Persistent halogenated compounds in two typical marine aquaculture zones of South China

Huan-Yun Yu [a,c], Ying Guo [a,c], Lian-Jun Bao [a,c], Yao-Wen Qiu [b], Eddy Y. Zeng [a,*]

[a] State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Science, Guangzhou 510640, China
[b] Key Laboratory of Tropic Marine Environmental Dynamics, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou 510301, China
[c] Graduate School, Chinese Academy of Sciences, Beijing 100049, China

ARTICLE INFO

ABSTRACT

Samples of two seawater farmed fish (crimson snapper (*Lutjanus erythopterus*) and snubnose pompano (*Trachinotus blochii*)), water, air, sediment, fish feed, macroalgae and phytoplankton were collected from two estuarine bays (Daya Bay and Hailing Bay) in South China. The concentrations of persistent halogenated compounds (PHCs) including polybrominated diphenyl ethers (PBDEs), organochlorine pesticides and polychlorinated biphenyls (PCBs) varied widely with the different sample matrices under investigation. The compositional patterns in fish, fish feed, macroalgae and phytoplankton, as well as the good correlations between the abundances of *p,p′*-DDT and BDE-209 and their metabolites (i.e., *p,p′*-DDD and *p,p′*-DDE for *p,p′*-DDT and BDE-47 for BDE-209) in fish indicated the occurrence of DDT and PBDE biotransformation in fish body. Finally, the marine aquaculture environment in South China is somewhat biologically impaired by DDT-contaminated water, sediment and fish feed, and there may be some cancer risk associated with fish consumption for humans, especially for urban residents.

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1. Introduction

The Pearl River Delta (PRD), located in South China within Guangdong Province, is one of the economically fastest growing regions in China and an important hub for imported e-wastes and electronic manufacturing ventures, as well as having the largest amounts of organochlorine pesticide (OCP) usage in history due to well-developed agriculture and fisheries. High levels of persistent halogenated compounds (PHCs) such as polybrominated diphenyl ethers (PBDEs) and OCPs were found in various environmental matrices (Guan et al., 2007; Guo et al., 2007, 2009a; Meng et al., 2007; Zhang et al., 2009). Besides, continuously growing fish production in Guangdong, the second-largest fishery province of China (Liu, 2008), provides increased animal protein consumption for coastal residents. On the other hand, health risk associated with consumption of contaminated fish has become a significant concern due to high concentrations of PHCs in aquatic products, one of which is seawater farmed fish (Meng et al., 2007; Guo et al., 2009a).

Our recent study (Meng et al., 2007) collected fish from local markets in 11 coastal cities of Guangdong Province. The results of this study indicated that the levels of PHCs in seawater farmed fish were significantly higher than those in freshwater farmed fish

and wild fish, which was a reflection of high contaminant loading in coastal seawater fish farms. Concentrations of PHCs in certain species, such as crimson snapper (*Lutjanus erythopterus*) and snubnose pompano (*Trachinotus blochii*), were at high enough concentrations to cause human health concerns (Meng et al., 2007). Furthermore, recent and previous studies found that PHCs, especially DDTs, were ubiquitous in fish feed including compound feed and trash fish (Hites et al., 2004; Minh et al., 2006; Guo et al., 2009b). These studies speculated that the presence of PHCs in farmed fish may be mainly derived from fish feed.

In order to track more clues to diagnose the potential input routes of PHCs in seawater farmed fish, the present study examined the occurrence of PHCs including polybrominated diphenyl ethers (PBDEs), organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in two seawater farmed fish species, the crimson snapper and snubnose pompano, collected from two estuarine bays (Hailing Bay and Daya Bay) in Guangdong Province (Fig. S1; "S" designates tables and figures in the Supporting Information thereafter), and also in the environment in which these fishes live (i.e. water, air, sediment, fish feed, macroalgae and phytoplankton). Besides, it is the hope of the present study that the results derived from health and ecological risk assessment will offer some constructive suggestions for marine aquaculture industry and responsible agencies to establish guidelines and recommendations for human consumption of seawater farmed fish.

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2. Materials and methods

2.1. Sample collection

Two species of fish (Crimson snapper and snubnose pompano) were collected from Daya Bay and Hailing Bay in July and December 2007 (Fig. S1). In addition, fish feeds including compound feeds and trash fish which the two species of fish feed on, phytoplankton and macroalgae growing around the seawater farmed cages, and nearby seawater and sediment samples were collected from seawater fishery farms in both Daya Bay and Hailing Bay. Besides, air samples around the fishery farms were also collected in the two bays (Table S1).

2.2. Sample preparation

All fish samples (except for the large bone of the fish head) were homogenized with a blender and then freeze-dried and ground into fine powders. After spiking with surrogate standards, PCB 67, ^{13}C-PCB-141, PCB-209 and ^{13}C-BDE 77, the samples were Soxhlet extracted with a 1:1 (v:v) acetone and hexane mixture for 48 h, and concentrated to 5 mL with a TurboVap II evaporator (Zymark, Hopkinton, MA). Twenty percent of the extract was used for determining lipid content using the gravimetric method (Meng et al., 2007) and the remainder was cleaned with a gel permeation column and a silica/alumina column by sequence. The sample was then concentrated to 100 μL under a gentle stream of N$_2$. Lastly, internal standards (PCB-82 for OCPs, PCB-82 and PCB-189 for PCBs, and ^{13}C-PCB-208 for PBDEs) were added before instrumental analysis. Methods used for processing other types of samples were similar to fish samples. The extraction methods for sediment, suspended particulate matters (SPM) and air samples were slightly different since determining and removing lipids were not necessary. Further descriptions and details of the methods for extraction have been described in previous studies (Guo et al., 2009b,c). Water samples were extracted and analyzed according to a previously published procedure (Guan et al., 2009).

2.3. Instrumental analysis

For OCPs, only DDT and its metabolites (o,p'- and p,p'-DDT, -DDD, and -DDE and p,p'-DDMU, sum of which is designated as DDXs and DDXs minus p,p'-DDMU is designated as DDTs) were detected and quantified, because concentrations of the other target analytes were low and rarely detectable. Concentrations of DDXs were determined with a Varian 3800 gas chromatograph interfaced with a Saturn 2000 mass spectrometer in the selective ion monitoring mode. A total of 42 BDE congeners, purchased from AccuStandard (New Haven, CT, USA), were used as target analytes and 23 congeners (BDE-17, -25, -28, -35,-47, -49,-66, -85, -99, -100, -116, -118, -119, -126,-138,-153, -154, -181, -183, -190, -196, -206, and -209, sum of which is designated as Σ_{23}PBDE thereafter) were detectable in fish, fish feed, macroalgae, phytoplankton, air and sediment samples while in water samples, only 14 congeners (BDE-15, -28, -47, -66, -85, -99, -100, -153, -154, -138, -183, -196, -206, -209, sum of which is designated as Σ_{14}PBDE thereafter) were detectable. Therefore, Σ_{14}PBDE in water samples and Σ_{23}PBDE in other samples were quantified in the present study. The procedures for the instrumental analysis of PBDEs has been detailed in a previous study (Guo et al., 2009b). Besides, 17 PCBs congeners (PCB-31/28, -52, -44, -99, -149/118, -153, -138, -180, -170, -194, -101, -110, -147, -146, and -187, sum of which is designated as Σ_{17}PCB thereafter) were detectable and quantified in fish and fish feed samples and were very low in the other samples. As a result, only the concentrations of Σ_{17}PCB in fish and fish feed are

reported in the following section. A Shimadzu Model 2010 GC coupled with a Model QP2010 MS (Shimadzu, Japan) was used for PCB analysis.

The reporting limits (RLs) for DDXs, PCBs, tri- to octa- BDEs, nona-BDEs, and BDE-209 were 1.25, 0.25, 0.05, 0.25, and 2.5 ng/g, respectively, for 1 g of wet fish sample weight. The RLs for DDXs, tri- to octa- BDEs, nona-BDEs, and BDE-209 were 1, 0.04, 0.2, and 2 ng/g for 1 g of sediment or 1 g of macroalgae or 1 g of phytoplankton on a dry weight basis, 1, 0.04, 0.2, and 2 ng/L for 1 L of water, and 500, 20, 100, and 1000 pg/m^3 for 1 m^3 of air sample. It should be noted that half of the RL was used for a value below the RL in correlation analysis on the abundances of BDE-47 and BDE-209 as the detection frequency of BDE-209 was low in fish.

2.4. Quality assurance/quality control

For each batch of 20 samples, a field blank, a procedural blank, a spiked blank, and a matrix spiked sample and a matrix spiked sample duplicate were processed. The mean recoveries of the surrogate and spiking standards varied from 58% to 118% and 58% to 121%, respectively, and the detailed QA/QC data are presented in Table S2. For BDE-116, -183, -190, -197, -206 and -209, the blank values were subtracted from the sample measurements. For the other PBDE congeners, DDXs and PCBs, concentrations in the procedural and field blank samples were close to or only slightly higher than the reporting limits; therefore blank values were not subtracted out. To ensure minimal degradation of p,p'-DDT during instrumental analysis, a standard solution of p,p'-DDT was analyzed once for every batch of 10 samples and instrumental analysis could proceed if thermal degradation rate was determined as <20%. Data comparison among groups was done using nonparametric tests (Mann–Whitney U) with statistical significance $p < 0.05$ under SPSS version 13.0.

3. Results and discussion

3.1. Occurrence of persistent halogenated compounds

Detailed concentrations are presented in Tables S3–S7 and Figs. S2–S6. The concentration levels of PCBs in all samples were lower than those of DDXs and PBDEs, attributable to the small amounts of PCBs manufactured and used historically in China (Breivik et al., 2002). DDTs were the most abundant constituents in all samples whereas p,p'-DDMU was rarely detected in air, water (dissolved phase) and macroalgae.

The concentrations of DDTs in Hailing Bay fish (12–512 ng/g wet wt. with a mean of 159 ng/g wet wt.) were significantly higher than those in Daya Bay fish (14–194 ng/g wet wt with a mean of 73 ng/g wet wt.) ($p = 0.000$). Besides, trash fish contained moderately higher DDT concentrations ranging from 2.5 to 421 (mean: 45) ng/g wet wt. than compound feed with DDT concentrations ranging from 0.33 to 37 (mean: 10) ng/g wet wt. ($p = 0.11$). In addition, phytoplankton contained significantly higher DDT concentrations (mean: 407 ng/g dry wt.) than macroalgae (mean: 8.4 ng/g dry wt.). Sediment concentrations of DDTs varied from 3.7 to 181 and 3.6 to 23 ng/g dry wt. with mean concentrations of 46 and 12 ng/g dry wt. in Hailing Bay and Daya Bay, respectively. The levels of DDTs in the water samples from Hailing Bay (0.14–1.4, mean: 0.50 ng/L) were higher than those in water samples from Daya Bay (0.033–0.40, mean: 0.17 ng/L) ($p = 0.000$). Additionally, concentrations of DDTs in SPM samples from Hailing Bay (0.069–2.2, mean: 0.52 ng/L) were comparable to those from Daya Bay (<RL-0.51, mean: 0.26 ng/L).

Levels of DDTs in the particulate and vapor phases of air samples were in the range of 2.2–59 and 16–187 pg/m^3 in Hailing

Bay and 1.3–9.4 and 8.7–105 pg/m^3 in Daya Bay with mean concentrations of 13, 90, 4.0 and 53 pg/m^3, respectively. Moreover, levels of DDTs in air vapor samples from Hailing Bay were significantly higher than those from Daya Bay ($p = 0.013$).

The concentrations of Σ_{17}PCB in Hailing Bay fish (0.72–12, mean: 2.8 ng/g wet wt.) and Daya Bay fish (0.39–10, mean: 3.3 ng/g wet wt.) were not significantly different from each other ($p = 0.14$). Fish feed samples showed similar levels of Σ_{17}PCB compared to fish, which ranged from 0.49 to 4.4 ng/g wet wt. in compound feed and from 0.68 to 9.1 ng/g wet wt. in trash fish. In addition, the levels of Σ_{17}PCB were significantly higher in trash fish than in compound feed ($p = 0.002$), which may be ascribed to a higher trophic level for trash fish (Fig. S4).

Levels of Σ_{23}PBDE varied from 0.23 to 23 and 0.30 to 14 ng/g wet wt. with mean values of 4.5 and 3.6 ng/g wet wt. in Hailing Bay and Daya Bay fish, respectively. Contrary to DDTs and Σ_{17}PCB, Σ_{23}PBDE were significantly more abundant in compound feed than in trash fish ($p = 0.004$) because of the presence of BDE-209. If BDE-209 was excluded, the relative abundances were reversed. Concentrations of Σ_{23}PBDEs in phytoplankton and macroalgae varied from 0.61 to 318 and 0.22 to 12 ng/g dry wt., respectively. In addition, the concentrations of Σ_{23}PBDE were significantly different in sediment from Hailing Bay (0.35–16, mean: 3.8 ng/g dry wt.) and Daya Bay (6.7–275, mean: 76 ng/g dry wt) ($p = 0.045$). Fig. S5 shows that one sediment sample from Daya Bay (marked as red square) contained an extraordinarily high level of Σ_{23}PBDE, probably reflective of point source pollution.

Concentrations of Σ_{14}PBDE in Hailing Bay water (0.039–0.29, mean: 0.16 ng/L) were in the similar range as that in Daya Bay water (0.015–0.36, mean: 0.085). However, Σ_{14}PBDE levels in Daya Bay SPM (0.048–1.3, mean: 0.39) were significantly higher than those in Hailing Bay SPM (0.0047–1.4, mean: 0.20) except one sample in Hailing Bay ($p = 0.032$). Wania and Dugani (2003) found that some of the low and intermediate-brominated BDE congeners have the long range transport potential; hence PBDE levels in air can provide important information on pollution sources. In the present study, Σ_{23}PBDE levels in air particulate and vapor phases were 4.7–542 and 6.3–47 pg/m^3 in Hailing Bay and 2.3–228 and 2.1–67 pg/m^3 in Daya Bay, respectively. Although the Σ_{23}PBDE levels in the vapor phase were not significantly different between the two bays, particulate Σ_{23}PBDE levels were significantly higher in Hailing Bay than in Daya Bay ($p = 0.024$) (Fig. S6).

Generally, concentration levels of DDTs in fish, water and air samples collected from Hailing Bay were much higher than those from Daya Bay (Figs. S2 and S3). However, SPM and sediment samples from Daya Bay contained higher levels of Σ_{23}PBDE than those from Hailing Bay (Fig. S5). These results are probably attributed to the different economic development patterns in the two regions. Daya Bay and Hailing Bay are located in the eastern and western coasts, respectively, of the Pearl River Delta. The city near Hailing Bay, Yangjiang, has been well developed in agriculture and fishery, but under developed in industry. On the other hand, Daya Bay is located near Shenzhen and Huizhou, one of the economically fast growing and urbanized regions in Guangdong Province. Thus, as the results suggest, the historical use of DDT in Yangjiang's agriculture and fisheries may be responsible for the occurrence of abundant DDTs in Hailing Bay compared to Daya Bay, while well-developed industry around Daya Bay should be the major source of higher residues of Σ_{23}PBDE in SPM and sediment from Daya Bay. It is worthwhile to note that air particulate from Hailing Bay contained much higher Σ_{23}PBDE than those from Daya Bay ($p = 0.005$) which was beyond our expectation. According to our sampling records, the wind direction of southeast or northeast prevail in Hailing bay, so it was very likely that pollutants were carried by wind from other places to Hailing Bay. In addition, there are still

several pharmaceutical, electric and metal companies around Hailing Bay which might also contribute to PBDE pollution.

3.2. Assessment of uptake routes based on congener compositional analyses

Fig. S7 shows that the compositional patterns of DDX were similar in various sample types from Daya Bay and Hailing Bay. Generally, p,p'-DDD, -DDE and -DDT dominated over other congeners. However, there were also some different patterns among different sample types. For instance, the abundances of p,p'-DDT were much higher in water than in fish ($p = 0.004$) from Daya Bay. Although p,p'-DDD dominated over other congeners in both fish and water, its relative abundances in fish and water were significantly different ($p = 0.001$ and 0.000 for Hailing Bay and Daya Bay). Besides, Daya Bay fish contained significantly higher abundances of p,p'-DDT and p,p'-DDE than Hailing Bay fish ($p = 0.013$ and 0.000, respectively). Abundances of p,p'-DDT in SPM and air vapor phase of Hailing Bay were also significantly higher than those of Daya Bay ($p = 0.029$ and 0.006). As shown in Fig. 1, the ratios of (DDE + DDD)/$\sum$DDXs in sediment, water and air samples were in the range of 0.24–1.0 with a mean value of 0.60 and 0.72 from Hailing Bay and Daya Bay, respectively, suggesting that historical residues have remained the main source of DDTs in these two estuarine bays which is also consistent with the results of a previous study (Xing et al., 2009).

Generally, seawater farmed fish mainly feed on compound fish feed made of fish powder, fish oil, flour, wheat protein powder and soymeal or trash fish which are constituted of various species of small wild fish captured in deep sea (Food and Agriculture Organization of the United Nation, 2007). According to the observations of local fishermen and some references (Meng, 2007; Southern Fisheries Website, 2010), snubnose pompano mainly feed on compound feed and crimson snapper mainly eat trash fish. As shown in Fig. 2a, the congener profiles of DDTs in snubnose pompano and compound feed were generally similar, with some minor differences. For example, snubnose pompano contained significantly higher abundances of p,p'-DDT than compound feed ($p = 0.000$). Besides, significantly different abundances of o,p'-DDT, o,p'-DDE and p,p'-DDE were also observed between compound feed and snubnose pompano ($p = 0.000$, 0.021 and 0.032). The abundances of p,p'-DDMU, o,p'-DDD, p,p'-DDD and o,p'-DDE were also significantly different between crimson snapper and trash fish
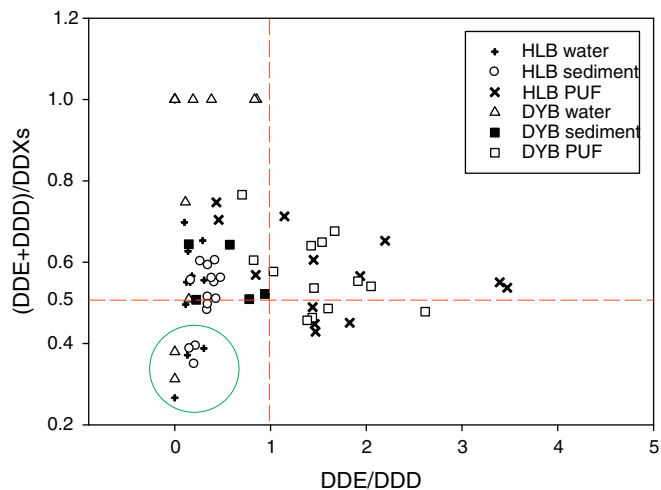

Fig. 1. Correlations between the ratios of (DDE + DDD)/DDXs and DDE/DDD in water, sediment and air samples from Hailing Bay (HLB) and Daya Bay (DYB).
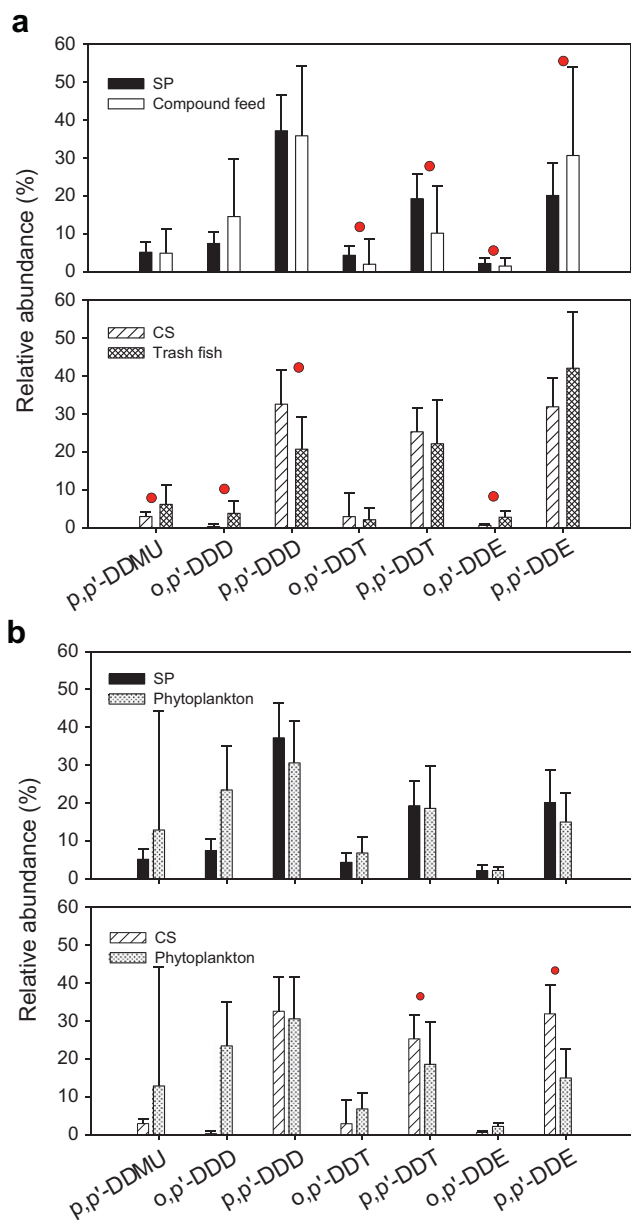
Fig. 2. Relative abundances of individual congeners normalized to DDXs in (a) snubnose pompano (SP), crimson snapper (CS), compound feed and trash fish; and (b) SP, CS and phytoplankton. The red solid circle above two related bars indicate significant difference ($p < 0.05$) between fish and fish feed and between fish and phytoplankton for the congener. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

($p = 0.015$, 0.016, 0.000 and 0.000, respectively) (Fig. 2a). All these results indicated that bioaccumulation of DDXs in fish was a considerably complex process under actual environmental conditions. In fact, large amounts of phytoplankton usually live in seawater around fish farming cages, so that seawater farmed fish may bioaccumulate PHCs from phytoplankton. Fig. 2b shows the compositional patterns of DDX in fish and phytoplankton. As described above, the relative abundances of p,p'-DDT in snubnose pompano were significantly higher than those in compound feed ($p = 0.000$) and abundances of p,p'-DDD in crimson snapper were significantly higher than those in trash fish ($p = 0.000$), but there was no significant difference between the abundances of p,p'-DDT in snubnose pompano and phytoplankton and between the abundances of p,p'-DDD in crimson snapper and phytoplankton. This indicated that snubnose pompano might bioaccumulate p,p'-

DDT from phytoplankton, confirming the above conjecture that snubnose pompano can bioaccumulate p,p'-DDT from other routes in addition to compound feed and crimson snapper might also bioaccumulate p,p'-DDD from phytoplankton or p,p'-DDT was more easily degraded to p,p'-DDD in crimson snapper.

Previous studies indicated that DDT can be metabolized to DDD or DDE in fish (Wang and Wang, 2005; Guo et al., 2008; Kwong et al., 2008). A good correlation between the abundances of p,p'-DDT and the sum of p,p'-DDD and DDE in crimson snapper from the present study (Fig. S8) partially confirmed the conclusion. Although there was no significant correlation between the abundances of p,p'-DDT and the sum of p,p'-DDD and DDE in snubnose pompano (Fig. S9), a good correlation was found in compound feed (Fig. S10), a major food source for snubnose pompano, indicating that degradation of DDT in compound feed might weaken the correlation between the abundances of p,p'-DDT and the sum of p,p'-DDD and DDE in snubnose pompano.

For PBDEs, the lower brominated congeners were generally more abundant in fish, water and air vapor phase, while the higher brominated congeners were more abundant in sediment, SPM and air particulate phase, which was mainly dictated by the strong affiliation of BDE-209 with the solid phase (Fig. S11). In detail, BDE-47 was the predominant congener in both fish and air vapor phase with the mean relative abundances of 38%, 44%, 35% and 34% in Hailing Bay fish, Daya Bay fish, Hailing Bay air vapor phase and Daya Bay air vapor phase, respectively. The mean relative abundances of BDE-209 in sediment, air particulate phase and SPM for both two bays were 90%, 73% and 60%, respectively. It was unexpected that BDE-209 was quite abundant (mean relative abundance of 42%) in Hailing Bay water (dissolved phase) samples, which was inconsistent with the previous finding that no BDE-209 was detected in the dissolved phase of water samples collected from the Pearl River Estuary (Luo et al., 2008). In fact, Daya Bay water samples also contained low levels of BDE-209 (with a mean relative abundance of 2.3%).

The relative abundances of most BDE congeners in fish and fish feed were significantly different as indicated by red solid circles (Fig. 3a). Besides, it is interesting to note that the relative abundances of BDE-209 in fish feed were significantly higher than those in fish, while abundances of BDE-47 in fish were significantly higher than those in fish feed ($p = 0.000$ and 0.016, respectively). It can be concluded that biotransformation of BDE-209 may have occurred in fish body, which was deemed feasible (Stapleton et al., 2006). In addition, the difference between the relative abundances of BDE-47 and BDE-209 in fish and another possible source of PHCs (i.e. phytoplankton) also exhibited a trend similar to that for fish and fish feed (Fig. 3b), indicating that biotransformation of BDE-209 did occur in fish. The significant correlation between the abundances of BDE-47 and BDE-209 in fish (Figs. S12 and S13) further supports the argument.

3.3. Potential biological effects from the occurrence of DDTs in marine-aquaculture zones

The ecological risk of DDTs in sediment can be assessed with two guideline values, effects range-low (ERL) and effects range-medium (ERM) (Long et al., 1995). The levels of DDTs in all sediment samples from Hailing Bay and Daya Bay were higher than the ERL value (1.58 ng/g dry wt.; Fig. S14). Also, levels of DDTs in two sediment samples from Hailing Bay were higher than ERM (46.1 ng/g dry wt.; Fig. S14). However, these DDT levels were much lower than the probable effect level (386.6 ng/g) proposed by the Canadian Council of Ministers of the Environment (Canadian Council of Ministers of the Environment, 2003).

In addition, 16 out of 18 Hailing Bay water samples and 6 out of 15 Daya Bay water samples contained DDTs at levels higher than
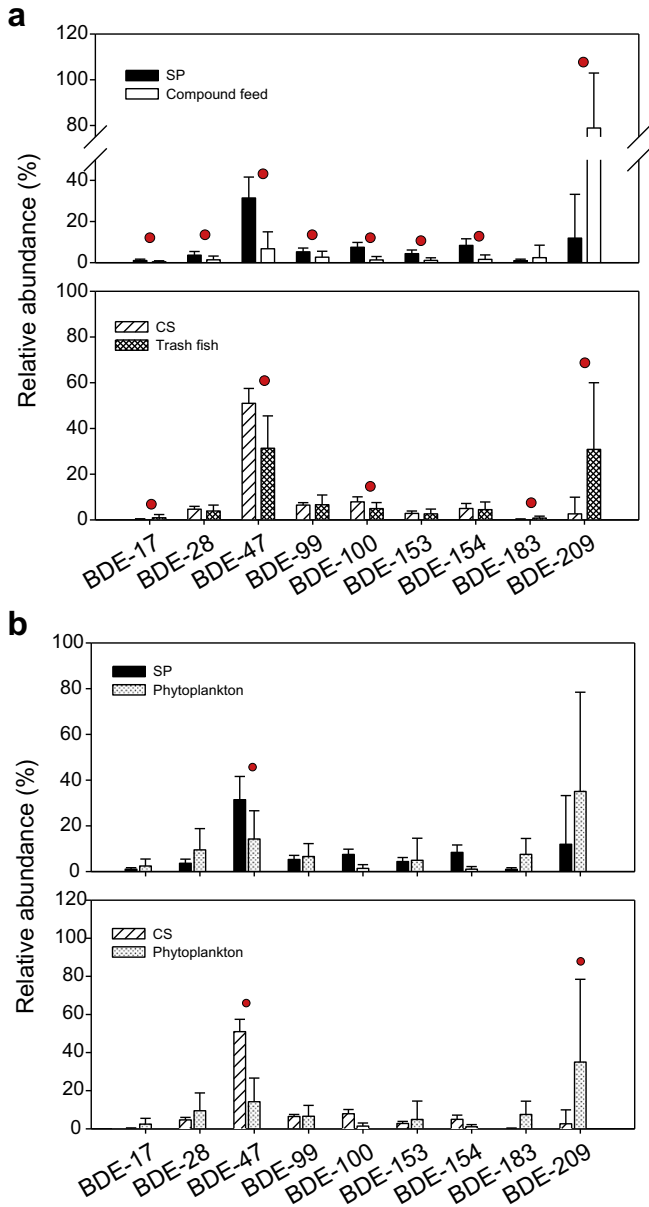
a

b

Fig. 3. Relative abundances of individual congeners normalized to Σ_{23}PBDEs in (a) snubnose pompano (SP), crimson snapper (CS), compound feed and trash fish; and (b) SP, CS and phytoplankton. The red solid circle above two related bars indicate significant difference ($p < 0.05$) between fish and fish feed and between fish and phytoplankton for the congener. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

the 30-day average concentration (0.17 ng/L) of DDTs permitted in receiving waters off California as regulated by the California Ocean Plan (Fig. S15) (California State Water Resources Control Board, 1997). However, levels of DDTs in all water samples were much lower than the maximum residue level (1000 ng/L) in sea water, which is the second grade standard and applicable to aquaculture set by the Chinese government (National Environmental Protection Agency and the State Oceanic Administration, 1997), and the limit (25 ng/L) recommended by the European Parliament and the Council of the European Union (The European Parliament and the Council of the European Union, 2008).

For fish feed, the mean level of DDTs in trash fish (45 ng/g wet wt.) was higher than the screening value (14 ng/g wet wt.) for wildlife consumption of aquatic biota established by the

Government of Canada (Canadian Council of Ministers of the Environment, 2003). Overall, the marine aquaculture environment in South China is somewhat biologically impaired by DDT-contaminated water, sediment and fish feed, an issue which deserves further monitoring.

3.4. Assessment of human health risk due to fish consumption

Twenty-two out of 32 Hailing Bay and eight out of 33 Daya Bay whole fish samples contained DDTs at higher levels than the screening value (100 ng/g wet wt.) for human consumption of fish developed by the Office of Environmental Health Hazard Assessment (Brodberg and Pollock, 1999) (Fig. 4). The cancer and non-cancer hazard risk derived from consumption of fish collected from these two estuarine bays was further assessed using the method developed by the US Environmental Protection Agency (2000).
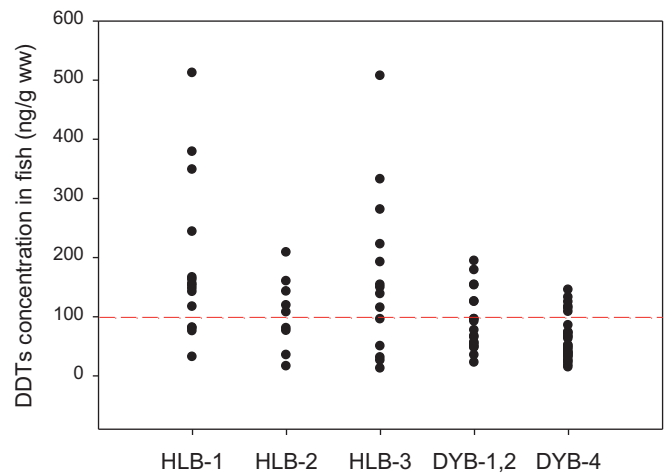

Fig. 4. Comparison of the screening value (100 ng/g wet wt.) for human consumption of fish developed by the Office of Environmental Health Hazard Assessment (Brodberg and Pollock, 1999) and the concentrations of DDTs in fish samples collected from various locations of Hailing Bay (HLB) and Daya Bay (DYB).
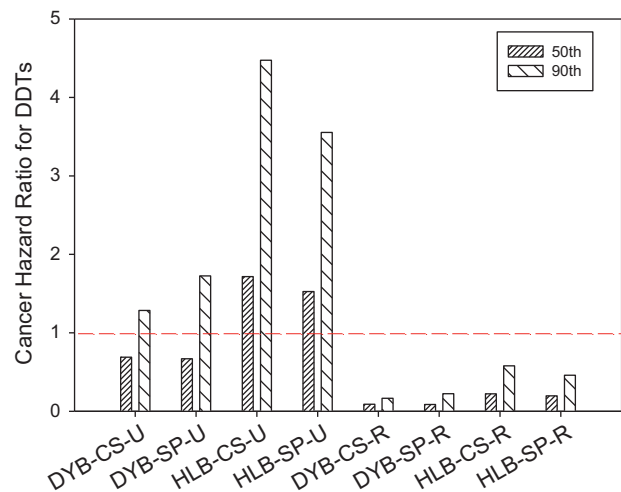

Fig. 5. Assessment of human health risk associated with consumption of snubnose pompano (SP) and crimson snapper (CS) collected from Hailing Bay (HLB) and Daya Bay (DYB) for urban (U) and rural (R) residents (The consumption rates are 34.2 g/day for urban residents and 12.3 g/day for rural residents (Meng, 2007) using the cancer hazard ratio developed by the US Environmental Protection Agency (2000) based on the 50th and 90th percentile concentrations of DDTs.

Figs. S16 and S17 show that all the non-cancer hazard ratio (HRn) values based on the 50th and 90th percentile concentrations of DDTs and PCBs and the cancer hazard ratio (HRc) values based on the 50th and 90th percentile concentrations of PCBs were considerably lower than unity, but several HRc values based on the 50th and 90th percentile concentrations of DDTs were higher than unity (Fig. 5), indicating that there may be some cancer risk for humans especially for urban residents through consumption of fish collected from the marine aquaculture zones of South China.

The results from the present study suggest that the marine-aquaculture environment in South China is somewhat impaired by the occurrence of PHCs, especially DDTs, resulting in possible human cancer risk through fish consumption. Identifying dominant input routes of PHCs to the marine aquaculture zones and consequently minimizing the pollution of seawater farmed fish by adopting shrewd farming practices should be the top priority for the scientific and management communities. Meanwhile, one of the best courses of action is to advise the public in coastal regions of China to limit consumption of seawater farmed fish to reduce exposure to PHCs.

Acknowledgments

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.marpolbul.2010.12.006.

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