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Bioaccumulation of Polychlorinated Biphenyls (PCBs) and Organochlorine Pesticides in Manila Clams (*Ruditapes philippinarum*) Collected from the Mid-western Coast of Korea

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Abstract : Bioaccumulation of polychlorinated biphenyls (PCBs) and organochlorine pesticides was studied in sediment dwelling bivalves, Manila clams (*Ruditapes philippinarum*), collected from the mid-western coast of Korea. As witnessed by the dominance of tetra- to penta-chlorinated congeners in sediments and the penta- to hexa-chlorinated congener dominance in clams, the profile of PCBs in the sediments and Manila clams differed. Lipid and organic carbon-normalized biota-sediment accumulation factors (BSAFs) were determined for organochlorine pesticides. BSAFs of β -hexachlorocyclohexane (β -HCH) and Σ DDTs were in the range of 0.06~1.36 and 0.31~1.06. No clear relationships were found between BSAFs of Σ DDTs in Manila clams and the concentrations of DDTs in the associated sediment. The accumulated PCBs and organochlorine pesticides were compared in Manila clams and oysters (*Crassostrea gigas*) collected from 3 sites. Highly chlorinated PCBs were more commonly found in oyster tissues than in clam tissues. The reasons for the different accumulation pattern of organic pollutants in the two organisms are discussed.

Key words : PCBs, Yellow Sea, Manila clam, DDT, HCH

1. Introduction

Polychlorinated biphenyls (PCBs) and organochlorine pesticides are hydrophobic and lipophilic substances, which persist in the environment for very long periods (Tolosa *et al.* 1996). PCBs are synthetic chemicals that were widely used until their production and applications were restricted or banned in developed countries during the late 1970s. Due to the extreme stability of many of their isomers, particularly those with 5 or more chlorines, PCBs have been used in various manufacturing processes and products (Erickson 1992). Organochlorine pesticides are well known as persistent organic pollutants (Jones and De Voogt 1999). The Stockholm Convention on POPs

(Persistent Organic Pollutants) acknowledged POPs, including polychlorinated biphenyls and organochlorine pesticides (i.e. DDT, aldrin, dieldrin, HCB, heptachlor, toxaphene), as a global problem in 2001. Since their ban, the release of PCBs and organochlorine pesticides to the environment has decreased. However, once released into the aquatic environment, these substances can either be immediately absorbed by organisms or adsorbed onto suspended particles. After particles settle on bottom sediments, benthic organisms may still accumulate these substances (Carvalho *et al.* 1992). Therefore, marine sediments will continue to serve as secondary sources of these contaminants, both to the inhabiting benthic communities and to other organisms through food web transfer. These contaminants also have the potential to enter the marine environment by leaching from stockpiles,

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landfills, waste deposits and so called 'closed systems'. Due to bioaccumulation through the food web and their highly toxic effects, PCBs and organochlorine pesticides represent a significant health problem. Furthermore, many of these chemicals are also carcinogenic substances and endocrine disruptors, which adversely affect hormone systems.

In the marine environment, marine organisms are used as bioindicators of chemical contaminants. Monitoring contaminant levels in marine organisms has many advantages over sediment monitoring. For example, bivalves may accumulate and record the contaminants present in the water column that may not be recorded in the sediment (Galceran and Santos 1993). Bivalves are widely used as environmental sentinels (Baumard *et al.* 1998). Bivalves, as sessile, filter-feeding and low metabolism organisms, may take up and concentrate contaminants to levels well above those present in the surrounding waters or sediment. Thus, bivalves are able to provide information on local pollution sources (Solé *et al.* 2000). Choosing a suitable species for monitoring depends on various criteria, with availability being the most restrictive limitation. Primarily, mussels and oysters, which are water-filtering bivalves, have been used for monitoring purposes (O'Connor 1996). As a part of the Mussel Watch Program in Korea, Kim *et al.* (2002) assessed the contamination levels and accumulation features of PCBs and OCPs along the entire coast of Korea. Ramu *et al.* (2007) analyzed mussels for PCBs and organochlorine pesticides from 20 coastal locations in Korean waters.

However, little attention has been paid to sediment dwelling bivalves that filter water and have sediment exposure (Dame 1996). Manila clams (*Ruditapes philippinarum*) are widely distributed along the coasts of Korea, China, Japan, Northwestern America, and several European countries (Nasci *et al.* 2000). Manila clams occur at high densities in intertidal areas where mussels and oysters are not largely distributed. This species is easily distinguishable from other species, which enhances its value for biomonitoring. Continuous exposure to contaminants from potentially polluted sediment and seawater directly affects clams because of their limited mobility. This species is thought to reflect not only sediment contamination, but also pollutants in the water column. Ji *et al.* (2006) evaluated the Manila clam as a sentinel species for metal pollution monitoring in estuarine tidal flats of Korea. The study found that most metal concentrations in Manila clams varied significantly

with body size, sex, and spawning status, and closely resembled variations in conventional sentinel organisms (i.e. mussels and oysters).

Thompson *et al.* (1999) studied selected PCB congeners, DDT and DDT degradation products in various bivalves including the Manila clam and sediment from Arcachon Bay, France. The study reported that water-column bivalves have higher concentrations of PCBs and DDTs than sediment dwelling bivalves like Manila clams. Binelli and Provini (2003) studied the organochlorine bioaccumulations in Manila clams bought from different Italian and European markets. They found, particularly in the Venice Lagoon, that concentrations of organochlorines in the soft tissues of mollusks varied considerably among source farming sites. Nasci *et al.* (2000) used the Manila clam to investigate water and sediment pollution in the Venice Lagoon (Italy), a heavily urbanized and industrialized area. In Manila clams cultured in the Venice Lagoon, Boscolo *et al.* (2007) reported that the highest level of Σ PCB detected was 4.01 ng g⁻¹ wet weight.

Although studies on contaminant bioaccumulation by mussels and oysters in Korea have been performed in the past, there have been no studies documenting sediment-based bioaccumulation of highly persistent trace organic contaminants in Manila clams. The main objective of this study was to acquire basic knowledge on the PCB and organochlorine pesticide accumulation of in the tissues of Manila clams. An attempt was made to use contaminant levels in this organism as an assessment tool for coastal area pollution monitoring programs. Thus, PCBs and organochlorine pesticides were analyzed in Manila clams, oysters, and sediments collected from the mid-western coastal area of Korea.

2. Materials and methods

Fig. 1 shows sampling sites from the mid-western coastal area of Korea. Over a 2-week period in May 2002, Manila clams and sediment samples were collected and analyzed for selected compounds of chlorinated pesticides and PCBs. Eight sampling sites were chosen for Manila clams: Matdolpo (MD), Daeho (DH), Samgilpo (SG), Anheung (AH), Chasok (CS), Kanwol (KA), Dogkot (DK) and Daechon (DE). Sites DH and SG are located near an important petrochemical complex and site MD is close to a big port facility. Even though there is no intense industrial activity near the other sites, continuous input of anthropogenic pollutants occurs from terrestrial sources due to patterns of intensive land use. The sampling sites

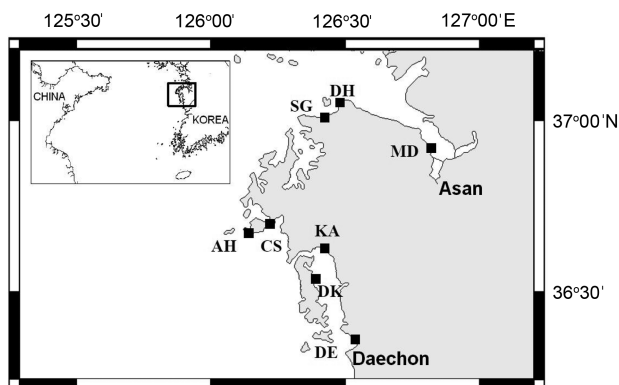


Fig. 1. Sampling sites on the mid-western coast of Korea.

were selected in order to provide a broad coverage of the intertidal zone on the mid-western coast of Korea.

Manila clams with shell lengths of 30~40 mm were hand-collected from the intertidal sediment. The mean size of clam samples was 34.8 ± 1.7 mm. In order to compare the accumulation pattern of organic contaminants in Manila clams with oysters, cultured oyster (*Crassostrea gigas*) samples with shell lengths of 60~70 mm were also collected at three sites (SG, DH, and DK) near the clam sampling sites. Local fishermen provided these cultured oyster samples. Intertidal sediment samples were collected with a stainless steel spoon. Sediment samples were collected in glass bottles and stored frozen until they were freeze-dried with Labconco Freezezone 6. Manila clam and oyster samples were stored frozen until analysis. The samples were analyzed for PCBs and organochlorine pesticides.

Sediments

Extraction and analyses of PCBs and organochlorine pesticides were conducted according to the method described by Lauenstein and Cantillo (1993). Ten grams of sediment samples were spiked with surrogate standards (PCB-103, PCB-198, DBOFB <4,4'-dibromooctafluorobiphenyl>). Samples were Soxhlet-extracted with 100 ml of hexane:acetone (1:1) for 18 hours. The extracts were concentrated to 2~3 ml in a rotary evaporator at 35~40°C. To remove sulfur, the extracts were then left to stand overnight with activated copper (20~30 mesh, J. T. Baker Co., U.S.A.). The extract was passed through a column containing 3 g of 5%-deactivated alumina (~150 mesh, Aldrich Chemical Co., U.S.A.) and was eluted with 60 ml hexane:acetone (1:1) for cleanup. The extract was concentrated to 5 ml, the solvent was then exchanged for hexane, and the extract was concentrated to 1 ml. To achieve further cleanup, the

extract was subjected to column chromatography using deactivated Florisil (60~100 mesh, J. T. Baker Co., U.S.A.).

Organisms

Whole shell tissue was used for the analysis. Soft tissues of 30 clams/sample were pooled and homogenized in a blender. Approximately 20 g of wet tissue was used for the extraction. After samples were dehydrated with 50 g of anhydrous Na_2SO_4 (anhydrous granular, Aldrich Chemical Co., U.S.A.), they were extracted with hexane:acetone (1:1).

The extract was passed through a column containing 3 g of 5%-deactivated alumina (~150 mesh, Aldrich Chemical Co., U.S.A.) and was eluted with 60 ml hexane:acetone (1:1) for cleanup of biological materials. To achieve further cleanup, the extracts were subjected to column chromatography using Florisil (60~100 mesh, J. T. Baker Co., U.S.A.) after cleanup on an alumina column.

Analyses

A high resolution gas chromatograph (split/splitless injection system) equipped with a ^{63}Ni electron capture detector (GC-ECD Hewlett Packard 5890) was used to identify and quantify PCBs and organochlorine pesticides. The GC column used for the analyses was a DB-5 (J&W Scientific Co. Ltd., U.S.A., 0.25 μm bonded phase). The column oven temperature was programmed at a rate of 5°C min^{-1} from an initial temperature of 100°C (1 min hold) to 140°C (1 min hold). Next, it was raised to 250°C at a rate of $1.5^\circ\text{C min}^{-1}$ (1 min hold). The temperature was then increased to a final temperature of 300°C at a rate of $10^\circ\text{C min}^{-1}$ (5 min hold). Helium and argon/methane (95:5) were used as the carrier and make-up gases (40 ml min^{-1}), respectively. The injector temperature was kept at 275°C , and the detector was maintained at 300°C .

Organic carbon in sediments and lipid contents in clams

Following the method of Hedges and Stern (1984), organic carbon contents were measured on dried sediment samples. Freeze-dried sediment samples were ground, and treatment of sample with 10% (v/v) HCl removed inorganic carbon. After the samples were dried at 50°C in an oven, the organic carbon was identified and quantified on a Carlo Erba CNS elemental analyzer 1106. Lipids in the organisms were determined by the method of Qian *et al.* (1998). An aliquot of 20 ml of the tissue extract was taken and brought to a final volume of 1 ml. An aliquot of 100 μl was evaporated to constant weight. The residual

weight of this dried portion was used to calculate the lipid percentage.

QA/QC

Procedural blanks were regularly performed and all results presented were corrected for blank levels. All glassware was rigorously cleaned with detergent and baked at 450°C. For PCBs and organochlorines, the percentage recovery varied from 75.4% to 116.2% for the sediments and 65.6% to 123.3% for the biological samples. The method detection limits of the procedure were 0.03–0.10 ng g⁻¹ dw (dry weight) for the sediment and 0.08–0.20 ng g⁻¹ dw for Manila clams. PCB concentrations were reported as the sum of 22 congener peaks (PCB-8, -18, -28, -29, -44, -52, -66, -87, -101, -105, -110, -118, -128, -138, -153, -170, -180, -187, -195, -200, -206, -209). To check for interference, a spiked sample consisting of all chemicals was run. For duplicate samples, the average reproducibility (1σ) of the method varied less than 10%. Quality assurance procedures were performed by co-analyzing reference materials of appropriate matrix, namely the sediment NIST 1941b and the mussel tissue homogenate IAEA-142.

3. Results and discussion

Organic contaminants in sediments

The concentrations of ΣPCBs ranged from 0.52 ng g⁻¹ dw (dry weight) at SG to 3.07 ng g⁻¹ dw at AH in the intertidal sediments collected from the mid-western coast of Korea. Fig. 2 shows the percentage of chlorine-based classes of PCBs. The most important congener of PCBs found in the intertidal sediments (22.53–48.33% of ΣPCBs) appeared to be tetra-CBs (PCB-44, -52, -66)

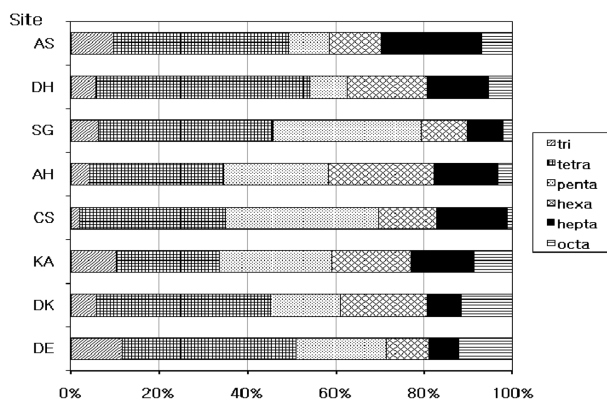


Fig. 2. The percentage of chlorine-based classes of PCBs in the intertidal sediments of the mid-western coast of Korea.

followed by penta-CBs (8.52–34.63%). The sediment samples were poor in heavier chlorinated PCB congeners. Total PCB concentrations in the sediments of Gyeonggi and Busan Bays, Korea were in the range of 0.55–16.0 ng g⁻¹ dw and 5.71 to 199 ng g⁻¹ dw, respectively, according to Hong *et al.* (2005). Concentrations of DDT, DDD and DDE in Masan Bay, Korea were reported to be in the range of not detected–39.6, 0.1–28.4 and 0.14–42.4 ng g⁻¹ dw, respectively (Hong *et al.* 2003). The results obtained in this study are much lower than those values measured in highly industrialized areas of Korea.

The concentrations of PCB compounds in the intertidal sediments of the study area were lower than 5 ng g⁻¹ dw, which was in agreement with the upper limit of a non-contaminated site suggested by Marchand *et al.* (1990). Moreover, the concentrations of these compounds were much less than the critical concentration (22.7 ng g⁻¹ dw) that can cause negative biological effects (Long *et al.* 1995). ΣHCHs (Hexachlorocyclohexanes) concentrations in the sediments were in the range of 0.10(KA)–0.75(CS) ng g⁻¹ dw. Although p,p'-DDE was present at low concentrations ranging from 0.10(DE) to 0.41(AH) ng g⁻¹ dw, it was widely found among the DDT metabolites in the intertidal sediments. ΣDDTs concentrations in the sediments of the study area ranged from 0.35(DE) to 1.18(AH) ng g⁻¹ dw and were lower than the critical concentration (1.58 ng g⁻¹ dw) that can cause negative biological effects (Long *et al.* 1995).

Organic contaminants in Manila clams

In Manila clams, ΣPCBs concentrations varied from 9.16 to 13.95 ng g⁻¹ dw. Tetra-CB concentration ranged from 1.06 ng g⁻¹ dw at site DE to 2.94 ng g⁻¹ dw at site DK. The highest concentration of penta-CBs was found at site AH (3.22 ng g⁻¹ dw), while site KA exhibited the lowest penta-CB concentration (2.28 ng g⁻¹ dw). The concentration of hexa-CBs was 2.10 ng g⁻¹ dw at site DH, while site DK showed the highest concentration (3.73 ng g⁻¹ dw). The profile of PCBs in the sediments and Manila clams differed because the sediments were dominated by tetra- to penta-chlorinated congeners while the Manila clams by penta- to hexa- chlorinated congeners. In Manila clams, tetra-CBs and penta-CBs (PCB-87, -101, -105, -110, -118) accounted for 11.58–25.75% and 18.79–33.79% of total PCBs, respectively (Fig. 3). Among total PCBs, the percentages of hexa-CBs (PCB-128, -138, -153) found in Manila clams were in the range of 20.57–34.61%.

Boscolo *et al.* (2007) found a clear predominance of hexa-CBs in Manila clams collected in the Venice Lagoon

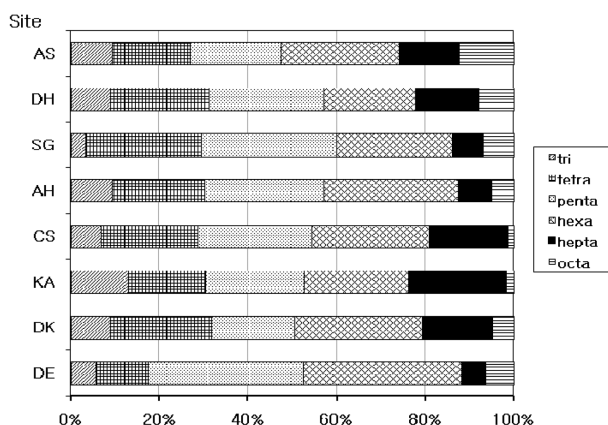


Fig. 3. The percentage of chlorine-based classes of PCBs in Manila clams collected along the mid-western coast of Korea.

(Italy), followed by penta-, hepta-, tetra- and tri-CBs. Binelli and Provini (2003) found that, in all the Manila clams collected in the different European aquatic ecosystems with a clear predominance of hexachlorinated congeners, the percentage of chlorine-based classes of PCBs is similar. Kim *et al.* (2002) reported that the dry weight based Σ PCBs and Σ OCPs in mussels collected from the entire coast of Korea ranged from 4.4–422.0 ng g⁻¹ and from 9.95–131.37 ng g⁻¹, respectively. High levels of PCBs, up to 1000 ng g⁻¹ lipid wt., were found in mussel samples from industrial areas near Ulsan and Masan Bays, Korea (Ramu *et al.* 2007). Monirith *et al.* (2003) reported that, in the Asia-Pacific region, higher levels of DDTs were found in mussels from China (830–54,000 ng g⁻¹ lipid wt.), Hong Kong (640–61,000 ng g⁻¹ lipid wt.), and Vietnam (220–34,000 ng g⁻¹ lipid wt.), while higher concentrations of PCBs were found in mussels from Japan (up to 12,000 ng g⁻¹ lipid wt.) and Russia (up to 3700 ng g⁻¹ lipid wt.). The values found in this study were much lower in Manila clams than in mussels.

Boscolo *et al.* (2007) reported Σ PCBs concentrations expressed on a dry weight basis in Manila clams cultured in the Venice Lagoon to be 15.27–28.64 ng g⁻¹ dw. These values were slightly higher than the values found in this study. Binelli and Provini (2003) reported a concentration of 18.8–93.8 ng g⁻¹ dw of Σ PCBs in all the clams collected from the different European aquatic ecosystems. These values were also higher than the values found in this study. During this study, Σ HCHs in Manila clams ranged from 0.62 ng g⁻¹ dw at site DE to 3.42 ng g⁻¹ dw at site DK. Σ DDTs in Manila clams ranged from 1.4 ng g⁻¹ dw at

site DE to 6.76 ng g⁻¹ dw at site KA. With percentages ranging from 23.5% to 55.1% of Σ DDTs, p,p'-DDE was the main component of DDT relatives. Σ DDTs values in this study were similar to those obtained by Nasci *et al.* (2000) in Manila clams collected in a less contaminated site of Venice (3.7–3.8 ng g⁻¹ dw).

Biota-sediment accumulation factor (BSAF)

The distribution of organochlorine compounds between the different phases in the aquatic environment is complex and depends on partitioning coefficients, which differ according to the properties of the organochlorine compound. Methods based on equilibrium partitioning have been used to study the bioaccumulation of PCBs and organochlorine pesticides. Introduced by Di Toro *et al.* (1991), lipid and organic carbon-normalized biota-sediment accumulation factor (BSAF) is a valuable tool for predicting bioaccumulation of lipophilic compounds that are primarily associated with tissue lipids and sediment organic carbon.

BSAF is defined as:

$$\text{BSAF} = \frac{C_B/F_{\text{lipid}}}{C_S/F_{\text{OC}}}$$

where C_B : contaminant concentration in biota (ng g⁻¹ wet weight)

C_S : contaminant concentration in sediments (ng g⁻¹ dry weight)

F_{lipid} : lipid concentration in biota (by weight)

F_{OC} : organic carbon fraction of sediments (by weight)

Factors governing the geographical variation in BSAF may include habitat difference and physiological conditions. In the associated sediments, BSAFs of β -HCH, γ -HCH, and Σ DDTs in Manila clams were estimated according to the concentrations of β -HCH, γ -HCH, and Σ DDTs, respectively (Table 1).

In this study, BSAF of β -HCH varied between 0.06 and 1.36. The highest BSAF of β -HCH was found at site DK (1.36) where β -HCH concentration in the sediment was relatively low (0.08 ng g⁻¹ dw). At CS, low BSAF of β -HCH (0.06) occurred with the highest concentration of β -HCH in the associated sediment (0.54 ng g⁻¹ dw). At site DK, BSAF of γ -HCH was the highest while γ -HCH concentration in the sediment was the lowest (0.08 ng g⁻¹ dw). BSAFs of Σ DDTs ranging from 0.31 to 1.06 was not clearly related to the concentrations of DDT in the associated sediment ($r^2=0.14$). The results of this study

Table 1. BSAFs and sediment characteristics of organochlorine pesticides in Manila clams from the study area

Site	BSAF			Concentrations in sediments			Organic carbon (%)
	β -HCH (ng g ⁻¹ dw)	γ -HCH (ng g ⁻¹ dw)	Σ DDTs (ng g ⁻¹ dw)	β -HCH (ng g ⁻¹ dw)	γ -HCH (ng g ⁻¹ dw)	Σ DDTs (ng g ⁻¹ dw)	
MD	0.25	0.45	0.49	0.37	0.15	0.69	0.50
DH	0.89	0.29	1.02	0.08	0.08	0.15	0.50
SG	0.66	0.59	0.61	0.11	0.10	0.29	0.41
AH	0.36	0.13	0.35	0.20	0.10	1.17	0.51
CS	0.06	0.57	0.31	0.54	0.08	0.30	0.47
KA	1.29	0.30	1.06	0.10	0.15	0.51	0.43
DK	1.36	0.85	0.40	0.08	0.08	0.39	0.42
DE	0.07	0.12	0.63	0.34	0.12	0.16	0.42

indicate that organic carbon contents in the sediment varied only from 0.41% to 0.51%. Our BSAF values are within the same range as in other studies. Zhou *et al.* (2008) measured BSAF values of 0.19~1.50 for Σ HCHs and 0.20~1.52 for Σ DDTs in the Asiatic Clam (*Corbicula fluminea*) collected from Qiantang River, China. Zhao *et al.* (2009) reported BSAF values for Σ HCHs and Σ DDTs in a freshwater benthic gastropod, *Bellamya aeruginosa*, of Taihu Lake, China to be in the range 0.06~0.16 and 0.23~0.74, respectively.

Yang *et al.* (2006) reported that higher BSAF of Σ HCHs and Σ DDTs in Asiatic clams (*Corbicula fluminea*), razor clams (*Sinonovacula constricta*), and Chinese marsh crabs (*Sesarma denaani*) collected from the Yangtze River estuary occurred at less polluted sites with lower organic carbon while lower BSAF was found at more contaminated sites with higher organic carbon. They explained that this effect was due to HCHs and DDTs having greater affinity for polluted, anthropogenic organic carbon. Thus, the hydrophobic organic contaminants would have less bioavailability to organisms. Ferraro *et al.* (1990) also found bivalve BSAF values to be lower in highly polluted, high organic carbon sediment and sometimes higher in low polluted, low organic carbon sediment.

The absence of highly polluted sites among our sampling sites is a possible reason why our results did not show the clear pattern of BSAF variation reported by Yang *et al.* (2006). Yang *et al.* (2006) collected some samples at polluted sites with Σ DDT concentration exceeding 20 ng g⁻¹ dw. Another possible reason is that the bioaccumulation potential in Manila clams may be different from that of the Asiatic clams and razor clams used by Yang *et al.* (2006). Magnusson *et al.* (2006) found that the biota/sediment partitioning of individual PCBs, expressed as BSAF, varied considerably between both congeners and

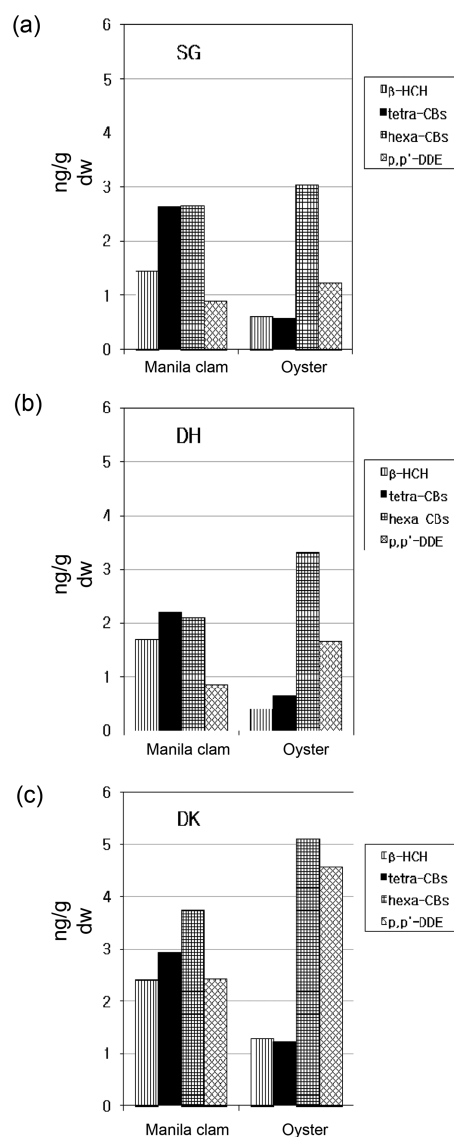


Fig. 4. The concentrations of β -HCH, tetra-CB, hexa-CB, and p,p'-DDE in Manila clams and oysters Collected along the mid-western coast of Korea (ng g⁻¹ dw) : (a) site SG, (b) site DH, and (c) site DK.

species for the same congener. They found that differences in biotransformation may cause interspecific variations in bioaccumulation as well as the age and size of the analyzed specimens. Morrison *et al.* (1996) also claimed that the feeding strategy of biota has a large influence on the degree of bioaccumulation.

Organic contaminants in Manila clams and oysters collected at 3 sites

In this study, oysters were collected to compare the accumulation potential of organic pollutants between two species. Various organic contaminants were measured in oyster tissues (*Crassostrea gigas*) collected from three sites. Fig. 4 shows the concentrations of β -HCH, tetra-CB, hexa-CB and p,p'-DDE in Manila clams and oysters collected from three sites. Relative abundance of organochlorine compounds in Manila clams differs to that of oysters. Heavily chlorinated PCBs and p,p'-DDE were more commonly found in oyster tissues than in clam tissues. While the concentration of hexa-CBs in Manila clams (2.09~3.73 ng g⁻¹ dw) was lower than in oysters (3.03~5.09 ng g⁻¹ dw), the concentration of tetra-CBs in Manila clams (2.21~2.94 ng g⁻¹ dw) was higher than in oysters (0.56~1.23 ng g⁻¹ dw). β -HCH, which has a low log K_{ow} (octanol-water partition coefficient) value, was richer in Manila clams than in oysters.

Differences in the accumulation pattern between Manila clams and oysters would be of interest. The removal of higher chlorinated PCB congeners in clam tissues may not be realistic since mollusks have a very low metabolic capacity for persistent, recalcitrant compounds (Kannan *et al.* 1995). One of the reasons for the different accumulation pattern of organic pollutants might be habitat differences. In this study, the oysters were collected from shellfish farms. Oysters usually grow in the water column, while Manila clams dwell on the sediment surface. Manila clams have more of a chance to be exposed to pore water than cultured oysters since Manila clams bury their bodies in sandy-mud bottoms and filter-feed using a siphon. Previous studies on PCBs in dissolved and particle phases of sea water suggest that the dissolved phase is dominated by lower chlorinated congeners (Schulz *et al.* 1988; Schulz-Bull *et al.* 1991, 1995). The uptake pattern in this study reveals domination of lower chlorinated PCBs in Manila clams occurring in the dissolved phase of water. This is a possible cause for the different accumulation pattern of chlorinated organic compounds between Manila clams and oysters.

4. Conclusions

This study provides bioaccumulation data of PCBs and organochlorine pesticides in Manila clams dwelling in the intertidal sediments of the mid-western coast of Korea. The intertidal sediments of the mid-western coast of Korea generally have low PCB and DDT concentrations. The profile of PCBs in the sediments and Manila clams differs because the sediment is dominated by tetra- to penta-chlorinated congeners, while penta- to hexa-chlorinated congeners dominate Manila clams. BSAFs of Σ HCHs and Σ DDTs in Manila clams were estimated according to the concentrations of Σ HCHs and Σ DDTs, respectively, in the associated sediments. No clear relationship was observed between BSAF of Σ DDTs in Manila clams and the concentration of Σ DDTs in the associated sediments. Heavily chlorinated PCBs and p,p'-DDE were more enriched in oyster tissues than in clam tissues in the study area. The study findings suggest that the different accumulation pattern of organic pollutants in Manila clams and oysters might result from habitat difference. Physiological behavior and metabolic activity of each species could also cause a difference in contaminant concentration in the organisms. These results demonstrate that, in the same location subjected to the same contamination sources, the availability and distribution of organic contaminants are highly variable between species. Biomonitoring using Manila clams has an advantage over other organisms in that they are widely distributed and can be easily collected. More intensive studies on the factors affecting the bioaccumulation potential of individual organic contaminants in Manila clams are needed to relate contaminant levels in clam tissues to the distribution of contaminants in the coastal environment.

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References

- Baumard P, Budzinski H, Garrigues P (1998) PAHs in the Arcachon Bay: Origin and biomonitoring with caged organisms. *Mar Pollut Bull* 36(8):577-586
- Binelli A, Provini A (2003) POPs in edible clams from

- different Italian and European markets and possible human health risk. *Mar Pollut Bull* **46**(7):886-897
- Boscolo R, Cacciatore F, Berto D, Giani M (2007) Polychlorinated biphenyls in clams *Tapes philippinarum* cultured in the Venice Lagoon (Italy): Contamination levels and dietary exposure assessment. *Food Chem Toxicol* **45**(5):1065-1075
- Carvalho FP, Fowler SW, Readman JW, Mee LD (1992) Pesticide residues in tropical coastal lagoons ; The use of ¹⁴C-labelled compounds to study cycling and fates of agrochemicals. In *Proceedings of the International Symposium on Application of Isotopes and Radiation in Conservation of the Environment* (Kahlsruhe, Germany 9-13 March, 1992), IAEA-Sm-325/106; IAEA, Vienna
- Dame RF (1996) *Ecology of Marine Bivalves; An Ecosystem Approach*. CRC Press, Florida, USA
- Di Toro DM, Zarba CS, Hansen DJ, Berry WJ, Swartz RC, Cowan CE, Pavlou SP, Allen HE, Thomas NA, Paquin PR (1991) Technical basis for establishing sediment quality criteria for nonionic organic chemicals using equilibrium partitioning. *Environ Toxicol Chem* **10**(6): 1541-1583
- Erickson MD (1992) Physical, chemical, commercial, environmental and biological properties. *Analytical Chemistry of PCBs*. Lewis publisher, Inc., Michigan, 62 p
- Ferraro SP, Lee IIIH, Ozretich RJ, Specht DT (1990) Predicting bioaccumulation potential: a test of a fugacity based model. *Arch. Environ Contam Toxicol* **19**(3):386-394
- Galceran MT, Santos FJ (1993) PCBs and chlorinated pesticides in shellfish of a Deltaic environment. *Chemosphere* **27**(7):1183-1200
- Hedges JI, Stern JH (1984) Carbon and nitrogen determinations of carbonate-containing solids. *Limnol Oceanogr* **29**(3):657-663
- Hong SH, Yim UH, Shim WJ, Oh JR (2005) Congener-specific survey for polychlorinated biphenyls in sediments of industrialized bays in Korea: Regional characteristics and pollution sources. *Environ Sci Technol* **39**(19):7380-7388
- Hong SH, Yim UH, Shim WJ, Oh JR, Lee IS (2003) Horizontal and vertical distribution of PCBs and chlorinated pesticides from Masan Bay, Korea. *Mar Pollut Bull* **46**(2):244-253
- Ji J, Choi HJ, Ahn IY (2006) Evaluation of Manila clam (*Ruditapes philippinarum*) as a sentinel species for metal pollution monitoring in estuarine tidal flats of Korea: Effects of size, sex, and spawning on baseline accumulation. *Mar Pollut Bull* **52**(4):447-468
- Jones KC, de Voogt P (1999) Persistent organic pollutants (POPs): state of the science. *Environ Pollut* **100**(1):209-221
- Kannan N, Reusch TBH, Schulz-Bull DE, Petrick G, Duinker IC (1995) Chlorobiphenyls: Model compounds for metabolism in food chain organisms and their potential use as ecotoxicological stress indicators. *Environ Sci Technol* **29**(7):1851-1859
- Kim SK, Oh JR, Shim WJ, Lee DH, Yim UH, Hong SH, Shin YB, Lee DS (2002) Geographical distribution and accumulation features of organochlorine residues in bivalves from coastal areas of South Korea. *Mar Pollut Bull* **45**(1):268-279
- Lauenstein GG, Cantillo AY (1993) Sampling and analytical methods of the national status and trends program, national benthic surveillance and mussel watch projects 1984-1992. *NOAA Technical Memorandum NOS ORCA* 71, 182 p
- Long ER, MacDonald DD, Smith SL, Calder FD (1995) Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments. *Environ Manage* **19**(1):81-97
- Magnusson K, Ekelund R, Grabic R, Bergqvist PA (2006) Bioaccumulation of PCB congeners in marine benthic infauna. *Mar Environ Res* **61**(4):379-395
- Marchand M, Abarnou A, Marcaillou-Lebaut C (1990) Les polychlorobiphenyles (PCB) en milieu marin, biogéochimie et écotoxicologie. *Rapport Scientifiques Techniques IFREMER* 18, Brest, France
- Monirith I, Ueno D, Takahashi S, Nakata H, Sudaryanto A, Subramanian A, Karuppiah S, Ismail A, Muchtar M, Zheng J, Richardson BJ, Prudente M, Hue ND, Tana TS, Tkalin AV, Tanabe S (2003) Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries. *Mar Pollut Bull* **46**:281-300
- Morrison HA, Gobas FAPC, Lazar R, Haffner GD (1996) Development and verification of a bioaccumulation model for organic contaminants in benthic invertebrates. *Environ Sci Technol* **30**(11):3377-3384
- Nasci C, Da Ros L, Nesto N, Sperti L, Passarini F, Pavoni B (2000) Biochemical and histochemical responses to environmental contaminants in clam, *Tapes philippinarum*, transplanted to different polluted areas of Venice Lagoon Italy. *Mar Environ Res* **50**(1):425-430
- O'Connor TP (1996) Trends in chemical concentrations in mussels and oysters collected along the US coasts from 1986 to 1993. *Mar Environ Res* **41**(2):183-200

- Qian Y, Serican JL, Wade TL (1998) Determination of Percent Lipid in Tissue. In : Lauenstein GG, Cantillo AA (eds) Sampling and analytical methods of the National Status and Trends Program Mussel Watch Project: 1993-1996 Update. NOAA Technical Memorandum NOS ORCA 130, pp 27-29
- Ramu K, Kajiwaru N, Isobe T, Takahashi S, Kim EY, Min BY, We SU, Tanabe S (2007) Spatial distribution and accumulation of brominated flame retardants, polychlorinated biphenyls and organochlorine pesticides in blue mussels (*Mytilus edulis*) from coastal waters of Korea. *Environ Pollut* **148**(1):562-569
- Schulz DE, Petrick G, Duinker JC (1988) Chlorinated biphenyls in north Atlantic surface and deep water. *Mar Pollut Bull* **19**(10):526-531
- Schulz-Bull DE, Petrick G, Duinker JC (1991) Polychlorinated biphenyls in North Sea water. *Mar Chem* **36**:365-384
- Schulz-Bull DE, Petrick G, Kannan N, Duinker JC (1995) Distribution of individual chlorobiphenyls (PCB) in solution and suspension in the Baltic Sea. *Mar Chem* **48**(3):245-270
- Solé M, Porte C, Barcelo D, Albaiges J (2000) Bivalves Residue Analysis for the Assessment of Coastal Pollution in the Ebro Delta (NW Mediterranean). *Mar Pollut Bull* **40**(9):746-753
- Thompson S, Budzinski H, Garrigues P, Narbonne JF (1999) Comparison of PCB and DDT Distribution between Water-column and Sediment-dwelling Bivalves in Arcachon Bay, France. *Mar Pollut Bull* **38**(8):655-662
- Tolosa I, Bayona IM, Albaiges J (1996) Aliphatic and polycyclic aromatic hydrocarbons and sulfur/oxygen derivatives in Northwestern Mediterranean sediments: spatial and temporal variability, fluxes and budgets. *Environ Sci Technol* **30**(8):2495-2503
- Yang Y, Liu M, Xu S, Hou L, Ou D, Liu H, Cheng S, Hofmann T (2006) HCHs and DDTs in sediment-dwelling animals from the Yangtze Estuary, China. *Chemosphere* **62**(3):381-389
- Zhao Z, Zhang L, Wu J, Fan C (2009) Distribution and bioaccumulation of organochlorine pesticides in surface sediments and benthic organisms from Taihu Lake. *China Chemosphere* **77**(9):1191-1198
- Zhou R, Zhu L, Kong Q (2008) Levels and distribution of organochlorine pesticides in shellfish from Qiantang River, China. *Journal of Hazardous Materials* **152**:1192-1200

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