

# Applicability of *Corbicula* as a bioindicator for monitoring organochlorine pesticides in fresh and brackish waters

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**Abstract** The applicability of *Corbicula* as a bioindicator for monitoring organochlorine pesticides (OCPs) in fresh and brackish waters is presented here. Differences in isomer compositions and OCP bioaccumulation levels were analyzed in western Japan and the Pearl River Delta (PRD) in China. Isomer compositions of DDTs, chlordanes, and HCHs were significantly different between the two areas because of their different historical uses and property of the chemicals. This is

represented by the (DDE + DDD)/DDT ratio in *Corbicula*, ranging 4.9–39 in western Japan and 1.1–2.4 in the PRD. However, isomer compositions in *Corbicula* reflected those in water, and the different patterns in *Corbicula* likely reflected the usage history. Concentrations of dissolved oxygen, suspended solids, and volatile suspended solids in water, and the difference in species did not influence OCP bioaccumulative levels in *Corbicula* when conducting biomonitoring. These levels are likely similar to those in *Mytilus galloprovincialis*. Therefore, *Corbicula* could be an appropriate bioindicator for monitoring OCPs in fresh and brackish waters.

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## Introduction

Organochlorine pesticides (OCPs) are persistent organic pollutants (POPs) used widely throughout the world since the 1950s. These pollutants have caused great concern because of their toxic effects, bioaccumulation, persistence, and long-range atmospheric transport (Jimenez 1997; Monirith et al. 1999; Zhang et al. 2002; Yamashita et al. 1993). Worldwide surveys of POPs, including OCPs, using mussels have been conducted in

seawaters (Boonyatumanond et al. 2002; Khaled et al. 2004; Munsch et al. 2008). Monitoring OCPs in fresh and brackish waters is important for understanding of local pollution and identifying pollution sources.

Field surveys of OCPs using various types of fish have been conducted in freshwaters to determine the pollution levels in fish, original sources of contamination, and risks to public health (Bordajandi et al. 2003; Jacobs et al. 2002; Monirith et al. 1999). However, when low environmental OCP concentrations are to be detected and monitored using bioindicators at specific points for long periods, fish have disadvantages due to their mobility. Thus, it is essential to establish biomonitoring methods using bivalves based on relationships between contaminant concentrations in bivalves and those in the environment.

Bioaccumulation is the uptake of chemical compounds by biota from either water or food. It is clearly known that lipids serve as the primary storage compartment of hydrophobic organic chemicals, and bioaccumulation factors on a lipid basis (BAFLs) were used to show the bioaccumulative levels of target chemicals and analyze bioaccumulation characteristics in aquatic organisms (Loonen et al. 1997; Mackay and Fraser 2000; Nichols et al. 2007). In addition, there are generally positive relationships between bioaccumulative levels in aquatic organisms and the hydrophobicities of the chemicals, and the relationship between bioaccumulative levels and  $\log K_{ow}$  are also often used (Mackay and Fraser 2000; Voutsas et al. 2002).

Because *Corbicula* which is sediment-dwelling bivalve exists worldwide (Byrne et al. 2000; Korniuschin 2004; Park and Kim 2003), it has the potential to be an effective worldwide bioindicator. Additionally, *Corbicula* has limited mobility and is easy to collect. *Corbicula leana* and *Corbicula japonica* are widely distributed in fresh and brackish waters in Japan, respectively, and *Corbicula sandai* occurs only in Lake Biwa (Nishino and Watanabe 2000). Meanwhile, *Corbicula fluminea* and *Corbicula fluminalis* are distributed in fresh and brackish waters, respectively, in China (Korniuschin 2004; Morton 1986). In addition, *C. fluminalis*, *C. japonica*, and *C. sandai*

are dioecious and *C. fluminea* and *C. leana* are hermaphroditic (Korniuschin 2004; Morton 1986; Takahashi 1989; Vidal et al. 2002).

Although field surveys have been conducted using *Corbicula* (Brown 1997; Zhou et al. 2008), only OCP concentrations in *Corbicula* were measured. Although bioaccumulation properties of OCPs and polychlorinated biphenyls (PCBs) in *Corbicula* have been studied (Colombo et al. 1995; Doherty 1990; Mcleod et al. 2008), it is not known whether concentrations in *Corbicula* reflect environmental concentrations in areas with different environmental conditions and pesticide use histories. Before conducting worldwide biomonitoring using *Corbicula*, it is important to understand its applicability in various environments.

In this study, field surveys to evaluate bioaccumulation levels were conducted using *Corbicula* in rivers and lakes in western Japan and in the Lake Biwa–Yodo River System. Sampling areas were later expanded to the Pearl River Delta (PRD) in China, where environmental conditions and OCP usage history differ from those in Japan.

The Lake Biwa–Yodo River System is located in the Kinki region of Japan. The upstream area consists primarily of arable fields, while the downstream area consists primarily of residential districts and land used for commercial and industrial purposes. Although the water system was polluted approximately 30 years ago, it was not comprehensively surveyed for OCPs.

The PRD is located in the developed southern part of China where water pollution has become a severe problem because of increasing urban wastewater discharges, resulting from rapid industrialization and urbanization (Zhai et al. 2005). The area has the highest pesticide use in China (Zhang et al. 2002). Previous studies in the PRD have focused on POP concentrations in water, sediment, and air (Li et al. 2006; Luo et al. 2004; Zhang et al. 2002); however, few surveys have been conducted using living organisms (Fu et al. 2003).

In this study, the applicability of *Corbicula* as a bioindicator for monitoring OCPs in the environment was analyzed using results from field surveys that reflected different environmental conditions and OCP usage histories. The study

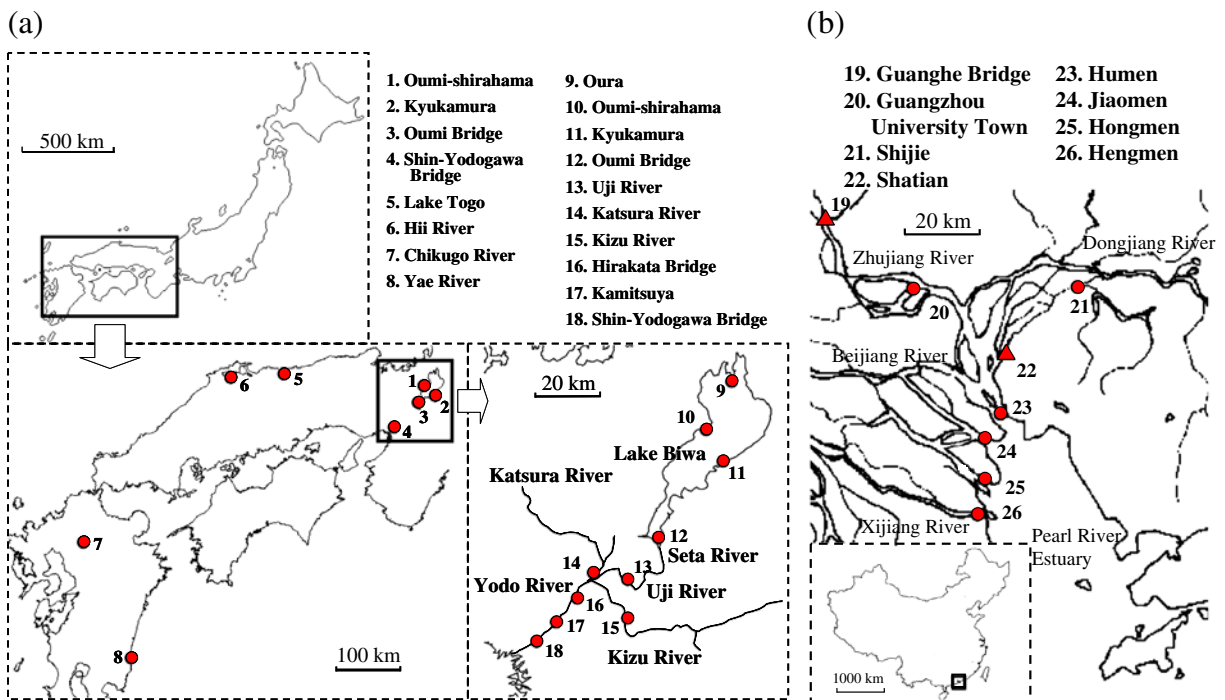
focused on differences in isomer compositions and bioaccumulation levels of OCPs. Additionally, the bioaccumulation levels of *Corbicula* were compared with those of *Mytilus galloprovincialis*, which has been widely used as a bioindicator in seawaters.

**Methods**

**Samples**

Sample selection complied with “Manual for monitoring surveys” of the Ministry of the Environment of Japan (2004). Approximately 50 pieces of single species *Corbicula*, which originally lived at each sampling point (*Corbicula sandai* (sampling points 1, 2, 3, 9, 10, 11, and 12 in Fig. 1), *C. leana* (6, 7, 13, 14, 15, 16, and 17), and *C. japonica* (4, 5, 8, and 18) in western Japan and *C. fluminea* (20 and 21) and *C. fluminalis* (23, 24, 25, and 26)

in the PRD), 12 L of water, and 100 g of surface sediment were collected at each point. *Corbicula* shells were washed with river or lake water at each sampling point in order to remove sediment from the shell. *Corbicula*, with a similar and dominant shell length, was collected at each point. The similar shell length was indicated by small variation coefficients (0.065–0.22 in western Japan; 0.068–0.11 in the PRD) at each point. In western Japan, the mean weight excluding shell was 0.28–2.4 g (wet) and shell length was 1.3–2.6 cm. In the PRD, mean weight excluding shell was 0.33–1.4 g (wet) and shell length was 1.3–2.3 cm. In accordance with a previous study (Colombo et al. 1995), *Corbicula* was not depurated. Collected *Corbicula* and sediment were transported in polyethylene (Ziplock) bags and glass bottles, respectively, and preserved at –20°C; the water was transported in glass bottles and preserved at 4°C. GF/B filters with pore size of 1.0 μm (Whatman, USA) were used to filter out soluble matter and suspended solids.



**Fig. 1** Maps showing sampling points in western Japan (a) and the Pearl River Delta (b). The triangles in (b) show points in which *Corbicula* could not be collected

## Sampling field

Samples were collected from eight points in western Japan between 28 October and 27 November 2005 (Fig. 1). The second field survey was conducted at 10 points in the Lake Biwa–Yodo River System between 7 June and 5 September 2007. Results from both surveys were combined and referred to as those from western Japan. Points at Oumi-shirahama, Kyukamura, Oumi Bridge, and Shin-Yodogawa Bridge were surveyed in both years; other points were surveyed once. Samples were collected once from eight points in the PRD between 24 October and 26 November 2007 (Fig. 1). As sufficient numbers of *Corbicula* clams for the pretreatment could not be obtained at two points (Guanghe Bridge and Shatian), only water and sediment were collected.

## Water quality

Water temperature, pH, dissolved oxygen (DO), and salinity concentrations were monitored using multiparameter monitoring system U-21 (Horiba, Japan) in western Japan. Water temperature and DO concentrations were measured using LDO HQ10 (Hach Company, USA), and pH was metered with pH meter sensION 3 (Hach Company, USA) in the PRD. Concentrations of suspended solids (SS) and volatile suspended solids (VSS) were determined at each point by combusting the filtration paper at 105°C for 2 h and 550°C for 30 min, respectively (Standard method, 2005).

## Sample processing and analysis

The ES-5261-1.2 Persistent Organic Pollutants Clean-up Spike (Cambridge Isotope Laboratories, USA) and the syringe spike MBP-101 ( $^{13}\text{C}_{12}$ -PCB #101) (Wellington Laboratories, Canada) were used to measure aldrin, dieldrin, endrin, mirex, hexachlorobenzene (HCB), dichlorodiphenyltrichloroethane (DDT), chlordane, heptachlor, nonachlor, hexachlorocyclohexane (HCH), and the possible isomers and metabolites of each chemical. The composition of the clean-up spike is shown in Table 1. The same pretreatment

**Table 1** Composition of the clean-up spike

Target chemical	Clean-up spike
Aldrin	Aldrin ( $^{13}\text{C}_{10}$ , 99%)
Dieldrin	Dieldrin ( $^{13}\text{C}_{10}$ , 99%)
Endrin	Endrin ( $^{13}\text{C}_{10}$ , 99%)
Mirex	Mirex ( $^{13}\text{C}_{10}$ , 99%)
HCB	HCB ( $^{13}\text{C}_6$ , 99%)
<i>o,p'</i> -DDT, <i>p,p'</i> -DDT	<i>p,p'</i> -DDT ( $^{13}\text{C}_{12}$ , 99%)
<i>o,p'</i> -DDE, <i>p,p'</i> -DDE	<i>p,p'</i> -DDE ( $^{13}\text{C}_{12}$ , 99%)
<i>o,p'</i> -DDD, <i>p,p'</i> -DDD	<i>p,p'</i> -DDD ( $^{13}\text{C}_{12}$ , 99%)
<i>cis</i> -chlordane, <i>trans</i> -chlordane	<i>trans</i> -chlordane ( $^{13}\text{C}_{10}$ , 99%)
<i>oxy</i> -chlordane	<i>oxy</i> -chlordane ( $^{13}\text{C}_{10}$ , 99%)
<i>cis</i> -nonachlor, <i>trans</i> -nonachlor	<i>trans</i> -nonachlor ( $^{13}\text{C}_{10}$ , 99%)
$\alpha$ -HCH	$\alpha$ -HCH ( $^{13}\text{C}_6$ , 99%)
$\beta$ -HCH, $\delta$ -HCH	$\beta$ -HCH ( $^{13}\text{C}_6$ , 99%)
$\gamma$ -HCH	$\gamma$ -HCH ( $^{13}\text{C}_6$ , 99%)
Heptachlor	Heptachlor ( $^{13}\text{C}_{10}$ , 99%)
<i>cis</i> -heptachlor-epoxide, <i>trans</i> -heptachlor-epoxide	<i>cis</i> -heptachlor-epoxide ( $^{13}\text{C}_{10}$ , 99%)

procedure used for soluble matter and SS, with the exception of the silica gel column (Varian, USA) clean-up process, was used as previously described PCB pretreatment (Takabe et al. 2009).

The following procedure was used for soluble matter. 400 ml of *n*-hexane (Wako, Japan) was added to 12 L of filtered water and the mixture was then agitated for 24 h. After the *n*-hexane was recovered, 2,000 pg of the clean-up spike was added. The solvent was dehydrated with sodium sulfate and concentrated to 0.5 mL with Turbo Vap 500 (Zymark, USA).

The following procedure was used for SS. The filter paper including the filter cake was cut into small pieces and added to 20 mL of acetone (Wako, Japan). The sample was ultrasonicated for 10 min followed by centrifugation at 3,000 rpm for 10 min. This procedure was repeated twice. The same procedure using *n*-hexane was done twice. The composite contained 2,000 pg of the clean-up spike. Then, 300 mL of 5% sodium chloride solution was added to the liquid solution, the mixture was then shaken for 1 min, and then the *n*-hexane layer was recovered. 60 ml of *n*-hexane was added to the remaining water layer to dissolve the acetone, and the mixture was shaken for 1 min. The *n*-hexane layer was recovered and added

to the earlier one. The solvent was dehydrated with sodium sulfate. It was filtered using the silica gel column, and concentrated to 0.5 mL with Turbo Vap 500.

The pretreatment process used for SS was applied for 20 g wet sediment samples. Concentrations on a dry weight basis were calculated using moisture contents. The median and range of the moisture in sediment were 20 (16–29) % in western Japan and 37 (19–49) % in the PRD.

The following procedure was used for *Corbicula*. Edible parts, excluding shell, of at least 20 pieces of *Corbicula* were homogenized in order to avoid sex bias among the samples. Twenty grams (wet) of homogenized sample were weighed. Then, 2,000 pg of the clean-up spike and 20 mL of acetone were added to the sample, which was homogenized and centrifuged at 3,000 rpm for 10 min. The supernatant was separated and recovered. This procedure was repeated using the resulting residue in order to collect 40 mL of acetone. Acetone was extracted with two portions of 60 mL *n*-hexane. The hexane solution was then dehydrated with sodium sulfate and concentrated to 20 mL with Turbo Vap 500. The concentrated solution was extracted using two 60-mL portions of saturated acetonitrile (Wako, Japan) with *n*-hexane. Acetonitrile was extracted with two 60-mL portions of *n*-hexane to eliminate impurities. Target OCPs were caught in the hexane layer. The solution was dehydrated with sodium sulfate, purified through the silica gel column, and then concentrated to 0.5 mL with Turbo Vap 500.

In 2005, analyses were performed using a 5890N high-resolution gas chromatograph (Agilent Technologies, USA) connected to JMS-700D high-resolution mass spectrometer (JEOL, Japan). In 2007, analyses were performed using a 6890N high-resolution gas chromatograph (Agilent Technologies, USA) connected to a JMS-800D high-resolution mass spectrometer (JEOL, Japan) operating at a resolution of >10,000. Splitless injection of 1  $\mu$ L was used; 500 pg of the syringe spike was added to samples prior to injection. Samples were analyzed in a fused silica capillary column (HP Ultra-2, 60  $\times$  0.25 mm, SGE, Australia). The column was heated from 120°C to 180°C at a rate of 20°C/min, to 210°C at a rate of 2°C/min and to 300°C at a

rate of 5°C/min. The temperature was held for 2 min. Mass spectrometry was performed using selected ion monitoring mode with an electron multiplier voltage of 10 eV.

#### Quality control for OCP analysis

Blank tests for each pretreatment were performed in western Japan and the PRD. Because significant HCB concentrations were found in the blank tests from the PRD compared to those in the field samples, findings on HCB in the PRD were omitted. Concentrations of all target chemicals were negligible in western Japan, and all concentrations of the other chemicals in the blank tests were negligible in the PRD.

Except for labeled aldrin, mean recoveries varied between 61% and 99% for soluble matter, 67% and 112% for SS, 53% and 98% for *Corbicula*, and 62% and 117% for sediment. The recoveries were in acceptable range according to the recovery ratio range of 50% to 120% given by Ministry of the Environment of Japan (2004). Also, the mean recovery ratio of the labeled aldrin was 30% for soluble matter, 34% for SS, 33% for *Corbicula*, and 30% for sediment.

Instrument detection limits ( $\sigma \times 2 \times t(6,0.05)$ , Table 2) and instrument quantification limits ( $\sigma \times 10$ ) were based on the standard deviations of seven repeated analyses with a 0.8 pg/L concentration of target chemicals. Instrument detection limits of sediment based on dry weight were calculated with moisture contents.

#### Analysis of lipid content in *Corbicula*

Edible parts (5 g-wet), excluding shell were homogenized with 80 mL of chloroform and 80 mL of methanol. After filtering through GF/B filters (Whatman, USA), 60 mL of ultrapure water was added to recover chloroform. Chloroform was dehydrated with sodium sulfate and dried at 30°C, and a ratio of the residue to the original weight was used as the lipid content (Ministry of the Environment of Japan 2002).

The lipid content in *Corbicula* varied between 0.35% and 1.9% in western Japan and between 0.75% and 1.1% in the PRD.



**Table 2** Concentrations of target chemicals in western Japan and the Pearl River Delta and instrument detection limits of target chemicals

Chemicals	Western Japan						The Pearl River Delta						Instrument detection limit			
	Water (pg/L)			Sediment (pg/g-dry)			Water (pg/L)			Corbicula (pg/g-wet)			SM	SS	Corbicula	Sediment
	Median	Min.	Max.	Median	Min.	Max.	Median	Min.	Max.	Median	Min.	Max.	(pg/L)	(pg/L)	(pg/g-wet)	(pg/g-dry)
Aldrin	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.46	0.91	0.27	0.32–0.60
Dieldrin	53	20	490	86	20	1,100	2.0	53	7.1	n.d.	43	15	0.41	0.83	0.25	0.30–0.56
Endrin	1.5	n.d.	30	4.7	n.d.	47	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.6	3.1	0.94	1.1–2.1
Mirex	n.d.	n.d.	n.d.	1.2	n.d.	9.9	n.d.	n.d.	19	8.0	86	62	0.17	0.34	0.10	0.12–0.22
HCB	67	4.4	250	79	14	1,400	22	300	n.d.	n.d.	n.d.	n.d.	0.16	0.33	0.098	0.12–0.22
DDTs	76	14	430	2,000	240	7,500	61	11	970	220	6,300	7,700	0.45	0.90	0.27	0.32–0.60
<i>o,p'</i> -DDT	n.d.	n.d.	n.d.	22	n.d.	210	n.d.	n.d.	79	23	660	850	0.79	1.6	0.47	0.56–1.0
<i>p,p'</i> -DDT	n.d.	20	81	n.d.	n.d.	740	n.d.	n.d.	260	73	3,600	2,400	4.5	9.0	2.7	3.2–6.0
<i>o,p'</i> -DDE	7.1	n.d.	32	15	n.d.	210	1.1	n.d.	110	12	59	200	0.21	0.41	0.12	0.14–0.27
<i>p,p'</i> -DDE	41	10	240	1,200	240	4,600	39	5.4	360	43	880	1,700	0.26	0.53	0.16	0.19–0.35
<i>o,p'</i> -DDD	3.6	n.d.	59	130	n.d.	650	11	n.d.	270	15	370	480	0.24	0.48	0.14	0.17–0.31
<i>p,p'</i> -DDD	19	n.d.	98	240	n.d.	1,100	24	4.2	340	54	2,000	2,200	0.45	0.90	0.27	0.32–0.60
Chlordanes	220	62	900	1,000	400	15,000	110	11	2,700	58	360	620	0.45	0.90	0.27	0.32–0.60
<i>cis</i> -chlordane	81	15	320	260	120	4,400	37	2.8	820	17	150	250	0.39	0.79	0.24	0.29–0.54
<i>trans</i> -chlordane	52	18	300	190	69	3,500	8.9	2.0	830	17	140	220	0.12	0.23	0.069	0.082–0.15
oxy-chlordane	6.3	n.d.	23	8	n.d.	220	1.1	n.d.	25	6.9	n.d.	6.0	0.48	0.95	0.29	0.34–0.64
<i>cis</i> -nonachlor	23	5.4	68	210	53	1,500	9.5	1.7	240	10	6.5	22	0.33	0.65	0.20	0.24–0.45
<i>trans</i> -nonachlor	69	16	190	390	140	5,100	15	3.1	740	14	41	100	0.35	0.70	0.21	0.25–0.47
HCHs	1,400	110	4,700	190	15	1,700	26	4.5	340	270	7,500	73	0.34	0.68	0.21	0.25–0.47
$\alpha$ -HCH	210	27	1,200	32	n.d.	180	7.0	n.d.	20	85	5,400	23	0.34	0.68	0.21	0.25–0.47
$\beta$ -HCH	1,000	42	4,200	130	3.8	1,500	23	1.7	310	97	1,100	20	0.60	1.2	0.36	0.43–0.80
$\gamma$ -HCH	100	17	410	12	n.d.	78	4.2	n.d.	10	63	650	22	0.55	1.1	0.33	0.39–0.73
$\delta$ -HCH	25	n.d.	570	7.0	n.d.	59	1.3	n.d.	13	15	400	8.4	0.41	0.82	0.24	0.29–0.54
Heptachlor	6.6	n.d.	54	7.0	n.d.	120	n.d.	n.d.	7.2	2.3	18	5.3	n.d.	n.d.	n.d.	n.d.
Heptachlor-epoxide	n.d.	n.d.	n.d.	2.8	n.d.	46	n.d.	n.d.	n.d.	n.d.	n.d.	2.3	0.40	0.79	0.24	0.29–0.54
<i>cis</i> -heptachlor-epoxide	6.6	n.d.	54	5.6	n.d.	79	n.d.	n.d.	3.8	2.3	18	2.6	0.21	0.43	0.13	0.15–0.29
<i>trans</i> -heptachlor-epoxide	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.20	0.40	0.12	0.14–0.27

n.d. not detected (below the instrument detection limit), SM soluble matter, SS suspended solids

**Results and discussion**

Hereafter, median, minimum, and maximum values are presented as median (minimum–maximum).

**Water quality**

There were no remarkable differences in water temperature and pH between the two areas, 27.6 (24.3–28.9)°C and 6.9–8.2, respectively, in western Japan, and 27.3 (23.9–29.3)°C and 7.1–8.1, respectively, in the PRD. DO concentrations in western Japan were 9.3 (8.4–12) mgO<sub>2</sub>/L, while the concentrations in the PRD were 2.2 (1.4–5.4) mgO<sub>2</sub>/L, significantly low by Wilcoxon tests with significant level of  $p < 0.05$  ( $p = 0.00019$ ). The concentrations of SS and VSS were 9.6 (1.6–25) mg/L and 3.0 (0.51–5.5) mg/L, respectively, in western Japan, and at 76 (28–180) mg/L and 27 (21–71) mg/L, respectively, in the PRD, significantly higher by Wilcoxon tests with significant level of  $p < 0.05$  (SS,  $p = 0.00019$ ; VSS,  $p = 0.00019$ ).

**OCP concentrations**

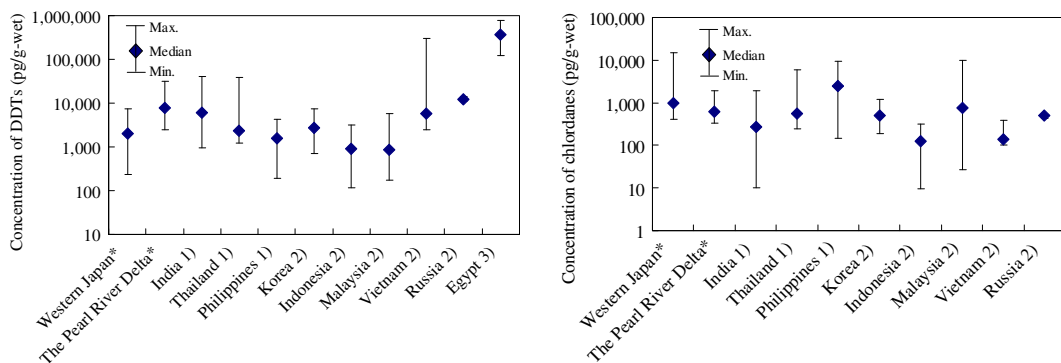
Concentrations of target chemicals in water, *Corbicula*, and sediment from western Japan and the PRD are summarized in Table 2. OCP concentrations in soluble matter and suspended solids were combined, and the concentrations in water were defined as the total concentrations. Concentrations of chlordanes are summations

of *cis*-chlordanes, *trans*-chlordanes, *oxy*-chlordanes, *cis*-nonachlor, and *trans*-nonachlor. DDTs, HCHs, and heptachlors signify a summation of respective isomers and metabolites.

In *Corbicula*, concentrations of DDTs and chlordanes were highest in western Japan. The highest concentrations of both chlordanes and DDTs were observed at Katsura River. Concentrations of DDTs were highest in *Corbicula* from the PRD. The highest concentrations of DDTs were observed at Hongmen. There were large differences in concentrations of dieldrin, mirex, and DDTs in *Corbicula* between western Japan and the PRD, but no remarkable differences were observed in concentrations of the other chemicals.

Field surveys using different species of bivalves have been reported in various seawater studies (Fig. 2; Khaled et al. 2004; Monirith et al. 2003; Tanabe et al. 2000). Concentrations of DDTs in *Corbicula* in western Japan were lower than those in bivalves in other Asian regions. Concentrations of DDTs in *Corbicula* in the PRD belonged to the high concentration group of bivalves in Asia; however, the concentrations were significantly lower than those found in Egypt by Wilcoxon tests with significant level of  $p < 0.05$  ( $p = 0.00046$ ), where recent DDT use has been suggested (Khaled et al. 2004). Compared with other Asian regions, concentrations of chlordanes were high in western Japan and at mid-range levels in the PRD.

In western Japan, concentrations of chlordanes and DDTs were highest in sediment, and concentrations of HCHs with low hydrophobicity were



**Fig. 2** Concentrations of DDTs and chlordanes in bivalves compared with study results (\*) and results from previous studies: (1) Tanabe et al. (2000), (2) Monirith et al. (2003), and (3) Khaled et al. (2004)

high in water. DDT concentrations were highest in sediment, and HCHs and DDTs were highest in water from the PRD. There were large differences in concentrations of dieldrin, mirex, and DDTs in water and mirex and DDTs in sediment, between western Japan and the PRD, but no remarkable differences were observed in concentrations of the other chemicals.

There were significant differences in concentrations in the soluble matter (pg/L) to those in the suspended solids (pg/L) ratios in water from western Japan and the PRD by Wilcoxon tests with significant level of  $p < 0.05$  (DDTs:  $p = 0.0011$ , chlordanes:  $p = 0.00015$ ). In western Japan, ratios for DDTs and chlordanes were 2.2 (0.24–10) and 1.7 (0.51–7.1); meanwhile, ratios were 0.41 (0.089–1.1) and 0.71 (0.39–0.85) in the PRD. OCPs were comparatively easily adsorbed onto the surface of SS probably because of the high SS and VSS concentrations in the PRD.

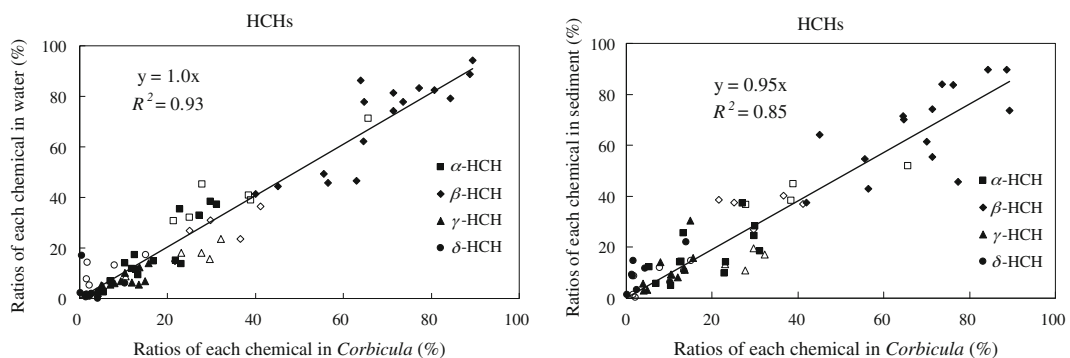
#### Composition of DDTs, chlordanes, and HCHs

Regression analysis was conducted between ratios of each chemical in DDTs, chlordanes, and HCHs in *Corbicula* and those in water, and between the ratios in *Corbicula* and those in sediment in order to compare the compositions of the chemicals in *Corbicula* to those in water and sediment. Because *o,p'*-DDT and *p,p'*-DDT were almost below the instrument detection limit in water and sediment samples in western Japan, the ratios of DDTs in western Japan were calculated using the other

metabolites. The results of the regression analysis of HCHs are shown in Fig. 3 as a representation. Consequently, a slope of almost 1 (DDTs, 0.90; chlordanes, 0.89; HCHs, 1.0) and high correlations (DDTs:  $R^2 = 0.83$ , chlordanes:  $R^2 = 0.72$ , DDTs:  $R^2 = 0.93$ ) were obtained in each line between the compositions in *Corbicula* and those in water. Also, a slope of almost 1 (DDTs, 0.90; chlordanes, 0.95; HCHs, 0.95) and high correlations (DDTs:  $R^2 = 0.86$ , chlordanes:  $R^2 = 0.77$ , DDTs:  $R^2 = 0.85$ ) were obtained in each line between the compositions in *Corbicula* and those in sediment. Therefore, it can be concluded that the compositions in *Corbicula* could well reflect those in water and sediment, regardless of the metabolism in *Corbicula*. This can also be demonstrated by the existing composition ratios in each media (Fig. 4).

*p,p'*-DDE accounted for the largest share of total DDT concentration in each media, followed by *p,p'*-DDD in western Japan; the PRD showed high proportions of *p,p'*-DDT and *p,p'*-DDD (Fig. 4).

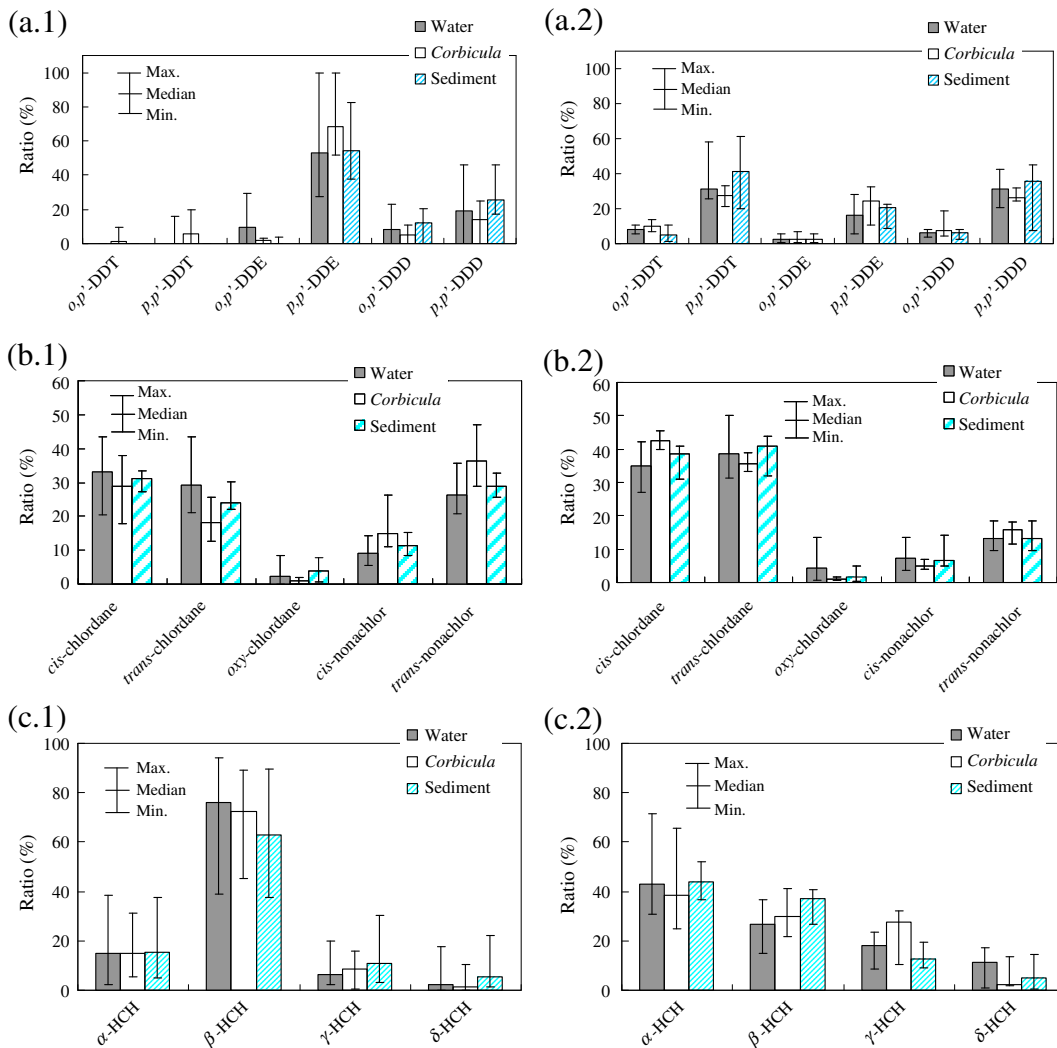
The (DDE + DDD)/DDT ratio has been suggested as an indicator to detect new DDT loads at low levels (e.g., Pereira et al. 1996). In western Japan, neither *o,p'*-DDT nor *p,p'*-DDT was detected in sediment samples. The ratios were 7.8 and 21 in water at two sampling points where DDT was detected. The ratio was 16 (4.9–39) in *Corbicula* at all sampling points, except for five points where neither *o,p'*-DDT nor *p,p'*-DDT was detected. The ratios from the PRD were as low as 1.5 (0.45–2.0) in water, 1.7 (1.1–2.4) in



**Fig. 3** Relationship between ratios of each isomer of chemicals of HCHs in *Corbicula* and those in water and between the ratios in *Corbicula* and those in sediment.

Black color marks and white ones mean the date from western Japan and the PRD, respectively





**Fig. 4** Compositions of DDTs (a), chlordanes (b), and HCHs (c) in water, *Corbicula*, and sediment. (1) Shows compositions in western Japan and (2) shows compositions in the Pearl River Delta

*Corbicula*, and 1.1 (0.61–3.3) in sediment. The 12-year gap in the prohibition of DDT use (from 1971 in Japan and from 1983 in China) (Zhang et al. 2002) likely contributed to the higher levels of residual DDT in the PRD.

Dicofol and technical DDT have been reported as DDT pollutants in China. Technical DDT contains less *o,p'*-DDT (≈15%) than *p,p'*-DDT (≈85%), while dicofol exhibits the opposite pattern (Qiu et al. 2005). Because the half-lives of *o,p'*-DDT and *p,p'*-DDT in soil are similar (Meijer et al. 2001), the high *p,p'*-DDT/*o,p'*-DDT ratios in water [4.6 (2.6–5.5)], *Corbicula* [2.5 (1.9–

4.3)], and sediment [5.6 (3.9–6.2)] indicate higher contributions of technical DDT in the PRD.

In western Japan, DDD/DDE ratios were 0.48 (0.092–1.1) in water, 0.31 (0.21–0.49) in *Corbicula*, and 0.44 (0.21–0.61) in sediment. The high proportion of DDE was likely attributed to reports that DDE is more persistent than either DDT or DDD (Aguillar 1984; Gong et al. 2004). In contrast, DDD/DDE ratios in water, *Corbicula*, and sediment were high in the PRD: 2.2 (0.98–3.8), 1.5 (0.78–3.8), and 2.4 (0.78–4.3). DDT metabolizes primarily to DDD under anaerobic conditions (Pereira et al. 1996), and low DO

concentrations were measured in this study. Therefore, there is a possibility that surveyed sediment may have been dominated by anaerobic conditions, and the metabolism of DDT into DDD may have contributed to the high proportion of DDD in the PRD.

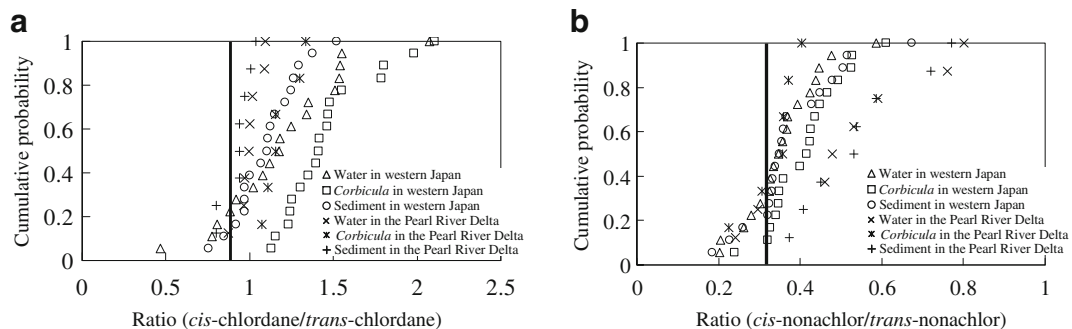
*Cis*-chlordane, *trans*-chlordane, and *trans*-nonachlor comprised the largest shares of chlordanes in western Japan; proportions of *cis*-chlordane and *trans*-chlordane were high in the PRD (Fig. 4). *Oxy*-chlordane was detected at extremely low levels in *Corbicula* in both western Japan and the PRD. Therefore, *Corbicula* appeared to have a poor ability to metabolize chlordane compounds.

Compositions of technical chlordane in Japan were measured in this study as *cis*-chlordane–*trans*-chlordane–*cis*-nonachlor–*trans*-nonachlor = 16:18:4.5:14 and were similar to those in technical chlordane in the international market (Buchert et al. 1989; Sovocool et al. 1977). In western Japan, *cis*-chlordane/*trans*-chlordane ratios in water, *Corbicula*, and sediment were higher than those in technical chlordane (0.89): 1.2 (0.50–2.1), 1.4 (1.1–2.1), and 1.1 (0.75–1.5) (Fig. 5). Because the production, import, and use of chlordane have been banned in Japan since 1981, *cis*-chlordane appears to be more persistent than *trans*-chlordane, which agrees with a previous study by Feroz and Khan (1979). *Cis*-nonachlor/*trans*-nonachlor ratios in water, *Corbicula*, and sediment were also higher than those in technical chlordane (0.32): 0.35 (0.20–0.59), 0.42 (0.24–0.61), and 0.35 (0.18–0.67) (Fig. 5). Probably *cis*-

nonachlor was thought to be more persistent than *trans*-nonachlor in western Japan.

A previous study compares *cis*-chlordane/*trans*-chlordane ratios in the atmosphere in the PRD to those in technical chlordane in the international market. The ratios in the international market were similar to those in technical chlordane from western Japan in this study (Yang et al. 2008). *Cis*-chlordane/*trans*-chlordane ratios in water, *Corbicula*, and sediment were 1.0 (0.87–1.1), 1.2 (1.1–1.3), and 0.94 (0.80–1.0), respectively, in the PRD, and these ratios were similar to those in soils from the PRD in a previous report (Li et al. 2006). In addition, the ratios in the PRD were closer to those in technical chlordane in the international market than the samples from western Japan (Fig. 5). It has been reported that chlordane has been widely used in China (Jaward et al. 2005). Therefore, it is likely that the recent usage of chlordane in China contributed to the ratio being close to technical chlordane. The ratios of *cis*-nonachlor/*trans*-nonachlor in water, *Corbicula*, and sediment in the PRD were 0.52 (0.24–0.80), 0.33 (0.23–0.40), and 0.54 (0.37–0.77), respectively (Fig. 5).

The  $\beta$ -HCH concentration was highest in western Japan, while  $\alpha$ -HCH dominated in the PRD (Fig. 4). Composition of technical HCH in Japan has been reported as  $\alpha$ -HCH (65–70%),  $\beta$ -HCH (7–10%),  $\gamma$ -HCH (14–15%), and  $\delta$ -HCH (approximately 7%), which is similar to that in China (Li et al. 2006; Takazawa et al. 2005).  $\beta$ -HCH is the least volatile and most persistent among HCHs (Buser and Mueller 1995; Willett et al.



**Fig. 5** Ratios of *cis*-chlordane/*trans*-chlordane (**a**) and *cis*-nonachlor/*trans*-nonachlor (**b**). Solid line shows ratios of technical chlordane in Japan in this study

1998). Besides, a long period of time has passed since the expiration of HCH registration as a pesticide in 1971 in Japan. These facts likely contribute to high percentages of  $\beta$ -HCH in western Japan.

Although high proportions of  $\beta$ -HCH in samples from the PRD suggest little influence by current input of technical HCH,  $\alpha$ -HCH existed at higher percentages in the PRD than in western Japan. In 1983, HCH production and use were officially banned in China (Zhang et al. 2002). Therefore, the 12-year gap between the prohibition of HCHs in Japan and China likely influenced the regional differences in HCH concentrations. Moreover, lindane is currently being used in China (Wang et al. 2007), and the  $\alpha/\gamma$  ratio has been used as an indicator for the presence of lindane (Gonzalez et al. 2003; Qian et al. 2006). The ratios in water, *Corbicula*, and sediment are lower than those in technical HCH (4.3–5.0), [2.5 (1.3–8.2), 1.3 (0.78–6.2), and 3.1 (2.3–5.7)], likely presented as a combined pollutant in the PRD.

The pollution characteristics of DDTs, chlordanes, and HCHs in western Japan were clearly different from the characteristics in the PRD because of different usage histories and property of the chemicals. The isomer compositions in *Corbicula* could well reflect those in water and sediment, regardless of the metabolism in *Corbicula*. Therefore, the extent of pollution by chemicals in water and sediment can be measured using *Corbicula* based on isomer compositions.

#### Bioaccumulation levels of OCPs in *Corbicula*

BAFLs [(pg/g lipid)/(pg/mL)] of each chemical in *Corbicula* were defined as the ratio of the concentration of each OCPs in *Corbicula* and the total concentration of each OCPs in soluble matter and suspended solids. BAFLs of some chemicals were not calculated because chemicals with concentrations below instrument quantification limits in water were omitted from this discussion so as not to overestimate their BAFLs.

Single species *Corbicula* was collected at each sampling point. As a result, the same species were collected at different points. Also, BAFLs of the same species from different points in western

Japan were compared. The maximum BAFL/minimum BAFL ratios of each chemical ranged from 1.1 to 7.3 in *C. sandai* taken from Lake Biwa, from 1.4 to 4.1 in *C. leana* in freshwater areas with no salinity (excluding Lake Biwa), and from 1.3 to 5.8 in *C. japonica* in brackish waters with salinity. The ratios were less than 10 in each species. When biomonitoring of OCPs is conducted for comprehending the fate in the environment, it is important that OCP concentrations in the environment should be obtained up to the same level of precision. Therefore, it can be suggested that bioaccumulative levels of each *Corbicula* species were similar regardless of sampling site.

In order to compare the bioaccumulative levels of OCPs between each *Corbicula* species in western Japan, data for each *Corbicula* species were combined, respectively. And then, regression analysis for logBAFL versus logKow was performed on the combined data (Eqs. 1, 2, and 3). Values of logKow were taken from ones recommended by Sangster Research Laboratories. Where recommended values were not fixed in the database, the most recent measured values were used (Sangster Research Laboratories 2222). The correlations in each regression equation were high (Fig. 6).

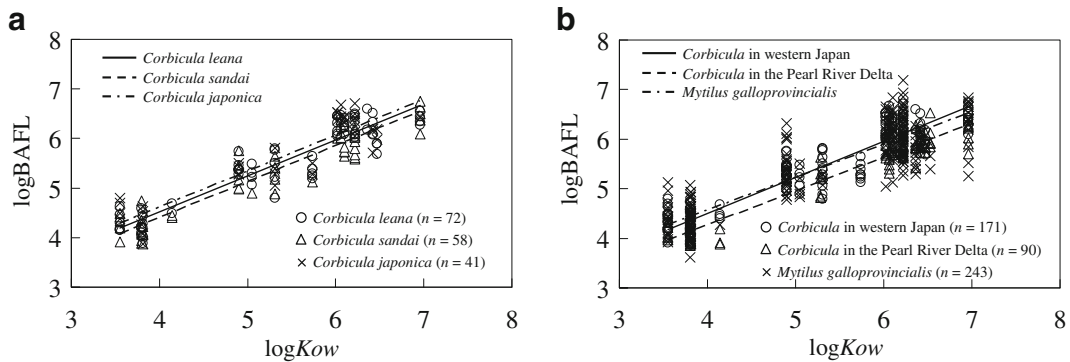
$$C. sandai : \log\text{BAFL} = 0.726 \times \log\text{Kow} + 1.50 \quad (R^2 = 0.890) \quad (1)$$

$$C. leana : \log\text{BAFL} = 0.726 \times \log\text{Kow} + 1.62 \quad (R^2 = 0.874) \quad (2)$$

$$C. japonica : \log\text{BAFL} = 0.723 \times \log\text{Kow} + 1.72 \quad (R^2 = 0.845) \quad (3)$$

*Corbicula* is a sediment dwelling bivalve and obtains food through filter feeding and pedal feeding within the streambed (Hakenkamp and Palmer 1999). However, the close relationship between logBAFL and logKow suggest that OCP concentrations in each *Corbicula* species reflect those in water.

In order to compare the three regression lines for each *Corbicula* species, analysis of covariance was conducted to test the parallelism with a significant level  $p < 0.10$  (Chazdon 1985; Okuno



**Fig. 6** Relationships between  $\log Kow$  and  $\log BAFL$  for *C. leana*, *C. sandai*, and *C. japonica* in western Japan (**a**), and for *Corbicula* in western Japan, *Corbicula* in the Pearl River Delta and *M. galloprovincialis* (Tsuno et al. 2007) (**b**)

and Shiga 1984). In the case when the hypothesis of the parallelism was not rejected, assuming the three regression lines were parallel, a second set of regression lines for each *Corbicula* species was calculated. Next, the intercepts of the second regression lines were compared to one another. In the case when the hypothesis of the parallelism of the first regression lines was rejected or the intercepts of the second regression lines were obtained up to a different level of precision, it would mean that bioaccumulation levels of OCPs are largely different among *Corbicula* species.

The following null hypothesis was used to test parallelism: the regression coefficients of Eqs. 1, 2, and 3 are equal. The null hypothesis was not rejected ( $p = 0.998$ ), concluding that there were no significant differences among the regression coefficients. Thus, assuming the three equations were parallel, a second set of regression equations (Eqs. 4, 5, and 6) was calculated.

$$C. sandai : \log BAFL = 0.725 \times \log Kow + 1.50 \quad (4)$$

$$C. leana : \log BAFL = 0.725 \times \log Kow + 1.62 \quad (5)$$

$$C. japonica : \log BAFL = 0.725 \times \log Kow + 1.71 \quad (6)$$

When biomonitoring of OCPs is conducted for comprehending the fate in the environment, it is important that OCP concentrations in the envi-

ronment should be obtained up to the same level of precision. Therefore, small differences among constant terms can be suggested. Based on the parallelism and similarity among constant terms, the differences among species and therefore the differences between fresh and brackish waters seem to make no digit difference on bioaccumulation levels of OCPs in *Corbicula*.

In order to compare the bioaccumulative levels of OCPs between *Corbicula* in western Japan and that in the PRD, data for *Corbicula* from western Japan and that from the PRD were combined, respectively (BAFLs of each chemical in *Corbicula* in western Japan and the PRD are shown in Table 3). And then, regression analysis for  $\log BAFL$  versus  $\log Kow$  was performed on the combined data (Eqs. 7 and 8). The regression analysis with data for *M. galloprovincialis* in Japan (Tsuno et al. 2007) was also shown (Eq. 9). High correlations were obtained in each regression equation (Fig. 6).

$$Corbicula \text{ in western Japan} : \log BAFL \\ = 0.726 \times \log Kow + 1.60 \quad (R^2 = 0.866) \quad (7)$$

$$Corbicula \text{ in the PRD} : \log BAFL \\ = 0.681 \times \log Kow + 1.55 \quad (R^2 = 0.900) \quad (8)$$

$$M. galloprovincialis : \log BAFL \\ = 0.677 \times \log Kow + 1.82 \quad (R^2 = 0.727) \quad (9)$$

The same test for parallelism with the same significance level was used. The null hypothesis was

**Table 3** Bioaccumulation factors on a lipid basis of organochlorine pesticides

Chemicals	Western Japan (×1,000)			The Pearl River Delta (×1,000)		
	Mean	Min.	Max.	Mean	Min.	Max.
Dieldrin	260	95	570	220	160	330
Endrin	170	78	300	biqlw		
Mirex	biqlw			190	66	430
HCB	300	62	640	biqlw		
<i>o,p'</i> -DDT	biqlw			1,300	710	3,400
<i>p,p'</i> -DDT	2,700	1,400	4,000	830	270	1,900
<i>o,p'</i> -DDE	910	490	1,500	820	540	1,300
<i>p,p'</i> -DDE	3,000	1,200	5,600	1,300	500	1,900
<i>o,p'</i> -DDD	1,700	510	3,200	1,100	540	2,100
<i>p,p'</i> -DDD	2,100	570	3,400	740	410	1,200
<i>cis</i> -chlordane	1,600	440	2,700	770	480	1,100
<i>trans</i> -chlordane	1,200	400	2,700	660	420	990
<i>cis</i> -nonachlor	2,300	1,000	4,700	470	390	590
<i>trans</i> -nonachlor	2,300	870	5,100	600	480	730
$\alpha$ -HCH	19	7.6	55	9.0	7.1	17
$\beta$ -HCH	18	7.4	43	14	7.6	22
$\gamma$ -HCH	27	8.1	62	13	9.4	21
$\delta$ -HCH	33	25	48	12	8.2	16
<i>cis</i> -heptachlor-epoxide	230	130	440	biqlw		

*biqlw* below the instrument quantification limit in water

not rejected ( $p = 0.109$ ), concluding that there were no significant differences among the regression coefficients. Thus, assuming that the three equations were parallel, a second set of regression equations (Eqs. 10, 11, and 12) was calculated.

$$\begin{aligned} & \text{Corbicula in western Japan : logBAFL} \\ & = 0.694 \times \log Kow + 1.77 \end{aligned} \tag{10}$$

$$\begin{aligned} & \text{Corbicula in the PRD : logBAFL} \\ & = 0.694 \times \log Kow + 1.48 \end{aligned} \tag{11}$$

$$\begin{aligned} & \text{M. galloprovincialis : logBAFL} \\ & = 0.694 \times \log Kow + 1.77 \end{aligned} \tag{12}$$

Small differences among the constant terms would have also been suggested. Based on these results, there seems to be no digit differences on bioaccumulation levels among *Corbicula* in western Japan, *Corbicula* in the PRD and *M. galloprovincialis*.

Although concentrations of DO, SS, and VSS were significantly different between the two areas, the covariance analysis revealed similar linear dependencies. Therefore, the range of environmental conditions seems to make no digit differences on the bioaccumulation levels in *Corbicula* used for biomonitoring.

The similar linear regressions between *Corbicula* and *M. galloprovincialis* whose feeding habit is filter feeding (Viarengo and Canesi 1991) support the idea that their bioaccumulation levels are similar despite pedal feeding of *Corbicula*. This result also means that under the same chemical conditions, *Corbicula* would accumulate the same levels of chemicals as would *M. galloprovincialis*.

Finally, by combining data for *Corbicula* in western Japan and the PRD with that for *M. galloprovincialis*, the following regression equation can be proposed:

$$\begin{aligned} & \text{Corbicula and M. galloprovincialis : logBAFL} \\ & = 0.684 \times \log Kow + 1.75 (R^2 = 0.782) \end{aligned} \tag{13}$$

### Conclusions

In this study, field surveys with *Corbicula* were conducted in western Japan and the PRD that reflected different environmental conditions and OCP usage histories. Applicability of *Corbicula* as a bioindicator for monitoring OCPs in the environment was analyzed. The study focused on differences in isomer compositions and bioaccumulation levels of OCPs. Additionally, the



bioaccumulation levels of *Corbicula* were compared with those of *M. galloprovincialis*, which has been widely used as a bioindicator in seawaters. Obtained results are presented below.

1. DO concentrations in western Japan were 9.4 (8.4–12) mgO<sub>2</sub>/L, while the concentrations in the PRD were significantly low, 2.6 (1.4–5.4) mgO<sub>2</sub>/L. The concentrations of SS and VSS were 11 (1.6–25) mg/L and 2.7 (0.51–5.5) mg/L, respectively, in western Japan and significantly higher at 68 (28–180) mg/L and 27 (21–71) mg/L, respectively, in the PRD.
2. Concentrations of DDTs [2,000 (240–7,500) pg/g-wet] and chlordanes [1,000 (400–15,000) pg/g-wet] were highest in *Corbicula* from western Japan; meanwhile, DDT concentrations [7,700 (2,500–31,000) pg/g-wet] were highest in *Corbicula* from the PRD. There were large differences in concentrations of dieldrin, mirex, and DDTs in *Corbicula* between western Japan and the PRD, but no remarkable differences were observed in concentrations of the other chemicals.
3. The isomer compositions in *Corbicula* could well reflect those in water and sediment, regardless of the metabolism in *Corbicula*. In addition, isomer compositions of DDTs, chlordanes, and HCHs were significantly different between the two areas because of their different historical uses and property of the chemicals. This is represented as follows: the (DDE + DDD)/DDT ratio in *Corbicula*, ranging 16 (4.9–39) in western Japan and 1.7 (1.1–2.4) in the PRD, the *cis*-chlordanes/*trans*-chlordanes ratios in *Corbicula*, ranging 1.4 (1.1–2.1) in western Japan and 1.2 (1.1–1.3) in the PRD, and high proportion of  $\beta$ -HCH in Japan and high proportion of  $\alpha$ -HCH in the PRD. Therefore, the extent of pollution by chemicals in water and sediment can be measured using *Corbicula* based on isomer compositions.
4. *Corbicula* is a sediment dwelling bivalve and obtains food through filter feeding and pedal feeding within the streambed; however, the close relationship between logBAFL and log *Kow* suggest that OCP concentrations in *Corbicula* reflect those in water.

5. It can be suggested that the difference in species and the range of environmental conditions did not seem to make digit influence on the bioaccumulative levels in *Corbicula* when conducting biomonitoring.
6. The bioaccumulation levels *Corbicula* and *M. galloprovincialis* are similar despite pedal feeding of *Corbicula*. This result also means that under the same chemical conditions, *Corbicula* would accumulate the same levels of chemicals as would *M. galloprovincialis*.

Based on the results, monitoring OCPs in fresh and brackish waters using *Corbicula* is possible regardless of the usage history when concentrations and isomer compositions are accounted for. A biomonitoring system using *Corbicula* is applicable regardless of the range of concentrations of DO, SS, and VSS in water and the *Corbicula* species. When monitoring OCP concentrations in water, concentrations in *Corbicula* can be compared to those in *M. galloprovincialis*.

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