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Marine Pollution Bulletin 46 (2003) 281–300

MARINE POLLUTION BULLETIN

www.elsevier.com/locate/marpolbul

Review

Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries

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Accepted 15 October 2002

Abstract

Contamination of persistent organochlorines (OCs) such as PCBs (polychlorinated biphenyls), DDT and its metabolites (DDTs), HCH (hexachlorocyclohexane) isomers (HCHs), chlordane compounds (CHLs), and HCB (hexachlorobenzene) were examined in mussels collected from coastal waters of Asian countries such as Cambodia, China, Hong Kong, India, Indonesia, Japan, Korea, Malaysia, Philippines, Far East Russia, Singapore, and Vietnam in 1994, 1997, 1998, 1999, and 2001 to elucidate the contamination status, distribution and possible pollution sources and to assess the risks on aquatic organisms and human. OCs were detected in all mussels collected from all the sampling sites investigated. Considerable residue levels of p,p'-DDT and α-HCH were found in mussels and the concentrations of DDTs and HCHs found in mussels from Asian developing countries were higher than those in developed nations suggesting present usage of DDTs and HCHs along the coastal waters of Asian developing countries. On the other hand, lower concentrations of PCBs detected in mussels from Asian developing countries than those in developed countries indicate that PCBs contamination in mussels is strongly related to industrial and activities. To our knowledge, this is a first comprehensive report on monitoring OCs pollution in the Asia-Pacific region.

Keywords: Organochlorine compounds (OCs); Mussels; Asia-Pacific region; Developing countries; Developed nations

1. Introduction

Environmental pollution by toxic chemicals is a global problem, particularly organochlorine compounds (OCs) representing "persistent organic pollutants (POPs)" are of great concern due to their bioaccumulative nature and toxic biological effects on wildlife and humans (Tanabe et al., 2000). Elevated concentrations of OCs have been detected in a wide range of environmental media and aquatic biota (Iwata et al., 1993; Kannan et al., 1997; Tanabe, 2000; Tanabe et al., 2000). The undesirable effects of some of these chemicals are linked to the occurrence of immunologic and teratogenic

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0025-326X/03/$ - see front matter © 2002 Elsevier Science Ltd. All rights reserved.
doi:10.1016/S0025-326X(02)00400-9

dysfunction, reproductive impairments and endocrine disruption in lower and higher trophic levels (Colborn and Smolen, 1996).

Several studies have reported the contamination of OCs in the air (Iwata et al., 1993), foodstuff (Kannan et al., 1997) and marine mammals (Tanabe et al., 1994; Minh et al., 1999, 2000) from Asian coastal areas indicating the presence of major emission sources of OCs in Asian region. Even though most of the developed countries have banned or restricted the production and usage of many of these OCs during 1970s and 1980s, these chemicals are still being used in some developing nations for agricultural and public health purposes (Dave, 1996; Li, 1999; Tanabe et al., 1994). Some of these chemicals are still being manufactured in Asian developing region (FCI Editorial Board, 1996). Considering the above facts, the present study was conducted to assess the levels of toxic contaminants such as OCs in green mussels collected from Asian coastal waters.

Bivalves such as mussels have been suggested as a suitable bioindicator for monitoring trace toxic contaminant levels in coastal waters due to their wide distribution, sessile lifestyle, easy sampling, tolerance to a considerable range of salinity, resistance to stress and high accumulation of a wide range of chemicals (Goldberg et al., 1978). In particular, the green mussels (*Perna viridis*) are widely distributed in the Asian coastal waters, and recognized as a commercially valuable seafood in this zone. Mussels are highly suitable for culture in the coastal areas. Being filter feeders, they occupy a low position in the food chain, making their exploitation a very economic utilization of the primary production available in coastal waters. Moreover, mussels have a high protein content, averaging 67% of the body weight (Cheong, 1982; Tanabe et al., 2000), which is comparable to the other food items of higher trophic levels and this underscores its importance as a source of inexpensive animal protein. It is for these reasons that mussels are an important part of Asian diet. Monitoring studies on the residues of toxic contaminants in this seafood is rather limited (Tanabe et al., 2000).

The Asia-Pacific mussel watch program (APMW), started in 1994, is under the umbrella of the International Mussel Watch-Asia Pacific Phase, a project that mainly involves coastal monitoring using sentinel organisms such as mussels and oysters as bioindicators in ascertaining the quality of coastal waters in the Asia-Pacific region (Tanabe, 2000; Tanabe et al., 2000). The present study is a part of the APMW project to exhibit the current status of contamination by OCs in coastal waters of Asian countries.

This study aims to assess the levels of contamination of OCs and their distribution in coastal waters of Asian developing countries.

2. Materials and methods

2.1. Sample collection

Green mussel (*Perna viridis*), blue mussel (*Mytilus gallorovincialis*), and Ezo mussel (*Crenomytilus grayamus*) were collected from various locations in the Asian countries such as Cambodia, China, Hong Kong, India, Indonesia, Japan, Korea, Malaysia, Philippines, Far East Russia, Singapore and Vietnam in 1994, 1997, 1998, 1999 and 2001 respectively. Mussel samples were kept in polyethylene bags, kept in ice box with ice or dry ice, and kept in a deep freezer immediately after reaching the laboratory. In the laboratory, the frozen mussel samples were thawed and biometric measurement were made. After shucking the whole soft tissues of mussels from each location were pooled, homogenized, transferred into clean glass bottles and frozen at −20 °C until chemical analysis. The details of sampling locations and biological data of mussels are presented in Fig. 1 and Table 1, respectively.

2.2. Chemical analysis

Polychlorinated biphenyls (PCBs) and organochlrorine (OC) insecticides such as DDT and its metabolites (DDTs: p,p'-DDT, p,p'-DDD, and p,p'-DDE), chlordane compounds (CHLs: *trans*-chlordane, *cis*-chlordane, *cis*-nonachlor and oxychlordane), hexachlorohexane isomers (HCHs: α-HCH, β-HCH, and γ-HCH) and hexachlorobenzene (HCB) were analyzed following the method described by Tanabe et al. (2000) and Kanatireklap et al. (1997). Briefly, samples were homogenized with anhydrous Na_2SO_4 and extracted using a Soxhlet apparatus with a mixture of diethyl ether and hexane (3:1, 400 ml) for 7 h. After concentrating the extract, lipid content was determined gravimetrically from an aliquot of the extract. A portion of the extract was added to dry Florisil column to remove lipid and then eluted with the mixture of 120 ml acetonitrile and 30 ml hexane-washed water. OCs in the eluate were then transferred to hexane. After concentration, the hexane extract was cleaned with sulfuric acid and separated into two fractions using Florisil packed glass column. The first fraction eluted with hexane contained PCBs, HCB and p,p'-DDE. The second fraction eluted with 20% dichloromethane in hexane contained HCH isomers (α-, β-, and γ-HCH), o,p'-DDT, p,p'-DDD, p,p'-DDT and chlordane compounds (*trans*-chlordane, *cis*-nonachlor, and oxychlordane). Final extracts were concentrated, cleaned up with sulfuric acid and subjected to quantification by capillary gas chromatography with a ^{63}Ni electron capture detector (GC-ECD). Chromatographic separation was performed on a Hewlett-Packard 5890 Series II gas chromatograph with a 30 m × 0.25 mm (i.d.) DB-1 capillary column coated with 0.25 μm film

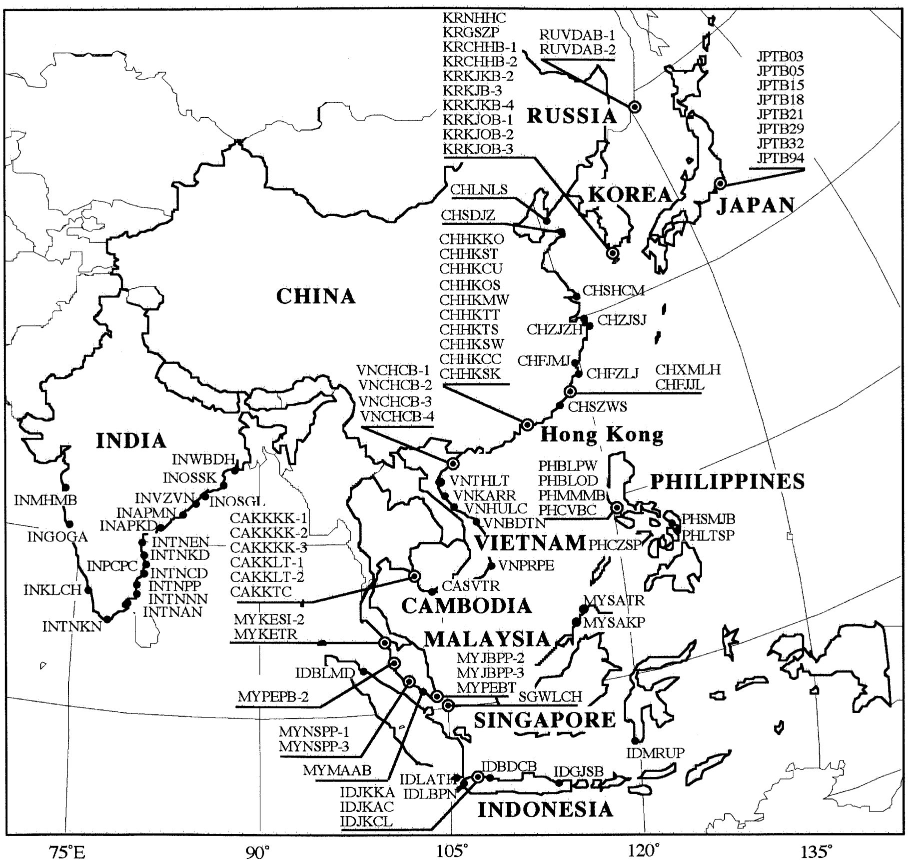


Fig. 1. Map showing sampling locations of mussels in coastal waters of Asia-Pacific countries.

thickness of 100% dimethyl-polysiloxane (J&W Scientific Co., Folsom, CA). PCBs were quantified by GC-ECD equipped with a fused silica capillary column (30 m length × 0.25 mm i.d., 0.25 μm film thickness) coated with SE-54 (Supelco, Inc., PA, USA).

The column oven temperature was programmed from 60 °C min^{-1} (hold 10 min) and increased from 160 to 260 °C at a rate of 2 °C min^{-1}, held for 15 min. Injector and detector temperatures were kept at 250 and 280 °C, respectively.

Total PCB concentrations in samples were quantified by summing up the concentrations of individually resolved peak areas relative to an equivalent mixture of Aroclor (1016:1242:1254:1260) with known PCB composition and content (Duinker et al., 1988). The concentrations of OC pesticides were quantified by comparing individually resolved peak areas with the corresponding peak areas of authentic standards.

Recoveries of OC pesticides and total PCBs through the analytical procedure were examined by spiking 40 ng of pesticide standard and 6.0 μg of PCB standard into corn oil. The results were 100 ± 12% for HCHs, 94 ± 5% for HCB, 103 ± 5% for CHLs, 100 ± 7% for DDTs and 102 ± 9% for PCBs. A procedural blank was run with every set of five samples to check for secondary contamination. Quality assurance for the measurement of OCs in our present technique was confirmed by analyzing Standard Reference Materials 1945 (Organics in Whale Blubber) provided by The National Institute of Standards and Technology (NIST), and the results agreed well with the NIST certified values. Concentrations of OCs were not corrected for the recoveries and are presented as nanograms per gram on a lipid weight basis.

3. Results and discussion

3.1. Status of contamination

PCBs and OC pesticides such as DDTs, HCHs, CHLs and HCB detected in all soft tissue homogenates of mussel samples from the coastal waters of Asian region are presented in Table 2. Contamination status of OCs in mussels varied depending on countries and the local sites of the sampling. Among the OCs analyzed in this study, DDTs (up to 61,000 ng/g lipid wt.) were the highest, and those of other OCs were in the order of PCBs > CHLs > HCHs > HCB (Table 2).

Table 1
Biological data of mussel samples collected from coastal waters of some Asian countries during 1994, 1997, 1998, 1999 and 2001

Location of sample	Code[a]	Species name (*Scientific name*)	Date	*n*[b]	SL (mm)[c]	STW(g)[d]	Site description
Cambodia							
Koh Kang-1, Koh Kong Province	CAKKKK-1	Green mussel (*Perna viridis*)	980721	46	71 (59–95)	4 (2–8)	Fishing village
Koh Kang-2, Koh Kong Province	CAKKKK-2	Green mussel (*Perna viridis*)	980721	52	68 (43–90)	4 (2–8)	Fishing village
Koh Kang-3, Koh Kong Province	CAKKKK-3	Green mussel (*Perna viridis*)	980721	59	66 (22–90)	3 (1–7)	Fishing village
Lo Tangao-1, Koh Kong Province	CAKKLT-1	Green mussel (*Perna viridis*)	980721	98	53 (35–75)	1 (0.4–3)	Fishing village
Lo Tangao-2, Koh Kong Province	CAKKLT-2	Green mussel (*Perna viridis*)	980721	75	58 (37–77)	2 (0.4–6)	Fishing village
Tachat, Koh Kong Province	CAKKTC	Green mussel (*Perna viridis*)	980721	75	58 (45–75)	1 (1–3)	Fishing village
Tomnup Rolork, Sihanouk Ville	CASVTR	Green mussel (*Perna viridis*)	980815	14	96 (54–96)	5 (2–27)	Internation port
China							
Xiamen Long Hai	CHXMLH	Green mussel (*Perna viridis*)	990515	24	(68–82)	(68–82)	Agriculture, small harbor
Shenzhen Westside	CHSZWS	Green mussel (*Perna viridis*)	990515	18	(78–91)	(78–91)	Agriculture, aquaculture
Fuzhou Lian Jian	CHFZLJ	Green mussel (*Perna viridis*)	990705	20	(62–79)	(62–79)	Harbor, industry, aquaculture, agriculture
Ling Shui Qiao, Liao Ning, Eastern China	CHLNLS	Blue mussel (*Mytilus edulis*)	010907	34	(42–64)	4 (1–7)	Aquaculture
Jiao Zhou Wan, Shan Dong, Eastern China	CHSDJZ	Blue mussel (*Mytilus edulis*)	010911	17	(51–70)	4 (1–6)	Industry, marine traffic, aquaculture, high population
Chong Ming Dao, Shang Hai, Eastern China	CHSHCM	Blue mussel (*Mytilus edulis*)	010925	33	(47–63)	3 (1–6)	Industry, ferry, aquaculture, high population
Shen Jia Men, Zhen Jian, South-eastern China	CHZJSJ	Blue mussel (*Mytilus edulis*)	011015	12	(53–82)	6 (4–9)	Ferry, aquaculture
Zhen Hai, Zhen Jian, South-eastern China	CHZJZH	Blue mussel (*Mytilus edulis*)	011002	8	(62–82)	5 (3–8)	Harbor, industry, aquaculture
Ming Jian, Fu Jian, South-eastern China	CHFJMJ	Green mussel (*Perna viridis*)	011004	20	(51–82)	4 (1–6)	Agriculture, aquaculture
Jiu Long Jian, Fu Jian, South-eastern China	CHFJJL	Green mussel (*Perna viridis*)	010925	9	(95–130)	8 (3–11)	Industry, ferry, aquaculture, high population
Hong Kong							
Kat O Chau	CHHKKO	Green mussel (*Perna viridis*)	981012	18	(80–96)	16 (11–18)	Agriculture
Sha Tau Kok	CHHKST	Green mussel (*Perna viridis*)	990315	20	(90–98)	17 (13–19)	Aquaculture, agriculture
CUMC	CHHKCU	Green mussel (*Perna viridis*)	981012	24	(82–95)	13 (11–16)	Small harbor, industry
Ma On Shan	CHHKOS	Green mussel (*Perna viridis*)	990315	25	(60–74)	7 (5–10)	Small harbor, industry
Ma Wan	CHHKMW	Green mussel (*Perna viridis*)	990315	21	(92–101)	17 (14–20)	Harbor, aquaculture
Tai Tau Chau	CHHKTT	Green mussel (*Perna viridis*)	990629	20	(90–114)	17 (13–22)	Aquaculture, agriculture
Tsim Sha Tsui	CHHKTS	Green mussel (*Perna viridis*)	990623	26	(60–78)	8 (5–11)	Ferry, shipping traffic, high populations
Shai Wan Ho	CHHKSW	Green mussel (*Perna viridis*)	980720	20	(80–94)	13 (11–15)	Harbor
Chaung Chau	CHHKCC	Green mussel (*Perna viridis*)	990625	21	(84–96)	14 (12–16)	Aquaculture
Sok Kwu Wan	CHHKSK	Green mussel (*Perna viridis*)	990623	25	(72–88)	10 (9–13)	Power station, aquaculture
India							
Bombay, Maharashtra	INMHMB	Green mussel (*Perna viridis*)	980319	31	73 (62–88)	(5–12)	Urban, harbor
Goa, Goa	INGOGA	Green mussel (*Perna viridis*)	980319	49	71 (53–86)	(4–12)	Urban, small harbor
Cochin, Kerala	INKRCH	Green mussel (*Perna viridis*)	980305	27	87 (72–125)	(8–24)	Harbor, aquaculture
Kanniya Kumari, Murram	INMRKN	Green mussel (*Perna viridis*)	980302	38	97 (87–105)	(10–16)	Fishing harbor
Akkaraipattinam, Tamil Nadu	INTNAN	Green mussel (*Perna viridis*)	980221	22	82 (37–105)	(1–16)	Fishing harbor
Nakgapattinam, Tamil Nadu	INTNNN	Green mussel (*Perna viridis*)	980202	16	91 (78–103)	(6–16)	Aquaculture
Porto Novo, Tamil Nadu	INTNPN	Green mussel (*Perna viridis*)	980304	26	69 (46–98)	8 (3–17)	Aquaculture
Cuddalore, Tamil Nadu	INTNCD	Green mussel (*Perna viridis*)	980121	29	102 (83–127)	(9–21)	Aquaculture

Pondicherry, Pondicherry	INPCPC	Green mussel (*Perna viridis*)	980121	46	74 (46–129)	(3–36)	Industry, small harbor
Kasimedu, Madras	INMDKD	Green mussel (*Perna viridis*)	980303	37	57 (36–69)	5 (3–9)	Fishing harbor
Ennore, Madras	INMDEN	Green mussel (*Perna viridis*)	980303	40	77 (55–102)	9 (3–16)	Industry
Machilipatinam, Anddhra Pradesh	INAPMN	Green mussel (*Perna viridis*)	980222	19	107 (96–124)	18 (13–24)	Fishing harbor
Kakinada, Andra Pradesh	INAPKD	Green mussel (*Perna viridis*)	980223	14	133 (108–144)	30 (18–63)	Aquaculture
Visakapatnam, Vizag	INVZVN	Green mussel (*Perna viridis*)	980224	10	132 (113–146)	35 (17–73)	Harbor
Gopalpular, Gopalbular	INGLGL	Green mussel (*Perna viridis*)	980220	24	(90–107)	13 (9–22)	Fishing harbor
Subarnarekha, Orissa	INOSSK	Green mussel (*Perna viridis*)	980112	15	111 (88–129)	30 (50–70)	Fishing harbor
Digha, West Bengal	INWBDH	Green mussel (*Perna viridis*)	980117	15	81 (64–104)	18 (6–29)	Fishing harbor
Indonesia							
Cilincing, Jakarta	IDJKCL	Green mussel (*Perna viridis*)	980728	49	60 (50–70)	5 (4–9)	Fishing port, industry
Ancol, Jakarta	IDJKAC	Green mussel (*Perna viridis*)	980728	51	45 (35–65)	3 (2–6)	Port, industry
Kamal, Jakarta	IDJKKA	Green mussel (*Perna viridis*)	980729	54	78 (60–94)	8 (4–13)	Fishing port, industry
T. Hurun Lampung	IDLATH	Green mussel (*Perna viridis*)	980723	40	83 (70–102)	7 (4–13)	Aquaculture
Maros, Ujung Pandang	IDMRUP	Green mussel (*Perna viridis*)	980716	24	105 (95–123)	20 (13–31)	Fishing landing
Lada Bay, Panimbang	IDLBPN	Green mussel (*Perna viridis*)	980701	56	57 (44–75)	4 (2–7)	Aquaculture
Belawan, Medan	IDBLMD	Green mussel (*Perna viridis*)	980712	48	90 (66–120)	11 (3–19)	Port
Genjeran, Surabaya	IDGJSB	Green mussel (*Perna viridis*)	980720	50	74 (48–91)	8 (3–13)	Port, industry
Bondet, Cirebon	IDBDCB	Green mussel (*Perna viridis*)	980803	51	81 (71–94)	9 (5–14)	Fishing
Japan							
Tokyo Bay	JPTB03	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPTB 05	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPTB15	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	PTB18	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPBT21	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPTB29	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPTB32	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Tokyo Bay	JPTB94	Blue mussel (*Mytilus gallorovincialis*)	1994	NA	NA	NA	
Korea							
Hangchon, Namhae	KRNHHC	Blue mussel (*Mytilus edulis*)	980513	48	46 (41–54)	2 (1–5)	Aquaculture site
Zyipo, Gosung	KRGSZP	Blue mussel (*Mytilus edulis*)	980218	31	49 (41–54)	3 (1–5)	Oyster farming
Haengam Bay, Chinhai	KRCHHB-1	Blue mussel (*Mytilus edulis*)	981104	20	43 (40–49)	2 (1–4)	Small harbor
Haengam Bay, Chinhai	KRCHHB-2	Blue mussel (*Mytilus edulis*)	981104	28	44 (40–49)	3 (2–5)	Island
Kohyonsong Bay, Kuje	KRKJKB-2	Blue mussel (*Mytilus edulis*)	980827	24	51 (47–55)	1 (2–7)	Shipyard
Kohyonsong Bay, Kuje	KRKJKB-3	Blue mussel (*Mytilus edulis*)	980827	38	49 (41–55)	4 (2–5)	Infront of shipyard
Kohyonsong Bay, Kuje	KRKJKB-4	Blue mussel (*Mytilus edulis*)	980513	23	50 (40–55)	4 (2–5)	Shipyard
Okpo Bay, Kuje	KRKJOB-1	Blue mussel (*Mytilus edulis*)	981027	42	46 (41–52)	5 (2–6)	Repairing shipyard
Okpo Bay, Kuje	KRKJOB-2	Blue mussel (*Mytilus edulis*)	981027	38	42 (40–50)	3 (2–7)	Infront of repairing shipyard
Okpo Bay, Kuje	KRKJOB-3	Blue mussel (*Mytilus edulis*)	981027	33	46 (41–51)	4 (2–6)	Infront of repairing shipyard
Malaysia							
Kuala Penyu, Sabah	MYSAKP	Green mussel (*Perna viridis*)	980829	25	80 (74–86)	6 (3–6)	Agriculture & aquaculture

Table 1 (*continued*)

Location of sample	Code[a]	Species name (*Scientific name*)	Date	*n*[b]	SL (mm)[c]	STW(g)[d]	Site description
Trayong, Sabah	MYSATR	Green mussel (*Perna viridis*)	980829	25	70 (64–74)	3 (1–4)	Agriculture & aquaculture
Sangkar Ikan, Lengkawi	MYKESI-2	Green mussel (*Perna viridis*)	980920	25	78 (52–90)	5 (3–6)	Urban, agriculture
Tanjung Rhu, Lengkawi	MYKETR	Green mussel (*Perna viridis*)	980920	25	71 (67–75)	5 (3–6)	Recreational beach, aquaculture
Penang Bridge, Panang	MYPEPB-2	Green mussel (*Perna viridis*)	980921	25	71 (66–74)	5 (3–8)	Industry, urban
Bagan Lalang, Selangor	MYSEBL	Green mussel (*Perna viridis*)	980608	25	87 (74–97)	6 (3–7)	Recreational beach, aquaculture
Lukut, Negeri Sembilan	MYNSLU	Green mussel (*Perna viridis*)	980808	25	86 (71–98)	9 (8–13)	Port, aquaculture
Pasir Panjang, Negeri Sembilan	MYNSPP-1	Green mussel (*Perna viridis*)	980819	25	91 (78–104)	10 (7–12)	Recreational beach, aquaculture
Pasir Panjang, Negeri Sembilan	MYNSPP-3	Green mussel (*Perna viridis*)	980922	25	89 (68–101)	8 (3–10)	Recreational beach, aquaculture
Tanjung Batu, Malacca	MYMAAB	Green mussel (*Perna viridis*)	980922	25	92 (84–101)	8 (5–11)	Agriculture & aquaculture
Pasir Puteh-2, Johor Bahru	MYJBPP-2	Green mussel (*Perna viridis*)	980923	25	73 (68–88)	6 (3–7)	Port, industry, urban
Pasir Puteh-2, Johor Bahru	MYJBPP-3	Green mussel (*Perna viridis*)	980530	25	64 (42–100)	6 (4–7)	Port, industry, urban
Butterworth, Penang	MYPEBT	Green mussel (*Perna viridis*)	990511	26	71[e]	NA	Industriay, urban
Philippines							
Bacoor, Cavite	PHCVBC	Green mussel (*Perna viridis*)	980327	43	69 (60–77)	7 (5–19)	
Pamarawan, Bulacan	PHBLPW	Green mussel (*Perna viridis*)	980326	15	81 (74–93)	8 (6–11)	
Obando, Bulacan	PHBLOD	Green mussel (*Perna viridis*)	980317	82	58 (17–89)	4 (2–10)	
San Pedro Bay, Leyte	PHLTSP	Green mussel (*Perna viridis*)	980321	15	104 (89–117)	17 (13–27)	Aquaculture site
Jiabong, Samar	PHSMJB	Green mussel (*Perna viridis*)	980322	51	66 (51–84)	4 (2–8)	Aquaculture site
Malabon, Metro Manila	PHMMMB	Green mussel (*Perna viridis*)	980316	87	63 (41–87)	6 (2–14)	
Sapia Bay, Capiz	PHCZSP	Green mussel (*Perna viridis*)	980408	87	64 (43–86)	4 (2–10)	
Russia							
Vladivostok, Amursky Bay	RUVDAB-1	Ezo mussel (*Crenomytilus grayamus*)	990719	10	79 (73–83)	15 (10–20)	
Vladivostok, Amursky Bay	RUVDAB-2	Ezo mussel (*Crenomytilus grayamus*)	990719	8	67 (61–72)	9 (6–12)	
Singapore							
Changi Airport, Woodland City	SGWLCH	Green mussel (*Perna viridis*)	990601	27	68[e]	NA	
Vietnam							
Cat Ba (Cat Hai Province)	VNCHCB-1	Green mussel (*Perna viridis*)	970929	38	13 (8–16)	26 (13–53)	Floating habitats
Cat Hai Province	VNCHCB-2	Green mussel (*Perna viridis*)	971027	34	9 (5–13)	12 (3–39)	Floating habitats
Cat Hai Province	VNCHCB-3	Green mussel (*Perna viridis*)	971027	8	12 (10–13)	23 (15–31)	Floating habitats
Cat Hai Province	VNCHCB-4	Green mussel (*Perna viridis*)	971026	12	5.42 (4–8)	3 (1–6)	Floating habitats
Lach Truong (Thanh Hoa Province)	VNTHLT	Green mussel (*Perna viridis*)	971025	33	12 (8–16)	21 (19–53)	Fishing, aquaculture
Ron River estuary, Ky Anh Province	VNKARR	Green mussel (*Perna viridis*)	971018	50	10 (8–13)	22 (9–119)	Fishing village
Lang Co (Hue City)	VNHULC	Green mussel (*Perna viridis*)	971912	143	7 (6–86)	4 (1–8)	Remote area
Thi Nai (Binh Dinh Province)	VNBDTN	Green mussel (*Perna viridis*)	971005	54	7 (6–9)	7 (3–15)	Urban, shipping traffic, aquaculture
Phan Ri estuary, Phan Ri	VNPRPE	Green mussel (*Perna viridis*)	970925	30	7 (5–11)	9 (2–25)	Urban, fishing village

NA: No data available.

[a] First, second and last two letters indicate the country, city/province or state and local name. The digits indicate the replicate time of sampling, excepting Tokyo Bay which indicates the sampling locations.

[b] Number of individuals homogenized.

[c] Shell length.

[d] Soft tissue weight.

[e] Average.

Table 2
Concentrations of organochlorines (ng/g lipid wt.) in mussel samples from Asian countries

Locations	Lipid (%)	PCBs	DDTs	CHLs	HCHs	HCB
Cambodia						
CAKKKK-1	1.2	<4.2	22	<0.80	<0.80	1.6
CAKKKK-2	1.2	<4.3	17	<0.70	<0.70	2.6
CAKKKK-3	1.1	<4.3	16	<0.90	<0.90	1.9
CAKKLT-1	0.90	<5.7	26	<1.1	<1.1	<1.1
CAKKLT-2	1.3	<3.8	16	<0.70	<0.70	1.4
CAKKTC	1.0	<5.1	22	<1.0	<1.0	<1.0
CAKSVTR	2.3	220	48	<0.40	<0.40	<0.40
China						
CHXMLH	0.56	55	23,000	360	110	<1.8
CHSZWS	1.1	25	30,000	130	30	<0.90
CHFZLJ	2.4	20	2400	40	21	2.0
CHLNLS	1.7	120	830	10	48	5.1
CHSDJZ	2.4	540	6200	130	77	540
CHSHCM	2.2	600	29,000	170	79	5.3
CHZJSJ	2.9	48	8600	120	41	5.5
CHZJZH	3.5	41	2200	38	16	2.2
CHFJMJ	1.9	69	7800	54	11	<0.50
CHFJJL	1.2	270	54,000	860	12	<0.70
Hong Kong						
CHHKKO	0.69	220	1400	18	12	<1.5
CHHKST	0.88	50	1500	460	12	<1.1
CHHKCU	2.2	180	700	31	29	<0.50
CHHKOS	1.1	200	640	130	19	<0.90
CHHKMW	1.7	370	1700	66	16	<0.60
CHHKTT	0.74	40	1100	<1.4	2.1	<1.4
CHHKTS	0.94	710	2800	750	27	<1.1
CHHKSW	0.68	500	4200	130	24	<1.5
CHHKCC	1.7	440	61,000	330	20	<0.60
CHHKSK	1.5	360	2400	500	18	<0.70
India						
INMHMB	0.50	600	3000	33	210	60
INGOGA	1.3	63	230	4.6	67	<0.80
INKRCH	1.8	420	770	NA	180	<0.60
INMRKN	1.1	150	160	37	20	1.4
INTNAN	1.6	100	130	11	87	<0.60
INTNNN	1.9	150	120	23	110	<0.50
INTNPN	1.8	140	71	8.0	430	<0.60
INTNCD	1.4	230	130	<0.70	100	1.5
INPCPC	2.0	2200	450	NA	58	<0.50
INMDKD	2.2	370	230	42	19	<0.50
INMDEN	2.4	370	78	40	22	<0.40
INAPMN	1.5	120	58	5.5	130	<0.70
INAPKD	2.3	83	50	11	120	<0.40
INVZVN	2.1	520	29	<0.5	100	<0.50
INGLGL	1.6	10	50	<0.60	120	<0.60
INOSSK	0.90	90	520	150	110	<1.1
INWBDH	2.1	240	280	160	140	<0.50
Indonesia						
IDJKCL	1.7	140	58	7.4	4.7	<0.60
IDJKAC	1.9	96	48	7.5	4.9	<0.50
IDJKKA	1.3	210	45	13	<0.80	<0.80
IDLATH	1.1	14	65	15	4.1	<0.90
IDMRUP	1.8	5.6	6.5	<0.60	<0.60	0.8
IDLBPN	1.1	85	110	<0.90	2.4	1.1
IDBLMD	1.4	13	15	5.3	5.3	0.8
IDGJSB	1.2	190	120	<0.80	<0.80	1.2
IDBDCB	2.0	30	160	16	2.1	1.5

(*continued on next page*)

Table 2 (*continued*)

Locations	Lipid (%)	PCBs	DDTs	CHLs	HCHs	HCB
Japan						
JPTB03	1.8	890	140	190	17	3.2
JPTB05	0.65	12,000	NA	1800	40	29
JPTB15	1.5	810	160	270	39	3.9
JPTB18	1.6	510	100	220	13	<0.60
JPBT21	1.2	620	70	150	18	<0.80
JPTB29	0.85	2600	500	440	50	6.6
JPTB32	1.1	920	110	210	32	5.7
JPTB94	1.5	5500	790	1100	17	17
Korea						
KRNHHC	1.1	70	81	40	80	<0.90
KRGSZP	5.0	30	14	3.7	1.9	<0.20
KRCHHB-1	2.1	340	350	30	6.8	5.4
KRCHHB-2	3.3	150	200	36	15	3.2
KRKJKB-2	2.1	80	150	22	5.5	<0.50
KRKJKB-3	2.0	90	310	18	10	1.8
KRKJKB-4	2.4	150	170	25	8.5	7.3
KRKJOB-1	2.0	210	60	10	3.9	3.2
KRKJOB-2	2.3	260	100	35	7.2	<0.40
KRKJOB-3	2.0	290	90	28	6.1	<0.50
Malaysia						
MYSAKP	0.73	7.5	100	8.7	<1.4	<1.4
MYSATR	0.65	8.3	32	4.1	3.1	<1.5
MYKESI-2	0.92	6.0	95	41	9.4	<1.1
MYKETR	1.1	5.1	16	2.5	4.9	<0.90
MYPEPB-2	1.0	60	71	180	<0.10	2.4
MYSEBL	1.2	<4.2	26	24	3.3	<0.80
MYSLU	1.5	54	53	50	1.0	2.2
MYNSPP-1	1.8	24	71	41	4.8	3.3
MYNSPP-3	1.6	11	93	60	3.5	<0.60
MYAAB	1.3	22	100	610	12	<0.80
MYJPP-2	2.1	250	130	470	5.2	<0.50
MYJPP-3	2.1	230	270	170	<0.50	<0.50
MYPPR99M	1.2	42	110	220	<0.80	<0.80
Philippines						
PHCVBC	2.2	550	30	130	2.1	<0.40
PHBLPW	1.3	290	22	50	4.0	<0.80
PHBLOD	1.9	320	19	57	2.6	<0.50
PHLTSP	1.9	170	12	5.1	1.1	<0.50
PHSMJB	1.6	51	24	6.3	1.1	<0.60
PHMMMB	2.2	640	38	120	0.90	<0.50
PHCZSP	1.8	22	3.3	6.6	0.60	<0.60
Russia						
RUVDAB-1	1.9	3700	730	28	57	7.4
RUVDAB-2	2.1	2700	520	29	34	1.5
Singapore						
SGWL	2.7	90	110	520	12	<0.40
Vietnam						
VNCHCB-1	1.1	86	530	14	3.6	<0.90
VNCHCB-2	0.90	20	300	12	12	<1.1
VNCHCB-3	0.70	450	2500	24	5.7	<1.4
VNCHCB-4	2.0	110	420	5.0	3.0	<0.50
VNTHLT	1.2	65	610	13	3.3	<0.80
VNKARR	0.60	190	470	20	5.5	<1.6
VNHHLC	0.90	380	34,000	NA	10	3.5
VNBDTN	1.1	26	220	36	6.3	2.3
VNPRPE	1.1	80	240	11	2.9	<0.90

NA: No data available.
DDTs: p,p'-DDE + p,p'-DDD + p,p'-DDT.
CHLs: *trans*-chlordane + *cis*-chlordane + *trans*-nonachlor + *cis*-nonachlor + oxychlordane.
HCHs: α-HCH + β-HCH + γ-HCH.

3.2. Global distribution

3.2.1. DDTs

The distribution and concentrations of DDTs in mussels collected from Asian coastal waters are shown in Table 2 and Fig. 2. The high residue levels of DDTs were found in mussels from China (830–54,000 ng/g lipid wt.), Hong Kong (640–61,000 ng/g lipid wt.) and Vietnam (220–34,000 ng/g lipid wt.) (Table 3). Elevated concentrations of DDTs have also been reported in mussels from China (Klumpp et al., 2002; Hong et al., 2000) and Hong Kong (Phillips, 1985, 1989). Several investigators found high concentrations of DDTs in river sediments from China (Hong et al., 1995, 1999; Wu et al., 1999). Wu et al. (1999) noted high concentrations of DDTs in the river sediments from northern China where a factory with high manufacturing capacity of DDT is located. Large amounts of technical DDT have been used (more than 10^5 tonnes) in China recently (Nhan et al., 1999). The present results clearly indicates the release of DDT at present to the environment in China, even though usage of DDTs have been officially banned in 1983. In Vietnam, higher levels of DDTs were also reported in fish (Kannan et al., 1995) and birds (Minh et al., 2002; Kunisue et al., in press). Over 20,000 tonnes of pesticides are currently used annually in Vietnam (Nhan et al., 1998). Our results indicate the recent usage of DDTs in Vietnam. In general, lower DDTs levels were observed in mussels from Phillipines, Cambodia, Indonesia and Malaysia (Fig. 2), which indicates less usage of DDTs in these countries.

The composition of DDT compounds in mussels from Asia-Pacific region are shown in Fig. 3. Mussels from China, Hong Kong, Vietnam and Far East Russia showed higher proportion of p,p'-DDT residues than p,p'-DDE. Sediments from Chinese coastal waters were

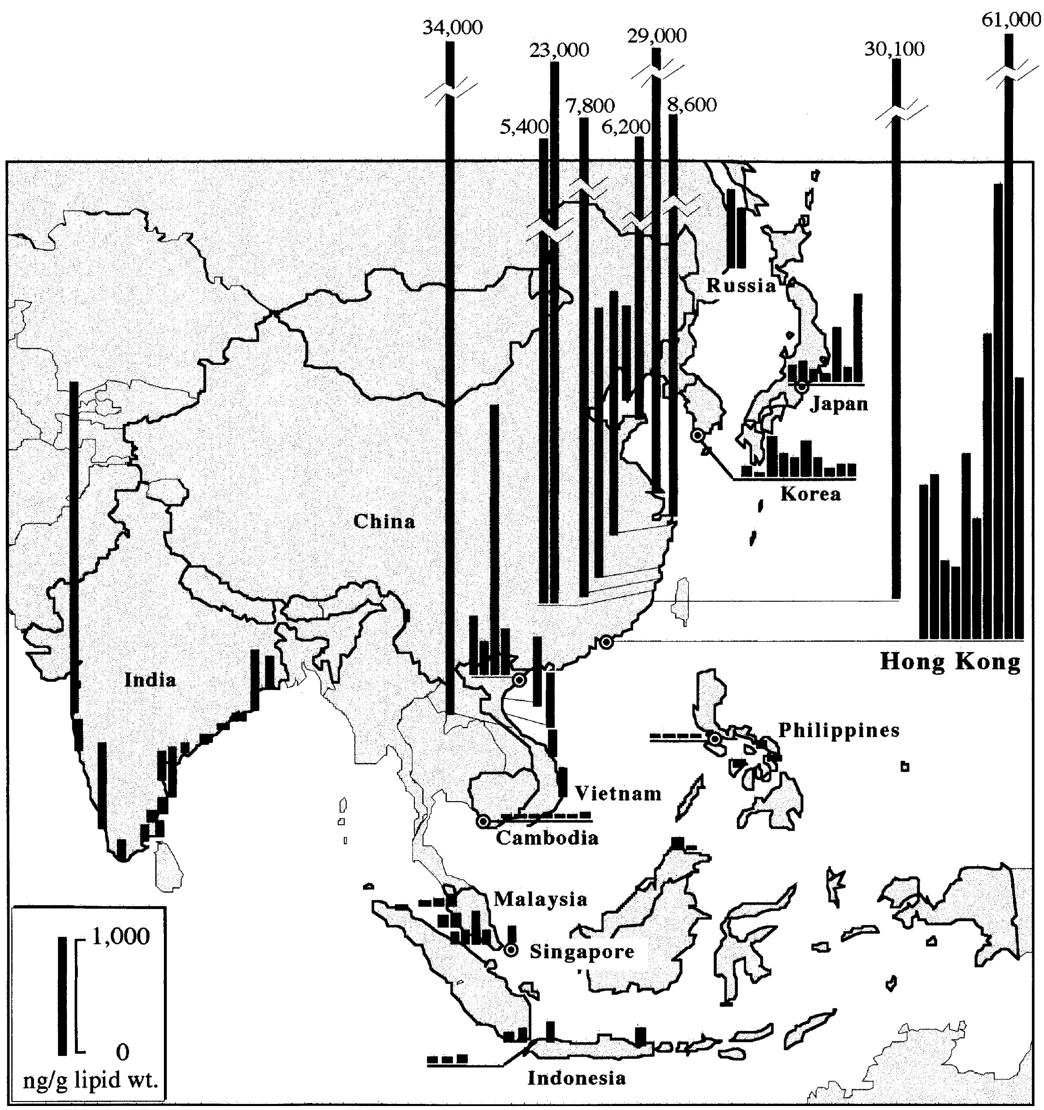


Fig. 2. Distribution of concentrations of DDTs in mussels collected from coastal waters of some Asian countries.

Table 3
Mean and range (in parentheses) of OC concentrations (ng/g) in mussels from Asian countries

Country (Location)	Fat (%)	PCBs		DDTs		HCHs		CHLs		HCB	
		Lipid wt.	Wet wt.	Lipid wt.	Wet wt.	Lipid wt.	Wet wt.	Lipid wt.	Wet wt.	Lipid wt.	Wet wt.
Cambodia	1.3	35	0.74	23	0.33	<0.30	<0.01	<0.30	<0.01	1.2	0.01
	(0.90–3.3)	(<3.8–220)	(<0.50–5.1)	(16–48)	(0.20–1.0)	<1.1		<1.1		(<0.40–2.6)	(<0.01–0.03)
China	2.0	120	2.5	16,000	240	44	0.80	190	3.0	56	1.3
	(0.60–3.5)	(15–540)	(0.30–13)	(830–54,000)	(15–640)	(11–110)	(0.10–2.0)	(10–860)	(0.20–10)	(<0.70–540)	(<0.01–13)
Hong Kong	1.2	310	3.7	7700	120	18	0.20	240	2.0	<1.5	<0.01
	(0.70–2.2)	(40–710)	(0.30–7.4)	(640–61,000)	(7.0–1000)	(2.1–30)	(0.02–0.60)	(<1.4–750)	(<0.01–4.1)		
India	1.7	340	3.8	380	4.2	120	2.0	35	0.60	4.0	0.02
	(0.50–2.4)	(9.8–600)	(0.20–11)	(29–3000)	(0.60–15)	(20–430)	(0.20–7.7)	(<0.50–160)	(<0.01–3.3)	(<0.40–63)	(<0.01–0.30)
Indonesia	1.5	87	1.3	70	1.0	3.0	0.04	7.3	0.10	0.70	0.01
	(1.1–2.0)	(5.6–210)	(0.10–2.7)	(6.5–160)	(0.10–3.1)	(<0.60–5.3)	(<0.01–0.10)	(<0.60–16)	(<0.01–0.30)	(<0.50–1.5)	(<0.01–0.03)
Japan	1.3	3000	30	270	3.5	28	0.32	550	6.0	8.2	0.08
	(0.60–1.8)	(510–12,000)	(7.4–84)	(70–790)	(0.80–12)	(13–50)	(0.20–0.60)	(150–1800)	(1.7–17)	(<0.60–29)	(<0.01–0.30)
South Korea	2.4	170	3.7	150	3.5	14	0.26	25	0.55	2.3	0.05
	(1.1–5.0)	(30–340)	(0.80–7.2)	(14–350)	(0.70–7.5)	(1.9–80)	(0.10–1.0)	(3.7–40)	(0.20–1.2)	(<0.40–7.3)	(<0.01–0.20)
Malaysia	1.3	56	1.0	90	1.4	3.7	<0.05	140	2.2	0.80	0.02
	(0.70–2.1)	(<4.2–250)	(<0.05–5.1)	(16–270)	(0.20–5.7)	(<0.80–12)	(<0.01–0.20)	(2.5–610)	(0.03–9.6)	(<0.50–3.3)	(<0.01–0.60)
Philippines	1.8	290	5.7	21	0.4	1.8	0.03	54	1.1	<0.80	<0.01
	(1.3–2.2)	(22–640)	(0.40–14)	(3.3–38)	(0.06–0.8)	(0.60–4.0)	(<0.01–0.05)	(5.1–130)	(0.10–2.8)		
Russia	2.0	3200	63	630	12	45	1.0	56	1.1	4.5	0.09
	(1.9–2.1)	(2700–3700)	(56–70)	(520–730)	(11–14)	(34–57)	(0.70–1.1)	(28–29)	(0.50–0.60)	(1.5–7.4)	(0.03–0.14)
Singapore	2.7	90	2.4	110	3.0	12	0.30	28	0.50	<0.40	<0.01
Vietnam	1.1	160	1.4	4400	40	5.8	0.06	17	0.33	1.0	0.01
	(0.6–2.0)	(21–450)	(0.20–3.4)	(220–34,000)	(2.4–310)	(3.0–12)	(0.03–0.10)	(5.0–36)	(0.1–1.4)	(<0.50–3.5)	(<0.01–0.03)

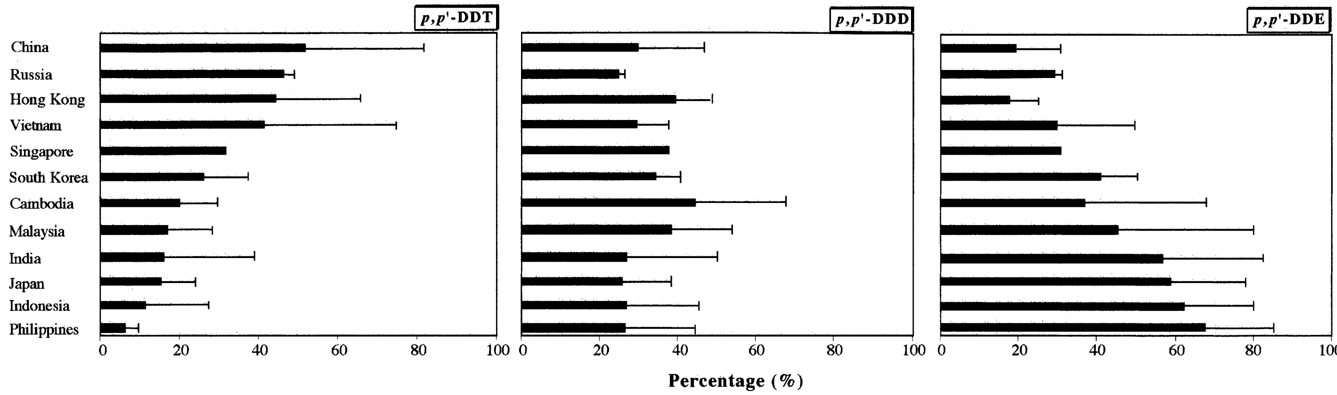


Fig. 3. Composition of DDT compounds in mussels collected from coastal waters of some Asian countries.

reported to contain very high ratio of p,p'-DDT (up to 80%) (Hong et al., 1995 and Wu et al., 1999). Sediments, clams and fish from Hong Kong, China and Vietnam (Kannan et al., 1995; McConnell et al., 1996; Nhan et al., 1999, 2000; Tanabe, 2000; Ueno et al., in press) also showed high proportion of p,p'-DDT. Considerably higher ratio of p,p'-DDT in total DDTs may again indicate the presence of current emission sources of DDTs in China, Hong Kong and Vietnam. The residue levels of DDTs in mussels from Russia were low, but the percentage of p,p'-DDT was high up to 50%. High proportion of DDT was also found in air, water, sediments and seals from Russia (Iwata et al., 1995; McConnell et al., 1996; Nakata et al., 1995). Lower concentrations of DDTs and higher ratio of p,p'-DDT in Russian mussels denote the recent usage of technical DDT. The compositions and residue levels of DDTs in mussels in this study indicate current usage of DDT for agriculture and public health purposes in some Asian developing countries.

3.2.2. HCHs

Concentrations of HCHs in mussels are shown in Table 2 and Fig. 4. Extremely higher levels of HCHs were observed in mussels from India (20–430 ng/g lipid wt.). Relatively high concentrations were also found in China (2.1–110 ng/g lipid wt.) and Russia (34–57 ng/g lipid wt.) (Table 3). The concentrations of HCHs in India in this study were comparable to the values detected in mussels from the same areas collected in our previous study (140–590 ng/g lipid wt.) (Kan-atireklap et al., 1998). India has been the largest user of technical HCH in the world (Li et al., 1998). The usage of technical HCH have been banned in India for agriculture such as vegetable, fruit and oil seed crops, and for preservation of grains since 1983, but the usage was allowed for public health purposes and on certain food crops (Li et al., 1998). Higher concentrations of HCHs were also reported in sediments from agriculture areas of China (Wu et al., 1999). China was also one of the largest producers and consumers of technical HCH in the world (Li, 1999). HCHs were also detected in air, water, sediments, soil (Iwata et al., 1995), Baikal seal (Nakata et al., 1995) and birds (Kunisue et al., 2002) from Lake Baikal, Russia. In the former Soviet Union, it was permitted to use residual stocks of pesticides (Zhulidov et al., 2000). It is quite possible that the active usage of these stockpiled amounts of HCH for agriculture, forestry and municipal purposes could have contributed to HCH contamination in mussels.

Compositions of HCH isomers in mussels varied depending on countries and the sampling sites (Fig. 5). Among HCH isomers, α-HCH was predominant (up to 45%) in mussel samples from India, Russia and China. Almost the same ratios of α-HCH were detected in mussels and fish from India and China (Kannan et al., 1995; Kan-atireklap et al., 1998; Ueno et al., in press). The predominant ratio of α-HCH may reflect the recent usage of technical HCH mixture (55–70%α-, 5–14%β-, 10–18%γ-, and 6–10%δ-isomer) in India, Russia and China.

Mussels from Singapore and Malaysia contained higher percentages of γ-HCH (up to 90%) (Fig. 5). Previous reports indicated the use of lindane (purified γ-HCH) in Malaysia (Tan and Vijayaletchumy, 1994; Kannan et al., 1995), supporting the result of the present study.

Higher concentrations of HCHs in mussels from India, China and Russia might be partly caused by continued usage of this chemical in these countries.

3.2.3. PCBs

The concentrations of PCBs in mussels collected from Asian coastal waters are shown in Table 2 and Fig. 6. Higher concentrations of PCBs were found in mussels from Japan (up to 12,000 ng/g lipid wt.) and Russia (up to 3700 ng/g lipid wt.) (Table 3). Higher concentrations of PCBs were also reported in marine mammals in the waters around Japan (Prudente et al., 1997), squids (Yamada et al., 1997) and skipjack tuna (Ueno et al., in press). These results suggest that emission sources of PCBs still exist in Japan. Even though production of

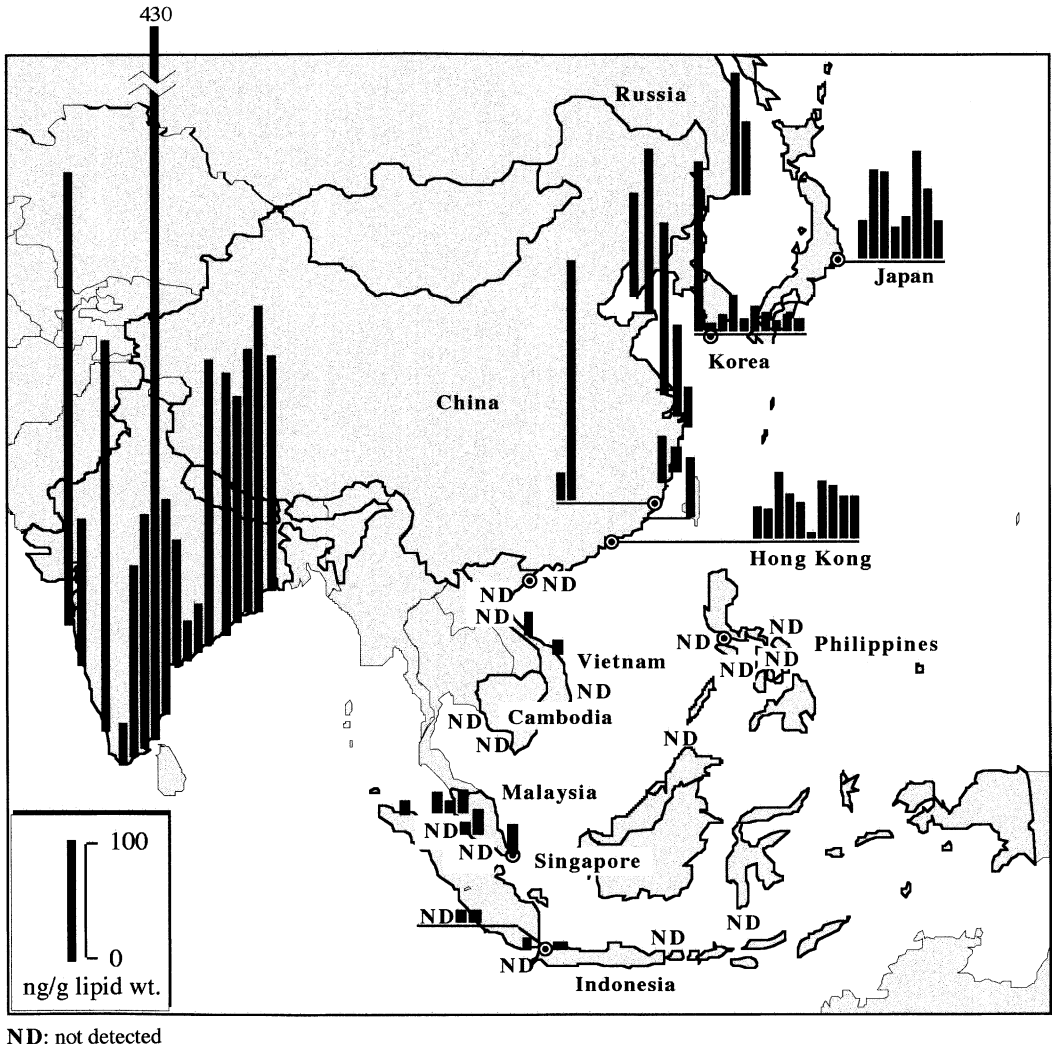


Fig. 4. Distribution of concentrations of HCHs in mussels collected from coastal waters of some Asian countries.

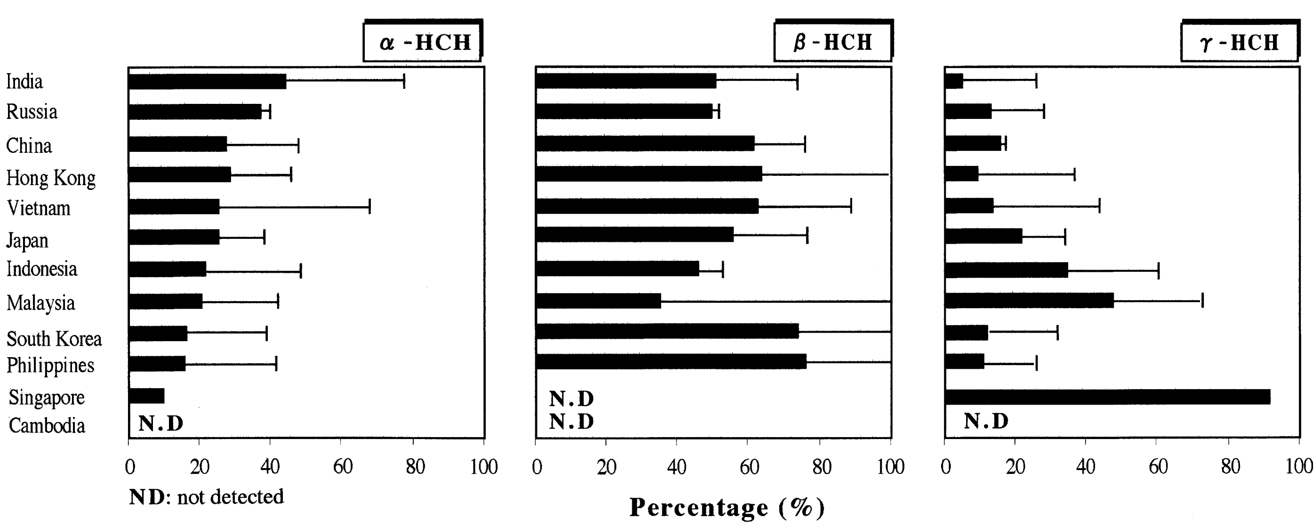
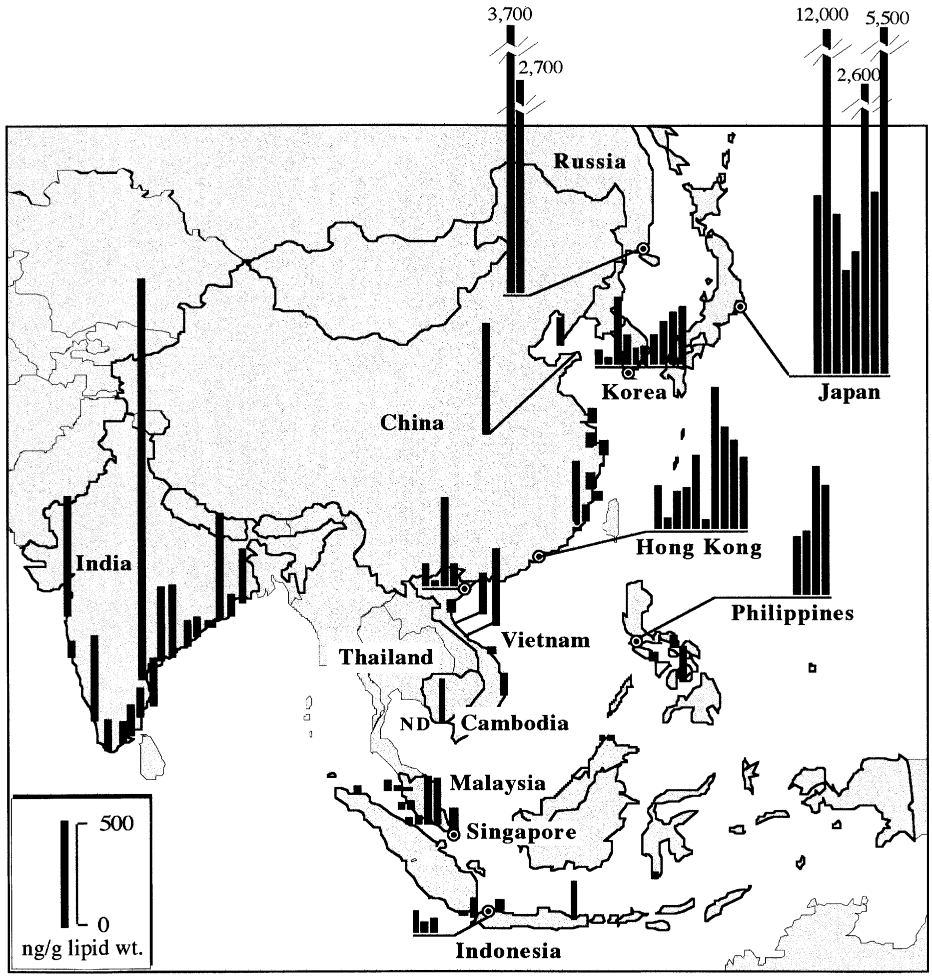


Fig. 5. Composition of HCH isomers in mussels collected from coastal waters of some Asian countries.

PCBs was prohibited in Japan since 1972, Japanese coastal waters have been contaminated by PCBs mainly due to continuous release from old transformers and capacitors still in use or in storage (Loganathan et al.,



ND: not detected

Fig. 6. Distribution of concentrations of PCBs in mussels collected from coastal waters of some Asian countries.

1993; Takahashi et al., 1998, 2000; Ueno et al., in press). PCBs were also detected in air, water, sediments, soil, fish, seals and birds from Russia (Kuklick et al., 1994; Iwata et al., 1995; Nakata et al., 1995; Kunisue et al., 2002). In the former USSR, technical PCBs mixtures (Sovol) have been produced and used as a dielectric fluid in the manufacture of power capacitors and transformers (Ivanov and Sandell, 1992). The present results suggest the presence of local sources of PCBs in Russia.

In general, low concentrations PCBs were found in mussels from Cambodia, Indonesia, Malaysia, Vietnam, India and China, which indicate fewer local sources. However, some urban/industrialized cities in these countries showed relatively higher concentrations, such as Bombay (INMHMB) and Cochin (INKRCH) in India, Fuzhou Lian Jian in China (CHFZLJ) and Manila (PHMMB, PHCVBC, PHBLOD) in the Philippines (Fig. 6 and Table 2). Such a pattern found in the present study was similar to our previous reports that showed relatively higher levels of PCBs in mussels from coastal waters in populated and industrialized cities in Asian developing countries such as Bangkok (Kan-atireklap et al., 1997), Cochin (Kan-atireklap et al., 1998) and Manila (Prudente et al., 1999). One of the major PCBs sources in metropolitan cities in developing nations might be considered as the release from electrical equipments, such as the accidental contamination by PCBs took place in Bangkok, Thailand, where imported older transformers and capacitors were dumped in the suburb (Watanabe et al., 1996).

3.2.4. CHLs

The residue levels of CHLs in mussels collected from Asian countries are given in Table 2 and Fig. 7. Higher levels of CHLs were observed in mussels from Japan (150–1800 ng/g lipid wt.), China (40–870 ng/g lipid wt.), Hong Kong (18–750 ng/g lipid wt.), Malaysia (17–630 ng/g lipid wt.) and Singapore (520 ng/g lipid wt.) (Table 3). China, Malaysia and Singapore showed higher concentrations of CHLs at sites in proximity to harbor, aquaculture, urban and densely populated areas (e.g. CHHKTS, CHHKST, CHHKOS, CHXMLH, CHFJL, CHSDJZ, CHLNLS, MYMAAB, MYJBPP-3 and SGWL). Higher levels of CHLs were also detected in

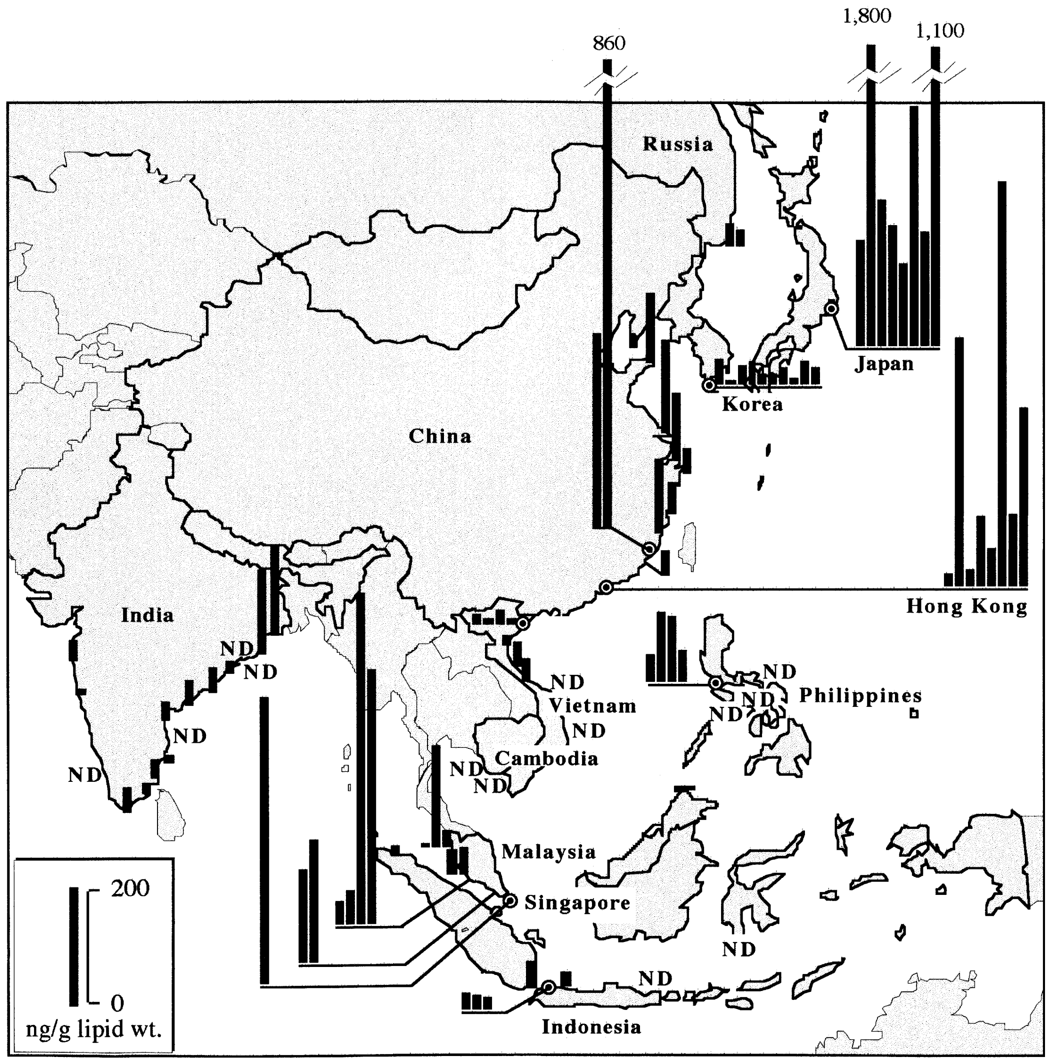


Fig. 7. Distribution of concentrations of CHLs in mussels collected from coastal waters of some Asian countries.

mussels from coastal waters of Japan (Ueno et al., 1999), cetaceans (Minh et al., 2000) and skipjack tuna (Ueno et al., in press). CHLs also had been used largely for termite control until 1986 in Japan (Loganathan et al., 1993) and still seem to be discharging into marine environment.

Despite lower concentrations of CHLs in mussels from Philippines and India, relatively higher concentrations of CHLs were found in certain locations in proximity to fishing harbors (e.g. INWBDH, INMDKD), urban and industrialized areas (e.g. PHCVBC, PHMMMB, INMHMB) in these countries (Table 2 and Fig. 7). Prudente et al. (1999) also found similar values of CHLs in mussels from coastal waters near to urbanized and industrialized areas in Philippines. This spatial distribution found in Asian developing countries suggest that CHLs may be still in use against termites in highly populated, industrialized and fishing harbor areas. Among Asian developing countries, concentrations of CHLs were lower than the detection limit in the mussels from Cambodia, which suggest very low usage of CHLs in this country.

The compositions of CHLs in mussels from Asia-Pacific region also varied depending on countries (Fig. 8). *Trans*-nonachlor and *cis*-nonachlor were the main constituents of total CHLs in mussels. Higher percentages of *trans*-nonachlor were found in mussels from Japan, Singapore, Malaysia, India, Philippines and China. The presence of *trans*-nonachlor in the present samples in these countries might suggest the recent usage of technical CHLs.

The present concentrations and compositions of CHLs found in the recent study in mussels from Japan, Singapore, Malaysia, India, Philippines and China might reflect the recent input of CHLs into coastal environment from these countries.

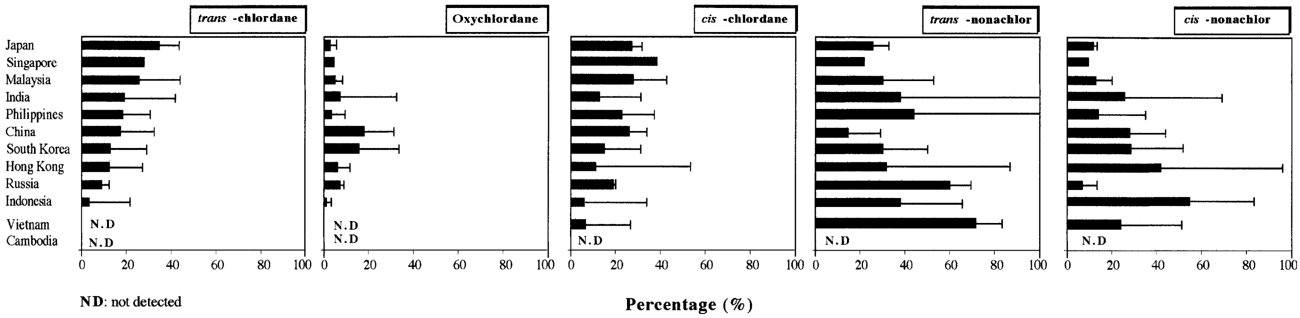


Fig. 8. Composition of CHLs compounds in mussels collected from coastal waters of some Asian countries.

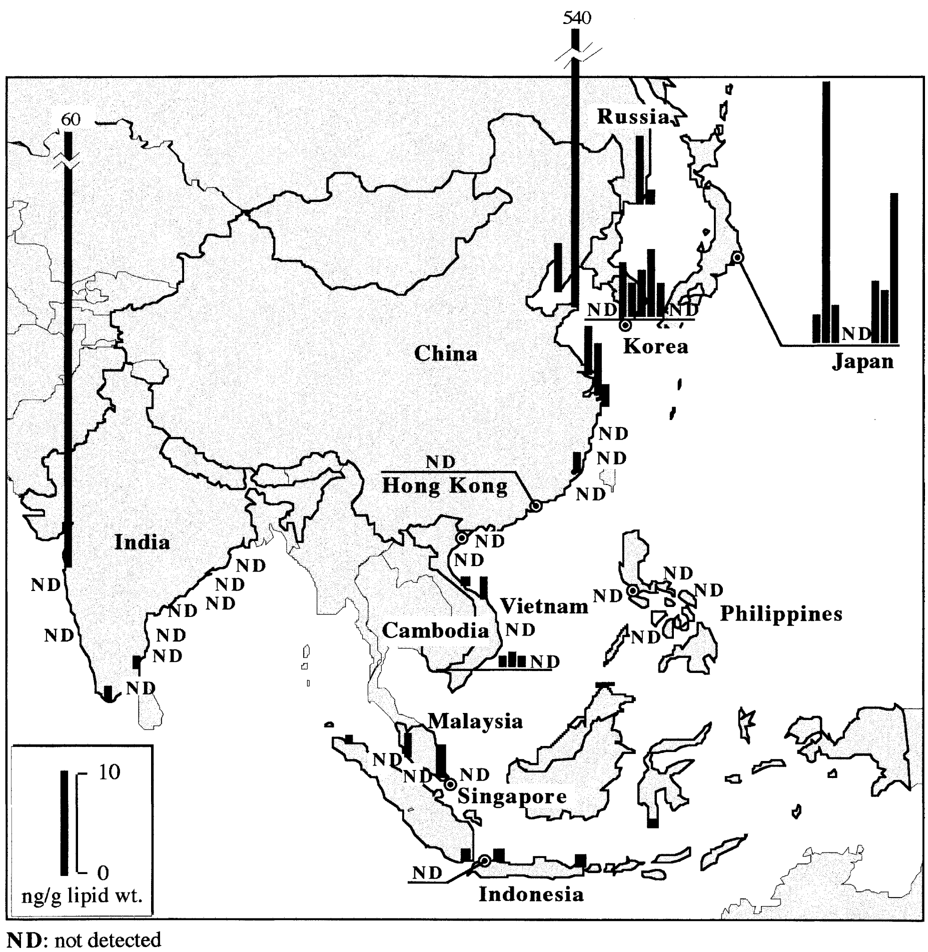


Fig. 9. Distribution of concentrations of HCB in mussels collected from coastal waters of some Asian countries.

3.2.5. HCB

Geographical distributions of HCB levels in mussel are shown in Table 2 and Fig. 9. Concentrations of HCB seems to be generally uniform in mussels from Asia-Pacific region, while relatively high concentrations were found in samples collected near the industrialized cities such as in Jiao Zhou Wan, Qing Dao City (CHSDJZ) in China, Bombay (INMHMB) in India and Tokyo Bay (JPTB 05, JPTB 94) in Japan (Fig. 9). HCB is not only used as a fungicide, but also generated as a by-product during the production and usage of several agrochemical and industrial chemicals. Furthermore, HCB has also been released into the environment by waste incineration (van-Birgelen, 1998). Hence present results may be reflecting the levels of HCB generated in industrial and thickly populated cities in this study. Additionally, HCB is known to have rather volatile nature (Calamari et al., 1991; Kannan et al., 1995). The residues of HCB in mussels of the present study might be the reflectin of limited source and the volatile nature of this compound.

3.3. International comparison

To compare the magnitude of contamination of OCs among Asian countries mean and 90th percentile values in mussels were calculated for each country (Table 4). Mussels from Japan, Hong Kong and the Philippines showed relatively high concentrations of PCBs and CHLs compared with those from other Asia-Pacific region. Interestingly, the order of mean and 90th percentile values of concentrations agree well with the per-capita gross national product (GNP) of each country (Table 4). Since value of GNP is an indicator of economic status, PCBs and CHLs contamination seem to be strongly related to industrial and human activities. The contamination of PCBs and CHLs may increase in those countries with high economic growth rate. Among Asian developing countries except China, Vietnam and India, DDT and HCH levels also agreed well with the GNP of the respective countries. DDTs and HCHs contamination relate not only with economic status but also with agricultural activities in each country, especially China, Vietnam and India.

Concentrations of OCs in mussels from some Asian countries were compared with the values in bivalves reported from various locations of the world (Table 5). Generally, concentrations of PCBs seemed to be lower in Asian developing countries than in developed nations. DDTs and HCHs were higher in Asian developing countries than those in developed nations, clearly suggesting that these insecticides are still being used in some Asian developing countries.

3.4. Probable emission sources estimated by mussel contamination

Based on the residue patterns and concentrations of OCs in mussels from Asian countries, it is clear that pattern of contamination of OCs in each country is different. As mentioned earlier, mussels from China, Hong Kong and Vietnam had higher concentrations of DDTs (Fig. 3). India, China and Russia had higher levels of HCHs (Fig. 4), while Japan, Russia and Hong Kong showed higher PCBs (Fig. 6), and Japan, China, Hong Kong and Malaysia had higher concentrations of CHLs (Fig. 7). Considering from the present results, the top three countries found with higher concentrations of different OCs in mussels are China, Hong Kong and Vietnam for DDTs and India, China and Russia for HCHs, while PCBs are the main contaminants in Japan, Russia and Hong Kong, and higher contamination of CHLs were found in Japan, China, and Hong Kong. These countries seem to play a role as probable emission sources of corresponding OCs in Asia, and play similar roles on their distribution and contamination on global terms.

Table 4
Mean, range, and 90th percentile values of OC concentrations (ng/g lipid wt.) in mussels from some Asian countries

Country	Sampling year	PCBs			CHLs			DDTs			HCHs			HCB			Per-capita GNP ($)[a]
		Range	Mean	90th per-centile	Range	Mean	90th per-centile	Range	Mean	90th per-centile	Range	Mean	90th per-centile	Range	Mean	90th per-centile	
Japan	1994	510–12,000	3000	10,000	150–1800	550	1600	70–2900	600	2300	13–50	28	47	<0.60–29	8.2	25	37,950
Hong Kong	1998–1998	40–710	307	610	18–750	240	610	640–61,000	7700	33,000	2.1–30	18	28	<1.5	<1.0	0.5	24,303
South Korea	1998	30–340	170	320	3.7–40	25	38	14–360	150	340	1.9–80	14	48	<0.40–7.3	2.3	6.4	9628
Malaysia	1997–1998	<4.2–250	56	230	17–630	140	520	16–270	90	160	<0.80–12	3.9	10	<0.50–3.3	0.90	2.6	3531
Thailand[b]	1994–1995	5.0–1100	170	600	10–510	80	180	48–3300	380	630	0.16–27	9.0	21	<0.50–8.0	3.3	7.4	1984
Philippines[c]	1994/ 1997–1998	22–2100	660	1700	5.1–460	120	340	4.0–200	69	340	0.50–10	4.5	8.5	<0.40–2.0	0.80	1.8	1035
China	1999–2001	15–540	120	370	10–870	190	610	830–54,000	17,000	42,000	11–110	44	95	<0.90–540	56	270	840
Indonesia	1998	5.6–210	87	202	8.3–45	19	37	6.5–160	70	140	<0.60–5.3	2.7	5.1	<0.50–1.5	0.80	1.4	692
India[d]	1994– 1997/1998	7.9–650	202	470	0.20–160	38	91	29–3000	430	950	20–590	205	380	<0.40–63	4.4	15	459
Vietnam	1997	21–450	160	420	7.0–160	35	110	220–34,000	4400	28,000	3.0–11	5.7	11	<0.50–3.5	0.92	1.8	398
Cambodia	1998	<1.4–220	33	180	<0.40–<1.1	<0.30	<0.70	16–42	23	39	<0.80–<1.2	<0.30		<0.40–2.6	1.2	2.5	280

[a] Data for 2001 cited from Asiaweek (2001).
[b] Data cited from Kan-atireklap et al. (1997).
[c] Data of this study combined with Prudente et al. (1999).
[d] Data of this study combined with Kan-atireklap et al. (1998).

Table 5
Global comparison of PCBs and organochlorine pesticides (ng/g wet wt.) in bivalve mollusks

Location	Survey year	Species	PCBs	DDTs	CHLs	HCHs	HCB	Reference
Developing nations								
Guanabara Bay, Brazil[a]	1986–1993	*Mytilus edulis*	34					Sericano et al. (1995)
Guanabara Bay, Brazil[a]	1996	*Perna viridis*	NA	1.1–10	NA	0.10–0.90	<0.01–0.7	de Brito and Bruning (2002)
Cambodia	1998	*Perna viridis*	<0.50–5.1	0.10–0.20	<0.01–<0.01	<0.01–<0.01	<0.01–0.03	Present study
China	1999–2001	*Perna viridis* & *Mytilus edulis*	0.30–3.1	58–630	1.0–10	0.10–0.60	<0.01–0.50	Present study
	1999	*Perna viridis*	NA	150–200	NA	NA	NA	Klumpp et al. (2002)
	1994–1995	*Perna viridis*	0.52–10	240–310	NA	0.70-5.4	NA	Hong et al. (2000)
India	1998	*Perna viridis*	0.20–11	0.60–15	<0.01–3.3	0.20–7.7	<0.01–0.30	Present study
India	1994–1995	*Perna viridis*	0.31–15	0.90–40	<0.01–2.0	1.5–12	<0.01–0.40	Kan-atireklap et al. (1998)
India	1988–1989	*Perna viridis*	0.70–7.1	3.0–40	NA	4.3–16	NA	Ramesh et al. (1990)
Indonesia	1998	*Perna viridis*	0.10–2.7	0.10–3.1	<0.01–0.30	<0.01–0.10	<0.01–0.03	Present study
Isla de Aserradores, Nicaragua	1986–1993	*Mytilus edulis*	NA	32	2.0	NA	NA	Sericano et al. (1995)
Malaysia	1998	*Perna viridis*	<0.05–5.1	0.06–0.80	0.10–9.6	<0.01–0.20	<0.01–0.60	Present study
Puerto Madero, Mexico	1986–1993	*Mytilus edulis*	NA	21	NA	NA	NA	Sericano et al. (1995)
Tampico, Mexico	1986–1993	*Mytilus edulis*	NA	18	2.0	NA	NA	Sericano et al. (1995)
Philippines	1998	*Perna viridis*	0.40–14	0.07–0.80	0.10–2.8	<0.01–0.05	<0.01–<0.01	Present study
Philippines	1994–1997	*Perna viridis*	0.70–36	0.20–4.2	0.15–9.5	<0.01–0.20	<0.01–0.04	Prudente et al. (1999)
Thailand	1994–1995	*Perna viridis*	<0.10–20	1.2–38	0.25–6.0	<0.01–0.33	<0.01–0.12	Kan-atireklap et al. (1997)
Vietnam	1997	*Perna viridis*	0.20–3.4	2.4–310	0.10–1.4	0.04–0.11	<0.01–0.03	Present study
Developed nations								
Lake Erie, Canada[a]	1995	*Dreissena polymorpha*	45–120	NA	NA	NA	NA	Roe and MacIsaac (1998)
Germany (South West Baltic Sea)	1990–1991	*Mytilus edulis*	4.7–97	1.0–18	NA	0.30–4.5	NA	Lee et al. (1996)
Hong Kong	1986	*Perna viridis*	49–330	50–520	NA	53–110	NA	Phillips (1989)
Hong Kong	1998–1999	*Perna viridis*	0.30–7.4	7.5–1000	0.10–7.0	0.02–0.60	<0.01	Present study
Hong Kong[a]	1983	*Perna viridis*	<9.6–300	<14–320	NA	<4.8–34	NA	Phillips (1985)
Italy (Baveno)[a]	1997	*Dreissena polymorpha*	NA	86–107	NA	NA	NA	Binelli et al. (2001)
Italy (Taranto)[a]	1997	*Dreissena polymorpha*	NA	14–64	NA	NA	NA	Binelli et al. (2001)
Tokyo Bay, Japan	1994	*Mytilus gallorovincialis*	7.4–84	0.80–12	1.7–17	0.20–0.60	<0.01–0.30	Present study
Tokyo Bay, Japan	1998	*Mytilus gallorovincialis*	94–164	5.0–8.0	5.0–6.5	0.06–0.30	<0.04–0.10	Ueno et al. (1999)
Mersey Estuary, England	1998	*Mytilus edulis*	32–110	0.20–17	NA	NA	<0.10–0.25	Connor et al. (2001)

Table 5 (*continued*)

Location	Survey year	Species	PCBs	DDTs	CHLs	HCHs	HCB	Reference
Russia	1999	*Crenomytilus grayanus*	56–70	0.50–0.60	1.0–1.2	0.70–1.1	0.03–0.14	Present study
Singapore	1999	*Perna viridis*	2.4	3.0	14	0.30	<0.01	Present study
South Korea	1998	*Mytilus edulis*	0.8–7.2	0.7.0–7.5	0.20–1.2	0.10–1.0	<0.01–0.20	Present study
Spain	1990–1991	*Mytilus edulis*	ND-620	ND-36	NA	ND-49	NA	Alvarez Pineiro et al. (1995)
Taiwan	1991–1998	*Crassostrea gigas*	NA	1.0–54	NA	NA	NA	Han et al. (2000)

NA: no data available, ND: not detected.
[a] Data reported was recalculated from dry weight basis to wet weight basic by using a conversion of 0.16 (Ramesh et al., 1990).

4. Conclusion

The results of the monitoring of marine pollution in Asian coastal waters using mussels as bioindicators showed clearly the status of contamination by OCs in this region, which suggest serious contamination by DDTs in China, Hong Kong and Vietnamese coastal waters, HCHs in Indian and Chinese marine environment, and PCBs and CHLs in the Philippines, Malaysia and Singapore marine waters. The occurrence of OC residues in mussels also pose a great concern on human health, as mussels are most valuable mariculture organisms and a commercially important seafood particularly in developing countries.

Based on our results, we suggest that continuous monitoring of OC residues including studies on ecotoxiclological risk assessment to elucidate the trend of contamination and toxic impact on humans and wildlife in Asian developing countries is necessary.

Acknowledgements

We wish to thank all the staff in counterpart countries for their cooperation in collecting samples. Sincere thanks are also due to Dr. K. Kannan (Michigan State University, USA), Dr. K. Kitazawa (UNESCO-IOC; present: Marine Science and Technology Center) and Dr. G. Kullenberg (UNESCO-IOC), Mr. G. Paoletto and Dr. J.I. Uitto (The United Nations University), Prof. E.D. Goldberg (Chairman of the International Mussel Watch Committee), Prof. J.W. Farrington and Mr. B.W. Tripp (Woods Hole Institute of Oceanography, USA) for their encouragement and support. This research was supported by Sumitomo Foundation, Toyota Foundation, Sotoshu Volunteer Association, the Japan Fund for Global Environment of Japan, Environment Corporation and Grant-in-Aid from the Scientific Research Program (Grants 09041163, 12308030 and 120555101) and "21st Century COE Program" from the Ministry of Education, Science, Culture, Sports and Technology in Japan.

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