in the air around the Taihu Lake, China. Environmental Science and Technology2004; 38: 1368–1374.

[61]Ramesh A, Tanabe S, Subramanian AN, Mohan D, Venugopa-lan VK, Tatsukawa R. Persistent organochlorine residues in green mussels from coastal waters of South India. Marine Pollution Bulletin1990; 21: 587–590.

[62]Ribas-Fito N, Cardo E, Sala M, Eulàlia de Muga M, Mazón C, Verdú A, Kogevinas M, Grimalt JO, Sunyer J. Breastfeeding, exposure to organochlorine compounds, and neurodevelopment in infants. Pediatrics2003; 111: 580–585.

[63]Sericano JL, Wade TL, Brooks JM, Elliot LA, Fay RR, Wilkinson DL. National status and trends and mussel watch program: chlordane-related compounds in Gulf of Mexico oysters. Environmental Pollution1993; 82: 23–32.

[64]Svensson BG, Nilsson A, Josson E, Schütz A, Åkesson B, Hagmar L. Fish consumption and exposure to persistent organochlorine compounds, mercury, selenium and methylamines among Swedish fishermen. Scandinavia Journal of Work Environmental Health 1995; 21: 96–105.

[65]Tanabe S, Tatsukawa R, Phillips DJH. Mussels as bioindicators of PCB pollution: a case study of uptake and release of PCB isomers and congeners in green-lipped mussels (*perna viridis*) in Hong Kong waters. Environmental Pollution1987; 47: 41–48.

[66]Tao S, Xu FL, Wang XJ, Liu WX, Gong ZM, Fang JY, Zhu LZ, Luo YM. Organochlorine pesticides in agricultural soil and vegetables from Tianjin, Environmental Science and Technology2005; 39: 2494–2499.

[67]UNEP (United Nations Environment Programme), Central and North East Asia regional report, regionally based assessment of PTS, UNEP/GEF. 2002.

[68]USEPA. Pesticide Environmental Fate on Line Summary: DDT (*p*, *p′*). Environmental Fate and Effects Division, USEPA, Washington, DC. , 1989.

[69]Van-Birgelen APJM. Hexachlorobenzene as a possible major contributor to the dioxin activity of human milk. Environmental Health Perspectives1998; 106: 183-188.

[70]Voldner EC, Li YF. Global usage of selected persistent organochlorines. Science of the Total Environment1995; 60/161: 201–210.

[71]Walker K, Vallero DA, Lewis RG. Factors influencing the distribution of lindane and other hexachlorocyclohexanes in the environment. Environmental Science and Technology1999; 33:4373–8.

[72]WB (World Bank) Demonstration of alternatives to chlordane and mirex in termite control, GEF-2359, Document of World Bank. 2005.

[73]Wei D, Kameya T, Urano K. Environmental management of pesticidal POPs in China: Past, present and future. Environment International2007; 07: 894-902.

[74]Wong MH, Choi K, Grosheva E, Sakai S, Shibata Y, Suzuki N, Wang J, Zhou H, Leung A. Central and North East Asia

regional report-Regionally based assessment of persistent toxic substances, UNEP/GEF. 2002.

[75]Wong MH, Leung AOW, Chan JKY, Choi MPK. A review on the usage of POP pesticides in China, with emphasis on DDT loadings in human milk. Chemosphere2005; 60(6): 740-752.

[76]Wu Y, Zhang J, Zhou Q. Persistent orgnochlorine residues in sediments from Chinese river/estuary systems. Environmental Pollution1999; 105: 143–150.

[77]Xu D, Zhong W, Deng L, Chai Z, Mao X. Regional distribution of organochlorinated pesticides in pine needles and its indication for socioeconomic development. Chemosphere2004; 54: 743–752.

[77]Yang N, Matsuda M, Kawano M, Wakimoto T. PCBs and organochlorine pesticides (OCPs) in edible fish and shellfish from China. Chemosphere2006; 63(8):1342-1352.

[78]Yang N, Matsuda M, Kawano M, Wakimoto T. PCBs and organochlorine pesticides (OCPs) in edible fish and shellfish from China. Chemosphere2006; 63(8): 1342-1352.

[79]Yang R Q, Yao Z W, Jiang G B. Zhou Q F, Liu J Y. HCH and DDT residues in molluscs from Chinese Bohai coastal sites. Marine Pollution Bulletin2004; 48(7-8): 795-799.

[80]Zhang G,Parker A, House A, Mai B, Li X, Kang Y, Wang Z. Sedimentary records of DDT and HCH in the Pearl River Delta, South China. Environmental Science and Technology2002; 36:3671–3677.

[81]Zhang G, Qi S, Parker A, Li X, Li J, Wang X, Distribution of organochlorine pesticides and polycyclic aromatic hydrocarbons in soils from the Pearl River Delta, South China. Abstract on the 3rd Asia–Pacific Sym. Environmental Geochemistry Guangzhou, China.2001; 93.

[82]Zhao G, Xu Y, Li W, Han G, Ling B. PCBs and OCPs in human milk and selected foods from Luqiao and Pingqiao in Zhejiang, China. Science of the Total Environment2007; 378(3):281-292.

[83]Zhou HY, Wong MH. Screening of organochlorines in freshwater fish collected from the Pearl River Delta, People's Republic of China. Archives of Environmental Contamination and Toxicology2004; 46: 106–113.

[84]Zhou K, Maskaoui Y, Qiu W, Hong HS, Wang ZD. Polychlorinated biphenyl congeners and organochlorine insecticides in the water column and sediments of Daya Bay, China. Environmental Pollution2001; 113: 373–384.

[85]Zhou, J., Hong, H., Zhang, Z., Maskaoui, K., and Chen, W. 2000. Multi-phase distribution of organic micropollutants in Xiamen Harbour, China. Water Research. 34 (7): 2132–2150.