

# Accumulation of trace elements and persistent organochlorines in resident and migratory waders from Calatagan Bay, Philippines

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**Abstract**—Concentrations of V, Cr, Mn, Co, Cu, Zn, Se, Rb, Sr, Mo, Ag, Cd, Sb, Cs, Ba, Hg, Tl and Pb were determined in the liver of six species of resident and migratory waders collected in December 1997 from Calatagan Bay, Philippines. Renal Cd concentrations in these species were also determined. Trace element levels were observed to have either decreased or remained intact in the winter, suggestive that trace element burdens for migratory waders could be minimal during wintering. It seems that the migration distance of waders is a dominant factor in the accumulation of toxic elements such as Cd, Hg and Pb. Hepatic Cd concentrations of pintail snipe are likely to be incritical levels. While concentrations of persistent organochlorines such as PCBs, DDTs, HCHs, CHLs and HCB were determined in the whole bodies and adipose tissues of migratory and resident waders collected in April and December 1994. Accumulation patterns of OCs found in resident birds suggested that PCBs and CHLs were the predominant contents. Among the OCs, It was noted that migratory birds retained mostly high concentrations of DDTs. This finding could be reflective of the presence of these contaminants in the stopover and breeding/wintering ground of these waders in China and Russia. Whereas residual concentrations found in resident birds reflect the pollution status of the sampling area, those found in migratory waders reflect both the pollution status of the sampling area and their migratory routes. Waders are therefore useful bioindicators to elucidate the contamination status of toxic metals and organochlorines in breeding grounds, stopover sites and wintering grounds.

**Key words:** Organochlorines, heavy metals, migratory waders, Philippines, East Asia

## Introduction

Waders are widely distributed in the world, and most of these waders spend their winter in tropical or subtropical areas and breed in the high latitudes of the northern hemisphere and the Arctic circle. In view of their long distance migration, these birds are affected by the local pollution not only in their wintering or breeding grounds but also in their stopover sites along their migratory routes. Furthermore, these waders drastically feed in stopover sites, such as wetlands and mudflats, in order to store enough energy during their short stay. Wetlands have the capacity to deposit both organic and inorganic pollutants including toxic substances. Thus, heavy metal pollution originating from industrial sources and mining activities, land use changes and the ap-

plication of pesticides could pose a serious threat to these avian species (White et al. 1983, Custer and Myers 1990). Consequently, exposure to toxic elements and organochlorine compounds (OCs) is an important consideration in the conservation and management of waders. Although studies on the accumulation of these in waders have been done in Europe (Senner and Howe 1984, De Voogt et al. 1985, Everaarts et al. 1991, Lambeck et al. 1991), such investigations have been scarcely done in Asian developing nations (Yasunaga et al. 2000, Kunisue et al. 2003).

The present study is part of a cooperative project on global monitoring of toxic chemicals using migratory shorebirds which aims primarily to investigate the accumulation and effects of toxic contaminants in migratory waterbirds collected from the wetlands of Asia and Oceania, and to develop a global monitoring system of contaminants using wa-

terbirds as bioindicators. The research area includes the Philippines, which has many ideal migratory grounds for waterbirds. Moreover, the Philippines is part of the east Asia flyway along which birds move to their non-breeding quarters. Those migrating from Japan pass through Taiwan and into the Philippines while those from the eastern seaboard of China will either pass through Taiwan and south into the Philippines or keep heading west into Thailand and Malaysia. Some 104 bird species including waders such as plovers and turnstones visit the country, where they either winter or pass through on their regular migration from northern Asia to the Malay archipelago and Australia (Gonzales and Rees 1988).

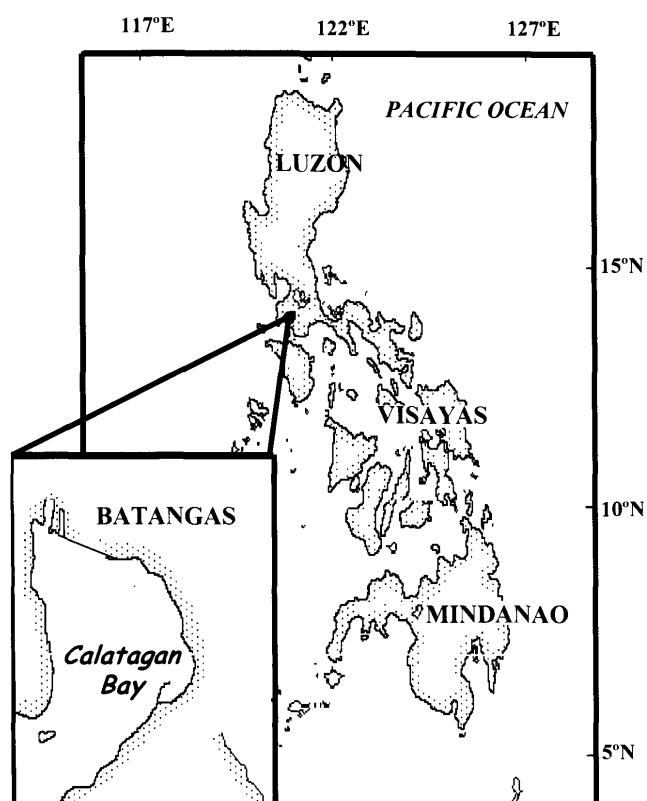


Fig. 1. Sampling location.

The wader samples in this study were collected from Calatagan Bay, a small coastal inlet of the South China Sea bordering the province of Batangas, located south of Metro Manila area (Fig. 1). The bay is often visited by a considerable number of birds especially waders due to its varied habitat consisting of extensive sand and mudflats and mangrove swamps.

An insight into the specific accumulation of trace elements and organochlorines (OCs) in resident and migratory waders is presented and comparison of residual composition of toxic elements in the liver and Cd in the kidney tissues, as well as OCs residues in their whole bodies is discussed. Moreover, residual concentrations detected in resident and migratory waders analysed here were compared with values from breeding, wintering and stopover sites in Asia to determine the degree of contamination in the study area.

## Materials and Methods

### Sampling

For the accurate elucidation of the accumulation features of these contaminants, the waders analysed in this study were grouped. According to Hoyo et al. (1996), waders could be classified into three groups namely: *resident*—which live almost in the same region all through the year for their entire life span such as painted snipe, chinese little bittern, Schrenck's little bittern and green-backed heron; *short-distance migrant*—which have their breeding grounds in central China to Japan such as little-ringed plover, Kentish plover and long-billed mongolian plover; and *long-distance migrant*—which have their breeding grounds in wide range of northeastern Europe to southeastern Russia such as short-billed mongolian plover and ruddy turnstone. The waders analysed for trace elements (Table 1) were collected by mist nets under government permission in September 1993, coinciding with the peak season of migration, while the birds

Table 1. Mean biometric data of waders analysed for trace elements (Ranges are given in parentheses).

Species	N	Body length (cm)	Migration type
Painted snipe ( <i>Rostratula benghalensis</i> )	10	23.6 (21.6–25.3)	resident
Little-ringed plover ( <i>Charadrius dominica</i> )	3	16.9 (16.4–17.5)	short-distance
Kentish plover ( <i>Charadrius alexandrius</i> )	5	17 (15.4–18.8)	short-distance
Great sand plover ( <i>Charadrius leschenautii</i> )	8	19.7 (17.7–21.5)	short-distance
Common sandpiper ( <i>Actitis hypoleucos</i> )	4	19.5 (18.8–20.3)	long-distance
Pintail snipe ( <i>Gallinago stenura</i> )	3	25.5 (23.6–27.2)	long-distance

N: no. of samples.

**Table 2.** Mean biometric data of waders analysed for OCs (Ranges are given in parentheses).

Species	N	Fat %	Body length (cm)	Body weight (g)
<b>Residents</b>				
Painted snipe ( <i>Rostratula benghalensis</i> )	6	5.9 (3.6–6.8)	22.5 (19.8–24.2)	128 (120–136)
Chinese little bittern ( <i>Ixobrychus sinensis</i> )	3	11 (6.2–18)	34.6 (33.9–35.7)	73.8 (72.7–75.5)
Schrenck's little bittern ( <i>Ixobrychus eurhythmus</i> )	2	11 (7.9–14)	37.9 (37.5–38.2)	181 (177–188)
Green-backed heron ( <i>Butorides striatus javensis</i> )	3	14 (13–14)	43.7 (42.0–46.2)	164 (125–197)
<b>Migrants</b>				
Ruddy turnstone ( <i>Arenaria interpres</i> )	3	9.7 (7.9–12)	24.7 (21.0–25.3)	95.5 (82.4–114)
Little-ringed plover ( <i>Charadrius dominica</i> )	3	12 (6.2–18)	15.5 (15.2–15.7)	45.1 (33.4–62.9)
Kentish plover ( <i>Charadrius alexandrius</i> )	13	15 (8.7–21)	16.3 (14.5–18.0)	34.2 (30.1–41.4)
Mongolian plover ( <i>Charadrius mongolius</i> )				
short-billed Mongolian plover	5	12 (8.4–17)	17.7 (17.1–18.4)	51.4 (42.2–56.2)
long-billed Mongolian plover	5	12 (5.1–29)	20.5 (18.8–21.4)	72.3 (61.4–84.4)

N: no. of samples.

considered for OCs residues (Table 2) were collected in April and December 1994. Samples were transported to Japan and stored at  $-20^{\circ}\text{C}$  until dissection and analyses. Biometric measurements were conducted prior to dissection and analysis. All wader samples collected for trace element analyses were dissected and the liver and kidney were taken for analyses, while those considered for OCs residues were homogenized for whole-body burden analysis and subsample of the adipose tissues of Ruddy turnstones and Kentish plovers were also examined.

## Chemical analyses

### Trace elements

The livers and kidneys were stripped externally to avoid contamination. Tissue samples were dried in an oven for 12 h at  $180^{\circ}\text{C}$  and digested in microwave using nitric acid in a PTFE (teflon) vessel. Seventeen trace elements (V, Cr, Mn, Co, Zn, Cu, Se, Rb, Sr, Mo, Ag, Cd, Sb, Cs, Ba, Tl and Pb) were measured by inductively coupled plasma-mass spectrometry (ICP-MS:Perkin-Elmer elan 5000) using external standard method with In as an internal standard within 24 h. Mercury concentrations were determined by cold vapor atomic absorption spectrometry (Shimadzu AA-680). Concentrations were given on a dry weight basis. Accuracy and precision of the methods were confirmed using bovine liver (NISTNo. 1577b). Accuracy of the element levels were assessed through standard addition method and correction factors were determined for external standard method. Probabil-

ity values less than 0.001 were considered as significant using Kruskal-Wallis tests in differences among habitats and migrant types.

### Organochlorine residues

The extraction, clean-up and separation procedures of OCs and analysis of lipid content were similar to those previously described by Tanabe et al., (1998). Quantification of organochlorines was made on a gas chromatograph (Hewlett Packard 6890 series) equipped with ECD (electron capture detector) and an automatic injector (Hewlett-Packard 7683 series) The GC column used was a fused silica capillary (DB-1; J&W Scientific, 30 m length,  $0.25\text{ }\mu\text{m}$  i.d. and  $0.25\text{ }\mu\text{m}$  film thickness). Helium was used as a carrier gas while nitrogen was the make-up gas. The concentration of individual organochlorines was quantified from the peak area of the samples to that of the corresponding external standard. The PCB standard used for quantification was an equivalent mixture of Kanechlor preparations (KC-300, KC-400, KC-500 and KC-600) with known PCB composition and content. Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain the total PCB concentrations. Recoveries through this analytical method were  $97\pm 4.2\%$  for PCBs,  $105\pm 5.7\%$  for DDTs,  $98.9\pm 6.3\%$  for HCHs,  $103.9\pm 4.3\%$  for CHLs and  $104.1\pm 7.9\%$  for HCB, respectively. Concentrations were not corrected for recovery rates. Standard reference material SRM 1945 was analyzed for selected PCB congeners and persistent

organochlorines. Reliable results were obtained by comparison of data from our laboratory with those from material reference values.

## Results and Discussion

### Accumulation of trace elements

The concentrations of 18 elements in the liver and Cd in the kidney of waders is presented in Table 3. It is well known that concentrations of essential elements in the liver are not relatively affected by several varying environments due to homeostasis. No significant difference in hepatic V, Cr, Mn, Co, Cu, Zn, Mo and Se concentrations was observed in the waders analysed, except for the pintail and painted snipes, which have different feeding habit. The levels of V, Mn, Co, Cu, Zn and Mo found were within the same range to the previously reported levels found in waders from Hong River in Vietnam and those birds collected from Porto Novo in India (Yasunaga et al. 2000). In the case of the toxic heavy metals, the concentrations of Cd, Hg and Pb were proximal in residual values found in the waders from Vietnam and India (Yasunaga et al. 2000). Hepatic levels of Ag, Sb, Cs and Tl were either equivalent to or below detection limit. Differences in element levels observed among different species remain unclear. Although the levels of Rb, Ba and Sr were found to be abundant, the environmental and biological behaviour of these elements remain unknown.

In order to discuss the variation of trace element concentrations according to locations, kentish plovers were considered due to their short-distance migration, which included three countries (India, Vietnam and Philippines). Subpopulations of kentish plovers collected from Vietnam and Philippines are known to breed in the southern Eurasian Continent (about 40°N) and those collected from southern India mainly breed on the Himalaya Mountains' foot (Hoyo et al. 1996). Significant differences in hepatic V, Cr, Co, Se, Mo, Cs, Tl, Pb, Mn, Zn, Cu and Rb concentrations were found among kentish plovers collected from India, Vietnam and Philippines. While concentrations of essential elements such as Co, Mo, Mn, Zn and Cu in kentish plovers analysed in this study were similar to those observed in waders from USA and Europe. However, Se levels in this wader species from the Philippines were closer in values to the waders from a polluted area in California (Yasunaga et al. 2000). On the other hand, hepatic Hg and Cd concentrations in kentish plovers from the Philippines were similar to those from Vietnam and India. The reason for the high Se levels in kentish plovers from the Philippines remain unclear, however, some oxidative stresses could be a plausible explanation. Hepatic Tl and Cs concentrations were generally low in birds from India, Vietnam and Philippines and these elements were of similar concentrations to the values of detection limit.

**Table 3.** Mean concentrations ( $\mu\text{g/g}$  dry wt) of trace and essential elements in the liver of waders Calatagan Bay, Philippines collected in December 1997.

Species	N	V	Cr	Mn	Co	Cu	Zn	Se	Rb	Sr	Mo	Ag	Sb	Cs	Ba	Tl
Painted snipe ( <i>Rostratula benghalensis</i> )	10	0.42	1.30	9.9	0.30	150	125	8.70	19.1	1.18	2.43	<0.001	0.082	0.036	1.30	0.020
Little-ringed plover ( <i>Charadrius dominical</i> )	3	0.29	1.50	14.6	0.25	18.1	102	7.10	6.95	2.20	2.83	<0.001	0.087	<0.01	1.70	0.007
Kentish plover ( <i>Charadrius alexandrius</i> )	5	0.17	1.30	12.2	0.22	22.9	169	16	8.36	4.03	2.47	<0.001	0.084	0.022	0.98	0.006
Great sand plover ( <i>Charadrius leschenautii</i> )	8	0.22	1.20	11.7	0.21	21.1	144	14	7.86	1.97	2.89	<0.001	0.085	0.025	6.20	0.003
Common sandpiper ( <i>Actitis hypoleucos</i> )	4	0.27	1.40	13.1	0.17	20.7	167	9.1	7.97	2.27	3.48	<0.001	0.083	0.028	0.51	0.007
Pintail snipe ( <i>Gallinago stenura</i> )	3	1.10	4.60	35.5	1.10	572	390	40	5.83	2.70	6.80	<0.001	0.084	0.022	0.65	0.010

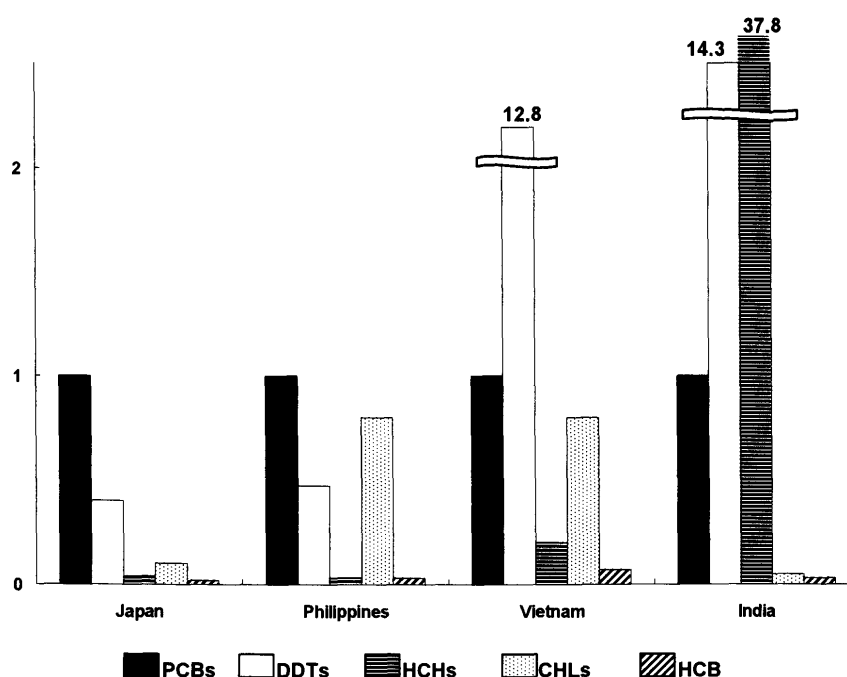
N: no. of samples.

**Table 4.** Mean concentrations ( $\mu\text{g/g}$  dry wt) of toxic heavy metals in the liver of waders from Calatagan Bay, Philippines.

Species	N	Cd	Cd-k	Hg	Pb
Painted snipe ( <i>Rostratula benghalensis</i> )	10	0.23	0.97	0.27	0.030
Little-ringed plover ( <i>Charadrius dominica</i> )	3	0.60	1.15	0.47	0.045
Kentish plover ( <i>Charadrius alexandrius</i> )	5	1.42	8.50	0.95	0.053
Great sand plover ( <i>Charadrius leschenautii</i> )	8	1.48	6.88	0.56	0.260
Common sandpiper ( <i>Actitis hypoleucos</i> )	4	3.72	23.70	0.57	0.055
Pintail snipe ( <i>Gallinago stenura</i> )	3	12.7	7.54	0.29	0.163

N: no. of samples.

Cd-k: Cd in kidney.

**Fig. 2.** Mean relative concentrations of OCs to PCBs in resident birds from Calatagan Bay, Philippines, Japan, India and Vietnam.

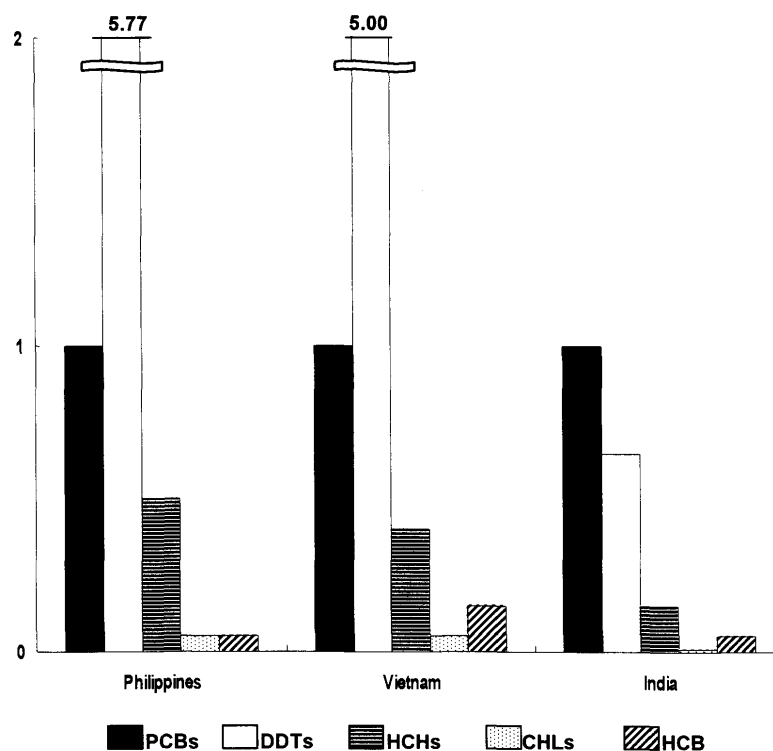
No regional significant differences were observed in hepatic Cd concentrations among the kentish plovers. Although the hepatic Cd concentrations in all the species from Vietnam and those from India were higher than those collected from the Philippines ( $p < 0.05$ ), the ratio of Cd kidney/liver concentrations in the Philippines were higher than those from India and Vietnam. This is suggestive that waders from the Philippines were principally exposed to Cd in breeding area or in stopover sites other than Philippines. It has been suggested (Yasunaga et al. 2000) that decreasing ratios of Cd kidney/liver concentrations are indicative of renal disease due to dysfunctional excretion of Cd from the kidney. The waders collected from the Philippines might then be potentially adversely affected as depicted by the slightly decreasing trend in the ratio of renal and hepatic Cd.

#### Accumulation of organochlorines

Examination of the range and mean concentrations of OCs in the whole bodies of migrant and resident waders col-

lected from the Philippines revealed substantial variations among the avian species analysed. Thus, we deemed it better to evaluate the accumulation features of OCs by estimating the relative concentrations of other organochlorines to PCBs in resident birds to make the range of absolute concentrations among species small and thereby making it easier to discern which OCs mainly remain in the avian species.

Among the OCs measured in the resident waders, PCBs were the prominent compounds followed by DDTs, CHLs, HCHs and HCB respectively (Fig. 2). This pattern was found to be similar to those waders collected from Japan, suggestive that notable PCBs contamination of biota in these countries is still occurring. It can be gleaned further that the relative concentrations of CHLs in waders from Calatagan was higher than that from other countries, while the residue levels of other OCs were generally low. In the Philippines, elevated concentrations of CHLs in dumpsite soils and sediments were reported and the use of CHLs for public health purposes was suspected (Lee et al. 1997).



**Fig. 3.** Mean relative concentrations of OCs to PCBs in short-distance migratory birds from Calatagan Bay, Philippines, India and Vietnam.

As for the migrant species, the composition of OCs followed the order: DDTs>PCBs>HCHs>CHLs>HCB. Such observation also holds true with the waders collected from Vietnam. This result seem to indicate that on their migratory routes, wader species collected from Calatagan Bay could be wintering and feeding in areas of high DDTs usage. These observations confirm the notion that DDTs and PCBs are highly persistent, less biodegradable, and are retained in the animal's body for a long time (Tanabe and Tasukawa 1991). In addition, accumulation patterns in birds of short-distance and long-distance migration were apparently different from those in resident birds. Thus, present results seem to suggest that the accumulation features of OCs in migratory waders reflect not only the pollution status in the area of collection, but also in stopover sites, breeding and wintering grounds.

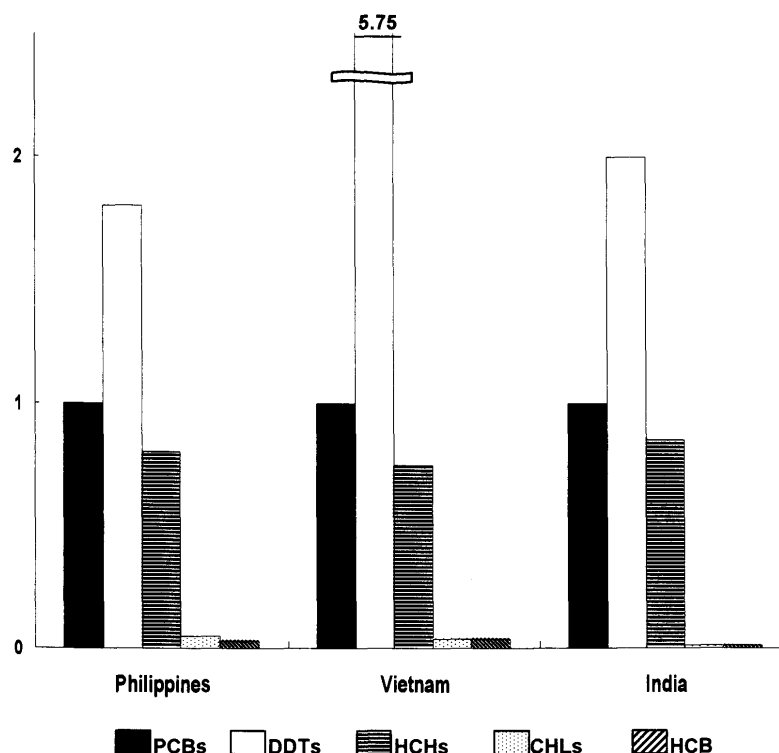
Concentrations of OCs and their mean relative concentrations to PCBs in short-distance migrants are presented (Fig. 3). It can be gathered that in short-distance migrants from the Philippines, DDTs and PCBs were the dominant contaminants. These findings also hold true for the avian samples from India. It is known that many species from India and the Philippines have their breeding grounds around Persian Gulf, Red Sea and Caspian Sea (Hoyo et al. 1996). High concentrations of PCBs were also detected in Caspian seals and fishes from Caspian Sea and PCBs' release into this environment could be continuing (Kajiwara et al. 2002, Watanabe et al. 1999). Moreover, the accumulation pattern in short-distance migrants from Calatagan Bay was almost similar to

those from Vietnam. In both countries, the relative concentrations of HCHs in short-distance migrants were slightly higher than those found in resident birds. These results are indicative that short-distance migrants from Calatagan Bay and Vietnam may have their breeding grounds or stopover sites in China, since China is known to have widely used HCHs and DDTs (Li et al. 1996, Wu et al. 1997, Zhu et al. 1999).

Long-distance migrants from Calatagan Bay showed similar trends to short-distance migrants (Fig. 4), indicating that the long-distance migrants might have been exposed to high levels of DDTs in their stopover sites and breeding grounds. This finding also holds true for the waders from Vietnam. In addition, relative concentrations of HCHs in long-distance migrants from the Philippines and Vietnam were slightly higher than those found in short-distance migrants. This finding could be suggestive that these long-distance migrant birds might have been exposed to HCHs in the Arctic regions, which are known to be polluted by HCHs due to the long-range atmospheric transport from the southern hemisphere (Muir et al. 1999).

## Conclusion

Significant differences among locations remain unclear in hepatic concentrations of trace elements collected from typical wintering areas such as the Philippines. The trace ele-



**Fig. 4.** Mean relative concentrations of OCs to PCBs in long-distance migratory birds from Calatagan Bay, Philippines, India and Vietnam.

ment levels of waders were observed to have either decreased or remained intact in the winter. Consequently it is suggestive that trace element burdens could be minimal during wintering. Results also suggest that migration distance of waders could be a dominant factor in the accumulation of toxic elements such as Cd, Hg and Pb, which could be attributed to the specific metabolism related to migration or mass feeding in stopover sites. Moreover, hepatic Cd concentrations in pintail snipes are likely to be in the critical levels. This finding could be indicative that some kind of heavy metal pollution may have reached an alarming stage especially in the case of long-distance migrant waders in Asia.

On the other hand, accumulation pattern of OCs in resident waders revealed that PCBs were the dominant contaminants, followed by DDTs>CHLs>HCHs>HCB. However, relative concentrations for CHLs among resident birds was found to be higher than those from other Asian countries, but the residue levels of other OCs were generally low. In migratory waders, DDTs were the most dominant OCs, indicating that many migratory species wintering in the Philippines are migrating and feeding in areas of high DDT usage.

This study also highlighted the use of avian species, such as waders, as useful bioindicators that could elucidate contamination status of toxic substances such as OCs, heavy metals and other trace elements in their breeding grounds, stopover sites and wintering grounds.

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