

Levels, Distribution And Risk Assessment of Hexabromocyclododecane (HBCD) In Fish In Xiamen, China

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Abstract

In this study, hexabromocyclododecane (HBCD) was detected in 114 fish samples collected from 6 administrative regions of Xiamen city, China. HBCD amounts ranged between ND (not detected) and 2.216 ng g⁻¹ ww (mean, 0.127±0.318 ng g⁻¹ ww). Besides, α-HBCD was the main diastereoisomer in these fish specimens, followed by β-HBCD. Meanwhile, γ-HBCD was not detected in any of the samples. Significant differences were recorded among fish species. The results indicated that the levels and detection rates of HBCD were higher in *Trachinotus ovatus* compared with other aquatic organisms. Therefore, *Trachinotus ovatus* could be used as a marine biological indicator of HBCD. Within the regions investigated, Siming was significantly different from Jimei, Haicang and Xiang'an. The spatial distribution of HBCD concentrations indicated higher mean levels in samples collected from Haicang, Jimei and Xiang'an, respectively, with the highest detection rates in Jimei and Xiang'an, which might be related to geographical location and intense industrial and urban activities. Estimation of daily HBCD intake was performed according to fish consumption in Xiamen residents. The medium bound HBCD amounts in fish were approximately 0.073 and 0.088 ng kg bw⁻¹ d⁻¹ for male and female residents of Xiamen, respectively. Exposure doses of HBCD indicated no health concern for Xiamen residents.

Introduction

Persistent organic pollutants (POPs) represent an important group of substances with the characteristics of high toxicity, persistence and bioaccumulation. Hexabromocyclododecane (HBCD) has been extensively used as a brominated flame retardant (BFR) for a long period of time in textiles, electronics, construction materials, thermal insulation materials, etc. γ-HBCD constitutes the main diastereoisomer of industrial HBCD, accounting for 75–89% of the overall weight, while α-HBCD (10–15%) and β-HBCD (1–12%) are two other diastereoisomers (Covaci et al., 2006). In 2011, about 31,000 tons of HBCD were produced worldwide (POPRC, 2011a, 2012). The Chinese HBCD production accounts for more than half of the global production (POPRC, 2012). Given the strong persistence, bioaccumulation, liver toxicity, neurotoxicity and immunotoxicity of HBCD (Tomy et al., 2008; Marvin et al., 2011; Zhu et al., 2016; Wang et al., 2016; Zhang et al., 2018), it was included in the list of POPs in 2013 (POPRC8.3, 2013). Besides, its production and utilization have been globally recommended to be banned since 2016. However, HBCD is still produced and applied in China, which allows its use in special building materials.

Additionally, HBCD is found in multiple environmental media, e.g., air, riverine water, sediments, sewage sludge and animal tissues (Ni et al., 2013; Feng et al., 2012; Gorga et al., 2013; Xia et al., 2018), as well as in humans (Kim et al., 2014) and biota in pristine regions, including the Antarctic Peninsula. Due to its low water solubility and high lipophilicity, HBCD can easily accumulate in aquatic organisms. Previous studies (Shi et al., 2009; Törnkvist et al., 2011; Barghi et al., 2016) demonstrated that HBCD levels are generally elevated in aquatic foods compared with other food products. Therefore, environmental pollution and human health problems caused by extensive application of HBCD have significantly attracted the attention of the international community.

Xiamen, in the southeast coast of China, currently undergoes rapid urbanization and industrialization; thus, seawater in this area tends to have poor quality. Furthermore, fish accounts for a great proportion of the dietary composition of Xiamen residents. This makes Xiamen residents even more vulnerable to exposure to persistent organic pollutants (POPs) via fish intake than inland inhabitants. Despite the importance of the abovementioned serious health problem in Xiamen, there are limited studies (Zhang et al., 2012; Qian et al., 2017) examining “classic” POPs, including DDT, HCH and PCB, in aquatic products in Xiamen, China. Hence, to date, data related to permissible levels of environmental exposure and associated risk factors in Xiamen are scarce.

No large-scale study focusing on HBCD quantitation in fish samples has been performed in Xiamen until now. Therefore, this work aimed to assess the contamination status of HBCD in fish collected from Xiamen, as well as species and regional distributions. Additionally, the related health risk for local residents consuming fish was estimated. We, for the first time, determined the estimated daily intake (EDI) of HBCD in Xiamen residents.

Materials And Methods

2.1. Sample collection

In the present research, as shown in Table S1, 114 fish samples were collected from 6 administrative regions (Siming, Huli, Haicang, Jimei, Xiang'an and Tong'an) of Xiamen between March 2017 and December 2018 (Fig. 1). Totally 14 fish species were regularly used by local residents as food. For a meaningful interpretation of data, we collected the same fish species during the same seasons in various regions. Specimens were preserved in a car refrigerator and transported within hours to the laboratory. In the laboratory, dorsal muscles were taken, homogenized, lyophilized, grinded and stored at -20 °C till analysis.

2.2. Chemicals and analytical methods

The chemicals and analytical methods used for HBCD have been described previously (Qian et al., 2021). The details of chemicals and analytical methods are provided in the Supporting Information.

2.3. Quality control (QC)

HBCD was identified according to relative retention time and the corresponding selected reaction monitoring (SRM) for various analytes. To prevent potential unexpected matrix effects, isotopically labeled standards were utilized for HBCD isomers. Then, a matrix-matched calibration curve was generated for quantification. A 5-point calibration curve spanning the concentration range of 1–100 ng·mL⁻¹ including internal standards was employed ($R^2 \geq 0.990$). The limit of detection (LOD) on the column, defined as a signal-to-noise ratio of 3:1, was 0.05 ng·g⁻¹. Procedural blank specimens were run after a block of 20 samples to rule out contamination. Recovery and precision rates were evaluated by spiking different matrix samples with HBCD at two concentration levels (0.25 ng·g⁻¹ and 0.5 ng·g⁻¹ ww).

Recovery rates were 70–110%. Intra-day reproducibility ($0.25 \text{ ng}\cdot\text{g}^{-1}$) for HBCD detection ($n = 5$) was 4.3–10.3%. Inter-day reproducibility ($0.5 \text{ ng}\cdot\text{g}^{-1}$) for HBCD detection ($n = 6$) was 8.2–11.2%. Spiking assay data are given in supplementary materials (Table S2). HBCD concentrations per wet weight (ww) were derived from Eq. (1) (Table 1).

$$C_i = C_j \times (100\% - \omega_{H_2O}), \quad (1)$$

where C_i is HBCD level in $\text{ng}\cdot\text{g}^{-1}$ (ww) in the wet specimen, C_j represents HBCD amounts ($\text{ng}\cdot\text{g}^{-1}$) in the lyophilized specimen, and ω_{H_2O} is the wet specimen's water content (%), derived from Eq. (2).

$$\omega_{H_2O} = \frac{m_w - m}{m_w} \times 100\%, \quad (2)$$

where m_w and m represent the weights of the wet and lyophilized samples, respectively (g).

The laboratory's performance was validated through participation in an interlaboratory comparison test for HBCD powered by the State Oceanic Administration People's Republic of China. Data reported by our laboratory were within consensual values.

2.4. Statistical analysis

SPSS 20.0 (SPSS, USA) was utilized for analysis. All concentrations in fish samples were presented on a ww basis. Levels below the LOD were set to half of the LOD. Data were presented as median, mean \pm standard deviation (SD) and range, respectively. Data normality and homogeneity of variance were assessed, followed by logarithm transformation. The data were all non-normally distributed. The nonparametric Kruskal-Wallis (KW) test was utilized for comparing HBCD concentrations among species and regions. $P < 0.05$ indicated statistical significance.

2.5. Daily intake calculations

Estimated dietary intake (EDI) of HBCD ($\text{ng kg}^{-1} \text{ d}^{-1}$) was assessed as follows:

$$EDI = \frac{C \times DR}{B_w}, \quad (3)$$

where C represents mean HBCD level (ng g^{-1} ww), DR is the daily consumption rate of fish (g d^{-1}), and B_w is mean human body weight (70.3 and 57.8 kg for adult males and females, respectively), according to the National Physique Monitoring Center of China (2014).

Results And Discussion

3.1. HBCD amounts in fish and isomer profile

In the present study, HBCD was detected in 30.7% of all studied specimens at amounts between ND to 2.216 ng g⁻¹ ww (mean, 0.127 ± 0.318 ng g⁻¹ ww). Of note, α-HBCD was the remarkably predominant diastereoisomer, whereas β-HBCD was only detected in one fish sample. Besides, unlike environmental samples and commercial HBCD, γ-HBCD was found in no fish specimen. α-HBCD predominance in fish specimens most likely results from elevated assimilation and reduced elimination rates of α-HBCD in comparison with β- and γ-HBCD (Law et al., 2006; Du et al., 2012; Zhang et al., 2013; Eljarrat et al., 2014). The bio-isomerization of β- and γ-HBCD to α-HBCD was suggested as another potential reason (Luo et al., 2013; Su et al., 2018). In addition, HBCD's bioavailability may influence the diastereoisomer profile of HBCD. Water solubility levels of α-, β- and γ-HBCD are 48.8, 14.7 and 2.1 μg·L⁻¹, respectively (Hunziker et al., 2004). This meant α- and β-HBCD are more easily enriched in aquatic organisms via the aquatic environment. Therefore, α-HBCD exhibited higher bioaccumulation than β- and γ-HBCD in aquatic organisms.

HBCD levels (range, ND-2.216 ng g⁻¹ ww; mean, 0.127 ± 0.318 ng g⁻¹ ww) detected in the present study were approximately 2 times higher than those reported by another work (range, ND-1.1 ng/g ww; median, 0.3 ng/g ww; mean, 0.41 ± .41 ng g⁻¹ ww) conducted in Beijing (Wang et al., 2014), and also higher than those observed in twelve fish species (range, ND-0.194 ng g⁻¹ ww; mean, 0.016 ng g⁻¹ ww) collected in 11 Chinese coastal cities (Meng et al., 2012). In addition, reduced HBCD amounts were found in the current fish specimens in comparison with those from fish caught in the Japanese coast (range, ND-77.3 ng g⁻¹ ww; Nakagawa et al., 2010) and a Japanese fish market (range, ND-21.9 ng g⁻¹ ww; Kakimoto et al., 2012). A great deal of HBCD-based studies have been conducted in European countries, where HBCD is used intensively. HBCD amounts found in this work were slightly elevated than described for seafood products in Belgium (range, ND-0.84 ng g⁻¹ ww; Goscinny et al., 2011), Sweden (range, 0.11–0.63 ng/g ww; mean, 0.145 ng g⁻¹ ww; Törnkvist et al., 2011) and France (range, 0.01–0.55 ng g⁻¹ ww; Munsch et al., 2013). However, HBCD levels in the current work were lower than reported for seafood products in Czech Republic (range, 0.02–11.6 ng g⁻¹ ww; median, 0.44 ng g⁻¹ ww; Hloušková et al., 2013), the Netherlands (range (marine), ND-7.3 ng g⁻¹ ww; range (freshwater eel), ND-230 ng g⁻¹ ww; Leeuwen et al., 2008) and Scotland (range, 0.03–12.1 ng g⁻¹ ww; Fernandes et al., 2008). Jointly, fish HBCD levels in this study were in the middle range of those previously reported.

Table 1

Concentrations and detection rates of HBCD in fish samples obtained from Xiamen (ng g⁻¹ ww)

Species	α -HBCD	β -HBCD	γ -HBCD	HBCD	
	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Detection rates (%)
<i>Psenopsis anomala</i>	ND	ND	ND	ND	—
<i>Larimichthys crocea</i>	0.210 \pm 0.246 0.161 (ND-0.580)	ND	ND	0.210 \pm 0.246 0.161 (ND-0.580)	50.0
<i>Scophthalmus maximus</i>	0.513 \pm 0.868 0.112 (ND-2.216)	ND	ND	0.513 \pm 0.868 0.112 (ND-2.216)	66.7
<i>Trichiurus lepturus</i>	ND	ND	ND	ND	—
<i>Nibea albiflora</i>	0.165 \pm 0.231 0.041 (ND-0.530)	0.040 \pm 0.098 ND (ND-0.239)	ND	0.205 \pm 0.313 0.041 (ND-0.770)	50.0
<i>Acanthopagrus latus</i>	0.037 \pm 0.074 ND (ND-0.219)	ND	ND	0.037 \pm 0.074 ND (ND-0.219)	25.0
<i>Nemipterus virgatus</i>	ND	ND	ND	ND	—
<i>Decapterus maruadsi</i>	0.111 \pm 0.146 0.041 (ND-0.333)	ND	ND	0.111 \pm 0.146 0.041 (ND-0.333)	50.0
<i>Lateolabrax japonicus</i>	0.029 \pm 0.072 ND (ND-0.232)	ND	ND	0.029 \pm 0.072 ND (ND-0.232)	16.7
<i>Trachinotus ovatus</i>	0.353 \pm 0.427 0.257 (ND-0.333)	ND	ND	0.353 \pm 0.427 0.257 (ND-0.333)	83.3
<i>Epinephelus awoara</i>	0.045 \pm 0.079 ND (ND-0.194)	ND	ND	0.045 \pm 0.079 ND (ND-0.194)	33.3
<i>Hapalogenys nitens</i>	0.239 \pm 0.586 ND (ND-1.435)	ND	ND	0.239 \pm 0.586 ND (ND-1.435)	16.7
<i>Pampus argenteus</i>	0.106 \pm 0.211 ND (ND-0.527)	ND	ND	0.106 \pm 0.211 ND (ND-0.527)	33.3

ND: Not detected SD□Standard deviation

Species	α -HBCD	β -HBCD	γ -HBCD	HBCD	
	Mean \pm SD	Mean \pm SD	Mean \pm SD	Mean \pm SD	Detection rates (%)
	Median (range)	Median (range)	Median (range)	Median (range)	
<i>Pagrus major</i>	0.070 \pm 0.181 ND (ND-0.602)	ND	ND	0.070 \pm 0.181 ND (ND-0.602)	16.7
Total	0.124 \pm 0.315 ND (ND-2.216)	0.002 \pm 0.023 ND (ND-0.239)	ND	0.127 \pm 0.318 ND (ND-2.216)	30.7

ND: Not detected SD Standard deviation

3.2. *Trachinotus ovatus* as a biological indicator of HBCD

The highest detection rates were found in *Trachinotus ovatus* (83.3%) and *Scophthalmus maximus* (66.7%), followed by *Larimichthys crocea* (50.0%), *Nibea albiflora* (50.0%) and *Decapterus maruadsi* (50.0%). The mean value above 0.100 ng g⁻¹ ww decreased stepwise in the following order: *Scophthalmus maximus* (0.513 \pm 0.868 ng g⁻¹ ww), *Trachinotus ovatus* (0.353 \pm 0.427 ng g⁻¹ ww), *Larimichthys crocea* (0.210 \pm 0.246 ng g⁻¹ ww), *Nibea albiflora* (0.205 \pm 0.313 ng g⁻¹ ww) and *Decapterus maruadsi* (0.111 \pm 0.146 ng g⁻¹ ww). In the present study, statistical analysis showed *Trachinotus ovatus* was markedly different from other fish species (KW test; p < 0.05). In addition, the pollution level and detection rate of HBCD in *Trachinotus ovatus* in Xiamen were higher than those of other aquatic organisms, as shown in Fig. 2. Selective HBCD bioaccumulation in *Trachinotus ovatus* may be related to the metabolism, habitat and diet of this organism. *Trachinotus ovatus* is widely distributed in the coastal areas of Xiamen, with strong disease resistance and endurance. Besides, it belongs to warm-water pelagic fish, and feeds on small fishes, plankton and crustaceans. To some extent, *Trachinotus ovatus* could be chosen as a marine biological indicator of HBCD, as it sensitively responds to HBCD exposure.

3.3. Regional distribution of HBCD in Xiamen

Table 2 shows the amounts and detection rates of HBCD in fish samples collected from 6 administrative regions (Siming, Huli, Haicang, Jimei, Tong'an, and Xiang'an) of Xiamen. These six regions showed marked geographical differences (KW test, p < 0.05; Fig. 3). Siming, where no HBCD was found in fish, is significantly different from Jimei, Haicang, Xiang'an. The average HBCD level (0.031 \pm 0.067 ng g⁻¹ ww) and detection rate (21.1%) in fish samples observed in Huli were the lowest among regions. This was consistent with the economic pattern of Siming and Huli, where the government has mainly developed commercial and cultural industries. The reason why pollution level was higher in Huli compared with Siming is serious pollution by sewage outlets in Dailiao as well as north of the Wuyuanwan Bridge (Xiamen Municipal Bureau of Ocean Development, 2017). This might have a serious adverse impact on

seawater quality, which needs strict supervision and management. The detection rates of HBCD in fish samples collected in Haicang, Jimei and Xiang'an were between 40% and 50%. However, the mean concentration of HBCD in Haicang, where industry is developed, was almost 2- and 3-fold higher than those of Jimei and Tong'an, respectively. These results can be explained in different aspects: (1) Haicang and Jimei are located in the west coast of Xiamen, where water quality is assessed as the fourth level, according to the single factor evaluation method (Xiamen Municipal Bureau of Ocean Development, 2018); (2) Electronics, machinery and biopharmaceutical industries are concentrated in Haicang; (3) as the earliest cultural and educational administrative area of Xiamen, with a dense population, the discharge of domestic sewage in Jimei in the past decades has caused a remarkable damage to the adjacent sea areas. Thus, discharge of land-based pollutants, including industrial sewage and domestic wastewater, beyond the self-purification capacity of bays, represented the major factor promoting accumulation and pollution of HBCD in Haicang and Jimei. Nevertheless, it is noteworthy that the maximum value and detection rate of HBCD was found in Xiang'an, demonstrating that rapid industrial development and urbanization in Xiang'an in recent years may lead to new environmental problems.

Table 2
Detection rates and levels of HB CD in fish specimens collected in 6 different regions of Xiamen

Sampling Regions	α -HBCD	β -HBCD	γ -HBCD	HBCD	
	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Mean \pm SD Median (range)	Detection rates (%)
Siming	ND	ND	ND	ND	—
Huli	0.031 \pm 0.067 ND (ND-0.215)	ND	ND	0.031 \pm 0.067 ND (ND-0.215)	21.1
Haicang	0.221 \pm 0.401 ND (ND-1.637)	0.013 \pm 0.055 ND (ND-0.239)	ND	0.248 \pm 0.407 0.112 (ND-1.637)	42.1
Jimei	0.127 \pm 0.181 ND (ND-0.602)	ND	ND	0.127 \pm 0.181 ND (ND-0.602)	47.4
Tong'an	0.085 \pm 0.168 ND (ND-0.580)	ND	ND	0.085 \pm 0.168 ND (ND-0.580)	26.3
Xiang'an	0.283 \pm 0.577 ND (ND-2.216)	ND	ND	0.283 \pm 0.577 ND (ND-2.216)	47.4

ND: Not detected SD□Standard deviation

3.4. EDI of HBCD through fish consumption

HBCD can enter the human body through a variety of ways, including diet, skin contact and respiration, of which dietary intake is the main pathway. Estimation of daily intake of HBCD was carried out based on seafood consumption (Qian et al., 2017). Values relative to mean bodyweight (70.3 and 57.8 kg for adult males and females, respectively) were obtained from the General Administration of Sport of China. Because HBCD amounts in some specimens were below LOD, values < LOD were considered to be zero ("lower bound" approach, LB), 0.5×LOD ("medium bound", MB) or equal to LOD ("upper bound", UB). Average intake levels in male Xiamen residents exposed to HBCD were 0.064 ng kg bw⁻¹d⁻¹ (LB), 0.073 ng kg bw⁻¹d⁻¹ (MB) and 0.081 ng kg bw⁻¹d⁻¹ (UB), respectively. Average daily intake levels of HBCD in Xiamen females were 0.078 ng kg bw⁻¹d⁻¹ (LB), 0.088 ng kg bw⁻¹d⁻¹ (MB) and 0.099 ng kg bw⁻¹d⁻¹ (UB), respectively, which were slightly higher than in men. These EDI values were elevated in comparison with those reported for residents of South China via fish consumption (12.5–16.0 ng kg bw⁻¹d⁻¹) (Meng et al., 2012). EDI values in this study were also elevated than 0.01–1.00 ng kg bw⁻¹d⁻¹ and 0.004–0.37 ng kg bw⁻¹d⁻¹ found in urban and rural residents of Chinese coastal areas through fish consumption (Xia et al., 2011). Additionally, the present values were lower than those reported for Taiwan China (0.252 ng kg bw⁻¹d⁻¹; Lee et al., 2019) and the Yangtze River Delta of China (0.181 ng kg bw⁻¹d⁻¹; Zhang et al., 2013b), and markedly reduced in comparison with those found in the Netherlands (0.12 ng kg bw⁻¹d⁻¹; van Leeuwen et al., 2008), Korea (males: 0.392 ng kg bw⁻¹d⁻¹, females: 0.252 ng kg bw⁻¹d⁻¹; Barghi et al., 2016) and Japan (1.3–3.7 ng kg bw⁻¹d⁻¹; Nakagawa et al., 2010).

A no-observed-adverse-effect level (NOAEL) of 10.2 mg kg bw⁻¹d⁻¹ in rats was recommended on the basis of a two-generation reproductive toxicity study (Ema et al., 2008). Considering interspecies extrapolation from animals to humans and potential intraspecies differences in sensitivity among humans, an uncertainty factor (100) was introduced to estimate equivalent effect levels in humans. Thus, our EDIs correspond to 0.064–0.098% of the NOAEL, suggesting low potential health risk to Xiamen residents through fish consumption. On the other hand, the average daily intake of fish by Xiamen residents was 35.5 g d⁻¹, which is much higher than the national aquatic product consumption (14.8 g d⁻¹) reported in the 2017 China Statistical Yearbook. In addition, Shi et al. (2017) demonstrated that the mean HBCD level in aquatic food in China showed an upward trend from 2007 to 2011. HBCD utilization in construction materials is still allowed in China, suggesting HBCD pollution might continue to increase in the future. Therefore, although HBCD amounts in Xiamen fish in this study may not threaten the health of Xiamen residents, it is important to monitor the HBCD concentration trend in fish collected from Xiamen.

Conclusions

This study provided the first data on isomer-specific HBCD levels in a variety of fish in Xiamen. HBCD was detected in > 30% of the examined fish samples, and the levels were comparable with those found in other studies. Significant differences in the amounts of HBCD were recorded among the species. This study found that *Trachinotus ovatus* could be chosen as a marine biological indicator of HBCD. Among

the regions investigated, the pollution levels of Haicang, Jimei and Xiang'an were relatively serious. Although the daily intakes in Xiamen from fish were below the proposed thresholds, it is important to monitor the HBCD concentration trend in fish collected from Xiamen due to higher daily intake of fish by Xiamen residents.

Declarations

Ethics approval and consent to participate: Not applicable.

Consent for publication: Not applicable.

Availability of data and materials: The datasets utilized or analyzed in this study are available from the corresponding author upon reasonable request.

Competing interests: The authors have no competing interests to declare.

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Authors' contributions: Zhuozhen Qian conceived the study, performed data analysis and drafted the manuscript. Zhiyu Liu provided critical points for discussion. Shuifen Tang and Fangfang Luo carried out additional analyses. Shaohong Wei collected the data.

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Figures



Figure 1

Geographical locations of sampling sites

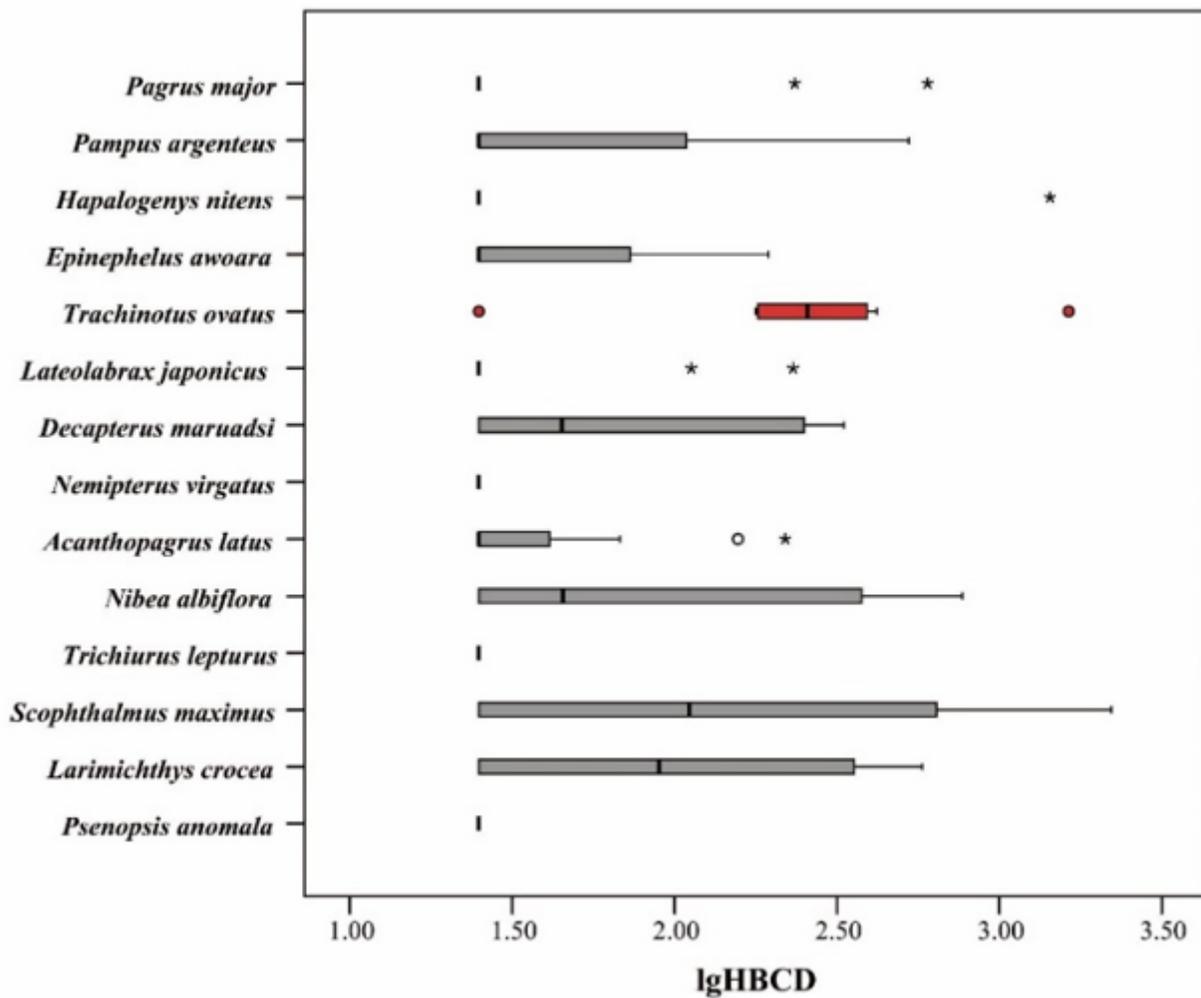


Figure 2

HBCD concentrations in different fish species

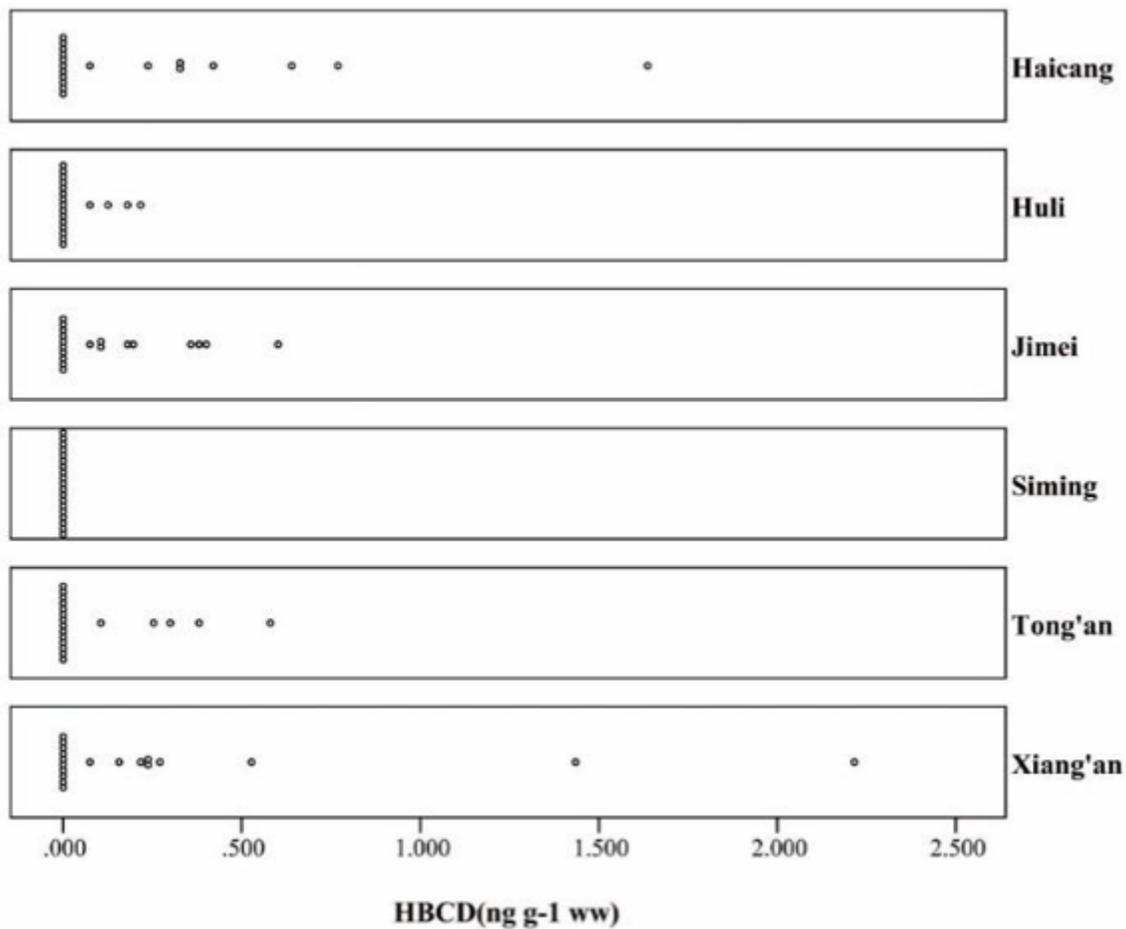


Figure 3

HBCD concentrations in the 6 administrative regions

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