Ecotoxicology and Environmental Safety 88 (2013) 26–34

Contents lists available at SciVerse ScienceDirect

Ecotoxicology and Environmental Safety

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

Accumulation profile and seasonal variations of polychlorinated biphenyls (PCBs) in bivalves *Crassostrea tulipa* (oysters) and *Anadara senilis* (mussels) at three different aquatic habitats in two seasons in Ghana

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ARTICLE INFO

Article history:
Received 1 May 2012
Received in revised form
9 October 2012
Accepted 16 October 2012
Available online 14 November 2012

Keywords:
Polychlorinated biphenyls
Bivalves
Congeners
TEQ
Risk assessment

ABSTRACT

Research has shown that some polychlorinated biphenyl congeners degrade slowly in the environment and build up in the food chain, causing a wide range of possible adverse effects to humans. In order to ascertain the nature of the situation in Ghana, polychlorinated biphenyls congener residues in *Crassostrea tulipa* (oysters) and *Anadara senilis* (mussels) at Narkwa, Ada and Anyanui in the coastal region of Ghana were determined. At Narkwa, both bivalves' species were collected; at Ada only *Anadara senilis* were collected while at Anyanui, only *Crassotrea tulipa* were collected. The number of each bivalve species collected from each site was 80 ($n=80$), making up a total of 320 for the dry and the wet seasons. The PCBs were extracted with (1:1) hexane-acetone mixture and analyzed with a gas chromatogram equipped with ^{65}Ni electron capture detector, model CP 3800 using the mixed PCBs standard of the ICES 7. Total PCBs in the bivalves ranged from 5.55 to 6.37 μg/kg wet weight in mussels and 2.95–11.41 μg/kg wet weight in oysters, respectively. The composition of the PCB homologues in the bivalves was dominated by tri-, hepta- and hexa-PCBs in descending order. Risk assessments conducted on the samples indicated that edible bivalves from Narkwa, Ada and Anyanui in Ghana might pose some health risk to the consumers.

1. Introduction

Polychlorinated biphenyl (PCB) is a generic term for a family of 209 chlorinated isomers of biphenyl. The different combinations are called congeners, each having a specific number of chlorine atoms located at specific positions (GreenFacts, 2006; Weis and Monosson, 2011). PCBs usually range from oily liquids to waxy solids. Due to their non-flammability, chemical stability, high boiling point and electrical insulating properties, PCBs have been used in hundreds of industrial and commercial applications including electrical, heat transfer, and hydraulic equipment; as plasticizers in paints, plastics and rubber products; in pigments, dyes and carbonless copy paper and many other applications (Agency for Toxic Substances and Disease Registry (ATSDR), 2000; UNEP Chemicals, 2004; Rudel et al., 2008; Weis, Monosson, 2011). More than 1.5 billion pounds of PCBs are known to have been manufactured in the United States prior to cessation of production in 1977 (UNEP Chemicals, 1999).

Concerns over the toxicity and persistence of PCBs in the environment led to the United States Congress in 1976 enacting Section 6(e) of the Toxic Substances Control Act (TSCA) that included among other things, prohibitions on the manufacture, processing, and distribution in commerce of PCBs. Despite the ban on production of PCBs in Western countries, a large proportion of PCBs remain in storage as well as in transformers and capacitors in most developing countries Nakata et al., 2002; Otchere, 2005; Centeno, 2010).

The most obvious signs of environmental harm caused by PCBs are in the aquatic ecosystems (Environment Canada, 2008). Once PCBs are released into the aquatic environment, they can be adsorbed onto suspended particles or taken up and concentrated by aquatic organisms. It is known, that, PCBs could bioaccumulate and biomagnified to about 200–70 000 times along the food chain and pose potential hazards to other organisms and human consumers (Ashley et al., 2000; Fontenot et al., 2000; Pruell et al., 2000; UNEP Chemicals, 2004). PCBs are known to be persistent in the environment and in living tissues with a half-life of 3 weeks to 2 years in air. They have been found to be insidious and toxic to mammals and are carcinogenic to animals and probably humans (IARC, 1987; EPA, 1990; World Federation of Public Health Associations (WFPHA), 2000). Except for occupational contact, human exposure is mainly through food. PCBs are known to cause chloroacne, skin discoloration, liver dysfunction, reproductive effects, dermatitis, dizziness, development toxicity

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http://dx.doi.org/10.1016/j.ecoenv.2012.10.013

and oncogenicity in exposed humans (Guo et al., 1999; Agency for Toxic Substances and Disease Registry (ATSDR), 2000; World Federation of Public Health Associations (WFPHA), 2000). Some PCBs are known to have the ability to alter reproductive processes in mammals (Winneke et al., 1998a, 1998b).

Indeed, there is a concern, based on extrapolation from animal studies that PCBs may be carcinogenic in humans (IARC, 1987; Agency fotToxic Substances and Disease Registry (), 2000). Human foetal exposures to PCBs are associated with neural and developmental changes, lower psychomotor scores, short-term memory and spatial learning effects, and long-term effects on intellectual function. Neurological dysfunction had been associated with prenatal PCB exposure in several Dutch studies (World Federation of Public Health Associations (WFPHA), 2000; Weis and Monosson, 2011). Prenatal exposure in animals can result in various degrees of developmental toxic effects (World Federation of Public Health Associations (WFPHA), 2000). PCBs are particularly toxic to fishes and invertebrates and are fatal to these animals in even small concentrations (McGraw-Hill, 1987; World Federation of Public Health Associations (WFPHA), 2000; The New Encyclopedia Britannica, 2003).

Similar to dioxin, toxicity of coplanar and mono-ortho-PCBs are thought to be solely mediated through binding to aryl hydrocarbon receptor, AhR (Safe and Hutzinger, 1984; Safe et al.,1985). Since AhR is a transcription factor, abnormal activation could disrupt cell function by changing the transcription of genes. The concept of toxic equivalency factors (TEF) is based on the ability of a PCB to activate AhR. Nevertheless, not all effects may be mediated by the AhR receptor, and PCBs do not alter estrogen concentrations to the same extent as other ligands of the AhR receptor, such as polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF), perhaps, reflecting the reduced potency of PCBs to induce CYP1A1 and CYP1B1(Wang et al., 2006). Examples of other actions of PCBs include di-ortho-substituted non-coplanar PCBs which is said to interfere with intra cellular signal transduction dependent on calcium which may lead to neurotoxicity (Simon et al., 2007). Ortho-PCBs could disrupt thyroid hormone transport by binding to transthyretin (Chauhan et al., 2000).

Biota could potentially acquire PCBs from three sectors in the environment: atmosphere, water and food. Because of their lipophilicity, changes in PCB concentration might also be related to changes in lipid content (Boon and Duinker, 1985; Nakata et al., 2002). For example, in aquatic organisms, uptake is said to involve adsorption/absorption/partitioning of PCBs in water through gills and epidermis and consumption of contaminated food. PCB levels in marine invertebrates are best explained by equilibrium partitioning between body lipids and ambient water. So, PCBs in tissues of bivalves such as oysters and mussels should reflect the PCB concentration in its environment (Phillips, 1980, 1986).

Bivalves are widely used as bio-indicators of organic pollution in coastal areas because they are known to concentrate these compounds, providing a time integrated indication of environmental contamination. In comparison to fish and crustaceans, bivalves have a very low level of activity of enzyme systems capable of metabolizing persistent PCBs. Therefore, contaminant concentrations in the tissues of bivalves more accurately reflect the magnitude of environmental contamination as stated by Phillips (1980, 1986).

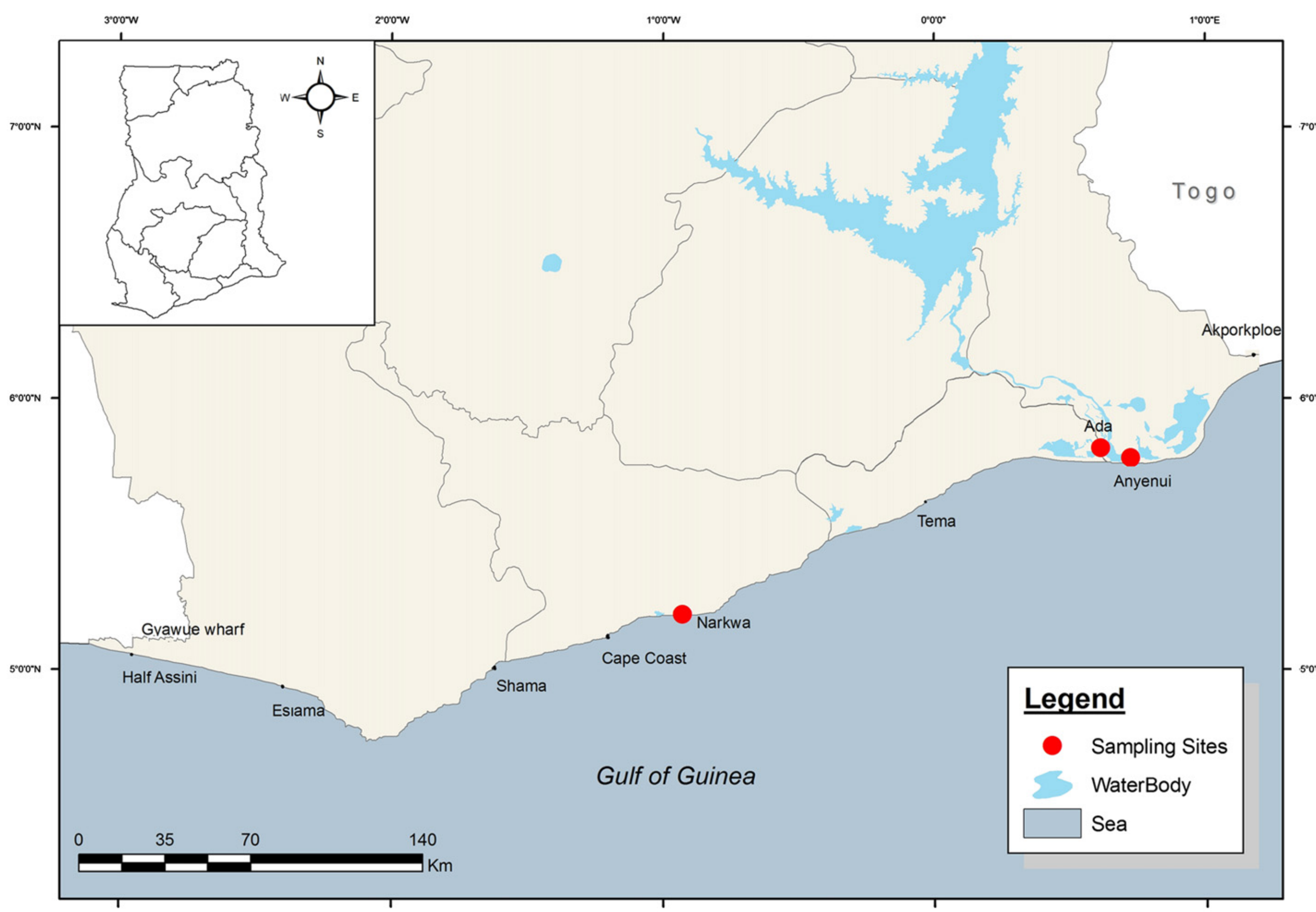

Fig. 1. A site map showing sampling sites.

Infiltration of PCBs into food chains poses a potential threat to top-level predators, including humans (Health Canada, 2005). Incidentally, this growing awareness does not seem to be the case in Ghana. Consequently, this study is intended to look at the levels and distribution pattern of PCBs in bivalves along the coastal region of Ghana where they serves as delicacies to the indigenes as well as their toxicity equivalent (WHO–TEQ) and hazard index (HI). The study will help provide additional information on the scope and environmental distribution of PCBs in Ghana.

2. Materials and methods

The species investigated in this study were bivalves *Crassostrea tulipa* (oysters) and *Anadara senilis* (mussels) collected from Narkwa, Ada and Anyanui along the coastal region of Ghana (Fig. 1). These locations were chosen due to accessibility of study materials (mussels and oysters) and also the fact that these bivalves serves as food for the indigenous people. The bivalves were collected into ice chest containers and labelled. The bivalves used in the study were of different sizes and were not purged.

The period of sample collection spanned over six months—from June, 2008 to February, 2009. Wet season samples were collected between June, 2008 and November,2008 while the dry season samples were collected between December 2008 and February 2009. In all, about 320 bivalves were collected; 160 pieces for each season. In the laboratory, the bivalves tissues were removed from their shells and blended after which sodium tetraoxosulphate (VI) was added to the homogenized sample.

Aliquots of wet tissues (10.0 g) were placed in flat bottom flasks. A 100 mL solvent mixture of 1:1 hexane–acetone was added to the samples in the flasks. The samples were subjected to cold maceration and wrist-shaker action for twenty four hours. The crude PCBs extracts were collected into an 80 mL clean glass vial and concentrated to about 2 mL using a rotary evaporator. The crude extract was transferred into a clean 12 mL vial for cleaning. Clean- up procedures followed that of US EPA Method 3630C. About 1 mL of concentrated tetraoxosulphate (VI) acid was added to the crude extract and the mixture vortexed for 30 s. This was done to remove any elemental sulphur and also to burn any organic carbon present. The hexane layer was transferred into another clean 12 mL vial after which the extract was washed with approximately 1 mL solution of saturated sodium tioxocarbonate (IV) in water. Afterwards, solvent-rinsed chromatographic columns (15 mm × 250 mm), packed with a plug of glass wool followed by 3 g deactivated silica gel and topped up with sodium tetraoxosulphate (VI) were prepared for the clean up. The columns were pre-rinsed with 15 mL hexane after which 2 mL of the analyte was added to the column and eluted with 60 mL hexane. The extracts were then concentrated to approximately 2 mL using a rotary evaporator and kept in a sample vial for gas chromatographic analysis. Cleaned-up extracts were analyzed using a gas chromatograph coupled with Electron Capture Detector (GC/ECD). The typical approximate reporting limit for individual PCBs is 1 μg/kg (ppm). PCB recovery standards at known concentrations were analyzed, after which the samples were also analyzed.

Target analytes in samples were tentatively identified and semi-quantitations made. Identifications were made by comparison of retention times, peak shapes and peak patterns of the samples with those of the recovery standards. Quantitations were based on sample peak areas or peak heights relative to standard peak areas or peak heights. In order to estimate the efficiency and precision of the extraction and analytical methods, certified reference material 1941b for PCBs in the marine environment from NIST, USA, were extracted and analyzed by the GC–ECD.

GC operating conditions for the determination PCB of congeners:
Detector:—ECD (Electron Capture Detector)
Injection point temperature: −270 °C
Column temperature: −70 °C (hold 2 min.) to 180 °C (hold 1 min) at a rate of 25 °C/min to 300 °C at a rate of 5 °C/min
Detector temperature: −300 °C
Carrier gas:—Nitrogen
Carrier gas flow rate: −1.0 mL/min
Make-up gas flow rate: −29.0 mL/min
Nature of column:—VF-5 m (40 m × 0.25 mm i.d × 0.25 μm film thickness

Once GC operating conditions were established, the same operating conditions were used for the analysis of standards, the QC (Quality Control) check samples, laboratory reagent blank (LRB) and samples.

3. Quality control

Quality control was undertaken to validate the method of extraction and analytical efficiencies. Certified reference material 1941b for PCBs in the marine environment from NIST, USA, was extracted and analyzed by the GC–ECD. In addition, duplicate samples were spiked with 0.2 ppm mixed PCBs standard to determine recovery and precision of extraction and analytical method employed.

4. Calculation of recovery and precision estimation

The percentage recovery of spiked sample was calculated according to the following formula:

$$\text{Recovery } (\%R) = \frac{Cs - Cu}{Cn}$$

where C_s=Measured concentration (peak area) of spiked sample aliquot, C_u=Measured concentration (peak area) of unspiked sample aliquot, C_n=Norninal concentration (peak area) of standard or spike.

The precision was estimated from the relative percent difference (RPD) of the concentration (peak areas) measured for spike duplicate pairs. The RPD was calculated according to the following formula

$$\text{RPD} = \frac{[C_1 - C_2] \times 100}{1/2[C_1 + C_2]}$$

where C_1=Measured concentration (peak area) of the first sample aliquot, C_2=Measured concentration (peak area) of the second sample aliquot.

5. Calculation of hazard index (HI) and toxic equivalents (TEQs)

The hazard index approach described in Environment Agency (2009b, 2009c, 2009d) was adopted for the calculation of the HI for the bivalves.

For the total PCB congeners analyzed, the average daily exposure of bivalves to PCBs, (ADE) was calculated using the relation:

$$\text{ADE} = \text{C} \times \text{EF} \times \text{TEF}$$

where

ADE: Average daily exposure of bivalves to PCB congeners in pg WHO–TEQ kg^{-1}bw day^{-1}
C: Concentration of total PCBs analyzed in ng kg^{-1} WW
TEF: Toxic equivalency factor for dioxin-like PCB 118=0.0001 pgWHO–TEQ pg^{-1}
EF: Exposure factor=5.57E-04 pg/kg BW/day/ng kg^{-1} WW
The Hazard Index, (HI), was calculated using the relation:

$$\text{HI} = \text{ADE}/\text{TDSI},$$

where

HI: Hazard Index of PCB congeners
ADE: Average daily exposure of bivalves tissues to PCBs in pg WHO–TEQ kg^{-1} BW day^{-1}
TDSI: Tolerable Daily Intake of PCBs=1.3 pg WHO–TEQ kg^{-1}bw $day^{-1}$for commercial scenario.
Toxic Equivalents (TEQs) are calculated values used to compare the toxicity of dioxin and dioxin-like compounds. The TEQs of PCB 118 in the study were calculated using the relation:

$$\text{TEQ} = \text{C} \times \text{TEF}$$

where,
C=concentration of PCB 118, and TEF having the same meaning as above.

6. Results and discussion

The percentage recoveries of the PCB congeners are shown in Tables 1 and 2. The NIST 1941b reference material gave recoveries of between 56.2 percent to 80.7 percent (Table 1). The values for the percentage recovery for this study were acceptable within the limits of experimental errors. In addition, duplicate sample pairs were spiked with 0.2 ppm PCB mixed standard and analyzed. The recovery of PCB congeners from this study ranged from 82 percent to 90 percent (Table 2). Results of the precision studies are also shown in Table 3 which were also within the framework of the standard set out by the international community.

In order to identify PCB residues pattern in the samples, a comparison was made based on 7PCB congeners (or ICES 7). Table 4 presents the results of the levels of PCB congeners in mussels and oysters at Narkwa, Anyanui and Ada during the dry and wet seasons; Table 5 presents mean PCB values in the bivalves during the dry and wet seasons while Table 6 lists results from other studies. Fig. 2 shows the distribution pattern of PCB congeners in these bivalves during the dry and wet seasons while Figs. 3 and 4 present the distribution pattern of the PCB congeners during the dry and wet seasons, respectively.

7. Distribution pattern of PCBs in bivalves from Narkwa, Ada and Anyanui

The results shown in Table 4 indicate that the mean total PCB congener concentrations in oysters and mussels at Narkwa during the dry and wet seasons, respectively, were 12.94 μg/kg, 9.88 μg/kg and 6.69 μg/kg, 4.41 μg/kg while those from Anyanui and Ada also recorded 3.3 μg/kg, 2.59 μg/kg and 5.77 μg/kg, 6.96 μg/kg, respectively within the same period. From the figures, the oyster at Narkwa appeared to be more contaminated with PCBs than those of the same species from Anyanui (i.e., 12.94 μg/kg at Narkwa as against 3.3 μg/kg at Anyanui). There was also the presence of all the PCB congeners with relatively higher concentrations during the period of investigation at Narkwa. This might be explained by the relative positions they occupy within the habitats. Those at Narkwa were stacked to the bottom sediments whereas those at Anyanui were permanently attached to mangroves growing by the sides of the lagoon, hence were not in direct contact with sediments. Available research information indicates that PCB concentration in sediments is higher than the surrounding water and this might have accounted for the above observation. This observation is in Thank you very much. It has been checked and accepted the changes made line with other research findings (Mckenzie et al., 1996).

Again, the mussels at Narkwa generally showed a higher rate of accumulation than those at Ada, though, the values were quite close (6.69 μg/kg as against 5.77 μg/kg). Considering the data for the PCB congener concentrations during the wet season, the values for oysters and mussels at Narkwa showed a general decreasing trend (though still higher) while the mussels at Ada showed an increasing trend.

Biota could potentially acquire PCBs from three sectors in the environment: atmosphere, water and food. Because of their lipophilicity, changes in PCB concentration might also be related to changes in lipid content of the bivalves (Boon and Duinker, 1986; Nakata et al., 2002). Differences in PCBs accumulation pattern between

Table 1
Percentage method recoveries of PCB congeners using NIST 1941b reference material.

PCB congener	Actual amount	Observed amount	Percentage recovery
28	4.52	3.68	81.4
52	5.24	4.05	77.3
101	5.11	3.92	76.7
118	4.23	3.20	75.7
138	3.60	2.34	65
153	5.47	3.54	64.7
180	3.24	1.82	56.2

Table 2
Recovery of 0.2 ppm PCBs mixed standards from spiked duplicate bivalves' samples.

PCB congener	Area count		$R=C_s-C_n$	$R\%=100R/C_n$
	C_s	C_n		
28	219413.8	114278	105135.8	92
52	211901.3	111527	100374.3	90
101	278858.5	148329	130529.5	88
118	368879.9	199394	169484.9	85
138	355429	190069	165360	87
153	373954	203236	170718	84
180	417299.4	231833	185466.4	80

Table 3
Precision of PCB analysis expressed as relative percent different (RPD%).

PCB	Area-count		$A=C_1-C_2$	$B=1/2(C_1+C_2)$	RPD% $=A/B$
Congener	C_1	C_2			
28	124563	114278	10285	119420	9.00
52	121564	111527	10037	116545	9.00
101	161678	148329	13349	155005	9.00
118	217339	199394	17945	208366	9.00
138	221527	203236	18291	212381	9.00
153	207175	190069	17106	198622	9.00
180	252698	231833	20865	242265	9.00
Total	3176544	1196666	107878	1232604	9.00

Table 4
Mean concentrations (μg/kg) of PCBs in mussels and oysters during the dry and wet seasons at Narkwa, Anyanui and Ada.

	Location							
	Narkwa				Anyanui		Ada	
Sample type	Oysters		Mussels		Oysters		Mussels	
PCB	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
28	3.46	2.26	2.19	2.20	0.86	0.80	1.41	0.76
52	0.38	0.46	1.60	0.94	0.24	0.24	1.10	1.96
101	1.30	1.18	0.62	0.62	ND	ND	0.52	0.62
118	1.30	0.94	ND	ND	0.5	0.35	0.24	0.18
138	1.32	0.72	0.22	ND	0.76	0.38	0.28	0.50
153	1.56	1.22	ND	ND	0.26	0.26	1.88	0.22
180	3.62	3.10	2.06	0.65	0.68	0.56	0.34	2.72
ΣPCB	12.94	9.88	6.69	4.41	3.30	2.59	5.77	6.96
Mean	11.40		5.55		2.95		6.37	

Table 5
Mean PCB congener profile in bivalves during the dry and wet seasons.

Town	**PCBs**	**28**	**52**	**101**	**118**	**138**	**153**	**180**
Narkwa (Oysters)		2.86	0.42	1.24	1.12	1.02	1.39	3.36
Narkwa (Mussels)		2.2	1.27	0.62		0.22		1.78
Ada (Mussels)		1.09	1.53	0.57	0.21	0.39	1.05	1.53
Anyanui (Oysters)		0.83	0.24	ND	0.43	0.57	0.26	0.62

Table 6
Concentrations of PCBs in bivalves collected worldwide.

Location	Bivalve	Range ng/g ww	References
Red sea	*M. brachiodontes*	7–66	Khaled et al. (2004)
Izmit bay (Turkey)	*M. galloprovincialis*	3–21	Telli-Karakoc et al. (2002)
Izmit bay (Turkey)	*M. galloprovincialis*	5–14	Tolun et al. (2001)
		1–36	" 1999
NW Med. Coast	*M. galloprovincialis*	10-700	Villenevue et al.(1999)
Ariake sea—Japan	*Mytilus edulis*	Av. 590	Nakata et al. (2002)
Korea	*M. edulis*	6–100	Khim et al. (2000)
Perth (Australia)	*M. edulis*	< 10	Burt and Ebell (1995)
U. S. A.	*M. edulis*	10-3,800	Sericano et al. (1996)
Denmark	*M. edulis*	3–328	Granby and Spliid (1995)
Hong Kong (China)	*Perna viridis*	1–152	Liu and Kueh (2005)
Cambodia	*Perna viridis*	0.5–5.1	Monirith et al. (2003)
Mainland (China)	*P. viridis*	0.3–13	"
India	*P. viridis*	0.2–11	"
Indonesia	*P. viridis*	0.1–2.7	"
Japan	*P. viridis*	7.4–84	"
South Korea	*P. viridis*	0.8–7.2	"
Malaysia		0.05–5.1	"
Philipines	*P. viridis*	0.4–14	"
Singapore	*P. viridis*	2.4	"
Vietnam	*P. viridis*	1.4	"
Thailand	*P. viridis*	0.01–20	Tanabe et al. (2000)
Nigeria		122	Azokwu, 1999
Ghana	*Anadara senilis*	101	Otchere 2005
Ghana	***Anadara senilis***	**5.55–6.37**	**This study**
"	***Crassotrea tulipa***	**2.95–11.41**	"

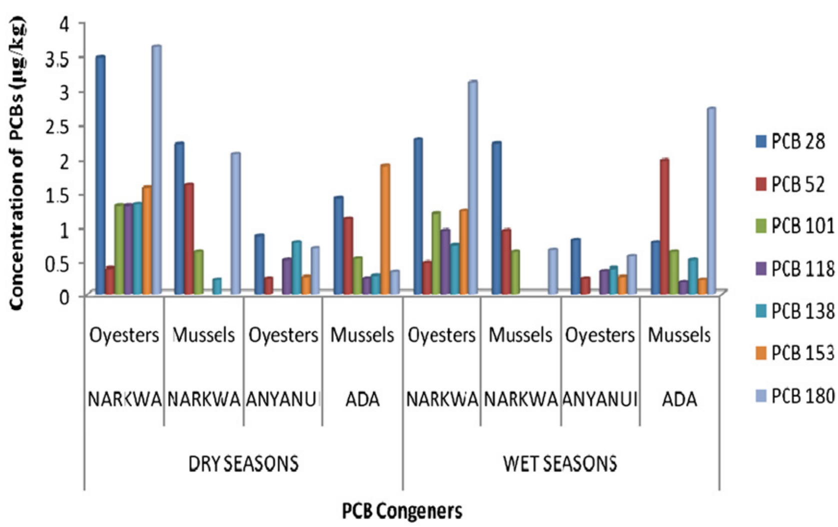

Fig. 2. Mean distribution of PCB congeners (µ/kg) in oysters and mussels at Narkwa, Anyanui and Ada during the dry and wet season.

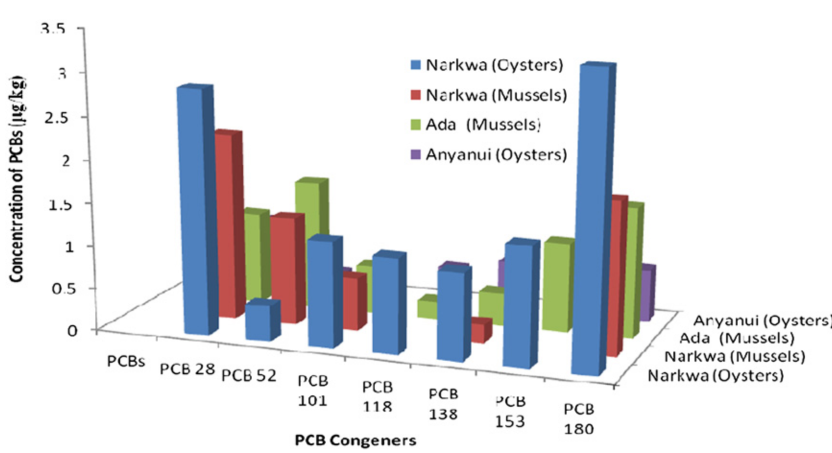

Fig. 3. Presents the mean PCB congener profile in oysters and mussels at Narkwa, Ada and Anyanui during the dry and wet seasons.

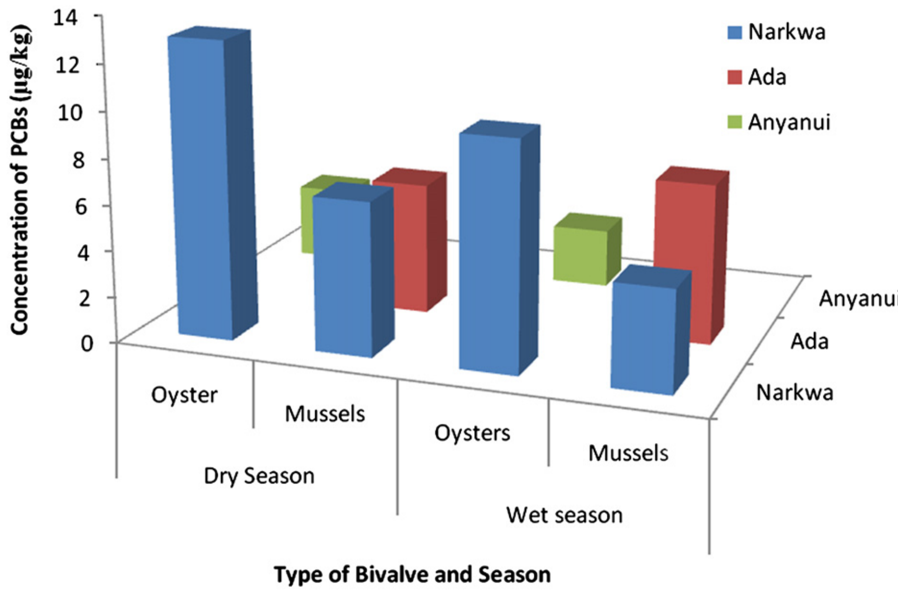

Fig. 4. Comparisons of total PCB congeners in bivalves at Narkwa, Ada and Anyanui during the dry and wet seasons.

organisms of the same species and between those of different species can occur as a result of the capacity to metabolize the congeners and the differences in diet, body condition, age, sex and possibly location (Boon et al., 1997; Tanabe et al., 1987; Storr-Hansen and Spliid, 1993; Storr-Hansen et al., 1995; Mckenzie et al., 1996). These situations may be responsible for the observed trends.

With exception of the mussels at Ada which showed a higher accumulation rate during the wet season, the mussels and oysters at Narkwa as well as the oysters at Anyanui showed a higher accumulation during the dry season. In contrast with the oysters at Narkwa which showed a higher accumulation of PCBs during both the dry and wet seasons, those species at Anyanui showed the lowest accumulation during both seasons. It is also significant to know that the oysters at Narkwa accumulated more of PCB 180 than all the other congeners during both seasons compared with their counterparts at Anyanui (i.e., 3.62 µg/kg or 27.97 percent and 3.10 µg/kg or 31.38 percent as against 0.68 µg/kg or 5.26 percent and 5.67, respectively). This might be due to the inability of these bivalves to metabolize the higher PCB congeners efficiently. Differences in the depuration rates and uptake of the different congeners in diet as well as biotransformation could explain these distributions (Porte and Albaigés, 1993). Furthermore, significant regional variations were observed between members of the same species e.g., oysters from Narkwa and those from Anyanui showed significant variation in PCB concentrations.

The pattern of accumulation of PCB congeners in oysters from the Narkwa estuary in the dry season followed the decreasing order: PCB 180 > PCB 28 > PCB 153 > PCB 118 > PCB 138 > PCB 101 and 118 > PCB 52. There was, however, slightly different distribution pattern of the congeners during the wet season, thus: PCB 180 > PCB 28 > PCB 153 > PCB 101 > PCB 118 > PCB 138 > PCB 52. It is clear from the results for both seasons that the oysters at Narkwa were highly enriched with PCB congeners 180, 28 and 153 than all the other congeners. This observation was, however, not in total agreement with some research findings (Sericano et al., 1996), that bivalves in general, preferentially accumulate congeners with 4, 5 and 6 chlorine substituents (i.e., PCBs 52, 101, 118, 138 and 153, respectively). It was observed that no residues of PCB 101 were detected in the oyster species from Anyanui in both seasons.

The mussels from Narkwa and Ada showed different patterns of accumulation of PCB congeners during both seasons. Whereas those at Ada accumulated all the congeners during both seasons, the species at Narkwa did not accumulate any residues of PCBs 118 and 138 during the dry season and none of 101, 118 and 153 during the wet season.

8. PCB congener profile in bivalves

The oysters and mussels in all the study areas were highly enriched with higher chlorinated isomers (i.e., PCB 180) as well as the less chlorinated isomers (PCB 28) with an intermediate distribution of the other isomers, giving almost a normal distribution curve, (Figs. 2–4). The composition of the PCB homologues were dominated by tri-PCBs (26.53 percent) > hepta-PCBs (26.13 percent) > hexa-PCBs (18.24 percent) > penta-PCBs (15.93 percent) > tetra-PCBs (13.17 percent), respectively. This observation indicates a higher level of contamination of the bivalves. The greater concentrations of higher PCB congeners detected in the bivalves might be due to the inability of the bivalves to metabolize these congeners efficiently or perhaps, their high lipid contents which facilitate the accumulation of the higher chlorinated isomers (US EPA, 1992; US EPA, 1995).

Phillips (1980) reported that, PCB concentrations in biological tissues correlate positively with the extractable lipids. Muncaster et al. (1990) found that PCB accumulation in the freshwater mussels *Lampsilisradiata* varied inversely with the body size, which was attributed to alteration in assimilation rates. Azokwu (1999) working on *Anadara senilis* did not find any relationship between lipid content and PCB concentration. All these features may imply that there were different mechanisms of accumulation or different metabolic capacities in aquatic biota (Boon et al., 1997; Joiris et al., 1997; Otchere, 2005).

The oysters at Narkwa had the highest amounts of all the congeners except PCB 52 (Fig. 2). No residues of PCB 118 and 153 were detected in mussels collected from Narkwa while none of PCB 101 was detected in oysters from Anyanui. Though maxima was generally observed for the dry season results (except mussels from Ada), no significant seasonal variation was observed among the bivalves except the oysters from Narkwa which showed some significant seasonal variation between the dry and wet seasons (Table 5).

The results of analysis showed that the bivalves were more enriched with all the PCB congeners, especially, the tri-PCBs, hepta-PCBs and appreciable concentrations of the penta-, hexa- and tetra-PCBs homologues, respectively.

9. Comparison of results with data from other regions of the world

Published data for PCB residues in bivalves from various regions of the world such as the U. S. A., Korea, Japan, Cambodia, Australia and many others are shown in Table 6. The mean total PCBs in the bivalves studied ranged from 5.55 to 6.37 μg/kg wet weight (in mussels) to 2.95–11.41 μg/kg wet weight (in oysters). It is clear from Table 6 that the levels of PCBs determined in the bivalves were not too high. The levels were, however, higher than those found in Vietnam, Indonesia, Singapore and Cambodia but far less than those found elsewhere such as the U. S. A., China, Japan, Denmark and many others.

The variability in the regions is large, often several orders of magnitude. This fact alone severely limits making intra- and inter-regional comparisons. Furthermore, the uses of different species, size difference, seasonal effect, different methods of quantification, etc. all interact to confound comparisons. In general, the highest values come from areas where there are known industrial input, and the lowest concentrations are found in organisms inhabiting remote areas. This is so because PCBs are industrial-based chemicals (Wong et al., 2000). In addition, PCB technical mixtures with specific congeners once in the environment undergo changes with a slow increase of the less chlorinated congeners, corresponding to de-chlorination of the higher ones. As a result, environmental PCB patterns can no longer be compared in some respect with the original pattern (Otchere, 2005).

10. Hazard index (HI) of bivalves

Hazard Index is a risk management strategy used to assess the degree of risks pose to humans by different levels of PCB exposures. An HI value of 1 or more represents a significant risk to human health while those less than 1 represent no significant risk (Environment Agency, 2009b). The Hazard Index approach described in Environment Agency (2009b, 2009c, 2009d) was adopted for the calculation of the HI. TEQs are calculated values that allow us to compare the toxicity of different combinations of dioxins and dioxin-like compounds. The TEQ values in the results were based only on PCB 118 since it is the only dioxin-like PCB among the congeners quantified. TEQs help people to understand the relative toxicity of the chemical release information. ADE refers to average daily exposure to PCBs.

The calculated total HI values for bivalves in Table 7 above were 0.14319, 0.03788, 0.06853 and 0.08197, respectively for Oysters at Narkwa and Anyanui and mussels at Narkwa and Ada. Since the HI values were all far less than 1 (i.e., HI $\ll$ 1), they may not pose any significant health risks to consumers (Environment Agency, 2009b, 2009c, 2009d.

Table 7
Hazard index (HI) of PCB in bivalves.

Region/Town	Sample	TEQ (Pg/g)	ADE Commercial	Hazard index (HI)	
	Oysters				
Narkwa	F1	0.1023	0.056981	0.043832	
	F2	0.0822	0.045785	0.03522	
	F3	0.0882	0.049127	0.03779	
	F4	0.0615	0.034256	0.02635	**0.14319**
Anyanui	AN1	0.0267	0.014872	0.01144	
	AN2	0.0308	0.017156	0.013197	
	AN3	0.0075	0.004178	0.003213	
	AN4	0.0234	0.013034	0.010026	**0.03788**
	Mussels				
Narkwa	K1	0.0324	0.018047	0.013882	
	K2	0.0587	0.032696	0.025151	
	K3	0.0359	0.019996	0.015382	
	K4	0.033	0.018381	0.014139	**0.06853**
	A1	0.0312	0.017378	0.013368	
Ada	A2	0.0503	0.028017	0.021552	
	A3	0.0654	0.036428	0.028021	
	A4	0.0444	0.024731	0.019024	**0.08197**

Table 8
Average PCB 118 TEQ Values of Bivalve Samples.

PCB 118 TEQ (μg/kg)			
Sample and location	Dry season	Wet season	Average
Oysters (Narkwa)	0.00013	0.000094	0.000112
Mussels (Narkwa)			
Oysters (Anyanui)	0.00005	0.000035	0.000043
Mussels (Ada)	0.000024	0.000018	0.000021

11. Toxicity equivalent (TEQ) concentrations (PCB 118)

Toxic equivalency factor (TEF) indicates a value to estimate the toxicity of a compound relative to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). TEF values, in combination with chemical residue data can be used to calculate toxic equivalent concentrations in various environmental samples including sediments and mussels (Ahlborg et al., 1994). In this study, the PCB 118 TEQ concentrations were calculated using the equation and TEF values of World Health Organization as given in Van den Berg et al. (2005). The concentrations of PCB 118 in the results were multiplied by the TEF of PCB 118 (i.e., 0.0001) to obtain the PCB 118 TEQs in the Table 8 below.

The relevance of the PCB 118 TEQ values in this study is that they enable us to compare the relative toxicity of PCB 118 in the bivalves and to ascertain whether it poses any significant health hazard to consumers.

12. Risk assessment of PCBs in bivalves and health implications us

Recently, a number of authorities have reassessed risks of PCBs and focused on developmental risks. Tolerable daily intake (TDI) values were described for risk assessments. The recommendation for TDI of WHO–TEQ of the UK Committee on Toxicity of Chemicals in Food, Consumer Products and Environment (COT) is 2 pg WHO–TEQ/kg bw (COT, 2001). Calculations using bivalves in this study, PCB 118 TEQ contents of 21.0–112.0 pg/kg showed that consumable bivalves from Narkwa, Ada and Anyanui in Ghana might pose some significant risks to health due to higher intake of the bivalves with high PCB 118 TEQ than the recommended TDI value. The TEQ results were, however, in sharp contrast with the HI values which indicated no significant health risk to consumers. However, due to limited data on levels of contamination in the study area, no definite conclusion can be drawn on the possible health hazards it may pose to consumers.

In general, PCB with higher chlorination in fatty tissues may increase the health risks because, the more highly chlorinated PCBs are retained longer in fatty tissues (US EPA, 1995), whereas PCBs with three or fewer chlorines are more readily biologically transformed (US EPA, 1992). The non-*ortho*substituted, co-planar PCB congeners and some mono-*ortho*-substituted congeners have been shown to have "dioxin-like" effects (US EPA, 1995). US EPA, 1995 has classified PCB mixtures as B2 carcinogens (probable human carcinogens). Besides potential carcinogenicity, the major toxic effect of PCBs in mammals is liver damage (US EPA, 1995).

13. Conclusions

The mean levels of PCBs detected in the bivalves ranged from 2.95 to 11.41 μg/kg (for Oysters) and 5.55–6.37 μg/kg (for mussels) for both the dry and wet seasons for all the locations. These levels compared favourably with those obtained from other parts of the world such as The Red Sea, 7–66 ng/g (Khaled et al., 2004); Izmit bay (Turket), 3.0–21 ng/g (Telli-Karakoc et al., 2002); 10.0–700.0 ng/g in the North-West Mediterranean Coast (Villenevue et al., 1999); the U. S. A., 10.- 3800.0 (Sericano et al., 1996) and many others as shown in Table 6 above. The Narkwa bivalves appeared to have accumulated more PCB congeners for both seasons than their counterparts at Anyanui and Ada i.e., 12.94 μg/kg and 9.88 μg/kg (for oysters); 6.69 μg/kg and 4.41 (for mussels) as against 3.3 μg/kg and 2.59(for oysters) and 5.77 μg/kg and 6.96 μg/kg (for mussels), respectively. The composition of the PCB homologues in the bivalves were dominated by tri-PCBs (26.53 percent) > hepta-PCBs (26.13 percent) > hexa-PCBs (18.24 percent) > penta-PCBs (15.93 percent) > tetra-PCBs (13.17 percent), respectively. There were no significant differences in the PCB congener concentrations in the bivalves between the dry and the wet season.

Moreover, a risk assessment was conducted on the results using HI and PCB 118 TEQ (WHO–TEQ) risk management strategy. The HI risk assessment gave 0.14319, 0.03788, 0.06853 and 0.08197, respectively for Narkwa and Anyanui Oysters and Narkwa and Ada mussels. Since the HI values were all far less than 1 (i.e., HI $\ll$ 1), they may not pose any significant health risks to consumers. The PCB 118 TEQ assessment, however, gave values far greater than the recommended 2 pg WHO–TEQ/kg bw (COT, 2001) by the U.K. Committee on Toxicity of Chemicals in food, Consumer Products and Environment. The PCB 118 TEQ contents of 21.0–112 pg/kg showed that consumable bivalves from Narkwa, Ada and Anyanui in Ghana might pose some significant risks to health due to higher intake of the bivalves with high PCB 118 TEQ than the recommended TDI value.

The study is significant because it will help provide additional information on the scope and environmental distribution of PCBs in Ghana. It will also contribute to the global inventory and pave way for adequate risk assessment where necessary. This will go a long way to augment the efforts of Ghana's EPA towards the elimination of PCBs in line with the Stockholm Convention.

Acknowledgments

We wish to express our gratitude to Mr. J. K. Bentum of the Chemistry Department, University of Cape Coast, for paying for the full cost of analysis and Mr. Paul Osei-Fosu of Ghana Standard Boards for running the samples. We also want to thank Auntie Efua of Narkwa and Togbui Kafui of Anyanui for assisting in the collection of the samples. Our final thanks go to Mr. Afful, a technician at the Chemistry Department of Ghana Atomic Energy Commission (GAEC) for the vital role he played towards analysis of the samples.

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