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Ecological and human health risk from polychlorinated biphenyls and organochlorine pesticides in bivalves of Cheonsu Bay, Korea

Jin Young Choi[1], Dong Beom Yang[1†], Gi Hoon Hong[1], Kyoungrean Kim[1], Kyung-Hoon Shin[2]

[1]*Korea Institute of Ocean Science and Technology, 787, Haeanro, Ansan 15627, Republic of Korea*
[2]*Department of Marine Sciences and Convergent Technology, College of Science and Technology, Hanyang University, Ansan 04763, Republic of Korea*

ABSTRACT

Cheonsu Bay, one of the most important in Korea as a coastal fishery is a semi-enclosed bay that is surrounded by large farmlands and industrial areas. This coastal environment has been affected by anthropogenic pollutants, such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs). The objectives of this study were to investigate the distribution of PCBs and OCPs in sediment, Manila clams, and mussels from Cheonsu Bay; the accumulation pattern of these chemicals in these bivalves in relation to seasonal changes; and the ecological risk from sediments and the risk to the Korean population from the consumption of these bivalves. The levels of ΣPCBs, ΣDDT, and ΣHCHs were 69.3-109, 40.3-49.3 and 6.25-17.8 ng/g lipid in Manila clams, and 70.6-159, 38.6-102 and 9.00-13.5 ng/g lipid in Mussels. Significant seasonal variations in PCBs and OCPs concentrations were observed in the two bivalves, suggesting that the accumulation of PCBs and OCPs in these species is related to their spawning times. The dietary intake of these two bivalves and the resulting lifetime cancer risk (LCR) and non-cancer risk were calculated for the human population. The consumption of these bivalves seemed to be safe in relation to human health with negligible LCR and non-cancer risk.

Keywords: Bio-accumulation, Ecological risk assessment, Human health risk assessment, Marine bivalves, Persistent organic pollutants (POPS)

1. Introduction

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), which are anthropogenic organic pollutants, have been considered important contaminants for several decades. PCBs and OCPs are hydrophobic and lipophilic substances that persist in the environment for very long periods [1]. Because of the extreme stability of many of their isomers, PCBs were widely used in many products until they were banned in developed countries during the late 1970s. OCPs were major agricultural pesticides until they were restricted because of their serious side effects. Chronic toxicity, reproductive, carcinogenicity, and develomental toxicity, as well the biochemical effects of comecial PCBs mixture and OCPs have been investigated in fish and wildlife species [2-6]. As well, ecological risk, including bio-magnification, bio-accumulation of those pollutants in food web in marine environment was also reported [7-8].

As a result of much effort, including restrictions, the release of PCBs and OCPs to the environment has decreased over the last few decades. However, because of the hydrophobic nature of these compounds, huge amounts have sedimented to the sea floor and have been adsorbed onto organic particles [9]. Contaminated marine sediment and suspended organic particles can be a secondary source of organic pollutants to marine organisms, particularly sediment-dwelling organisms [10-12]. Because of their high octanol/water partition coefficient (K_{ow}), these hydrophobic organic pollutants accumulate in the bodies of marine organisms [9]. Even extremely low concentrations of bioaccumulative pollutants detected in water or bottom sediments may result in their accumulation in marine organisms such as fish and shellfish [13]. Bivalves have been widely used to obtain information about local pollution [14, 15], and were recommended as bio-indicators by

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† Corresponding author
Email: dbyangh@naver.com
Tel: +82-31-400-6157 Fax: +82-31-408-4493

international conventions such as the OSPAR Commission, the Barcelona Convention, and the Helsinki Commission [16-18].

Manila clams (*Ruditapes philippinarum*) and mussels (*Mytilus coruscus*) are widely distributed throughout the intertidal zones along the western coast of Korea [19]. Mussels live on rocky shores, attaching themselves to rocks or to each other by means of byssal threads. As part of the Mussel Watch Program in Korea, Kim et al. assessed the contamination levels and accumulation features of PCBs and OCPs along the entire coast of Korea [20]. Ramu et al. analyzed mussels from 20 coastal locations in Korean waters to determine the concentrations of PCBs and OCPs [21]. Manila clams are also regarded as a useful bioindicator of sediment pollution because they live in sandy-mud bottoms and extrude their siphons to filter feed from near-bottom water [22]. The bioaccumulation of organic pollutants in Manila clams has been reported in recent studies [14, 23, 24].

Bioaccumulated hydrophobic organic pollutants are associated with the lipid content of biota. Lipids and fats change throughout the seasons in relation to an animal's physiological condition, such as spawning and gametogenesis [25, 26]. Thus, the accumulated concentrations in biota of lipophilic contaminants, including organochlorine contents, can be influenced by seasonal changes and age-grade. Both Manila clams and mussels are commercially important bivalves, because they are a fishery food resource in Korea. Thus, understanding the bioaccumulation mechanisms in these aquatic organisms is critical to predicting their potential toxicity to human health [27]. However, the effects of season and size on organochlorine concentrations in *R. philippinarum* and *M. coruscus* collected in Korea have not been previously reported.

Cheonsu Bay, which is one of the most important bays in Korea, has a long tidal beach with a length of about 2,000 km. Both sandy-mud flat and tidal rock areas, the habitats of Manila clams and mussels, respectively, are well developed along the shoreline of Cheonsu Bay. This area is surrounded by agricultural and industrial areas [28], and the environmental pollution and ecological changes by anthropogenic activities have been reported [29]. Park et al. reported the environmental impacts on water quality of Cheonsu Bay derived from the input of adjacent fresh waters [30].

In order to understand the accumulation characteristics of PCBs and OCPs in Manila clams and mussels, surface sediment, Manila clams, and mussels from Cheonsu Bay, Korea, were collected. The objectives of this study were thus (1) to investigate the current distributions of PCBs and OCPs in sediment, Manila clams, and mussels; (2) to determine how the accumulation patterns of these chemicals in Manila clams and mussels are related to seasonal changes; and (3) to assess the ecological risk from the sediment and the risk to the Korean population posed by consuming these two bivalves.

2. Materials and Methods

2.1. Sample Collection

Fig. 1 shows three sampling sites from Cheonsu Bay, Korea. These sampling sites were selected in order to provide full coverage of this bay. Surface sediments, Manila clams, and mussels were collected from these three sampling sites in different seasons.

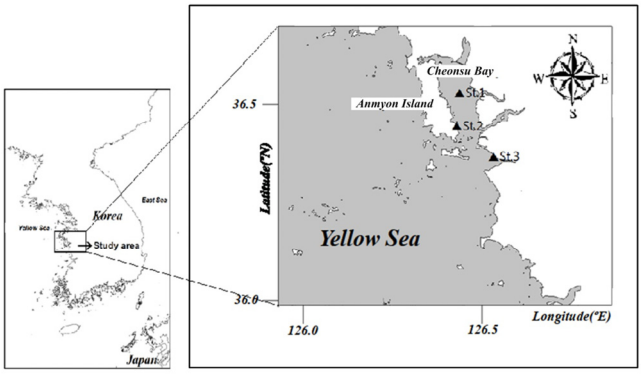


Fig. 1. Sampling sites for sediments, Manila clams (*Ruditapes philippinarum*), and mussels (*Mytilus coruscus*) in Cheonsu Bay, Korea.

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 using Labconco Freezone 6.

Manila clams were hand collected in December, May, and August in 2001-2002, and mussel samples were collected in December, May, and July in 2001-2003, in order to compare the accumulation patterns of PCBs and OCPs in the tissues of these two bivalves according to the sampling season and size. The collected bivalves were wrapped in aluminum foil and stored frozen until analysis. The shells of the bivalves were removed, and the soft tissues, including the viscera, were pooled and homogenized for analysis. All of the samples were freeze-dried using Labconco Freezone 6. The samples were analyzed for PCBs and OCPs.

2.2. Chemical Analysis

The extraction and analysis of PCBs and OCPs were conducted according to the method described by Lauenstein and Cantillo [31], with some modifications. Freeze-dried sediment and pooled clam tissue samples were spiked with 2,2′,4,5′,6-pentachlorobiphenyl (PCB 103), 2,2′,3,3′,4,5,5′,6-octachlorobiphenyl (PCB 198), and 4,4′-dibromo octafluoro biphenyl (DBOFB) as surrogate standards, and a Soxhlet extraction was carried out using hexane:acetone (1:1, v/v). Sulfur was removed from the extracts of the sediments using activated copper (Granular, 20-30 mesh, J.T. Baker Co., USA). Each extract was passed through a column containing deactivated silicagel (~60 mesh; Merck, Germany)/alumina (~150 mesh; Aldrich Chemical Co., USA) withhexane:acetone (1:1, v/v) for cleanup to remove the polar/non-polar interference from the extracts. The extracts was also cleaned using a deactivated Florisil® (60-100 mesh; J.T. Baker Co., USA) column with hexane for further separation of PCBs from the most extracted components. Samples were concentrated to 0.5 mL under nitrogen and sealed in vials until analyzed.

2.3. Instrumental Analysis and Quality Control

A gas chromatograph equipped with a 63Ni electron capture detector (GC-ECD Hewlett Packard 5890) was used to identify and quantify the PCBs and OCPs. The GC column used for the analysis was a DB-5 (25-μm bonded phase; J&W Scientific Co Ltd., USA). The organic carbon in the sediment sample was quantified using

a Carlo Erba CNS elemental analyzer 1106 following the method described by Hedges and Stern [32]. The lipid weight percentages (weight/weight basis) in the tissues of the organisms were operationally determined using the method of Qian et al. [33]. The same process and analytical method were used for blanks without samples to determine the contamination during the analytical process, and all the presented results were corrected for the blank levels. All of the glassware was cleaned with detergent, baked at 450℃ for 4 h, and rinsed with hexane before use. The recovery percentages of the spiked PCBs and OCPs varied from 75.4% to 116.2% for the sediments, and 65.6% to 123.3% for the biological samples. The method detection limits (MDL) of the procedure for each compound ranged between 0.01 and 0.03 $ng \cdot g^{-1}$dw. Concentrations detected at less than the MDLs were expressed as not detected (ND). A reagent blank was used to determine laboratory contamination. To check for interference, a spiked sample consisting of all the chemicals was run. For duplicate samples, the average reproducibility of the method varied by less than 10%. Quality assurance procedures were performed by co-analyzing standard reference materials (SRM) of the appropriate matrix, namely, the sediment NIST 1941b and mussel tissue homogenate IAEA-142, with recoveries ranging from 62% to 123%. The 18 congeners (PCBs 8, 18, 28, 29, 44, 52, 66, 87, 101, 105, 110, 118, 128, 138, 153, 170, 180, and 187), including 13 major components in environmental mixtures [34], were analyzed. ∑PCB estimates were derived from the sum of the measured 18 congeners multiplied by a standard factor of two used by NOAA NS&T in 1987 and Howell et al. (2008). The ∑DDT and ∑HCH concentrations were found using the sums of six metabolites (*o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT, and *p,p′*-DDT) and four isomers (α-HCH, β-HCH, γ-HCH, and δ-HCH), respectively.

2.4. Calculation of Human Health Risk from Dietary Intake

To assess the human health risk from ingesting Manila clams with PCBs and OCPs, the exposure levels (expressed as average daily intakes (ADI)) for PCBs and OCPs were calculated. The average daily consumptions of Manila clams (0.80 g/d) and mussels (0.99 g/d) for the general population in South Korea were obtained from the Food Balance Sheet Korea published by the Korean Rural Economic Institute [35]. Lifetime cancer risk (LCR) and hazard indices (HI) were derived using the calculated ADI in this study, the cancer slope factor (CSF_o), and the oral reference dose (RfD_o) for each compound, following the method of health risk assessment suggested by the National Research Council (NRC) and National Academy of Science (NAS) of the USA [36].

$$ADI = Cc \times IR_c / BW$$
$$LCR = Cc \times IR_c \times ED \times EF \times CSF_o / BW \times AT$$
$$HI = \Sigma \text{ Hazard Quotients (HQ) (sum of hazard quotients)}$$
$$\text{Hazard Quotient (HQ)} = C_c \times IR_c \times ED \times EF / RfD_o \times BW \times AT$$

Here, C_c is the concentration of organochlorine pesticide ($ng \cdot g^{-1}$ww) of the clams, IRc is the ingestion rate per unit of time ($kg \cdot d^{-1}$), BW is the body weight of a human (65 kg), ED is the exposure duration (70 y), EF is the exposure frequency (365 y^{-1}), and AT is the average time for exposure (365 $d \cdot y^{-1}$ for 70 y). The cancer slope factor and oral reference dose were obtained using an integrated risk information system (IRIS) reported by the US EPA [37].

2.5. Statistical Analysis

All of the statistical analyses were performed using the PASW 18. One-way ANOVA, Hierarchical cluster analysis (HCA) and principle component analysis (PCA) were used to explain the spatial distributions of the PCBs and OCPs in the sediment samples. Pearson's correlation test was applied to consider the correlation between the contaminants in the sediments and contaminants in the clams, and between Log K_{ow} and Log BSAF for organic pollutants. The levels of significance were 0.05.

3. Results and Discussion

3.1. PCBs and OCPs in Sediment from Cheonsu Bay

Organochlorine pollutants such as PCBs, DDTs, and HCHs were detected in all of the sediment samples collected in Cheonsu Bay. During the sampling period, the mean concentrations of ΣPCBs, ΣDDTs, and ΣHCHs in the sediments collected from sites 1, 2, and 3 were 0.86 ± 0.12, 0.22 ± 0.035, and 0.20 ± 0.051 ng/g dry weight, respectively (Table 1). The organic carbon contents in the sediments were 0.32%, 0.23%, and 0.42%, respectively. In order to verify spatial and temporal homogeneity of chemical sediment parameters, including the concentration of contaminants during the sampling period, one-way ANOVA was performed for sediment groups collected from all sampling stations. There was no statistical difference in the organochlorine pollutant levels in the sediments collected at the three sampling stations, indicating no seasonal or special difference in the organochlorine pollutant levels in the sediments during the period ($F = 0.41$, $p = 0.67$) (Supplementary data 1). Therefore, collected sediments from these sampling stations were suitable for representing the pollution level of Cheonsu Bay with PCBs and OCPs.

Anthropogenic pollutants, including industrial and agricultural chemicals from the watershed, have been potential sources of PCBs and OCPs in Cheonsu Bay. According to the Korea Agricultural and Rural Infrastructure Corporation [39], the numbers of industrial factories and oil consumption in the area surrounding Cheonsu Bay were much higher than those of other areas in Korea. The total amount of DDT applied onto the areas adjacent to Cheonsu Bay from 1946 to 1974, was approximated to be 36.72 tons [40, 41].

In Korea, the use of PCBs in dielectric fluid was restricted in 1979, and the import of PCBs was prohibited in 1984 [42, 49]. Despite this ban, high levels of PCBs have been found in some coastal areas near industrial complexes or large harbors, as listed in Table 2a. However, according to the total PCB estimates (Table 1), none of the sites in this study exceeded the minimum values reported for the sediment quality guidelines, including the effects-range-low (ERL) for marine sediments (22.7 ng/g dry weight) [43]. As listed in Table 2a, the PCB and OCP levels in the sediments of this study area were comparably lower than those reported in other polluted coastal areas [50, 51].

Table 1. Mean Concentrations of ΣPCBs, ΣDDTs, and ΣHCHs in the Sediments Collected from St.1, 2, and 3 at the Cheonsu Bay, Korea, during the Sampling Period (Unit. ng/g dry weight)

Compounds	Unit	Mean ± SD	(Min-Max)
PCB8	ng/g	0.008 ± 0.006	<MDL-0.015
PCB18		0.017 ± 0.012	0.010-0.003
PCB28		0.070 ± 0.020	0.050-0.090
PCB29		0.032 ± 0.026	0.040-0.050
PCB52		0.080 ± 0.062	0.030-0.15
PCB44		0.093 ± 0.016	0.080-0.11
PCB66		0.085 ± 0.028	0.06-0.12
PCB87		0.16 ± 0.057	0.11-0.22
PCB101		0.027 ± 0.022	0.010-0.052
PCB105		< MDL	-
PCB110		0.022 ± 0.017	0.0065-0.040
PCB118		< MDL	-
PCB128		0.053 ± 0.015	0.040-0.069
PCB138		0.058 ± 0.034	0.023-0.090
PCB153		0.021 ± 0.008	0.014-0.030
PCB170		0.032 ± 0.013	0.020-0.045
PCB187		0.034 ± 0.017	0.020-0.053
PCB180		0.050 ± 0.035	0.029-0.090
Σ PCBs		0.86 ± 0.12	0.75-0.98
Total PCBs estimateda		1.73 ± 0.23	1.5-2.0
o,p'-DDE	ng/g	0.035 ± 0.028	0.04-0.06
p,p'-DDE		0.039 ± 0.011	0.03-0.05
o,p'-DDD		0.018 ± 0.014	0.01-0.04
p,p'-DDD		0.061 ± 0.036	0.02-0.26
o,p'-DDT		0.029 ± 0.033	0.01-0.07
p,p'-DDT		0.042 ± 0.011	0.03-0.050
Σ DDTs		0.22 ± 0.035	0.20-0.26
p,p'-DDT/p,p'-DDE+p,p'-DDD	-	0.46 ± 0.24	0.25-0.71
α-HCH	ng/g	0.008 ± 0.003	0.008-0.010
β-HCH		0.108 ± 0.037	0.070-0.14
γ-HCH		0.057 ± 0.015	0.040-0.070
δ-HCH		0.031 ± 0.012	0.020-0.040
Σ HCHs		0.20 ± 0.051	0.17-0.26
α-HCH/γ-HCH	-	0.16 ± 0.095	0.070-0.25
Organic carbon (%)	%	0.32 ± 0.095	0.23-0.42

Total PCBs[a] estimates derived from the sum of 18 congeners multiplied by a standard factor of two [38], and < MDL is 'below method detection limit'.

Supplementary data 1. One-way ANOVA Results for HCHs, DDTs, and PCBs Concentrations in Sediment from Cheonsu Bay (α = 0.05)

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	0.00144	2	0.0007	0.41	0.67
Within Groups	0.14205	81	0.0018		
Total	0.14349	83			

Fig. 2 presents the profiles of the PCBs and OCPs in the sediments from Cheonsu Bay. The most important homologue group of PCBs found in the sediments appeared to be tetra-CBs, which accounted for about 30% of the PCB contamination, followed by penta-CBs, which accounted for 27%. In order to consider the source of PCB pollution, the congener profiles of PCBs in this area were compared with those of commercial PCB mixtures. Typical chlorination level plots from the sediment samples and PCB commercial mixtures such as those of Aroclor 1248, 1254, and 1260 are illustrated in Fig. 2a. All the profiles indicated that the PCBs in the sediments

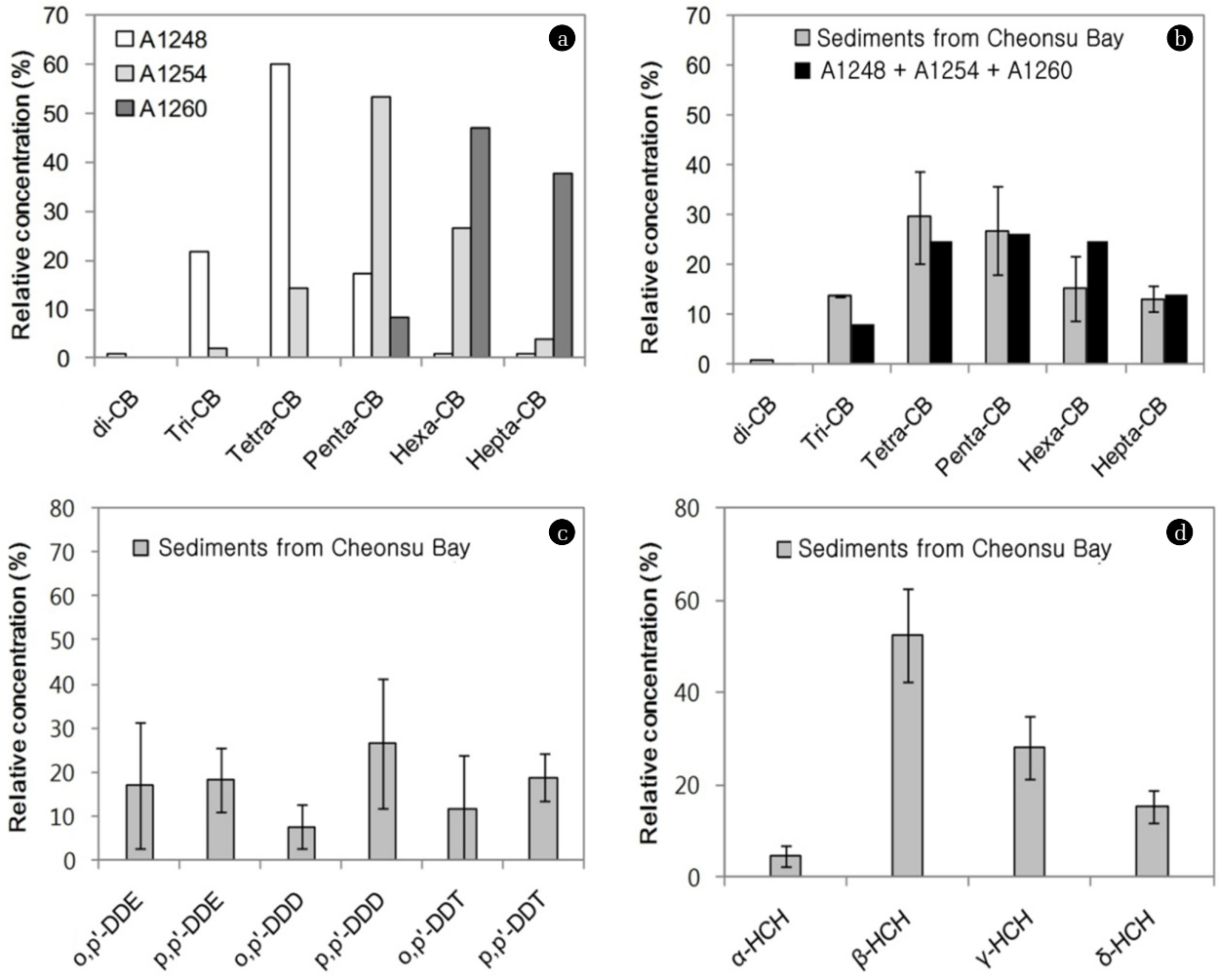


Fig. 2. a) PCB composition of commercial PCBs, Aroclor 1248 (A1248), -1254 (A1254), and -1260 (A1260), b) composition of PCB homologues, c) DDT metabolites, and d) HCH isomers in sediment samples from Cheonsu Bay.

were of a composite mixture and that they derived from more than one technical formulation with different degrees of chlorination (Fig. 2b). Similar PCB profiles were also estimated in other areas of Korea [24], and their results showed that the PCBs in Nakdong River sediment were influenced by Aroclor 1254 and 1260.

As shown in Fig. 2c and 2d, *p,p'*-DDD and β-HCH were the dominant pesticides among the OCP compounds in the sediment. As listed in Table 1, the DDT compositional ratio (*p,p'*-DDT/*p,p'*-DDE+*p,p'*-DDD) of the sediment samples had a range of 0.25-0.71 in this area. This result suggests that the DDTs found in the studied sediment were mainly weathered DDTs from past use. The α-HCH/γ-HCH ratios in the sediment samples were below one, indicating the previous use of lindane (γ-HCH) in this area [56]. The Stockholm conventions classified HCHs, including α-HCH, β-HCH, and lindane (γ-HCH), as "Additional POPs" in 2009, because of their bioaccumulation, their potential for long-range transport, and their adverse effects. The technical HCH mixture has been banned in many countries since the 1970s and 1980s, while lindane has been used until recently.

Distributions and seasonal changes in PCBs and OCPs in Manila clams and mussels collected in Cheonsu Bay.

The lipid-normalized concentrations of PCBs and OCPs in the Manila clams and mussels from Cheonsu Bay, Korea, are summarized in Table 3. The PCB levels were the highest in the tissues of the Manila clams and mussels, with concentrations of 91.2 ± 20.2 ng/g and 118 ± 36.4 ng/g lipid weight, respectively. The concentrations of DDTs in the Manila clams and mussels were 45.5 ± 4.68 ng/g and 68.8 ± 31.8 ng/g lipid weights, respectively. The HCH concentrations in the Manila clams and mussels were 12.9 ± 5.95 ng/g and 11.2 ± 2.25 ng/g lipid weights, respectively, showing lower levels than the DDTs. These levels were relatively lower than those previously reported for Manila clams and mussels collected in other coastal areas in the world, showing that seafood in Korea is less contaminated than that in other coastal areas (Table 2b). Although the consumption of HCHs was higher than

Table 2. The Levels of PCBs, DDTs and HCHs in a) Sediment and in the Literatures and Sediment Quality Guidelines Considered for Organochlorine Contaminants in Marine Sediments, and b) Bivalves Collected from Polluted Coastal Areas in References

(a) (unit. ng/g dry weight)

Locations	ΣPCB	ΣDDT	ΣHCH	Year	References
Venice Lagoon, Italy	2-2049 (12 cong)			1996-1998	[45]
Singapore	1.4-329.6 (37 cong)	2.2-11.9	3.4-46.1	2003	[46]
Osaka Bay, Japan	63-240 (Aroclor and Kaneclor)	2.5-12		1989-1991	[47]
Osaka Bay, Japan	-	2.5	1.3	1990	[47]
Busan Bay, Korea	5.71-199 (23 cong)	-	-	2000	[48]
Masan Bay, Korea	2.48-75.0 (18 cong)	0.27-89.2	nd-1.33	1997	[49]
Minjian River Estuary, China	15.8-57.9	1.6-13.1	3-16.2	1999	[50]
Bohai sea, China	0.30-14.95 (10 cong)	0.38-1417.08	-	1991	[51]
Guaratuba bay, Brazil	n.d-6.06 (194 cong)	n.d-0.49	-	2010	[52]
Sea Lots, Trinidad and Tobago	62-601 (136 cong)	6.1-29	-		[53]
Mersin Bay, Turkey	n.d-26.07 (Ar1254+Ar1260)	n.d.-17.3	-	2009	[54]
Mediterranean Coast	2.29-377.35 (96 cong)	0.07-81.5	-	2005	[55]
Mid-western coast, Korea	1.08-3.5 (36 cong)	0.12-0.35	0.090-0.30	2001-2003	[24]
ERL (effects range low after)	22.7	1.6	-		[44]

-: not measured

(b) (unit. ng/g)

Location	ΣPCBs	ΣDDTs	ΣHCHs	year	References
Masan Bay, Korea	1000 (62 cong)	400	6.4	2005	[21][a]
Ulsan Bay, Korea	500-1000 (62 cong)	240-250	5.2-8.9	2005	[21][a]
Busan Harbor, Korea	16.4 (104 cong)	5.31	0.25	1999	[20][c]
Pohang, Korea	5.6-10.6 (104 cong)	2.95-7.56	0.35-1.02	1999	[20][c]
Chonsoo Bay, Korea	69.3-159 (18 cong)	38.6-102	9.00-17.8	2002	This studya
Hong Kong	49.0-330 (74 cong)	50.0-520	53.0-110	1986	[57][c]
The NW Mediterranean coast	20-630 (Aroclor1254)	130	-	1988-1989	[58][b]
Red Sea coast	6.7-66.4 (10 cong)	98.1-629.8	-	1973-1974	[59][c]
Riá of Ferrol	-	2.17-26.9	n.d.-0.97(γ-HCH only)	1998-2002	[60][b]
Svalbard waters	25.57-171.61 (16 cong)	-	1.93-20.73(α-HCH only)	2009	[61][a]

-: not measured
n.d.: not detected
a lipid weight based level
bdry weight based level
c wet weight based level

that of DDTs in northeast Asian countries [62], the accumulation levels of HCHs were lower than those of the DDTs in the Manila clams and mussels collected from the study area. This may be the result of the higher degradability and lower lipophilicity of HCHs than those of DDTs [63].

The values of PCBs and OCPs in the Manila clams that were 25-45 mm in size and in the mussels that were 30-70 mm in size, collected at different sampling times are compared in Fig. 3. The mean of the lipid normalized ΣPCB concentrations in the Manila clams collected in May and in mussels collected in July were significantly lower than those collected in December (one-way ANOVA, $p < 0.05$). The lowest lipid normalized ΣHCH and ΣDDT concentrations in the Manila clams and mussels were also observed in May and July, respectively. In addition, the mean of the lipid normalized ΣHCH concentration in the Manila clams collected in May was significantly lower than those for clams collected in other periods (one-way ANOVA, $p < 0.05$).

This result might be related to a seasonal physiological change in these organisms, and its possible influencing factor was reproduction. The ΣPCB, ΣDDT, and ΣHCH values in the Manila clam tissues in May decreased by 36.6%, 14.3%, and 56%, respectively, compared with those for December. In the case of the mussels, the ΣPCB, ΣDDT, and ΣHCH values in the tissues collected in July decreased by 38.4%, 58.3%, and 23.7%, respectively, compared to those obtained in May. In this area, the spawning period of Manila clams is from April to June [64], and that of mussels is from May to July. This suggests the possibility of release of organochlorines (OCs) from the bodies of Manila clams and mussels

Table 3. Lipid Contents and Lipid-normalized Concentrations of PCBs and OCPs in Tissues of Manila Clams and Mussels Collected from Cheonsu Bay, Korea

	Sampling time	Lipid (%)	ΣPCBs	ΣDDTs (ng/g lipid weight)	ΣHCHs
Ruditapes philippinarum	December, 2001	0.99	109	47.0	14.5
	May, 2002	1.45	69.3	40.3	6.25
	August, 2002	1.03	95.3	49.3	17.8
	Mean	1.16	91.2	45.5	12.9
	SD	0.252	20.2	4.68	5.95
Mytilus coruscus	December, 2001	2.00	159	65.8	11.0
	May, 2002	0.88	124	102	13.5
	June, 2003	1.92	70.6	38.6	9.00
	Mean	1.60	118	68.8	11.2
	SD	0.625	36.4	31.8	2.25

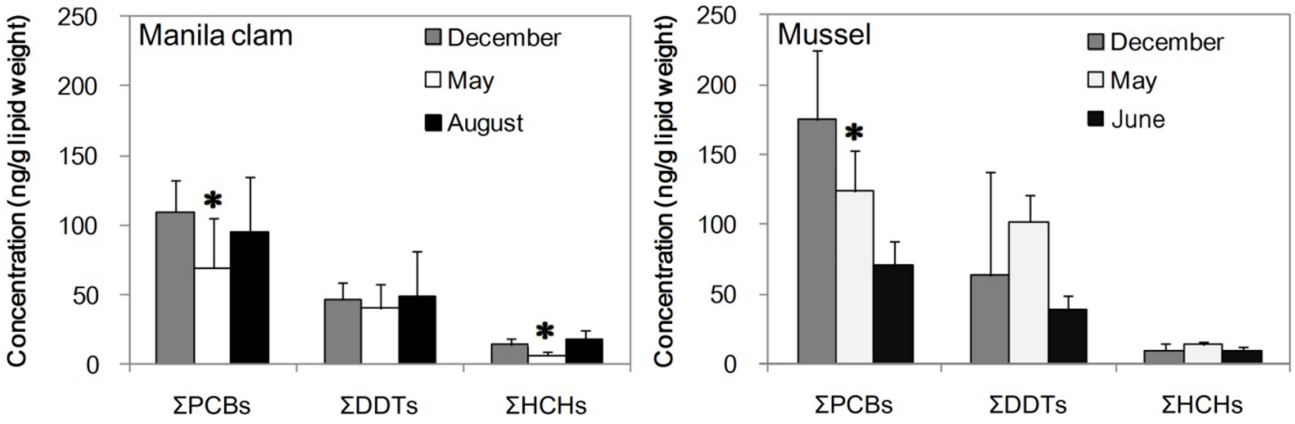


Fig. 3. Seasonal changes in ∑PCBs, ∑DDTs, and ∑HCHs concentrations in Manila clams and mussels. Asterisks (*) indicate a significant difference value. One-way ANOVA: * $p < 0.05$.

through spawning behavior.

Other studies found similar seasonal variation in OC accumulation in the bodies of aquatic organisms, including fish [65, 66], cetacean [67], rainbow trout [68], and the copepod Acartia tonsa [69]. Moreover, Hummel et al. reported that mussels might "lose" one-half to two-thirds of their PCB content by means of reproductive output [70]. Because the concentrations of OCs in Manila clams and mussels is able to be varied by their breeding behaviors, seasonal consideration in using these bivalves as an indicator species for risk assessment is more required. So we investigated that how risks to human health change with consuming these two bivalves as a result of seasonal variation of OCs in those bivalves.

Comparison of organic pollutant accumulation potentials of Manila clams (*Ruditapes philippinarum*) and mussels (*Mytilus coruscus*) and analysis of human health risk posed by consuming these two bivalves

The organic pollutant accumulation potentials of Manila clams, *Ruditapes philippinarum*, and mussels, *Mytilus coruscus*, were compared, and their PCB and OCP concentration profiles are shown in Supplementary data 2. The tetra-, penta-, and hexa-CBs were the major homolog groups in the Manila clams and mussels, constituting 67% and 77% of the total PCBs in Manila clams and mussels, respectively. The predominant congeners of PCBs were PCB 138 and 153 in both Manila clam and mussel tissues. In previous studies of other organisms in Korea, similar profiles were reported for porpoise, fish, and human serum [67, 71, 72]. The other major congeners were PCB 52, 87, and 44 in Manila clams and PCB 110, 52, and 44 in mussels. Among the OCPs, the major compound was *p,p'*-isomers of DDT followed by β-HCH, indicating long-term degradation from the use of those OCPs.

The accumulation patterns of the OCs in Manila clams were slightly different from those in mussels. In order to compare the accumulations of OCs in these two bivalves, a hierarchical cluster analysis (HCA) of the important pollutants in Manila clams and mussels collected at different sampling times, and of the sediments, was carried out (Fig. 4). As a result of this HCA, Manila clams and sediment were included in Group 1, and the other cluster, Group 2, consisted of mussel samples. This result indicated that the distributions of PCBs and OCPs in Manila clams were more closely related to those in sediment than those in mussels. This result might be caused by the habitat difference between these two species. Mussels inhabit the reefs of coastal regions, attached

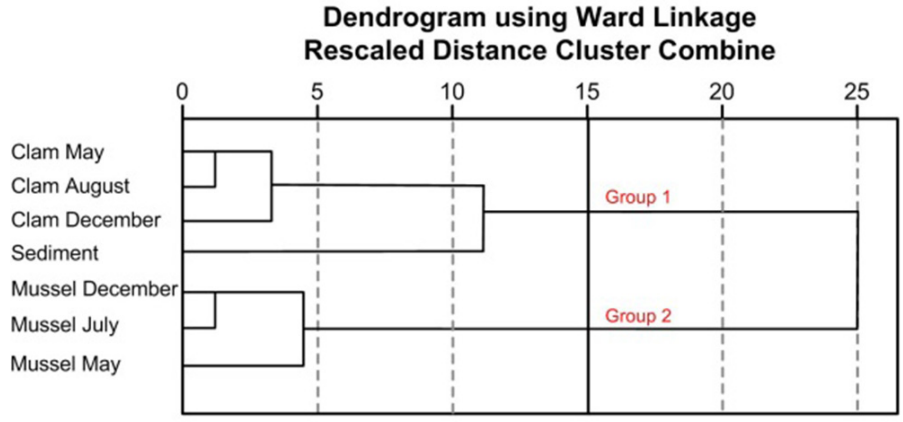
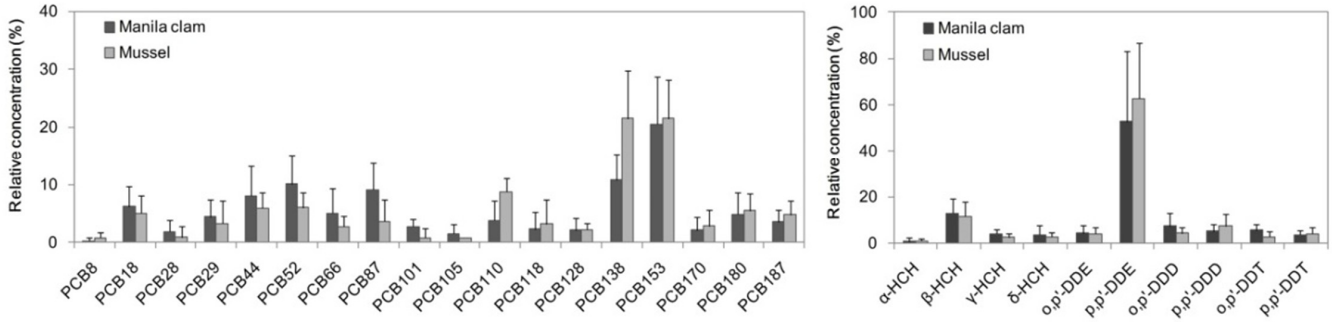


Fig. 4. Dendrogram of hierarchical cluster analysis of important PCB congeners (PCB 44, 52, 87, 110, 138, and 153), *p,p'*-DDE, and β-HCH in sediment, Manila clam, and mussel samples.



Supplementary data 2. Profiles of a) PCBs and b) OCPs concentrations in Manila clams and mussels from Cheonsu Bay.

Table 4. Life-time Cancer Risk (LCR) and Non-cancer Risk Posed by Consumption of Manila Clams and Mussels by Korean Population

Compounds		*Ruditapesphilippinarum*				*Mytiluscoruscus*			
		DEC	MAY	AUG	Annual mean	DEC	MAY	JUN	Annual mean
	CSfoa (mg/kg/d)	Life-time cancer risk (LCR)							
ΣPCBs	2	5.8E-08	4.9E-08	6.2E-08	5.6E-08	2.1E-07	7.1E-08	9.0E-08	1.3E-07
ΣDDTs	0.34	1.4E-09	2.4E-09	2.6E-09	2.1E-09	7.1E-09	5.0E-09	4.2E-09	5.5E-09
α-HCH	6.3	-	-	-	-	-	1.2E-09	-	-
β-HCH	1.8	1.3E-08	1.1E-09	2.5E-09	5.4E-09	3.8E-09	2.5E-09	3.3E-09	3.2E-09
	RfDob	Non-cancer risk (Hazardous quotients)							
ΣPCBs	2.E-05	1.5.E-03	1.2.E-03	1.5.E-03	1.4.E-03	5.2.E-03	1.8.E-03	2.3.E-03	3.2.E-03
γ-HCH	5.E-04	5.5.E-07	4.5.E-07	1.2.E-06	7.4.E-07	5.6.E-07	4.1.E-07	1.2.E-06	7.5.E-07
p,p'-DDT	3.E-04	2.3.E-06	2.7.E-07	2.8.E-06	1.8.E-06	2.3.E-06	2.8.E-07	1.3.E-06	1.4.E-06
HIc		1.5.E-03	1.2.E-03	1.6.E-03	1.4.E-03	5.2.E-03	1.8.E-03	2.3.E-03	3.2.E-03

[a] CSfo: Oral cancer slope factor [36]
[b] RfDo: Oral reference dose [36]
[c] HI : Hazard Index = Sum of hazard quotients

to rocks. On the other hand, Manila clams dwell on the sediment surface. Because Manila clams bury their body in a sandy-mud bottom and filter-feed using a siphon [22], they have a greater chance of being exposed to the bottom sediment than do mussels.

To estimate the human health risk from the ingestion of Manila clams and mussels collected from this area, the dietary intake and resulting LCR and non-cancer risk were calculated. The LCRs from ΣPCBs, ΣDDTs, and HCHs posed by the consumption of Manila clams and mussels during the sampling period are reported in Table 4. During the sampling period, the highest LCR and HI for these OCs in Manila clams were observed in August. On the other hand, the highest LCR and HI for these OCs in mussels were observed in December. The LCR and HI of these OCs, except for β-HCH, induced by the annual consumption of Manila clams

were lower than those of mussels. Although there was a sharp fluctuation in the human health risk value, the risk levels did not exceed the acceptable risk (1 in 100,000), indicating that the consumption of these two bivalves from Cheonsu Bay seems to be safe in relation to human health, with negligible LCR and non-cancer risk.

4. Conclusions

We investigated the concentrations of PCBs and OCPs in sediments and bivalves (Manila clams and mussels) collected from Cheonsu Bay, Korea. PCBs and OCPs were detected in most samples, but their levels were relatively lower than those in the contaminated marine areas. Seasonal variations of these OC levels in the two species were observed. The lowest levels of these OCs were found in Manila clams in May and in mussels in June. The accumulation patterns of these chemicals in both bivalves were related to their habitats and behavior, such as spawning. The results of this study showed that the consumption of Manila clam and mussels collected in Cheonsu Bay seems to be safe in relation to human health according to the calculated LCR and HI.

Although this bio-monitoring study gave information on the behavior and fate of PCB and OCPs in the environment of Cheonsu Bay, the long-term monitoring would need to be continued in order to understand the recent temporal changes in this estuarine environment and the biological fate of these chemicals in Manila clams and mussels.

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