



Organochlorine pesticide and heavy metal residues in some edible biota collected from Quanzhou Bay and Xinghua Bay, Southeast China

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Received 18 April 2009; revised 22 June 2009; accepted 16 October 2009

Abstract

In order to assess the contamination of edible marine organisms in Quanzhou and Xinghua Bays, Fujian Province, China, concentrations of organochlorine pesticides (hexachlorocyclohexane isomers or HCHs and dichlorodiphenyltrichloroethane isomers/derivatives or DDTs) and heavy metals (Cu, Pb, Zn, As, Cr, Cd, Ni and Hg) in 13 species collected from different sites in the bays were analyzed by GC-ECD and ICP-MS. The concentration of the sum of DDTs exceeded that the sum of HCHs in the samples. Most of the organisms showed higher levels of DDTs than the first level criterion (10 ng/g) for marine biological quality in China (GB 18421-2001) but conformed to the first level criterion (20 ng/g) for HCHs. The estimated mean daily uptake of pesticides was below the level for minimal risk to the consumers. The levels of Cd, As and Pb in most organisms from both bays were markedly elevated and above the recommended legal limits for human consumption. Our results showed the need for routine monitoring of marine species contamination to ensure public and environmental health.

Key words: organochlorine pesticides; residue levels; technical HCH; heavy metals; Fujian Province

DOI: 10.1016/S1001-0742(09)60110-8

Introduction

Anthropogenic chemical pollutants such as organochlorine pesticides (OCPs) and heavy metals have been reported to contaminate the marine environment. The lipophilic nature, hydrophobicity and low chemical and biological degradation rates of OCPs have led to their bioaccumulation and subsequent magnification in organisms of the food chain (Vassilopoulou and Gregoriades, 1993). Some heavy metals such as copper, zinc, cobalt, manganese and iron are essential for biological processes, but only at low concentrations. Nevertheless, metals such as cadmium, lead and mercury have no known essential biological functions and are toxic even at low concentrations. Humans, as a final link in the food chain, are mostly affected, and consequently the public has become increasingly concerned about the potential risk to human health from the consumption of such polluted biota (Mwvura, 2002). From 1950s to 1983, the total production of technical hexachlorocyclohexanes (HCHs) (4.9 million tons) and dichlorodiphenyltrichloroethanes (DDTs) (0.4 million tons) in China accounted for 33% and 20% of the total world production, respectively (Hua and Shan, 1996). In addition, the ongoing industrialization and urbanization

in both the inland and coastal areas, have resulted in the discharge of heavy metals into the coastal environments.

Quanzhou Bay (118°36'E–118°48'E, 24°46'N–24°51'N) and Xinghua Bay (119°15'E–119°37'E, 25°23'N–25°37'N) in Fujian Province, Southeast China, are major fishing grounds for bivalves, which are favorite seafood of local people (Chen et al., 2002). There are mainly two considerable rivers flowing into Quanzhou Bay, namely Jinjiang and Luoyang while Mulan is the major stream entering into Xinghua Bay. The pollution states of the bays are to some extent dependent on anthropogenic inputs from the adjacent industrialized cities. Consequently, sediment cores from Quanzhou Bay were found with OCPs dating back to 1940s (Gong et al., 2007) and with heavy metals in concentrations higher than the permissible levels for marine environmental quality in China (Yu et al., 2008). Due to these concerns, continuous monitoring of contaminant levels in marine organisms is desirable. This study was carried out to measure the concentrations of some OCPs and heavy metals in selected marine organisms from Quanzhou Bay and Xinghua Bay to assess the seafood consumption safety. Our results were evaluated against National and International standards to identify potential public health risk.

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1 Materials and methods

1.1 Chemicals

All chemicals used were of high quality analytical grade. Chemical reagents, dichloromethane and *n*-hexane were purchased from Tedia Co., (USA). Acetone was purchased from Fisher Scientific (USA). Anhydrous sodium sulphate, alumina gel (0.15 mm mesh) and silica gel (0.15 mm mesh) were obtained from Sinopharm Medicine Holding Co., Ltd., China. Nitric acid and sulphuric acid were purchased from Merck, USA. The mixed standard sample of organochlorine pesticides, 2,4,5,6-tetrachloro-*m*-xylene (TCMX), decachlorobiphenyl (PCB 209) and pentachloronitrobenzene (PCNB) were purchased from Ultra Scientific (USA).

1.2 Sample collection

Samples were collected from the estuary of Jinjiang River and Luoyang River to the mouth of Quanzhou Bay and from Mulan Stream to the mouth of Xinghua Bay, respectively, in January 2005. The locations of study area and sampling sites are shown in Fig. 1.

Shells of bivalves were carefully removed with a sterilized plastic knife. After washing with deionized water, each sample was stored in a well sealed polyethylene bag, frozen and transported to the Key Laboratory of Biogeology and Environmental Geology of Ministry of Education, Wuhan, China and stored at -20°C till analysis.

Samples for chemical analysis were removed from the freezer, allowed to thaw. Fish flesh (muscle tissue) was carefully dissected then ground to a homogeneous paste in a mixer. Ten grams of tissues were weighed, and mixed with anhydrous sodium sulphate to extract OCPs. The rest was stored at -20°C again and then freeze-dried at -45°C for 3 days. One gram of the freeze-dried portion of each sample was carefully weighed and digested for heavy metal determination.

1.3 Extraction and analysis of OCPs

The extraction and clean up of the OCPs were similar to those previously described by Yang et al. (2004). In brief, 10 g of pretreated samples were spiked with 20 ng of 2,4,5,6-tetrachloro-*m*-xylene (TCMX) and decachlorobiphenyl (PCB 209) as recovery surrogates and were soxhlet-extracted for 48 hr with 120 mL mixture of 2:1 (V/V) dichloromethane and acetone. The concentrated extract was transferred into a 100-mL separation flask and 3 drops of 98% H_2SO_4 were added. The mixture was mixed well and kept for several minutes to settle down. The lower layer (fat in H_2SO_4) was carefully removed. This procedure was repeated 3 times before the remaining organic layer was reduced to 2–3 mL using a rotary evaporator at 40°C . The extract was transferred to an aluminum/silica gel clean up column (1:2, V/V, and both 3% deactivated). The column was eluted with 25 mL of *n*-hexane and dichloromethane (3:2, V/V) to yield the OCPs fraction. The fraction was concentrated to 0.2 mL under a gentle stream of nitrogen gas. A 4- μL of 5 mg/L pentachloronitrobenzene (PCNB) was added as an internal standard prior to analysis.

Analysis of OCPs was carried out with a gas chromatograph equipped with a micro-cell ^{63}Ni electron capture detector (GC-ECD) (Agilent 6890, USA). The separation was performed on a fused silica capillary column (HP-5, 30 m \times 0.32 mm ID, and 0.25 μm film thickness). The carrier gas was nitrogen with a constant flow rate of 2.5 mL/min. The injector and the detector temperatures were 290 and 300°C , respectively. The GC oven temperature was programmed as follows: initial temperature 100°C held for 4 min, increased to 200°C at a rate of $4^{\circ}\text{C}/\text{min}$, then increased to 230°C at a rate of $2^{\circ}\text{C}/\text{min}$, followed by $8^{\circ}\text{C}/\text{min}$ till 280°C which was maintained for 15 min. A 2- μL sample was injected into the GC-ECD for analysis. A six-point response factor calibration curve was established to quantify the target analytes (according to a

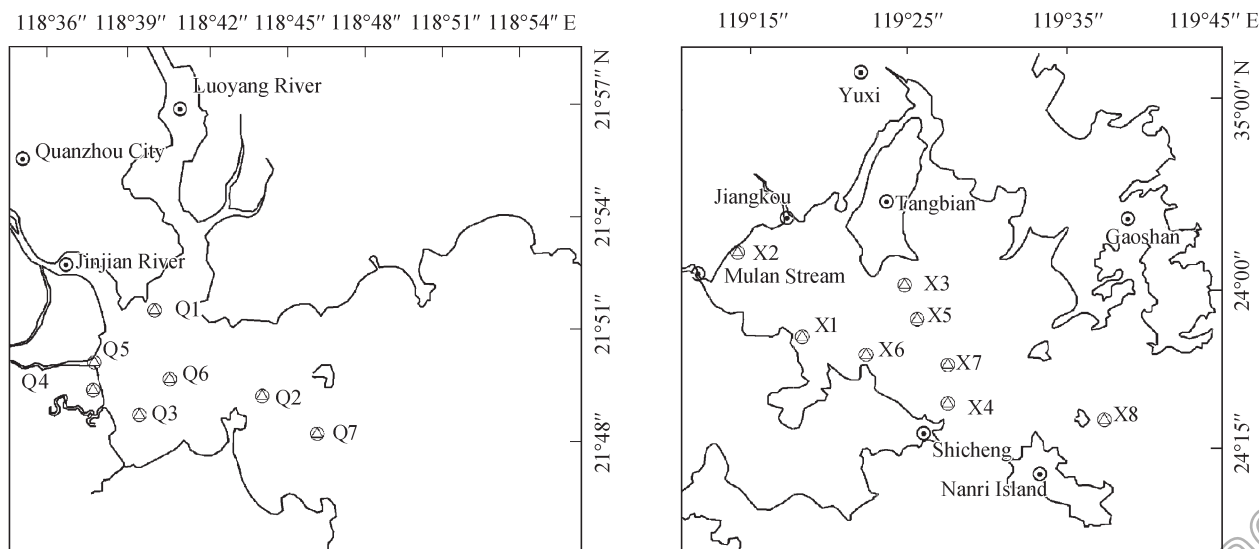


Fig. 1 Locations of sampling sites in Quanzhou Bay (Q1–Q7) and Xinghua Bay (X1–X8).

series of standard samples with 10, 20, 50, 100 and 200 ng/mL concentrations). Then the quantity of OCPs was corrected by surrogates. Data were processed using the HP-3365 Chemstation software.

1.4 Digestion and analysis for heavy metal

The digestion procedure adopted has been described by Alam et al. (2002) taking precautions to prevent cross contamination of the samples. About 1 g of ground tissue sample was transferred into 100 mL teflon beakers and then 10 mL ultra pure concentrated nitric acid was added slowly. Each teflon beaker was covered with a watch glass, and heated at 200°C on a hot plate for 3 hr or until the solution evaporated slowly to near dryness. Two milliliters of 1 mol/L nitric acid was added to the residue and the solution was evaporated again on the hot plate. The digestion was repeated until all organic materials in each sample were completely digested. After cooling, 2.5 mL of 1 mol/L nitric acid was added to the digested residue and was transferred to 25 mL volumetric flasks, then diluted to the mark with deionized water. The samples were then filtered through a 0.45- μ m nitrocellulose membrane filter to yield the final solution for analysis. The samples were then spiked with an internal standard mixture of Scandium, Germanium and Rhodium, in HNO₃ matrix. Sample blanks were subjected to a similar procedure. Samples were then aspirated into an inductively coupled plasma-mass spectrometer (ICP-MS) (Perkin Elmer Sciex Elan 6100 DRC^{plus}, USA) to quantify Cd, Cr, Cu, As, Ni, Pb, Zn and Hg. All metal concentrations obtained were converted into wet weight basis.

1.5 Quality assurance and quality control

The OCP concentrations were quantitatively determined by comparing the area under each peak with the area under the standard peak. For each sample, a procedural blank and a spiked sample consisting of all reagents were run to check for interference and cross contamination. The resulting correlation coefficients (γ) for the calibration curves of OCPs were all greater than 0.998. The limits of detection (LODs) of OCPs were described as three of signal-to-noise ratio (S/N). LODs of OCPs ranged from 0.02 to 0.08 ng/g wet weight (ww). The spiked recoveries of OCPs using 50 ng of composite standards were in the range of 76%–104%. The relative standard deviation (RSD) ranged from 4% to 10%.

For heavy metals, standard solutions were prepared from stock solutions (multi element standard, Merck, USA). Replicate blanks and the reference materials DORM-2 (dogfish muscle, the National Research Council, Canada) and NIST SRM 1566a (oyster tissue, the National Bureau of Standards, USA) were used for quality control. The blanks were not detected with any analyte. The recovery rates for the standard reference materials DORM-2 and NIST SRM 1566a were around 80%–125%. The results showed good agreement between the certified and the analytical values. The recovery of elements being partially complete for most of them. Sample analyses were carried out in duplicates for both OCPs and heavy metals.

1.6 Statistical analysis

Statistical analysis of data was carried out using MINITAB 14 statistical package program. One-way analysis of variance (ANOVA) and Turkey's pair wise comparison tests were used to assess whether OCPs or metal concentrations varied significantly among species.

2 Results and discussion

2.1 OCPs residue levels

As shown in Table 1, the concentrations of the sum of HCHs in the samples from Quanzhou Bay ranged from 0.19 to 0.45 ng/g ww (mean 0.27 ng/g ww), while total DDT levels were in the range of 0.56–53.77 ng/g ww (mean 23.76 ng/g ww). The concentration ranges for HCHs and DDTs in samples collected from Xinghua Bay were 0.18–0.93 ng/g ww (mean 0.48 ng/g ww) and 0.83–109.32 ng/g (mean 26.50 ng/g ww), respectively.

For the species taken from both bays such as *Sinonovacula constricta* and *Meretrix meretrix*, the mean Σ HCHs values were higher in samples from Xinghua Bay than that from Quanzhou Bay, whereas the mean Σ DDTs in *S. constricta* and *M. meretrix* were both higher in the samples from Quanzhou Bay than that from Xinghua Bay. However, the results imply that the bioaccumulation of OCPs in fish is species-specific due to their ecological characteristics (Yang et al., 2006). The greatest concentration of OCPs was recorded in *Cephalopholis sonnerati* (Groupers), which feeds on fishes and crustaceans. Therefore, it appears that OCPs tend to bioaccumulate more in higher trophic levels in an aquatic ecosystem. Pelagic organisms were found to be more subjected to OCPs bioaccumulation than biota relying upon benthic primary production (Kidd et al., 1998).

With respect to the average concentrations of pesticides in marine organisms the proportions of the HCH isomers, α -, β -, γ - and δ -HCH were found to be 13.5%, 17.5%, 37.3% and 31.7% respectively in the samples from Quanzhou Bay while in those from Xinghua Bay the distribution was 20.2%, 14.0%, 35.1% and 30.7% (Fig. 2a), respectively.

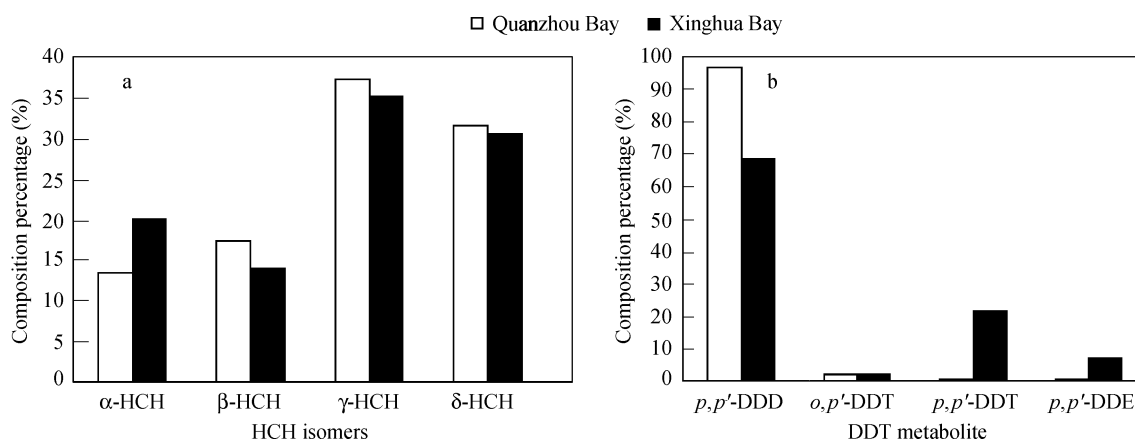
Commercial HCHs are produced mainly in two forms, technical HCHs (α -HCH (55%–80%), β -HCH (5%–14%), γ -HCH (8%–15%) and δ -HCH (2%–16%)) and lindane (pure γ -HCH) (Gong et al., 2007). Therefore, the much lower percentages of α - and β -HCH in the samples than that of technical HCH reflects that technical HCH contamination was mainly due to historical usage. Similarly, the predominance of γ -HCH over α -HCH may suggest that there were some recent inputs of lindane in the study areas.

The mean p,p' -DDD, o,p' -DDT, p,p' -DDT and p,p' -DDE were 96.61%, 2.12%, 0.64% and 0.63%, respectively in the samples from Quanzhou Bay while in those from Xinghua Bay were 68.51%, 2.47%, 21.65% and 7.37%, respectively (Fig. 2b). The precise reasons for the predominance of p,p' -DDD is uncertain because it has been reported that metabolism of DDT in fish is generally accomplished through dechlorination to DDE but generally

Table 1 Concentrations (ng/g ww) of HCHs and DDTs in marine organisms collected from Quanzhou Bay (Q1–Q7) and Xinghua Bay (X1–X8)

Sample	Q1 (n = 2)	Q2 (n = 8)	Q3 (n = 3)	Q4 (n = 6)	Q5 (n = 2)	Q6 (n = 4)	Q7 (n = 5)	Quality STD
Species	<i>S. constricta</i>	<i>P. viridis</i>	<i>M. edulis</i>	<i>M. meretrix</i>	<i>P. japonicus</i>	<i>C. antiquata</i>	<i>G. filamentosus</i>	
α-HCH	0.05 a	0.05 a	0.04 a	0.04 a	0.05 a	0.04 a	0.03 a	
β-HCH	0.06 a	0.05 a	0.05 a	0.04 a	0.13 b	0.05 a	0.03 a	
γ-HCH	0.08 a	0.06 a	0.07 a	0.31 b	0.07 a	0.08 a	0.06 a	
δ-HCH	0.10 a	0.09 a	0.03 a	0.06 a	0.04 a	0.06 a	0.10 a	
∑HCHs	0.29	0.25	0.19	0.45	0.29	0.23	0.22	20
<i>p,p'</i> -DDE	0.15 a	0.16 a	0.13 a	0.14 a	0.18 a	0.13 a	0.16 a	
<i>p,p'</i> -DDD	28.0 bc	50.94 c	45.14 c	16.62 b	0.12 a	19.18 b	0.68 a	
<i>o,p'</i> -DDT	0.16 a	2.54 c	ND	0.60 b	0.09 b	ND	0.13 a	
<i>p,p'</i> -DDT	0.16 a	0.13 a	0.18 a	0.11 a	0.17 a	0.15 a	0.17 a	
∑DDTs	28.47	53.77	45.45	17.47	0.56	19.46	1.14	10
(DDE+DDD)/ DDTs	0.98	0.95	0.59	0.95	0.53	0.99	0.73	
Sample	X1 (n = 8)	X2 (n = 8)	X3 (n = 4)	X4 (n = 7)	X5 (n = 3)	X6 (n = 3)	X7 (n = 5)	X8 (n = 6)
Species	<i>H. discus hannai</i>	<i>C. gigas</i>	<i>Battillaria</i> sp.	<i>E. mordax</i>	<i>C. sonnerati</i>	<i>N. japonicus</i>	<i>S. constricta</i>	<i>M. meretrix</i>
α-HCH	0.05 a	0.07 a	0.04 a	0.04 a	0.29 b	0.11 a	0.11 a	0.07 a
β-HCH	0.05 a	0.03 a	0.04 a	0.06 a	0.12 b	0.09 a	0.07 a	0.08 a
γ-HCH	0.41 b	0.04 a	0.07 a	0.06 a	0.32 b	0.30 b	0.05 a	0.10 a
δ-HCH	0.08 a	0.04 a	0.04 a	0.05 a	0.20 b	0.21 b	0.24 b	0.32 b
∑HCHs	0.59	0.18	0.19	0.21	0.93	0.71	0.47	0.57
<i>p,p'</i> -DDE	0.41 b	0.41 b	0.15 a	1.34 b	13.18 c	0.06 a	0.02 a	0.07 a
<i>p,p'</i> -DDD	ND	7.74 b	43.14 c	0.22 a	54.77 c	16.99 b	11.96 b	10.47 b
<i>o,p'</i> -DDT	0.17 a	1.59 b	3.34 c	0.12 a	ND	ND	ND	ND
<i>p,p'</i> -DDT	0.25 a	0.33 a	0.15 a	0.18 a	41.36 c	2.39 b	ND	1.26 b
∑DDTs	0.83	10.07	46.78	1.86	109.32	19.44	11.98	11.80
(DDE+DDD)/ DDTs	0.49	0.80	0.92	0.83	0.62	0.87	1.00	0.89

a, b, c mean in rows with the same letter were not significantly different ($P > 0.05$). ND: not detected. Quality STD: Marine Biological Quality Standard in China (GB 18421-2001).

**Fig. 2** Percentage compositions of HCHs (a) and DDTs (b) of biological samples of Quanzhou Bay and Xinghua Bay.

not to DDD (Schmitt et al., 1999). However, the relative total concentration of the metabolites to the parent DDT has been used for assessing pollution sources (Doong et al., 2002). Ratios of (DDD + DDE)/DDTs > 0.5 show that DDTs have been subjected to long term weathering. In this study, the ratios DDE+DDD/DDTs were in the range of 0.49–1.00 (Table 1) with most being above 0.80 suggesting that DDTs contaminating marine biota in the study areas may have weathered.

Using the consumption rate of fish and seafood (Du et al., 2004), and assuming the average body weight of 60 kg, the estimated daily intakes (EDI, ng/(kg ww-day)) of the

OCPs were calculated as follows:

$$EDI = C_f \times C_{OCP} / W_b$$

where, C_f (g/day) is the daily fish consumption; C_{OCP} (ng/g ww) is OCP concentration; W_b (kg) is human body weight. The EDIs of HCHs and DDTs in marine organisms collected from Quanzhou Bay were 0.13 ng/(kg ww-day) and 12.00 ng/(kg ww-day) respectively while those values from Xinghua Bay were 0.24 ng/(kg ww-day) and 13.47 ng/(kg ww-day) respectively, indicating that the EDIs were lower than the established acceptable daily intakes (ADIs) issued by FAO/WHO (1993). The results further revealed

that the EDIs were either low or comparable to the EDIs in marine products in some regions of China (Yang et al., 2006; Guo et al., 2007).

2.2 Heavy metals

The average concentrations of heavy metals in the samples from Quanzhou Bay were in the order of Zn > Cu > As > Pb > Ni > Cd > Cr > Hg, while the samples from Xinghua Bay were in the order of Zn > As > Cu > Cd > Cr > Ni > Pb > Hg (Table 2).

The average concentrations of Zn, Cu, Cd, As and Cr in different marine species exceeded the threshold values recommended by national and/or international guidelines, indicating potential risk to both the organisms and consumers. *Sinonovacula constricta* (Razor clam) showed 3.6 folds of maximum permitted limit for Zn (30 mg/kg ww) issued by FAO (1983). Some other species *Meretrix meretrix* (Hard clam), *Crassostrea gigas* (Pacific oyster)

and *Cephalopholis sonnerati* (Grouper) also exceeded the proposed limits for Zn. In contrast, Cu contents in species studied were lower than the FAO limit (30 mg/kg ww). However, Cu contents in *C. gigas*, *M. meretrix*, *H. discus hannai*, *C. gigas* and *Batillaria* (sand snail) were higher than the guideline issued by China (5 mg/kg ww).

The Cd and Pb analyzed in tissues of organisms collected from Quanzhou and Xinghua Bays showed higher concentrations than the permitted limits declared by FAO (0.05 mg/kg ww for Cd and Pb) and China (0.05 mg/kg ww for Cd and 1.0 mg/kg ww for Pb). Among the species, the highest Cd concentration was found in the tissues of *Batillaria* species. Lead concentrations in most of species tissues were much greater than those of Cd. Lead is usually weakly associated with air particles and can be easily dissolved in water (Foster and Charlesworth, 1996). They are therefore highly reactive and biologically available.

The average As concentration analyzed in these tissues

Table 2 Heavy metal concentrations (mg/kg ww) in marine organisms collected from Quanzhou Bay (Q1–Q7) and Xinghua Bay (X1–X8)

Sample	Q1 (n = 2)	Q2 (n = 8)	Q3 (n = 3)	Q4 (n = 6)	Q5 (n = 2)
Species	<i>S. constricta</i>	<i>P. viridis</i>	<i>M. edulis</i>	<i>M. meretrix</i>	<i>P. japonicus</i>
As	1.55 ± 0.02 b	2.44 ± 0.01 b	2.36 ± 0.13 b	1.38 ± 0.10 b	1.98 ± 0.02 b
Cd	0.14 ± 0.01 a	0.18 ± 0.01 a	0.41 ± 0.03 ab	0.40 ± 0.01 ab	0.25 ± 0.02 a
Cr	0.06 ± 0.01 a	0.09 ± 0.01 a	0.20 ± 0.01 a	0.21 ± 0.01 a	0.12 ± 0.02 a
Cu	9.72 ± 1.12 b	3.68 ± 0.53 b	3.95 ± 0.02 b	7.13 ± 0.88 b	3.73 ± 0.16 b
Ni	0.47 ± 0.01 a	1.34 ± 0.02 b	0.86 ± 0.13 a	0.56 ± 0.11 a	0.11 ± 0.10 a
Pb	0.55 ± 0.01 a	0.95 ± 0.01 a	1.54 ± 0.11 ab	2.78 ± 0.01 b	1.33 ± 0.21 ab
Zn	22.32 ± 0.21 a	16.66 ± 1.11 a	22.45 ± 0.10 a	18.21 ± 0.06 a	13.95 ± 0.01 a
Hg	0.0010 ± 0.0001 a	0.0038 ± 0.0003 a	0.0016 ± 0.0002 a	0.0009 ± 0.0001 a	0.0015 ± 0.0003 a
Sample	Q6 (n = 4)	Q7 (n = 5)	All samples		
Species	<i>C. antiquata</i>	<i>G. filamentosus</i>			
As	1.73 ± 0.11 b	0.45 ± 0.01 a	1.69 ± 0.12		
Cd	0.58 ± 0.02 b	0.12 ± 0.01 a	0.30 ± 0.02		
Cr	0.25 ± 0.12 a	0.05 ± 0.03 a	0.14 ± 0.01		
Cu	3.29 ± 0.11 b	0.34 ± 0.01 a	4.55 ± 0.23		
Ni	1.08 ± 0.23 b	0.11 ± 0.01 a	0.67 ± 0.12		
Pb	2.82 ± 0.01 b	1.51 ± 0.11 a	1.64 ± 0.15		
Zn	22.82 ± 2.32 a	9.04 ± 0.53 a	17.92 ± 0.56		
Hg	0.0001 ± 0.0002 a	0.0003 ± 0.0001 a	0.0001 ± 0.0001		
Sample	X1 (n = 8)	X2 (n = 8)	X3 (n = 4)	X4 (n = 7)	X5 (n = 3)
Species	<i>H. discus hannai</i>	<i>C. gigas</i>	<i>Batillaria</i> sp.	<i>E. mordax</i>	<i>C. sonnerati</i>
As	2.70 ± 0.03 b	1.35 ± 0.11 ab	38.65 ± 0.02 d	0.59 ± 0.11 a	6.11 ± 0.20 bc
Cd	2.62 ± 0.01 b	0.97 ± 0.03 b	6.55 ± 0.11 c	0.20 ± 0.03 a	0.03 ± 0.01 a
Cr	1.93 ± 0.02 b	0.71 ± 0.01 ab	0.61 ± 0.05 ab	0.03 ± 0.01 a	1.70 ± 0.08 b
Cu	13.07 ± 0.32 c	18.24 ± 0.23 c	18.55 ± 0.21 c	0.35 ± 0.02 a	1.47 ± 0.08 a
Ni	2.90 ± 0.04 b	0.36 ± 0.02 a	0.22 ± 0.01 a	0.11 ± 0.03 a	0.13 ± 0.01 a
Pb	0.62 ± 0.01 a	0.60 ± 0.01 a	1.73 ± 0.03 ab	1.54 ± 0.02 ab	0.41 ± 0.01 a
Zn	25.67 ± 1.31 ab	35.71 ± 2.18 ab	25.40 ± 1.06 ab	9.22 ± 0.16 a	60.90 ± 3.16 b
Hg	0.0017 ± 0.0002 a	0.0011 ± 0.0001 a	0.0010 ± 0.0003 a	0.0004 ± 0.0001 a	0.0003 ± 0.0005 a
Sample	X6 (n = 3)	X7 (n = 5)	X8 (n = 6)	All samples	
Species	<i>N. japonicus</i>	<i>S. constricta</i>	<i>M. meretrix</i>		
As	10.59 ± 0.02 c	19.40 ± 0.01 c	10.25 ± 0.31 c	11.2 ± 0.20	
Cd	0.01 ± 0.01 a	1.79 ± 0.02 b	0.45 ± 0.04 ab	1.58 ± 0.22	
Cr	1.67 ± 0.02 b	4.06 ± 0.02 c	1.69 ± 0.01 b	1.55 ± 0.02	
Cu	1.29 ± 0.10 a	16.67 ± 0.12 c	7.48 ± 0.11 bc	9.64 ± 0.02	
Ni	0.61 ± 0.02 a	1.73 ± 0.20 b	3.67 ± 0.01 b	1.21 ± 0.01	
Pb	0.25 ± 0.01 a	2.42 ± 0.04 b	0.12 ± 0.01 a	0.96 ± 0.01	
Zn	17.78 ± 1.21 a	108.73 ± 5.12 c	64.84 ± 2.23 b	43.53 ± 0.80	
Hg	0.0954 ± 0.0005 b	0.0394 ± 0.0002 b	0.0701 ± 0.0003 b	0.026 ± 0.0001	

a, b, c mean in rows with the same letter were not significantly different ($P > 0.05$).

from Xinghua Bay showed extremely high values compared to the Australian permissible limit (1.0 mg/kg ww) (Ikem and Egiebor, 2005). The highest arsenic concentration was found in *Battillaria* species (38.65 mg/kg). Some other benthic organisms also showed high levels. In samples the Hg concentrations (from 0.0003 to 0.0954 mg/kg) were far below the proposed limit (0.2 mg/kg ww) issued by WHO (1990) (Table 2).

The concentration of Cr in tissue samples of *H. discus hannai*, *C. sonnerati*, *N. japonicus*, *S. constricta* and *M. meritrix* exceeded the proposed concentration (1 mg/kg). The highest concentration of Cr was observed in *S. constricta* (4.06 mg/kg). However, Cr concentration in tissues of different organisms from Quanzhou Bay showed relatively low values which were below the permissible limit. The mean Ni concentrations ranged from 0.11 to 3.67 mg/kg (mean 1.21 mg/kg) in the samples collected from Xinghua Bay and ranged from 0.11 to 1.34 mg/kg (mean 0.64 mg/kg) in the samples collected from Quanzhou Bay. There is no information about maximum permissible intakes of Ni concentration. However, if an adult who consumed 30.50 g/day (Du et al., 2004) of seafood daily from Xinghua Bay, he would take in approximately 0.036 mg (1.21 mg/kg \times 30.50 g) of Ni each day. Similarly, this would be 0.019 mg for Quanzhou Bay. The calculated results reveal that these values are far below the recommended limit for the upper tolerable intake level of Ni (40 mg/day) for males/females (19–70 years old) issued by Agency for Toxic Substances and Disease Registry (2004). Therefore, the contamination for Ni from the two bays were negligible.

The results implied that, the accumulation of trace elements is higher in shellfish than those in pelagic fish. Shellfish are benthic organisms, and are usually relatively sessile. Based on the feeding mode and living habits, the trace metal content of shellfish most likely reflects the quality of the water and sediment in the aquatic environment (Ip et al., 2005). The significantly elevated concentrations of Zn, Cr, Pb and As found in shellfish likely resulted from the fact that their primary source of food is suspended matter, near or on the sea floor.

This study revealed that the average concentrations of Cd, As and Pb in most organisms from Quanzhou and Xinghua Bays were markedly elevated and were above the recommended legal limits for human consumption (Table 2). The markedly high concentrations of As, Cr, Pb and Zn were also noted in other parts of the world (Ip et al., 2005; Wei et al., 2002; Yap et al., 2004; Yilmaz, 2003; Kavun et al., 2002; Fattorini et al., 2004; Maanan, 2008).

3 Conclusions

Sea foods are a cheap and the most accessible source of animal protein for the coastal inhabitants. However, there is a growing amount of evidence that seafood could be potentially harmful to human health as they are contaminated from pesticides and heavy metals. This study revealed that the commercially important marine organisms in Quanzhou Bay and Xinghua Bay in Fujian Province,

Southeast China have been contaminated by heavy metals to a greater degree and exceeds the recommended safety levels for consumption. The composition distributions of OCPs in marine organisms showed that recent applications of lindane in the catchment areas of the bays while current DDT applications were low in the catchments. Strict regulations for the usage of heavy metals and frequent monitoring and controlling programmes are needed to check heavy metal contamination of marine organisms in the area that may potentially cause greater threats to human and ecological health.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 40473043). Authors are grateful to the Geological Survey of Fujian Province, China.

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