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ORGANOCHLORINE RESIDUES IN COASTAL BIRDS FROM THE VELLAR RIVER WATERSHED AREAS, PARANGIPETTAI, SOUTHERN INDIA

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Organochlorine contaminants **(HCHs,** DDTs and **PCBs)** in the muscle tissues of 12 bird species collected from Vellar River watershed areas **(S.** India) were determined. The accumulation pattern of organochlorine residues in birds of different feeding groups for **HCHs** and **PCBs** was: scavengers > inland piscivores > coastal piscivores > insectivores. The pattern for DDTs was: coastal piscivores > scavengers > insectivores > inland piscivores. Scavengers accumulated all the three organochlorines to a higher degree than the other groups of birds. Marked variations in the accumulation pattern by different species and individuals of birds to the same and different organochlorine residues were observed. The differences in the accumulation pattern of residues in different species of birds with similar feeding habits could be attributed to different feeding areas and because migrants are exposed to contaminants in different geographical locations. **HCH** residues were generally found in higher concentrations than DDTs, reflecting increasing use of **HCH** pesticides since banning of DDT for agriculture. Levels of PCBs were lower than in birds in developed countries. Results are discussed in relation to future pest control and industrialisation in India.

Keywords: Pesticides; bird species; contamination; India

1. INTRODUCTION

Chlorinated hydrocarbons occur throughout the terrestrial (Fishbein, 1972; Kaphalia and Seth, 1981) and aquatic ecosystems (Jensen *et al.,* 1969; Portmann, 1975; Tanabe *et al.,* 1984; Berg *et al.,* 1992) even

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including polar regions (Sladen *et a/.,* 1966; Risebrough *et al.,* 1976; Subramanian *et a/.,* 1986; Wania and Mackay, 1993). These synthetic compounds, including pesticides such as DDT and HCH and the industrial pollutants, polychlorinated biphenyls (PCB), have been of great concern because their relative chemical and biological stability, their mobility, low solubility in water and high solubility in lipids, mean they persist in the environment and accumulate *via* food webs and can be biomagnified (Bennington *et a/.,* 1975). Although many of the biotic components from Indian environments have been analysed for organochlorines (Kaphalia and Seth, 1981; Ramesh *et al.,* 1992), few include extensive and systematic monitoring of organochlorines in the different biotic compartments of coastal environments (Rajendran, 1984; Shailaja and Sen Gupta, 1990; Ramesh *et al.,* 1990; Tanabe *et al.,* 1993). The main sources of pesticide residues to the coastal environment of Bay of Bengal, including the Vellar River estuary, Southern India and other watershed areas of Parangipettai, are from agriculture. The coastal biotopes of Parangipettai comprising estuarine, mangrove forests and watershed and catchment areas, support a large number of birds. Since birds are an integral part of the biosphere with wide range of activities and are exposed to those persistent contaminants, an attempt was made to study the distribution and accumulation pattern of organochlorine residue levels in wild coastal birds of Parangipettai.

2. MATERIAL AND METHODS

Birds representing four groups (scavengers, inland piscivores, coastal piscivores and insectivores) based on their habitats, food and feeding habits were collected in and around the Parangipettai coastal environment, including the Vellar River watershed (Fig. 1 and Tab. I). Immediately after collection, length and weight were recorded, flight muscles were removed and stored in a deep-freezer. Biometry and ecological notes of the samples are summarised in Table **I.**

Extraction and quantification of the HCH (BHC) isomers (α, β, γ) and δ), DDT and its metabolites (o, p' -DDT, p, p' -DDT, p, p' -DDD and p, p' -DDE) and PCBs were carried out following the methods of Ramesh *et al.* (1992).

FIGURE **¹** Map showing the **Vellar** River and Parangipettai coastal study **(S.** India) area.

Twenty five to thirty grams of the sample were homogenised with anhydrous sodium sulphate to remove moisture and subjected to Soxhlet extraction for 7 hours in a mixture of 300 ml diethyl ether and 100 ml n-hexane. The extract was made up to a required volume and a known quantity was used to estimate the extractable fat content. The remaining extract was concentrated to 5 ml in a KD (Kuderna-Danish) evaporator.

The concentrate was added on to 20g of florisil packed in a glass column (15 mm i.d. to 260 mm length). Organochlorine compounds adsorbed on to the florisil were eluted using a mixture of 120ml acetonitrile and 30 ml hexane-washed glass distilled water. The eluate was thoroughly shaken in a separating funnel containing 600 ml water and 100ml hexane. After partitioning, the hexane layer containing the organochlorines was washed with water, concentrated to about *5* ml and added to 1.5 **g** of silica gel packed in a glass column (12mm i.d. to 300 mm length). During fractionation, the first fraction eluted with 180 ml of n-hexane contained the **PCBs** and p,p'-DDE and the second fraction eluted with 20 ml dichloromethane and 80 ml of *n*-hexane contained the HCH isomers, o, p' -DDT, p, p' -DDD and p, p' -DDT. Each fraction was concentrated, cleaned with *5%* fuming sulphuric

acid in concentrated sulphuric acid to remove interfering substances and then washed with hexane-washed water.

Quantification of HCH isomers and DDT and its metabolites and total PCBs was performed by injecting aliquots of the final extracts into a gas chromatograph (Hewlett Packard Model 5890 - Series **XI)** equipped with ⁶³Ni electron capture detector (GC-ECD). Glass columns (2mm i.d. to 1.8m length) packed with 3% OV-25 WHP 100/120 were employed.

The operating conditions of the GC were as follows:

The injector and detector temperatures were 275° and 300° C respectively. Column temperature was 185°C held for 15 minutes and programmed to rise at a rate of 2° C min⁻¹ to 215^oC and held for 10 minutes. Nitrogen (IOLAR Grade **I)** was used as the carrier gas at a flow rate of $30 \text{ ml} \text{min}^{-1}$. Standard mixtures and spiked samples were used frequently to check the accuracy of quantification. Organochlorine insecticide residues were quantified by comparing the individually resolved peak heights with those of the corresponding peak heights of the standards which were obtained from Ultra Scientific Co., **USA.**

For PCBs, the column consisted of fused silica capillary (0.25 mm i.d. to 25 m length) with chemically bonded silicone OV-101 equipped in HP 5890 series **1.** Column temperature was 140°C held for 2 minutes and programmed to rise at a rate of 1.5°C min⁻¹ to 230°C and held for *5* minutes. Nitrogen was used both as a carrier and a makeup gas (flow rate about $1.5 \text{ m} \text{ l} \text{ min}^{-1}$). Total PCB concentrations in the bird samples were calculated by adding concentrations of the individually resolved peaks of different PCB isomers and congeners. The detection limit of the analysed organochlorine compounds were 0.01 ng g^{-1} . The recovery efficiency of HCH isomers and DDT compounds varied between 89 and 93 percent. Results are reported on wet weight basis and were not corrected for recovery.

3. RESULTS

The range and mean concentrations of major organochlorine residues (HCHs-sum of α , β , γ and δ isomers, DDTs-sum of p, p'-DDE, *p,p* '-DDD, *0, p* '-DDT and *p, p* '-DDT, and PCBs) in the bird tissues

FIGURE 2 Percentage of extractable fat content (%) and range and mean concentrations (ngg-' wetwt) *of* organochlorine residues in bird species collected in and around Parangipettai.

are shown in Figure 2. Concentrations of HCHs ranked first among all the three organochlorine residues estimated, followed by DDTs and PCBs. The levels of HCHs ranged from 27 ng g^{-1} wet wt in red-wattled lapwing to $3,800$ ng g⁻¹ wet wt in Pariah kite. DDT concentrations varied between 5.7 and 1920 ng g^{-1} wet wt in red-wattled lapwing and blackheaded gull respectively. PCBs were found to range from 12 ng g⁻¹ wet wt in whitebreasted kingfisher to 180 ng g⁻¹ wet wt in Brahminy kite.

In general, the levels of HCHs were higher than DDTs in most of the bird species except for blackheaded gull and Kentish plover. Concentrations of PCBs were lower than HCHs and DDTs in all the bird species, except in the case of red-wattled lapwing where the mean concentrations of DDTs and PCBs were more or less similar (Fig. 2). Higher levels of HCHs in these bird species could be due to the higher exposure to HCH contaminated food than to other residues. Blackheaded gull contained higher quantities of DDTs than other birds. This might be due to their migrant and feeding behaviour in this coastal environment.

According to the feeding habit of birds, the accumulation pattern of organochlorine residues (Tab. IT) can be rated for HCHs and PCBs in the order:

Trophic group (n)	Fat $(\%)$	HCH _s	DDTs	PCBs
Scavengers (Brahminy kite, Pariah kite, Sparrowhawk) (31)	$2.3 - 5.7$ (3.72)	$320 - 3800$ (1960)	$54 - 1040$ (380)	$16 - 180$ (75)
Inland piscivores (Pond heron, Indian river tern) (22)	$1.8 - 7.2$ (4.28)	$280 - 1800$ (1108)	$64 - 240$ (143)	$31 - 87$ (62)
Coastal piscivores (Blackheaded gull, Brownheaded gull, Whitebreasted kingfisher) (35)	$1.8 - 13.5$ (7.8)	$94 - 1650$ (590)	$55 - 1920$ (633)	$12 - 110$ (62)
Insectivores (Kentish plover, Red-wattled lapwing, Indian roller, common snipe) (40)	$1.7 - 7.8$ (3.0)	$38 - 930$ (351)	$5.7 - 720$ (157)	$14 - 96$ (43)

TABLE II Range and mean concentrations (ngg^{-1} wet wt) organochlorines in Indian wild birds according to feeding habits *(n)*

scavenger > inland piscivores > coastal piscivores > insectivores whereas, DDTs recorded a different pattern of accumulation: coastal piscivores > scavengers > insectivores > inland piscivores.

4. DISCUSSION

The accumulation of organochlorine residues by birds in the higher trophic levels in the food chain, such as kites, observed in the present study, was supported by the observations of Ramesh *et al.* (1992). Moreover, the wide range of concentration pattern observed in birds of different trophic levels in the food chain suggest that feeding ground is a vital factor in the accumulation of these organochlorine residues. For example, inland piscivores, like pond heron and Indian river tern, accumulated larger quantities of **HCH** residues than the insectivores like Kentish plover and lapwing. Both pond heron and river tern feed mainly in the vicinity of agricultural fields, canals and ditches, exposing themselves to pesticides applied there, resulting in higher residue levels (Ramesh *et al.,* 1992). The changing pattern of consumption (use) of pesticides from DDT to HCH for agriculture (Fig. 3) could also be the reason for the enhanced levels of HCH residues in the tissues of pond heron and Indian river tern. Scavengers and other fisheating groups like inland piscivores and coastal piscivores accumulated high residue levels of HCH resulting from the application of insecticides for vector control and agriculture as reported by

FIGURE 3 Consumption $(-)$ and demand $(-)$ of pesticides in India from 1965 to 1990 (Gupta, 1986).

Dubrawski and Falandysz (1980) from Poland, Matthiessen (1985) from Zimbabwe and Mora and Anderson (1991) from Mexico.

In the present investigation, marked variations in the accumulation pattern by different species and individuals of the same species of birds to different organochlorine residues were observed. Among the coastal piscivores, the levels of HCH residues were found to be higher in whitebreasted kingfisher $(350 - 1650)$ ng g⁻¹ wet wt) than in the other two species of the same group (blackheaded gull $(220 - 860$ ng g⁻¹) and brownheaded gull $(94-270 \text{ ng g}^{-1})$. The higher levels of HCHs in whitebreasted kingfisher could be due to a tendency to feed in riversides and lakes in cultivated areas. The levels of DDT residues were higher in blackheaded gull $(390 - 1920)$ ng g⁻¹) than in the other two species $(169 - 310$ ng g⁻¹ in brownheaded gull and $55 - 490$ ng g⁻¹ in whitebreasted kingfisher), possibly because it is a migrant species which is exposed to DDT in different geographic locations. Moreover, blackheaded gulls inhabit sea coasts and harbour areas, and in India, most of the harbours are located in the urban regions where consumption of DDT for vector control in the 'Malaria Eradication Programme' is still being continued. This should be shown since the fish collected from urban areas of India accumulated higher levels of DDTs than HCHs (Kannan *et al.,* 1992; Ramesh *et al.,* 1992). Further studies *(e.g.,* by tagging) are needed to throw more light on the accumulation pattern of pesticides in migrating species visiting India.

Although insectivores, (Indian roller, Kentish plover) occupy lower trophic levels than other birds, in the present study, they contained considerable quantities of insecticide residues (Tab. **11).** Exposure of these birds to such insecticides is possibly relatively high because they inhabit open cultivated areas, ploughed fields and grazing lands.

Range and mean concentrations of organochlorine residues in males and females of birds are shown in Tables **I11** and IV. The residue accumulation as a function of sex did not depict a distinct pattern, but comparing the groups by feeding habits suggests that females of inland piscivores and coastal piscivores accumulated slightly lower residue levels than the males (Tabs. I11 and **IV).** The differences in residue

TABLE III Range and mean concentrations $(\text{ng} g^{-1} wet wt)$ of organochlorines in Indian wild birds according to sex (male) *(n)*

Trophic group (n)	Weight	Fat $(\%)$	HCHS	DDTs	PCBs
Scavengers (16)	450.8	$2.3 - 5.7$ (3.76)	$320 - 3800$ (1992)	$65 - 1040$ (397)	$21 - 180$ (78)
Inland piscivores (12)	206.3	$2.4 - 7.2$ (4.5)	$510 - 1800$ (1242)	$85 - 240$ (152)	$45 - 87$ (68)
Coastal piscivores (19)	370.3	$1.8 - 12.8$ (8.1)	$94 - 1650$ (626)	$92 - 1920$ (712)	$19 - 104$ (70)
Insectivores (20)	115.2	$1.7 - 5.9$ (3.01)	$47 - 780$ (372)	$7.2 - 720$ (167)	$17 - 96$ (45)

TABLE IV Range and mean concentrations (ngg^{-1} wet wt) of organochlorines in Indian wild birds according to sex (female) *(n)*

levels might be due to loss through biological behaviours like egg laying and variations in exposure to the contaminants or both.

As far as PCBs are concerned, levels were lower than HCHs and DDTs, but slightly higher than the levels observed by Ramesh *et al.* (1992) for 14 species in the same area. In a few species such as lapwing, Indian roller and common snipe, both PCBs and DDTs were found to be similar. The present levels of PCBs are generally lower than those found in more industrially developed countries (Tab. **V).** PCBs, at first, were found to have distributed near the coasts of industrial areas (Keith and Gruchy, 1973), but it was soon found that they were the major organochlorine component in pelagic sea-birds in most of the world oceans around developed nations (Bourne and Bogan, 1972; Risebrough and Carmignani, 1972; Fisher, 1973) whereas in less developed and developing nations, the levels of PCBs were much lower than other organochlorine compounds (Kannan *et al.,* 1992; Ramesh *et al.,* 1992; Tanabe *et al.,* 1993; Iwata *et al.,* 1993). This accords with the mainly industrial sources of PCBs.

The variations of organochlorines estimated in the present study showed HCHs as predominant compound in all the birds except one or two species. This could be attributed to the increasing consumption of HCHs over other organochlorines in India particularly since the banning of the use of DDT for agriculture in 1984. This changing pattern of distribution and accumulation of insecticide residues *(ie.,* higher levels of HCHs than DDTs) in the abiotic and biotic components of the Indian environment can be seen in the observations of Ramesh *et al.* (1989-1992) and Kannan *et al.* (1992).

Among the four isomers of HCH, β isomers dominated, followed by α , γ and δ isomers (Fig. 4). The mean percentage composition of α -HCH isomer ranged from 3.6 to 23.8% in Indian roller and Indian tern. The maximum mean percentage of β -HCH was observed as 93.5% in common snipe and a minimum of 64.7% in Indian tern. The lowest and highest mean compositions of γ -HCH are 1.4 and 6.9% observed in Indian roller and Indian tern. The mean composition of δ -HCH varied between 1.1% in common snipe and 7.4 percent in sparrowhawk. Most of the higher trophic organisms, including birds, are capable of degrading the synthetic organic chemicals accumulated in their body tissues, but β -HCH is more stable than α and γ -HCH to enzymatic degradation. The low water solubility, vapour pressure, and

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 $\overline{}$ $\overline{}$ TABLE V Comparison of concentrations (µgg⁻¹ wet wt) of organochlorine residues in birds from different locations TABLE **V** Comparison of concentrations (pgg-l wet wt) *of* organochlorine residues in birds from different locations j. ~ ~ ~

*SCF-Subcutaneous fat. * **SCF-Subcutaneous fat** resistance to microbial degradation of β -HCH also makes it the most persistent isomer in the environment, subsequently magnifying its levels in the food chain (Tanabe *et al.,* **1984;** Ramesh *et al.,* **1992;** Kannan *et al.*, 1992). Moreover, isomerization of γ -HCH to β -HCH in the ambient environment (Hayes, **1982)** could also contribute to the higher accumulation of β -HCH in higher trophic organisms like birds.

Regarding the variations in the composition of DDT and its metabolites in the tissues of bird samples, the percentage of p, p' -DDE was higher than the other DDT compounds (Fig. 5). The lowest and highest values of p,p'-DDE ranged between **75.2%** and **96.7%** in Indian roller and blackheaded gull. The maximum value of p, p' -DDD was 11.7% in common snipe and a minimum of **1.8%** was observed in blackheaded gull. The composition of o, p' -DDT varied between 0.3% in whitebreasted kingfisher and **6.4%** in common snipe whereas in Kentish plover o, p' -DDT was not observed. The observed values of p,p'-DDT composition ranged from 0.9% in Kentish plover to **6.3%** in Indian roller. DDT, the first organochlorine pesticide, is broken down by biological activity into the related compounds DDD, TDE

FIGURE 4 Percentage composition of **HCH** isomers in bird species.

FIGURE *5* Percentage composition of DDT compounds in bird species.

and DDE. The DDD and TDE are soon broken down further, but DDE is very persistent, so the 'chronology of DDT pollution' can be assessed by determining the proportion of DDE in total DDT (Bourne, 1976). The transformation of DDT into DDE is greater in birds than in fishes (Ramesh *et al.,* 1992) suggesting higher metabolic capacity in birds. Exposure to DDE enriched food materials may also contribute to such a higher percentage of DDE in birds.

5. CONCLUSIONS

Fat soluble compounds generally include the important classes of pollutants such as pesticides, PCBs, organomercury compounds, hydrocarbons of all kinds and plastics and their by-products which once introduced into the food web may be passed on, concentrated (Bourne, 1976) and magnified in the higher trophic organisms (Portmann, 1975). An assessment of the magnification of organochlorine residues in the Indian bird wildlife generally shows that the levels of HCH are higher than other organochlorines. In future, the levels may increase further in birds and other higher trophic animals due to the increasing consumption of this insecticide for agriculture (Verma, 1990; Singh and Singh,

1990). Subsequently the levels of DDTs are likely to decline further in the near future because of the recent ban on its consumption for agriculture, which came into force during 1986 (Nair, 1998). Consequently, the production and use of HCH has been steadily increasing (Mehrotra, 1985; Ray *et al.,* 1985). Moreover, the levels of **PCBs** may increase because of the rapid industrialisation in recent times. In this juncture of the factors mentioned, all the man-made organics should be used carefully by implementing alternative approaches for pest control and industrial purposes.

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