Marine Pollution Bulletin 63 (2011) 516–522

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Distribution and source apportionments of polychlorinated biphenyls (PCBs) in mariculture sediments from the Pearl River Delta, South China

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ARTICLE INFO

Keywords:
PCBs
Mariculture
Enrichment percentage
Fish feed

ABSTRACT

Surface and core sediments collected from six mariculture farms in the Pearl River Delta (PRD) were analyzed to evaluate contamination levels of polychlorinated biphenyls (PCBs). The ∑PCBs (37 congeners) concentrations ranged from 5.10 to 11.0 ng g^{-1} (mean 7.96 ng g^{-1}) in surface and 3.19 to 22.1 ng g^{-1} (mean 7.75 ng g^{-1}) in core sediments, respectively. The concentrations were significantly higher than that measured in the sediments of their corresponding reference sites, whereby the average enrichment percentages were 62.0% and 42.7% in surface and core sediments, respectively. Significant correlations ($R^2 = 0.77$, $p < 0.05$) of PCB homologue group proportions between fish feeds and surface mariculture sediments suggested that fish feed input was probably the main source for the enrichment of PCBs. Due to the fact that PCBs could be transferred along food chains, PCB contamination in fish feeds and mariculture sediments should not be overlooked.

1. Introduction

Polychlorinated biphenyls (PCBs) were first manufactured commercially in 1929 and used widely as electric insulators in transformers, hydraulic fluids and paint additives (Safe, 1994). Serious concerns about the distribution of PCBs were raised since they were found to be ubiquitous and persistent in environmental and biota samples such as soil, water, animal and human tissues (Jansson et al., 1993). Although the production of PCBs has been banned since the early 1970s (Harrad et al., 1994), PCBs persist as legacy pollutants in which the chronic toxicity still represents a serious environmental risk.

High concentrations of PCBs have been reported in the coastal food web such in fish and shellfish collected from all over the world (Domingo and Bocio, 2007), including South China (Nie et al., 2005). As PCBs in sediments can be bioaccumulated in benthic organisms and transported further up the food chain to higher level consumers (such as fish) (Magnusson et al., 2006; Parnell et al., 2008), the assessment of sediment PCB contamination is an important issue for the evaluation of food safety, especially with regards to fish cultured along the coastal areas. Nutrient enrichment in mariculture sediments is common worldwide such as in Japan (Yokoyama et al., 2006), Turkey (Alpaslant and Pulatsue, 2008), Spain (Mendiguchia et al., 2006) and Hong Kong (Gao et al., 2005). The enriched organic matter could enhance the accumulation capacity of heavy metals (Mendiguchia et al., 2006) and persistent organic pollutants (POPs) (Wang et al., 2010a) in mariculture sediments. The unconsumed fish feeds containing trace organic pollutants would further promote this contamination. Although high PCB concentrations have been observed in fish feeds manufactured in various parts of the world including China (Guo et al., 2009), Canada (Kelly et al., 2008) and Europe (Jacobs et al., 2002), no data is currently available to differentiate the levels and sources of PCBs between natural and mariculture sediments.

The aim of the present study was to evaluate the level of enrichment of PCBs in mariculture surface and core sediments. The temporal trends, potential source apportionments and sediment burden of PCBs were also investigated. To our knowledge, this is the first study to investigate and compare the differences in PCB contamination in mariculture sediments with levels found in natural sediments.

2. Methods and materials

2.1. Study area and sampling

Six mariculture farms located at Xixiang (XX), Tsing Yi (TY), Sam Mun Tsai (SMT), Mirs Bay (MB), Sai Kung (SK) and Tung Lung Chau

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doi:10.1016/j.marpolbul.2011.02.009

(TLC) were chosen for this study (Fig. 1). All sampling sites were located along the coast of Hong Kong and mainland China, representing the typical subtropical fish farming regions in the Pearl River Delta (PRD). The major fish feeds used in the mariculture were trash fish (mainly anchovies *Thryssa* sp.), and moist and dry pellet feeds. The detailed information about each sampling site is listed in Table 1. During July–September 2008, at least three surface (0–5 cm, using a stainless steel grab sampler) and core (using a KC Kajak sediment core sampler, ø60/52 mm, length 100 cm, Denmark) sediment samples (including sediments beneath mariculture cages and reference sediments about 1–2 km away) were collected at each site. Each sediment core was sectioned into 2.5 cm intervals for the first 10 cm, then 5 cm intervals to 40 cm, and then 10 cm intervals to the end. Fish feeds including trash fish (*Thryssa* sp., $n = 6$) and dry pellet feeds ($n = 9$), originally bought from local markets, were collected from TLC fish farms. Fish farmers of other sampling sites confirmed that their feeds were also bought from local markets, but they were reluctant to give us the feeds. All samples were wrapped in aluminum foil and kept at −20 °C until further analyses.

2.2. Chemical analyses

The collected sediment samples were freeze-dried, homogenized and stored in desiccators prior to chemical analyses. The samples were Soxhlet extracted according to US EPA Standard Method 3540 C (USEPA, 1996a) using 120 ml mixture of acetone, dichloromethane (DCM) and *n*-hexane (1:1:1, v:v:v) for 16–18 h at 68 °C. Acid washed copper powder was added into the extracts to remove sulfur. The extract solution was then concentrated to 2 ml with a rotary evaporator. About 10 ml of *n*-hexane was added and was rotary evaporated to remove acetone and DCM. The concentrated extract was then cleaned up through a multilayer silica gel column containing, from top to bottom, 1 g anhydrous sodium sulfate, 2 g of deactivated silica (3% organic-free reagent water, w/w), 8 g of acidic silica (44% concentrated sulfuric acid, w/w), 1 g of

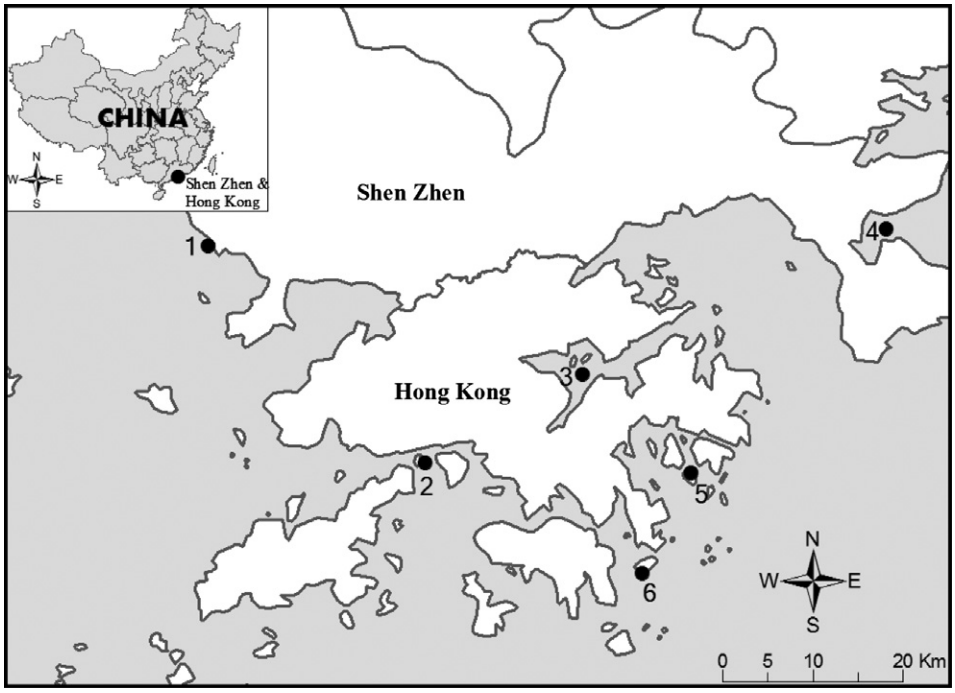

Fig. 1. Sampling sites. 1: Xixiang (XX); 2: Tsing Yi (TY); 3: Sam Mun Tsai (SMT); 4: Mirs Bay (MB); 5: Sai Kung (SK); 6: Tung Lung Chau (TLC).

Table 1
Detailed information of mariculture sites.

No.	Site	Ab.	Latitude	Longitude	WD. (m)	OM (% LOI)[a]	Descriptions
1	Xixiang	XX	22°33.243'N	113°51.738'E	5–10	7.25 ± 0.91	Located at Pearl River Estuary, which constantly receives discharges from the Pearl River. Black mud sediment
2	Tsing Yi	TY	22°21.047'N	114°03.366'E	40–50	7.97 ± 0.12	Located beside the Kwai Tsing Container Terminal, the third busiest container port in the world. Participant of the "Accredited Fish Farm Scheme"
3	Sam Mun Tsai	SMT	22°27.288'N	114°13.481'E	30–40	18.5 ± 1.34	Located at a semi-enclosed bay in the inner region of Tolo Harbor with a long history of receiving heavy pollution. Black mud sediment
4	Mirs Bay	MB	22°33.870'N	114°31.260'E	20–25	11.1 ± 0.79	A semi-enclosed bay located in the east coast of Guangdong Province. Relatively unpolluted marine environment (Liu and Hills, 1998)
5	Sai Kung	SK	22°20.483'N	114°19.043'E	20–30	10.9 ± 0.74	Near the open-sea. Relatively unpolluted marine environment around culture cages
6	Tung Lung Chau	TLC	22°15.400'N	114°17.228'E	20–30	6.38 ± 0.42	A small island located off the peninsula of Clear Water Bay

Ab. = Abbreviation; WD. = Water depth.
[a] The organic matter (loss of ignition) in mariculture surface sediments of each sampling sites.