PERSISTENT ORGANOCHLORINE RESIDUES AND THEIR BIOACCUMULATION PROFILES IN RESIDENT AND MIGRATORY BIRDS FROM NORTH VIETNAM

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ABSTRACT

Concentrations of persistent organochlorines were determined in whole body homogenates of resident and migratory birds collected from the Red River estuary, North Vietnam during March and October 1997. Contamination pattern was in the order of DDTs > PCBs > HCHs > CHLs > HCB in both resident and migratory birds. Bioaccumulation feature according to the feeding habit showed little variability, which may reflect relatively similar trophic levels of bird species. Resident birds accumulated greater concentrations of DDTs as compared to migrants. In contrast, HCH residues were apparently higher in migratory species. Higher proportions of *p,p'*-DDT in total DDT were found in many species of both residents and migrants, indicating recent exposure to technical DDT in North Vietnam. Congener-specific PCB analysis showed the predominance of penta- and hexachlorobiphenyls in all species analyzed. Estimated hepatic microsomal enzyme activities suggest higher metabolic capacity for PCB congeners in shore birds from Vietnam as compared to high trophic predator birds and marine mammals. Comparison of OC residues in avian system in Asia-Pacific revealed that resident birds in North Vietnam accumulated the highest DDT residues, whereas contamination in migratory birds was lower than other countries, suggesting recent usage of DDT in Vietnam. Compilation of available data for birds and fish and bivalve from recent Mussel Watch Program in Asia-Pacific suggests that Vietnam may become a potential emission source of DDT among Asian developing countries. To our knowledge, this is the first study of the OC accumulation in avian species from Vietnam.

Key words: Persistent organochlorines, birds, Vietnam, DDTs, emission source

INTRODUCTION

Organochlorine compounds such as DDTs, hexachlorocyclohexane isomers (HCHs) and polychlorinated biphenyls (PCBs) are well known to be potentially one of the most toxic groups of environmental pollutants for humans and wildlife. Extensive usage of these chemicals has led to human health hazard as well as to deterioration of environmental quality. A large number of studies have been focused on the accumulation of persistent organochlorines (OCs) in various species of flora and fauna over the world. Abnormalities in occurrence of eggshell thinning, reproductive failure and mortality of embryo and chicks in avifauna have been observed and possible link between these abnormalities and high degree of exposure to OCs, particularly DDTs and PCBs, has been suggested [1-5]. Despite this fact, some OCs are still being used in tropical and sub-tropical regions, particularly in Asian developing countries for agricultural purposes and vector-borne disease eradication program. Our recent comprehensive monitoring surveys in Asia-Pacific have also suggested the role of this region as one of the major sources of OC contamination in a global perspective, particularly for pristine areas such as the Arctic and the Antarctic [6-8].

Vietnam, a developing Asian country of interest, is located in the tropical region. Many agricultural chemicals including DDTs and HCHs have been consumed in the country until very recently [9]. Despite the continuing usage of OC pesticides in Vietnam, there has been little information regarding OC exposure in higher trophic animals, such as birds living and wintering in this country. It is well known that birds have been widely used as bioindicators for environmental pollution, particularly by persistent OCs, which have been considered as potential endocrine disrupting chemicals in wildlife. Earlier studies have indicated widespread occurrence of persistent OCs in abiotic and biotic samples including air, water, soils, sediments, foodstuffs and human breast milk collected from various locations in Vietnam [7,8,10-12]. From the environmental health point of view, understanding of OC exposure in higher trophic animals is of major importance because organochlorines posed high bioaccumulation potential and cause adverse effects in these animals. However, no investigation of OC pollution in avian species in Vietnam has been conducted so far and therefore, accumulation features as well as possible toxic impacts of persistent OCs to avian species from Vietnam have not been characterized. The magnitude of exposure to OCs in birds from Vietnam is of concern.

In the present study, we carried out an extensive sampling survey to collect different species of birds living and wintering in the Red River estuary, North Vietnam during March and October, 1997, and determined the concentrations of persistent OCs such as DDT and its metabolites (DDTs), hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB), chlordane compounds (CHLs) and polychlorinated biphenyls (PCBs). Residue levels and bioaccumulation profiles of OC insecticides and PCB congeners were discussed. Bioaccumulation features according to the feeding habit and migratory behavior of birds were also examined. Further, OC residues in fish and bivalve from the present study and those reported in our recent Mussel Watch Program in Asia-Pacific region were compiled and analyzed to understand the magnitude of OC contamination in Vietnamese biota and suggest the role of Vietnamese environment among Southeast Asian countries as emission source of pollution.

MATERIALS AND METHODS

Sampling

Resident and migratory birds (n = 101) were collected from the wetland (Con Lu Island) in the outer estuary system of the main Red River estuary, during March and November, 1997 (Fig.1). Birds were trapped by mist nets and immediately after collection, birds were iced, transported to laboratory, and shipped to Japan with dry ice. The data of biological characteristics and ecological information on birds analyzed are presented in Table 1. The whole body, except the feathers, was homogenized and stored at -20°C until analysis. Diet samples including fish (5 to 10 specimens) and shrimp (20 specimens) were also pooled and analyzed.

According to ecological studies by de Hoyo *et al.* [13], most of the bird species analyzed in this study were classified into 2 main groups, namely residents living in the same region all year for their entire life span, and migrants which have their breeding grounds located in other locations in the East and central Asia, Russia, Siberia, and Australia. Ecological studies have shown that most of the migratory birds that winter in Vietnam originate from eastern Asian countries such as Korea, Japan, Hong Kong and Philippines, central Russia and Russian Arctic

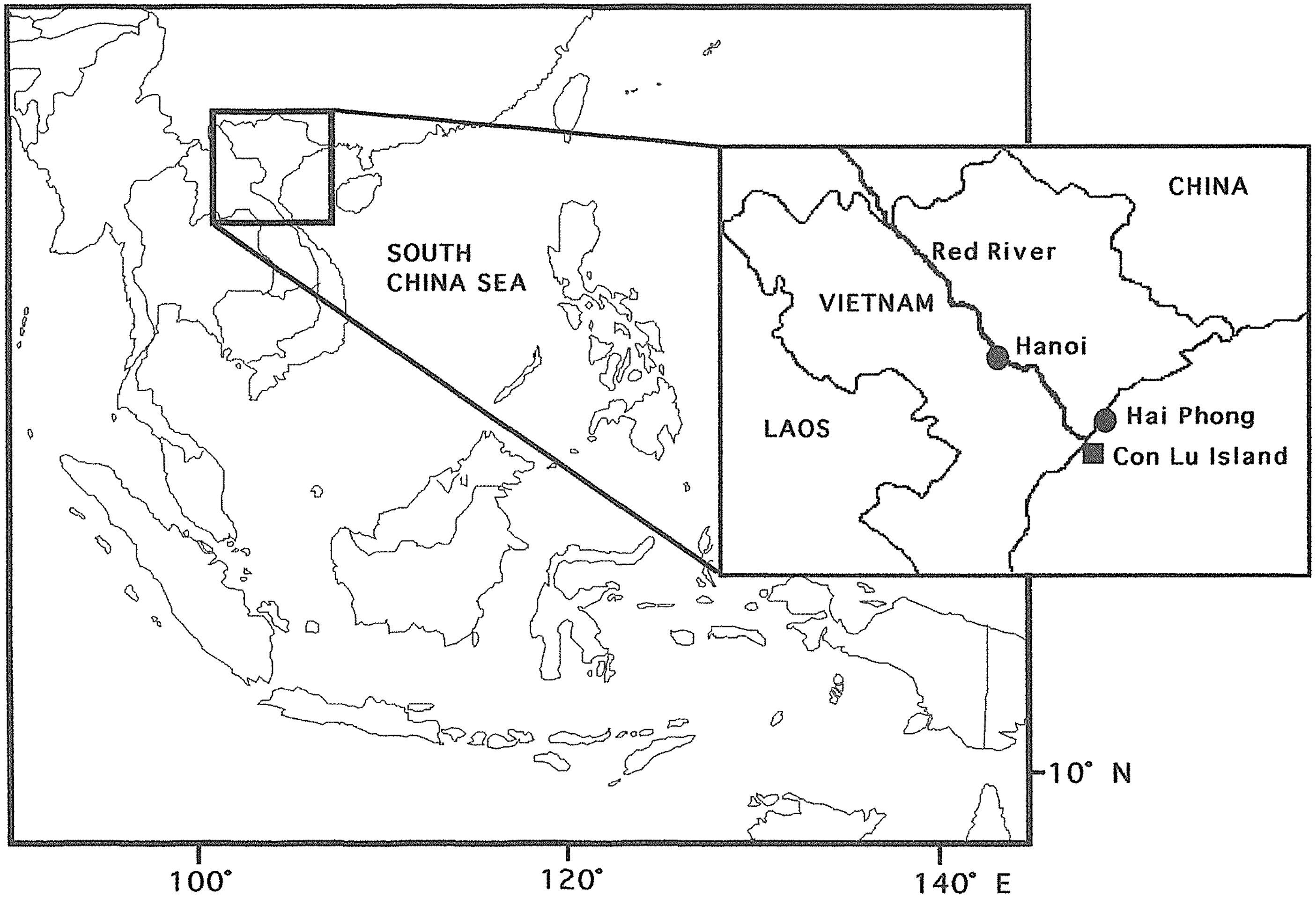

Figure 1. Map showing sampling location in Red River delta, North Vietnam.

Table 1. Biometric data of resident, migratory birds and diet analyzed in the present study.

Species (Scientific name)	*n*	Sex	Body weight (g)	Standard length (cm)
Resident				
Black-capped kingfisher (*Halcyon pileata*)	2	2F	100 (99-101)	28 (27-30)
Common kingfisher (*Alcedo atthis*)	5	(3M, 2F)[a]	16 (14-18)	27 (23-29)
Common moorhen (*Gallinula chloropus*)	1	1F	134	28
Cinnamon bittern (*Ixobrychus cinnamomeus*)	1	1F	120	39
Slaty-breasted rail (*Rallus striatus*)	2	2F	133 (120-146)	25 (24-26)
White-breasted waterhen (*Amaurornis phoenicurus*)	3	(2M, 1F)	156 (120-198)	27 (27-28)
White-throated kingfisher (*Halcyon smyrnensis*)	1	1M	106	28
Migrant				
Common redshank (*Tringa totanus*) (S)[b]	10	(6M, 4F)	122 (103-141)	31 (29-33)
Common redshank (*Tringa totanus*) (A)[b]	5	(2M, 3F)	108 (102-114)	25 (23-26)
Kentish plover (*Charadrius alexandrinus*)	10	(7M, 3F)	20 (16-23)	67 (55-85)
Gull-billed tern (*Sterna nilotica*)	1	1M	184	33
Little tern (*Sterna albifrons*)	1	1F	71	25
Long-billed Mongolian plover (*Charadrius mongolus* ; sub species *C. atrifrons*) (S)	2	2M	79 (78-80)	20 (20-20)
Long-billed Mongolian plover (*Charadrius mongolus*)(A)	2	2M	83 (81-85)	21 (20-21)
Whimbrel (*Numenius phaeopus*)	1	1M	348	39
Whiskered tern (*Sterna hybrida*) (S)	2	2F	76 (74-77)	27 (26-28)
Whiskered tern (*Sterna hybrida*) (A)	2	2M	72 (69-75)	24 (24-24)
Bar-tailed godwit (*Limosa lapponica*)	1	1F	294	39
Dunlin (*Calidris alpnia*) (S)	4	(1M, 3F)	58 (53-63)	20 (19-21)
Dunlin (*Calidris alpnia*) (A)	4	(2M, 2F)	55 (46-62)	21 (20-22)
Great knot (*Calidris tenuirostris*) (S)	2	2M	234 (214-261)	26 (25-28)
Great knot (*Calidris tenuirostris*) (A)	2	(1M, 1F)	200 (181-223)	29 (28-29)
Grey plover (*Pluvialis squatarola*)	3	3M	210 (195-236)	29 (28-30)
Marsh sandpiper (*Tringa stagnatilis*) (S)	10	(7M, 3F)	24 (22-26)	66 (60-73)
Marsh sandpiper (*Tringa stagnatilis*) (A)	4	4M	24 (24-25)	61 (56-65)
Red knot (*Calidris canutus*)	5	5M	130 (100-165)	25 (23-28)
Rufous-necked stint (*Calidris ruficollis*)	5	(3M,2F)	24 (22-26)	15 (14-16)
Short-billed Mongolian plover (*Charadrius mongolus* ; sub species *C. schaeferi*) (S)	3	(2M,1F)	62 (60-64)	19 (18-19)
Short-billed Mongolian plover (*Charadrius mongolus*)(A)	2	2M	60 (59-60)	20 (19-20)
Spotted redshank (*Tringa erythropus*)	1	1M	117	29
Terek sandpiper (*Xenus cinereus*)	2	2M	65 (64-65)	24 (23-24)
Diets				
Fish species (*Mugil sp* & *Chlorophthalmus sp*)	10	unknown	15 (11-18)	22 (9.7-54)
Shrimp	20	unknown	not measured	not measured

[a]M: male, F: female. Figure indicate number of sample.

[b]S, A: samples collected in spring and autumn, respectively.

[13,14]. Some species such as common redshank, kentish plover, gull-billed tern, little tern, long-billed Mongolian plover, short-billed Mongolian plover, whimbrel, whiskered tern have their breeding ground mainly in China, Korea and Japan. Other species includes bar-tailed godwit, dunlin, great knot, grey plover, marsh sandpiper, rufous-necked stint, spotted redshank, red knot and terek sandpiper breed chiefly in Eastern Russia, Siberia and Russian Arctic. In addition, these migratory species may migrate and have stopover sites in Indian sub-continent and Southeast Asian region.

Chemical Analysis

Chemical analyses of OCs followed the method previously described [15]. Briefly, 10–15g whole body homogenate samples were homogenized with anhydrous Na_2SO_4 and OCs were extracted in a Soxhlet apparatus with a mixture of hexane and diethyl ether. The fat content was gravimetrically determined from an aliquot of the extract. The extract was then added into a dry Florisil column to remove fat. OCs were eluted with 150 ml of 20 % water in acetonitrile to a separatory funnel containing hexane and water. After partitioning, the hexane layer was concentrated and then passed through a 12g activated Florisil column for fractionation. The first fraction eluted with hexane contained HCB, PCBs, *p,p'*-DDE, *trans*-nonachlor; the second fraction eluted with 20 % dichloromethane in hexane contained *p,p'*-DDD, *p,p'*-DDT, HCH isomers (α,β,γ isomers), CHL compounds (*cis*-chlordane, *trans*-chlordane, *cis*-nonachlor and oxychlordane). Each fraction was concentrated and injected into a gas chromatograph with electron capture detector (GC-ECD) for quantification.

OCs were quantified by a Hewlett Packard 5890 series II GC-ECD (Wilmington, DE) equipped with a moving needle-type injection port. The GC column employed was DB-1 fused silica capillary column (0.25 mm x 30 m; J & W Scientific Inc., Folsom, CA) coated with 100 % dimethylpolysiloxane at 0.25 μm film thickness. The column oven temperature was programmed from 60 to 160°C, held for 10 min, then increased to 260°C at a rate of 2°C/min and held for 20 min. Injector and detector temperatures were set at 260 and 280°C, respectively. Helium and nitrogen were used as carrier and make up gases, respectively. OC concentrations were calculated from the peak area of the sample to the corresponding external standard. The PCB standard used for quantification was an equivalent mixture of Kanechlor preparations (KC-300, KC-400, KC-500, KC-600) with known PCB composition and content. Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain total PCB concentrations. Recoveries of target contaminants through this analytical method ranged from 95 to 105 %. Concentrations were not corrected for recovery rates. A procedural blank was analyzed with every set of 6 samples to check for interfering compounds and to correct samples values, if necessary. DDTs represent the sum of *p,p'*-DDT, *p,p'*-DDD and *p,p'*-DDE, while CHLs include *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane. HCHs include α, β and γ- isomers.

Isomer-specific analysis of PCBs was similar to that described previously [16]. Briefly, 10-15 g of whole body homogenates was refluxed in 1 N KOH in ethanol for 1 h, and the solution was then transferred to a separatory funnel containing hexane and hexane-washed water. After partitioning, the hexane layer was concentrated and cleaned up on 1.5 g of silica gel (Wako-gel S-1, Wako Chemical Co., Japan) packed in a glass column. PCB congeners were eluted with 200 ml hexane. The hexane was then concentrated, treated with 5 % fuming sulfuric acid and rinsed with hexane-washed water. The final solution was micro-concentrated and injected into a gas chromatograph with a mass selective detector (GC-MSD) for quantification.

Quantification of PCB congeners was carried out using a Hewllet-Packard 5890 Series II GC-MSD coupled with a Hewllet-Packard 5972 Series MS (Wilmington, DE) having an electron impact (EI) at 70 eV. The GC column employed was DB-1 fused silica capillary column (0.25 mm x 30 m; J & W Scientific Inc., Folsom, CA) coated with 100 % dimethylpolysiloxane at 0.25 μm film thickness. The column oven temperature was programmed from 70 to 160°C, held for 20 min, then increased to 260°C at a rate of 2°C/min and held for 30 min. Injector and ion source temperatures were kept at 250 and 280°C, respectively. An equivalent mixture of Kanechlors 300, 400, 500 and 600 was used as a standard for quantification. Concentrations of individual PCBs were quantified from the peak area of the sample to that of the corresponding external standard. PCB homologues were determined by selective ion monitoring. Data was acquired by a Hewlett-Packard 5972C data system, in which cluster ions were monitored at m/z 254 and 256, 290 and 292, 324 and 326, 358 and 360, 392 and 394 and 428 and 430 for tri-, tetra-, penta-, hexa-, hepta-, and octachlorobiphenyls, respectively.

Recoveries of total PCBs were examined by spiking 3.0 μg of Kanechlor standard to corn oil. The recoveries ranged from 100 ± 3 % for total PCBs. PCB congeners are referred using their IUPAC number throughout the manuscript.

For quality assurance and quality control, our laboratory participated in the Intercomparison Exercise for Persistent Organochlorine Contaminants in Marine Mammal Blubber organized by the National Institute of Standards and Technology (Gaithersburg, MD) and Marine Mammal Health and Stranding Response Program of the National Oceanic and Atmospheric Administration's National Marine Fisheries Service (Silver Spring, MD). Standard reference material SRM 1945 was analyzed for selected PCB congeners and persistent OCs. Reliable results were obtained by comparison of data from our laboratory with those from material reference values.

RESULTS AND DISCUSSION

Residue levels and feature of accumulation

OCs were detected in all resident and migratory birds in both spring and autumn seasons (Table 2). Residue pattern in most species of resident and migratory birds analyzed followed the order of DDTs > PCBs > HCHs > CHLs > HCB. Similar pattern was observed in fish and shrimp diet, with elevated concentrations of DDTs (up to 170 ng/g wet wt). DDT concentrations in resident and migratory species were approximately one order of magnitude greater than those of PCBs and HCHs. These results indicate high degree of exposure to DDTs in biota from Vietnam and extensive usage of this insecticide in tropical Asian countries including Vietnam. This accumulation pattern is different from that found in resident and migratory birds collected from south India and Lake Baikal, Russia, which showed HCHs and PCBs as predominant contaminants, respectively [15,17]. Technical HCH is still being used in India in huge quantity for agriculture purposes and elevated contamination of HCHs has been common in most of environmental samples collected from India [7,8,15,18]. In Lake Baikal region, higher PCB source was allocated due to extensive pulp and paper mill industries in southern lake [16,17,19]. Thus, residue patterns of OC accumulation observed in resident birds from Vietnam, India and Lake Baikal reflect the status of OC usage in these areas in recent years.

Among residents, higher OC concentrations were found in black-capped kingfisher, cinnamon bittern and white-breasted waterhen. In particular, white-breasted waterhen carried elevated amounts of DDTs (mean 3100 ng/g wet wt), at least 5 times greater than those in other species (Table 2). Higher residues of PCBs and HCHs were found in common and black-capped kingfisher, while lowest levels were recorded in slaty-breasted rail. Kingfisher and cinnamon bittern are piscivorous species, which feed mainly on small fish and insects, contained higher concentrations of OCs. Slaty-breasted rail, feed on plants, accumulated lower residues. White-breasted waterhen eats small insects and plants, but accumulated high concentrations of DDTs. Ecological studies indicated that this species often occurs close to human habitation, such as village ponds, sewage ponds, in addition to agricultural areas such as rice fields and sugar cane [13]. This probably a plausible explanation for elevated OC accumulation in white-breasted waterhen. From ecotoxicological viewpoint, this species may

Table 2. Concentrations of persistent organochlorines in resident and migratory birds collected from North Vietnam.

Species	*n*	Fat content	Concentration (ng/g wet wt.)				
		(%)	PCBs	DDTs	HCHs	CHLs	HCB
Resident							
Black-capped kingfisher (S)[a]	2	8.4 (6.7-10)	53 (46-60)[b]	340 (300-380)	27 (17-36)	1.5 (1.4-1.5)	2.1 (2.1-2.1)
Common kingfisher (S)	5	11 (6.7-15)	59 (25-84)	140 (100-170)	10 (2.6-32)	1.9 (0.80-4.7)	0.68 (0.40-1.6)
Common kingfisher (A)[a]	2	3.2 (2.3-4.0)	62 (42-82)	400 (350-450)	3.9 (3.6-4.2)	0.77 (0.54-1.0)	1.2 (1.0-1.4)
Common moorhen (A)	1	1.9	16	200	5.2	2.3	2.1
Cinnamon bittern (A)	1	6.7	32	640	16	37	1.7
Slaty-breasted rail (S)	2	4.2 (1.7-6.7)	22 (21-23)	370 (360-370)	1.8 (1.5-2.0)	1.9 (0.58-3.3)	2.3 (1.3-3.2)
White-breasted waterhen (A)	3	14 (5.8-24)	31 (20-44)	3100 (24-9000)	6.5 (0.39-12)	2.7 (0.29-5.5)	0.93 (0.13-1.8)
White-throasted kingfisher (S)	1	16	40	280	3.7	0.80	1.7
Migrant							
Common redshank (S)	10	8.2 (3.6-16)	26 (9.5-41)	120 (56-200)	31 (4.0-120)	0.92 (0.20-3.6)	1.2 (0.26-3.7)
Common redshank (A)	5	7.1 (3.6-8.8)	28 (18-36)	400 (310-520)	18 (4.7-44)	0.97 (0.63-1.5)	0.91 (0.70-1.3)
Gull-billed tern (A)	1	6.6	89	440	110	8.3	15
Kentish plover (S)	10	15 (10-30)	36 (18-96)	310 (230-530)	21 (6.5-88)	2.5 (0.62-8.0)	1.5 (0.37-2.5)
Little tern (A)	1	6.5	66	370	6.3	1.2	2.5
Long- billed Mongolian plover (S)	2	13 (11-14)	73 (46-100)	570 (510-630)	29 (28-30)	2.2 (2.1-2.3)	2.3 (1.5-3.1)
Long- billed Mongolian plover (A)	2	21 (19-23)	18 (12-24)	150 (120-180)	35 (31-39)	2.0 (1.6-2.4)	1.0 (0.30-1.7)
Whimbrel (A)	1	17	18	130	3.4	1.8	0.58
Whiskered tern (S)	2	6.7 (6.2-7.2)	110 (96-120)	230 (170-290)	8.7 (7.5-9.8)	0.77 (0.53-1.0)	1.5 (1.0-2.0)
Whiskered tern (A)	2	12 (6.3-17)	190 (100-270)	550 (490-600)	27 (18-36)	4.7 (1.9-7.5)	25 (6.5-43)
Bar-tailed godwit (A)	1	10	34	79	18	2.3	2.1
Dunlin (S)	4	22 (5.8-32)	31 (19-36)	240 (170-330)	6.5 (1.7-13)	1.9 (0.82-2.6)	0.80 (0.41-1.2)
Dunlin (A)	4	9.3 (6.2-16)	40 (29-71)	290 (130-450)	39 (21-57)	2.2 (1.5-2.6)	1.9 (1.5-2.7)
Great knot (S)	2	17 (12-22)	45 (44-45)	340 (210-470)	150 (140-160)	1.3 (0.62-2.0)	3.1 (1.5-4.6)
Great knot (A)	2	33 (27-38)	77 (43-110)	340 (110-560)	180 (55-300)	2.3 (1.4-3.1)	8.8 (2.5-15)
Grey plover (A)	3	7.6 (3.9-13)	23 (11-29)	110 (51-230)	6.6 (2.3-14)	1.6 (0.81-2.5)	0.87 (0.50-1.1)
Marsh sandpiper (S)	10	7.3 (2.0-11)	25 (10-49)	210 (80-280)	24 (7.0-72)	1.5 (0.44-3.3)	1.4 (0.72-3.9)
Marsh sandpiper (A)	4	11 (4.8-16)	45 (28-71)	160 (89-210)	29 (3.3-53)	0.58 (0.26-0.90)	2.7 (0.82-5.6)
Red knot (A)	5	11 (8.1-15)	28 (13-58)	170 (92-210)	28 (3.7-97)	1.3 (0.66-2.7)	3.6 (0.62-12)
Rufous- necked stint (S)	5	8.6 (6.2-11)	14 (10-17)	320 (200-400)	3.7 (2.9-5.2)	1.3 (0.82-2.7)	1.4 (1.1-1.9)
Short- billed Mongolian plover (S)	3	11 (8.8-13)	44 (19-89)	240 (140-300)	13 (10-19)	1.2 (0.49-1.8)	1.0 (0.75-1.5)
Short-billed Mongolian plover (A)	2	8.6 (7.4-9.8)	46 (40-51)	180 (160-200)	40 (8.2-71)	3.0 (1.9-4.0)	1.1 (0.10-2.1)
Spotted redshank (A)	1	4.1	40	51	16	1.5	0.84
Terek sandpiper (A)	2	12 (10-13)	49 (40-57)	630 (620-640)	9.0 (6.0-12)	3.2 (1.9-4.4)	1.0 (1.0-1.0)
Diet							
Fish 1 (*Mugil sp*)	5	3.1	2.5	120	3.5	3.2	0.18
Fish 2 (*Chlorophthalmus sp*)	5	2	2.7	170	0.53	4.1	0.11
Small fishes	10	4.5	4.5	15	9.3	1.3	0.10
Shrimp	20	2.8	2.8	4.6	4.2	0.11	0.15

[a]S and A: samples collected during spring and autumn, respectively.

[b]Values in parenthesis indicate the range.

experience harmful effect due to DDT accumulation. Assuming the transfer rate of 20 % from mother to eggs weighing 20 % of body mass, residues in whole body birds may reflect concentrations in eggs [15]. Considering this, few individuals of white-breasted waterhen analyzed in this study contained mean DDT level of 3.1 μg/g wet wt in the eggs, which approached the level associated with reduced breeding success (3 μg/g wet wt) as suggested by Newton [20]. Thus, this species may be at risk due to high extent of exposure to DDTs and deserves particular attention, considering the fact that this insecticide has been used until very recently in Vietnam.

As for migratory birds, residue levels were varied among species. Greater concentrations of DDTs and PCBs were observed in some piscivorous species such as Mongolian plover, whiskered tern, and common redshank. Whiskered tern have breeding ground in northeastern China, southwest and central Europe, which may explain noticeable DDT and PCB concentrations in samples collected in autumn (mean concentrations: 190 and 550 ng/g wet wt, respectively). Great knot, contained relatively high levels of HCHs in both spring and autumn, which could be due to the migratory behavior along coast lines of India and Australia [13]. Accumulation in stopover sites in India may elevate HCH concentrations in this species. Concentrations of CHLs and HCB were uniformly low in all the species examined, indicating minimal exposure to these chemicals in resident and migratory birds from Vietnam. In general, except for few species, concentrations of OCs in migratory birds from North Vietnam were generally lower than those observed in birds from India. A possible reason is that birds employed in this study may probably migrate and winter in stopover sites mainly in Southeast Asian region, where status of OC contamination deemed to be lesser than other countries such as India, Japan and China. Descriptions regarding comparative assessment of OC pollution in Asian developing region are discussed later in this paper, based on compilation and analysis of monitoring data in birds and fish from Asia-Pacific Mussel Watch Program.

To further understand bioaccumulation characteristics of OC in birds from North Vietnam, we attempted to assess residue levels in birds according to their feeding habit, habitat and migratory behavior. It should be noted that feeding habit of these shore birds were relatively similar. Ecological studies regarding food and feeding habit by de Hoyo *et al.* [13] indicated that most of the species employed in this study feed on small insects, and other low trophic organisms such as invertebrates, molluscs, crustaceans, etc. A few species have small fish as main diet (kingfishers, cinnamon bittern, redshanks, terns, Mongolian plovers, marsh sandpiper), while some species feed extensively on small insects and molluscs (kentish plover, bar-tailed godwit, dunlin, terek sandpiper, red knot). Other birds eat mainly insects and plant, which are considered as "insectivore/granivore". Two species, which have relatively more opportunistic feeding habit, were classified as omnivores (common moorhen and gull-billed tern). Bioaccumulation pattern of OCs in these birds were described in Figure 2. Among residents, PCBs and HCHs residues were higher in piscivores than those in omnivores and granivores, which is somewhat similar to those observed in birds from India [15,21]. An exception was observed in white-breasted waterhen, which accumulated elevated DDT residues. As discussed earlier, habitat areas close to human activities and agricultural fields of this species seems to be more profound reason for the observed result. In migratory birds, in general, piscivores and omnivores accumulated higher levels of OCs than insectivores and granivores, but no significant trend was found. It has been pointed out in earlier studies that fish-eating birds are capable of accumulating elevated OC concentrations [15,21,22]. This feature was characterized by relatively low hepatic microsomal monooxygenase activities, which are responsible for metabolism/detoxification of xenobiotics in birds [23,24]. Accumulation capacity is largely influenced by trophic levels of birds. Accordingly, fish-eating birds and species having opportunistic feeding habit accumulated to a greater extent than birds that feed on lower trophic organisms [15,17]. Looking at the accumulation pattern of OCs in birds according to their feeding habit in this study, it can be said that variations in

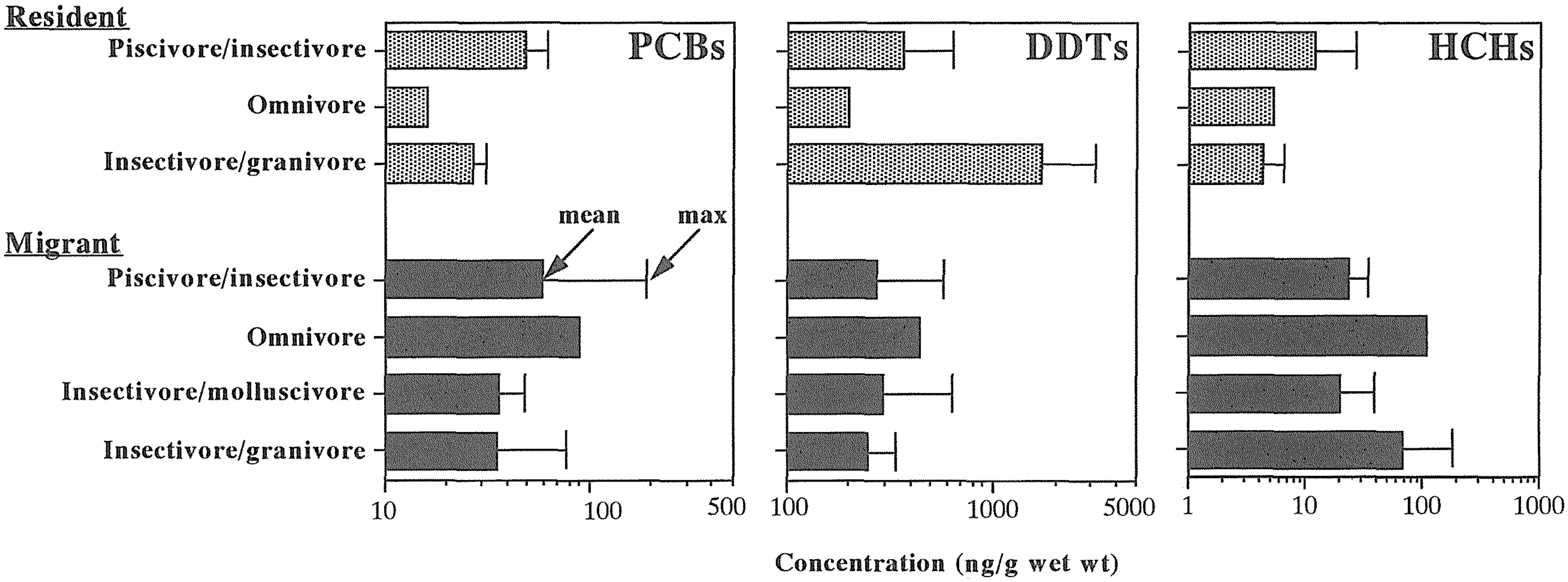

Figure 2. OC concentrations in birds according to the feeding habit. Residents: piscivore/insectivore (black-capped kingfisher, common kingfisher cinnamon bittern, white-throasted kingfisher); omnivore (common moorhen); insectivore/granivore (white-breasted waterhen, slaty-breasted rail). Migrants: piscivore/insectivore (common redshank, little tern, short- and long-billed Mongolian plover, whiskered tern, spotted redshank, marsh sandpiper); omnivore (gull-billed tern); insectivore/molluscivore (kentish plover, bar-tailed godwit, dunlin, terek sandpiper, red knot); insectivore/granivore (rufous-necked stint, grey plover, whimbrel, great knot). See text for further details.

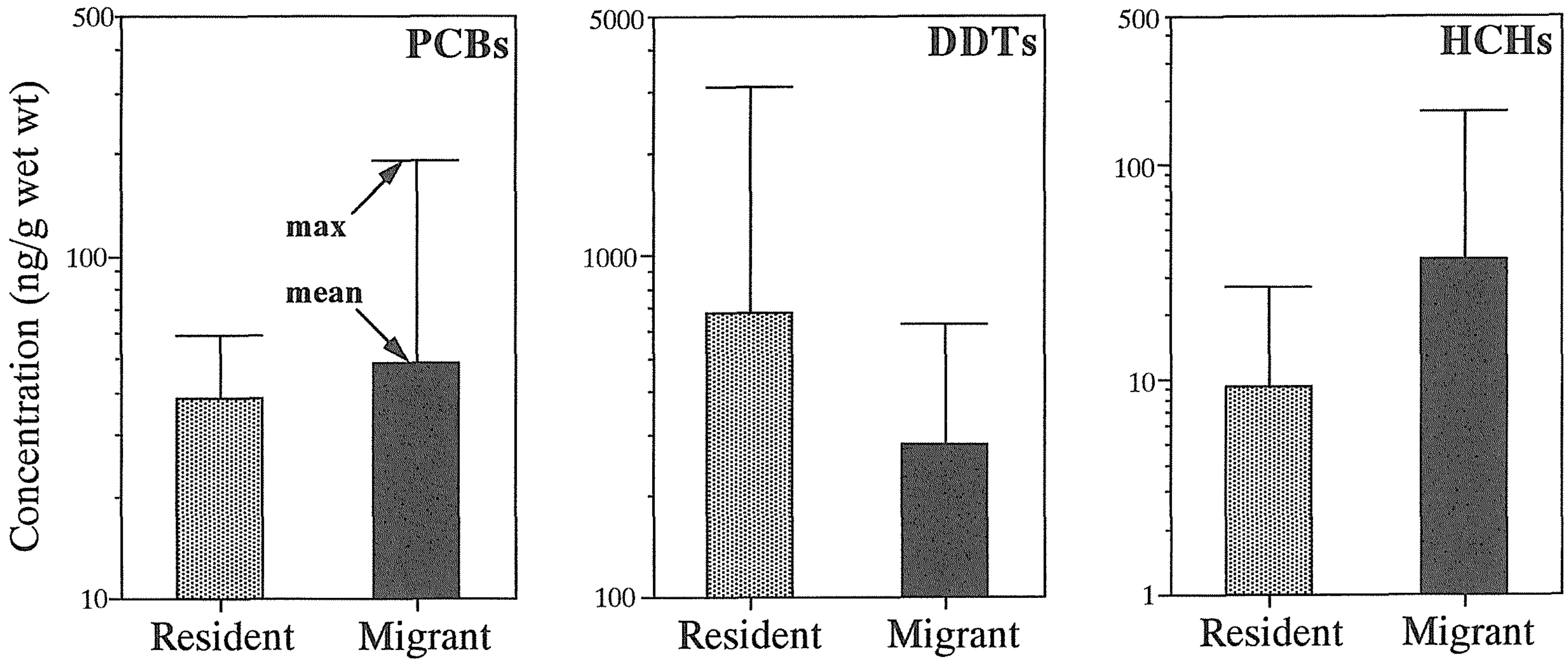

Figure 3. Comparison of OC concentrations in resident and migratory birds from North Vietnam.

OC resides were not so significant. This may partly reflect similar trophic levels of the species examined.

Unlike accumulation pattern according to the feeding habit, differences in OC accumulation between resident and migratory birds were more apparent (Figure 3). Concentrations of DDTs in resident birds were significantly higher than those in migrants, indicating recent exposure to DDTs in North Vietnam. Earlier studies have also suggested fresh input of DDTs in coastal areas and rivers of northern region as well as in industrial urban city [25,26,27]. Interestingly, accumulation of HCHs revealed contrast pattern, showing apparently greater concentrations in migratory birds. This could be due to accumulation in stopover sites during migration of migratory species in some polluted areas such as India, southern China and Japan. The role of these countries as potential source of HCH accumulation in wintering migrant birds breeding in Lake Baikal has been also suggested in our recent study [17]. Mean concentrations of PCBs were similar in residents and migratory species, with few species accumulated relatively higher levels. However, PCB accumulation in birds from Vietnam was generally low, indicating small source of PCBs in North Vietnam in recent years. Overall, bioaccumulation profile of OCs in birds from North Vietnam can be characterized by elevated accumulation of DDTs due to the recent usage of this insecticide. Local source of PCBs in North Vietnam seems to be less pronounced; they therefore accumulated to only minor extents in avian species.

Composition of HCHs and DDTs

Among HCH isomers, β-HCH was predominant isomer (Figure 4) reflecting its more stable nature than other isomers toward enzymatic degradation. Moreover, α-HCH occupied relatively high percentage in the diets than almost bird species, suggesting that bird from North Vietnam may have ability to metabolize α-HCH. Some species such as common kingfisher, Mongolian plover, dunlin and great knot showed relatively higher percentage of α-HCH than other species, probably reflecting accumulation pattern in their fish diet or recent exposure in stopover sites during migration.

Composition of *p,p'*-DDE was highest among DDT compounds, indicating greater ability to transform *p,p'*-DDT to *p,p'*-DDE (Figure 4). Interestingly, some residents and migrants such as white-breasted waterhen, common kingfisher, kentish plover, long-billed Mongolian plover, great knot and red knot comprised relatively larger proportion of *p,p'*-DDT than those in other birds, again suggesting recent exposure to DDTs in Vietnam. Particularly, white-breasted waterhen contained greatest proportion of *p,p'*-DDT, about 50 % of total DDT. Considering the fact that this species also accumulated elevated residues of DDTs, it can be suggested that waterhen may have specifically low capacity to metabolize DDT compounds, in addition to high degree of recent exposure. In the case of south India, composition of DDT compounds in relatively related species exhibited different pattern, in which *p,p'*-DDT comprised only minor proportions [15]. Despite the efficient capacity to transform *p,p'*-DDT to *p,p'*-DDE in avian species, high composition of *p,p'*-DDT was found in many species of both residents and migrants collected from North Vietnam, clearly indicating extensive exposure to technical DDT used in Red River watershed. From ecotoxicological point of view, this fact is a matter of concern since avian system living and wintering in Vietnam may experience harmful effects such as eggshell thinning and population decline as observed in some highly industrialized areas in North America and Western Europe.

Isomer-specific accumulation profile and metabolic capacity of PCBs

Isomer-specific analysis revealed the presence of about 50 chlorobiphenyl (CB) congeners. PCB isomers and congener patterns in some representative species of resident and migratory birds and their diet are shown in Figure 5. Penta- and hexachlorobiphenyls, particularly CB-99, 105, 118, 138, 153 and 180 were predominant congeners.

In general, congener CB-153 was abundant in almost species, which further supported to

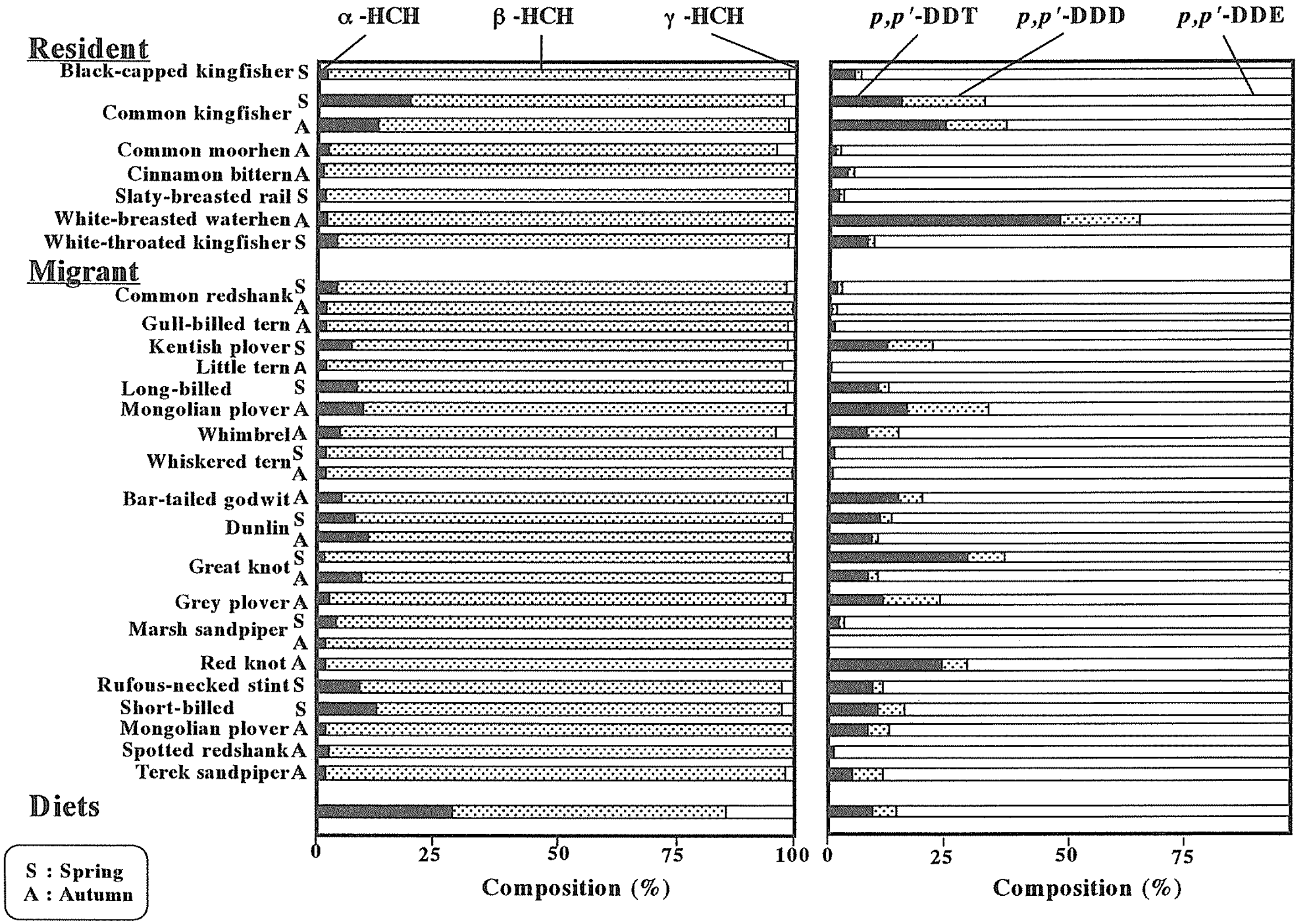

Figure 4. Compositions of HCH isomers and DDT compounds found in resident and migratory birds from North Vietnam.

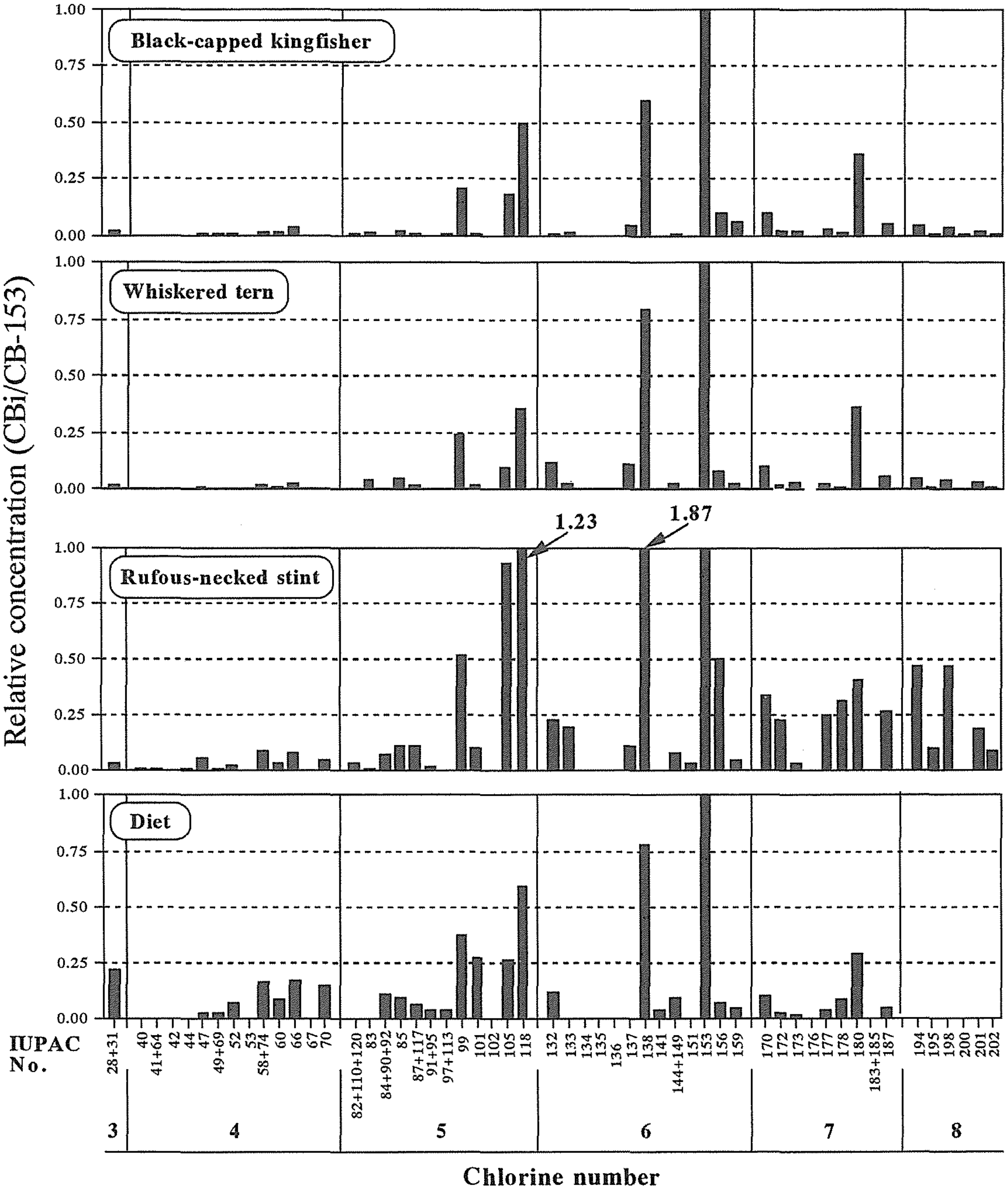

Figure 5. PCB isomer and congener compositions of some species of resident and migratory birds and their diet collected from North Vietnam. Black-capped kingfisher: resident. Whiskered tern, rufous-necked stint: migrants. Vertical bars represent concentrations of individual congeners relative to the most abundance congeners (CB-153), which were treated as 1.0.

the observations regarding the preferential persistence of this congener in birds. Similar results were also observed in other studies with aquatic birds [28-30]. Among migrants, most of birds showed similar patterns. However, specific pattern was observed in rufous-necked stint, showing congener CB-138 and CB-118 as predominant congeners. In addition, higher chlorinated congeners (hexa- and heptachlorobiphenyls) were also accounted for relatively high proportion in this species. This may probably be due to species-specific accumulation of this species. In general, accumulation of PCB isomers in birds from Vietnam was somewhat similar to that observed in related species from south India [30].

To further understand the degradation capacity and explain the specific isomer pattern of shore birds analyzed in this study, we attempted to estimate the capacity of metabolism using metabolic index proposed by Tanabe *et al.* [31].

$$MI_i = \mathrm{Log}\,(CR_{180}/CR_i)$$

Where MI_i is metabolic index of PCB isomer i; CR_{180} is the concentration ratio of CB-180 in bird and the diet; CR_i is the concentration ratio of the congener i. Details regarding this concept were described in the previous study [31].

PCB congeners are metabolized by hepatic microsomal drug-metabolizing enzyme systems, and metabolism process is enhanced by Phenobarbital (PB) and 3-methylcholanthrene (MC) type enzymes. Congeners possess vicinal nonchlorinated *meta-para* carbons and *ortho-meta* ones are metabolized by PB- and MC-type enzymes, respectively. CB-52 and 66, having 2 adjacent nonchlorinated *meta-para* carbons and *ortho-meta* carbons, respectively, were selected for calculation of MI. MI values of these 2 congeners can be used to estimate the extent of activities of PB- and MC-type enzymes [31]. Accordingly, estimated PB- and MC-type enzyme activities of birds from Vietnam in comparison to other high trophic waterbirds and animals were described in Figure 6. Since diet of these species comprises a variety of items and only small fishes and shrimps were obtained, we estimated MI values for only some representative species whose have small fish as main diet, such as black-capped kingfisher, whiskered tern, long-billed Mongolian plover and common redshank. Interestingly, estimated PB-type enzyme activities of some species such as black-capped kingfisher and whiskered tern were comparable to those in common cormorant, but higher than those in other higher trophic species such as kite, puffin and gull. While MC-type enzyme activities seem comparable or slightly higher in these species. Although MI values were estimated based on assumption that fish comprise 100 % of bird diet, higher PB- and to some extents, MC-type enzyme activities in shore birds from Vietnam suggest that these species may have higher capacity to metabolize PCB congeners, which may further explain relatively low levels of PCBs and other OCs in shore birds as compared to high trophic top predator species.

The estimated enzyme activities can be considered to further clarify the specific isomer pattern observed in rufous-necked stint, which have specific pattern with CB-118 and CB-138 as predominant congeners. Since this species did not feed on fish, but small insects, plant and crustaceans, data for small shrimps were used for estimating enzyme activities. Based on this assumption, the estimated MI values representing PB-type and MC-type enzyme activity would be 0.62 and 0.51, respectively. These values were apparently lower than those in other species, indicating weaker ability to metabolize PCB congeners in this species. Metabolism of CB-138 and 118 involves induction of both PB- and MC-type enzymes, while CB-153 metabolized by PB-type enzyme [32]. The predominance of CB-118 and 138 over CB-153 observed in rufous-necked stint can be supported by the less active MC-type enzyme in this species. It has been reported that aquatic mammal such as Ganger river dolphin with inactive MC- and PB-type enzymes, higher abundance of CB-138 was also observed [18].

International comparison and the role of the Southeast Asian region as emission source of contamination

To understand the magnitude of contamination in avian species from Vietnam, OC residues in birds reported for countries in Asia-Pacific were compared (Table 3). Since data

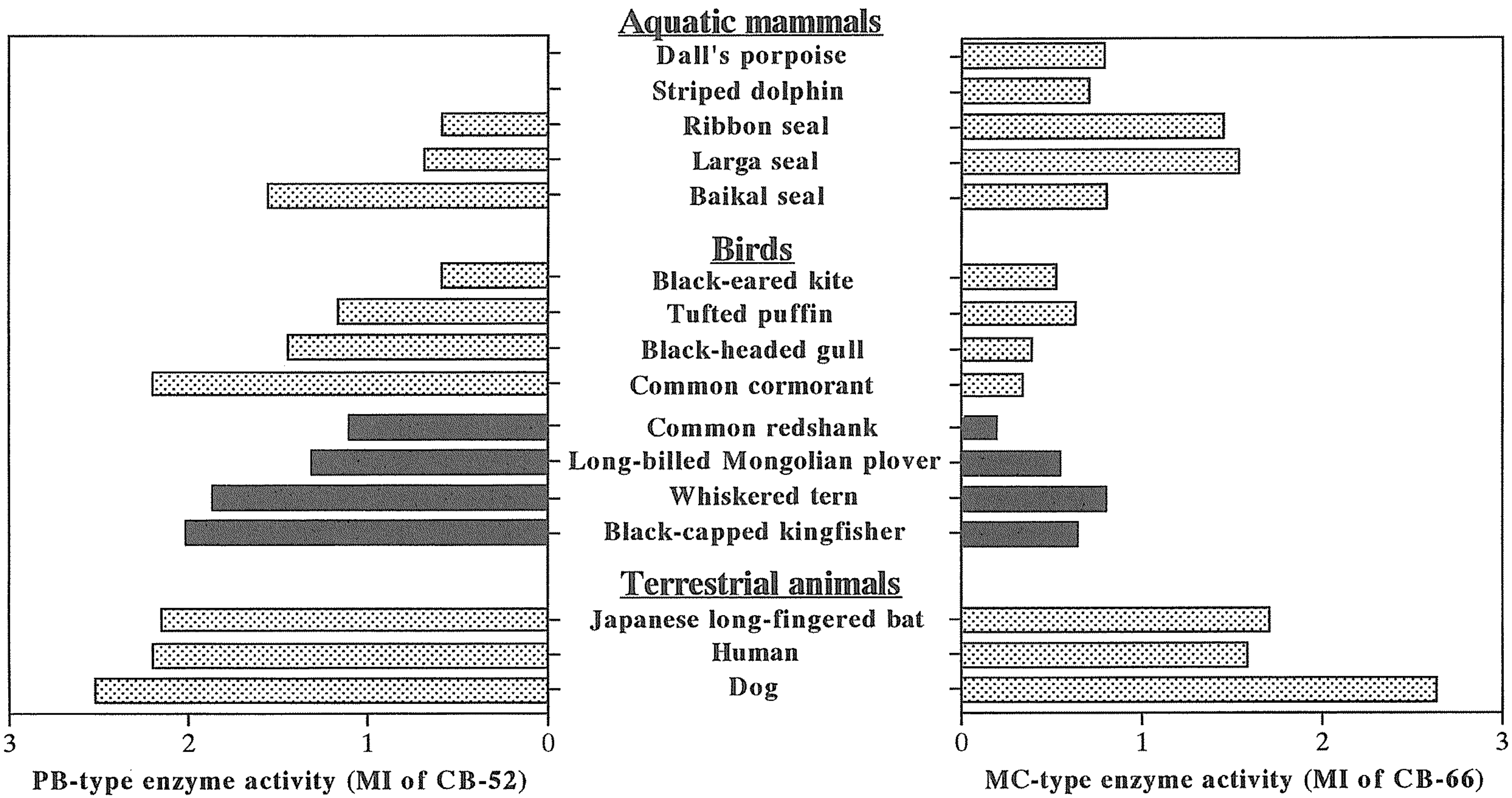

Figure 6. Comparison of estimated PB- and MC-type enzyme activities in higher trophic animals by metabolic indices of CB-52 and 66. Black bars represent enzyme activities of Vietnamese birds. Data for other animals were cited from Tanabe *et al.* [31], Nakata *et al.* [16] and Guruge and Tanabe [29].

Table 3. Comparison of recent OC residues (ng/g lipid wt) in birds from different countries in Asia-Pacific region.

Location	Species	Analytical tissue	Collected year	PCBs	DDTs	HCHs	Reference
Lake Baikal, Russia	Resident						
	House sparrow	Breast muscle	1996	1000	190	60	Kunisue *et al* . [16]
	Carrion crow	Breast muscle	1996	5000	2700	240	Kunisue *et al* . [16]
	Grey heron	Breast muscle	1996	240	840	75	Kunisue *et al* . [16]
	Migrant[a]	Breast muscle and whole body	1996	7100[b] (20-78000)[c]	3900 (42-17000)	730 (7.9-5100)	Kunisue *et al* . [16]
Chubu, Japan	Resident						
	Carrion crow	Liver	1991-1993	980	200	24	Hoshi *et al* . [36]
	Common cormorant	Liver	1991-1993	6400	1900	100	Hoshi *et al* . [36]
	Black kite	Liver	1991-1993	2100	340	75	Hoshi *et al* . [36]
North Vietnam	Resident[d]	Whole body	1997	780 (250-2400)	10000 (1400-27000)	150 (23-310)	Present study
	Migrant[e]	Whole body	1997	530 (82-1600)	2900 (750-6800)	330 (20-1700)	Present study
South India	Resident[f]	Whole body	1995	490 (150-1900)	5300 (6-26000)	13000 (190-73000)	Tanabe *et al* . [14]
	Migrant[g]	Whole body	1995	5900 (190-40000)	11000 (86-58000)	6200 (270-26000)	Tanabe *et al* . [14]
Australia	Resident						
	European starling	Whole body	1986	not reported	0.2 - 2.0[h]	not reported	Olsen *et al* . [37]
	Peregrine falcons	Egg	1980s	not reported	15000[h]	not reported	Olsen *et al* . [37]

[a]Including 13 species. See Kunisue *et al* . [16] for details.
[b]Mean concentrations of the analyzed species.
[c]Range concentrations of the analyzed species
[d]Including 7 species. See Table 2 for details.
[e]Including 17 species. See Table 2 for details.
[f]Including 11 species. See Tanabe *et al* . [14] for details.
[g]Including 12 species. See Tanabe *et al* . [14] for details.
[h]Concentrations in lipid wt were calculated assuming a lipid content of 10 %.

were reported for different tissue, we used lipid-normalized concentrations for comparison. PCB concentrations in resident birds from North Vietnam were in the range to those reported for Indian birds, but lower than those in birds from Lake Baikal, Russia and Japan. Extent of PCB contamination in each location is consistent with known current status of usage. Accordingly, smaller source of PCBs in developing countries is a reason for less degree of exposure, while recently local source in eastern Siberia and historically heavy usage in Japan elevated PCB concentrations in biota. HCH concentrations in Vietnamese birds were similar to those reported for Lake Baikal and Japanese birds, but several orders of magnitude lower than those in Indian birds. Interestingly, Vietnamese birds accumulated greatest DDT residues, higher than those in birds from India by a factor of 2 and at least 10-fold for lake Baikal and Japanese birds. Although DDT levels in migratory birds were still lower than those in India and lake Baikal, elevated residues found in resident species again indicate recent usage of DDT in Vietnam. DDT residues in birds from Vietnam were even comparable or higher than those in avian species recently reported in some locations in the United States and western Europe, which are known as heavily polluted areas due to the huge historical production and usage. For examples, migrant passerine species from Illinois, US collected in 1996 accumulated *p,p'*-DDE levels in the range of 1.4 – 390 ng/g wet wt, which were less than those in Vietnamese birds [33]. Little tern collected from the Baltic Sea in 1995-1996 contained a mean *p,p'*-DDE level of 400 ng/g wet wt, which was in the range to that reported for birds from North Vietnam [34]. These data indicate the fact that despite OC residue concentrations in avian systems in developed nations have declined gradually due to the ban of these chemicals, recent DDT contamination found in birds from developing countries located in tropical region such as Vietnam may continuously be elevated due to current application of this insecticide for malaria eradication program as well as agricultural purposes.

The present accumulation pattern in birds as discussed above suggests that developing countries in Southeast Asian region may serve as potential emission source of OC pollution in recent years. To further make clear this observation, we compiled data in fish and mussels derives from the Asia-Pacific Mussel Watch Program, particularly in Asian developing countries recently conducted in our laboratory (Table 4). Concentrations of PCBs in Vietnamese fish and mussels were lower than those in developed nations in region but comparable to those from India and Philippines. Particularly, these levels were higher than those reported for other developing countries in Southeast Asia, such as Cambodia, Thailand, Indonesia and Malaysia, indicating greater source of PCB in Vietnam, probably due to the release from weapons used during Indochina War [12]. Interestingly, as for HCHs and DDTs, concentrations of these chemicals in fish and mussels from Vietnamese coastal waters are among the highest values reported for the countries surveyed. HCH residues were highest in India, a main HCH user in the world, followed in Vietnam. Similar to the result observed in birds, Vietnamese fish and mussels contained greatest DDT concentrations. Survey conducted during 1989-1993 and recent years have also indicated elevated DDT concentrations in atmospheric, hydrospheric and biotic samples from both north and south Vietnam [7,8,12,26]. From the temporal point of view, DDT residues in fish analyzed in the present study were higher than observed in 1989 (Table 4). Nhan *et al.* [27] found high levels of DDTs in industrialized areas in capital Hanoi, and suggested recent usage of DDT for sanitary purposes, particularly mosquito eradication. Information of DDT usage in Vietnam is rather limited. According to the domestic statistical data, DDT was imported and used in Vietnam with a quantity of approximately 25,000 tons during 1957-1995. Despite the use of DDT was officially banned in 1993, large amounts of DDT have been in storage until recently [35,36]. As for comparison, cumulative production of DDT in India until 1995 was estimated about 500,000 tons [8]. In fact, residues in birds and river dolphins from India did not appear to have declined until recently [15,18]. Compared to India, although usage quantity of DDT in Vietnam is substantially lower, illegal application of this insecticide is probably the main reason for high DDT levels found in various environmental compartments in Vietnam. Thus,

Table 4. Comparison of OC residues (ng/g lipid wt) in fish and bivalve from different countries in Asia-Pacific region.

Location/country	Species	Analytical tissue	Collected year	PCBs	DDTs	HCHs	Reference
Lake Baikal, Russia	Fish	Whole body	1992	1700	1100	19	Nakata *et al* . [15]
Lake Biwa, Japan	Fish	Whole body	1993	3700	1900	240	Guruge *et al* . [38]
Tokyo Bay, Japan	Blue mussel	Whole body	1998	5100	250	8.3	Ueno *et al* . [39]
India	Fish	Muscle	1989-1993	150	630	1200	Kannan *et al* . [8]
	Fish	Whole body	1998	100	88	390	Senthilkumar *et al* . [40]
	Green mussel	Whole body	1998	240	260	120	Monirith *et al* . [41]
Vietnam	Fish	Muscle	1989-1993	530	1400	95	Kannan *et al* . [8]
	Fish	Whole body	1997	110	4200	120	Present study
	Green mussel	Whole body	1997	130	4000	5.5	Monirith *et al* . [41]
	Clam	Whole body	1997	820	840	600	Nhan *et al* . [25]
Cambodia	Fish	Whole body	1998	13	300	2	Monirith *et al* . [41]
	Green mussel	Whole body	1998	57	38	1.5	Monirith *et al* . [41]
Thailand	Fish	Muscle	1989-1993	30	120	15	Kannan *et al* . [8]
	Green mussel	Whole body	1994-1995	170	310	11	Kan-Atireklap *et al* . [42]
Philippines	Green mussel	Whole body	1998	320	28	2.2	Monirith *et al* . [41]
Indonesia	Fish	Muscle	1989-1993	86	930	24	Kannan *et al* . [8]
	Green mussel	Whole body	1998	93	73	2.7	Monirith *et al* . [41]
Malaysia	Green mussel	Whole body	1998	77	110	7.7	Monirith *et al* . [41]
Australia	Fish	Muscle	1989-1993	1600	650	10	Kannan *et al* . [8]

monitoring studies based on both spatial and temporal terms were somewhat consistent with statistical data regarding the present status of usage; and this fact suggests that beside India, Vietnam has become a potential source of DDT pollution in Southeast Asian region.

Conclusion and recommendation for future research

Recent monitoring surveys in birds and Mussel Watch Program conducted in our laboratory have revealed that contamination by OC insecticides, particularly DDTs was apparent in Vietnamese environment. In the previous study, we have suggested that India, Japan and south China are major source of pollution for birds breeding in Lake Baikal and wintering in southern region [17]. Our results of Vietnamese birds have further highlighted the role of tropical developing countries as emission source of semivolatile persistent OCs. In this context, India can be considered as a major origin of HCHs while Vietnam may become a potential emission source of DDTs in a global perspective. Result from this study also indicated that despite relatively rapid reduction of OC residues in developing nations during the last decades, the status of contamination in developing countries seems to become more serious in future. Further comprehensive investigations regarding temporal trend of OC contamination in Asian developing environments are therefore needed to trace the source and assess possible long-term impacts of OCs in tropical ecosystem. Ecotoxicological studies on adverse effects of insecticide, particularly DDE, known as potential contaminant caused egg shell thinning in avian species, are also required, considering the fact that very little information is available regarding this aspect in tropical developing countries.

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