

Organochlorine Pesticide Compounds in Organisms from the Bay of Bengal

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Zooplankton and bottom-feeding fish (four species) from the coastal Bay of Bengal were analysed for residues of DDT, DDD, DDE and aldrin. Concentrations of Σ -DDT (DDT+DDD+DDE) ranging from 1.31 to 115.90 ng g⁻¹ wet weight in different fish tissues and 4.00 to 1587.76 ng g⁻¹ wet weight in zooplankton were found. Aldrin levels were of the order of 0.32–4.23 ng g⁻¹ in the fish tissues and 'not detected' to 0.78 ng g⁻¹ in zooplankton. The concentration levels are discussed in relation to the distribution of the compounds in different fish tissues, differences in the proportion of DDT and its metabolites DDE and DDD in the various organisms and the possible influence of suspended particulate matter on the availability of DDT residues to organisms in the water column.

Introduction

A large number of Indian rivers flow into the Bay of Bengal, many of them draining vast agricultural areas *en route*. DDT and other persistent chlorine-containing pesticides are still widely used, both in agriculture and for malaria vector control, in India and many other developing countries. Studies carried out previously have indicated the presence of considerable amounts of organochlorine pesticide residues in the sediments from the east coast of India which were mostly made up of DDT and its metabolites, aldrin and isomers of HCH (Sarkar & Sen Gupta, 1988). There is evidence that highly contaminated bottom sediment can act as a reservoir of DDT residues to near-shore fishes (Young *et al.*, 1977). Hence, studies were undertaken to determine the pesticide-residue levels in some species of bottom-feeding fish sampled from the Bay of Bengal. A few samples of zooplankton obtained from surface waters were also examined in order to get a comprehensive picture of the distribution of these compounds in the Bay of Bengal.

An analysis of the results obtained is presented in this paper.

Materials and methods

The samples were collected from different locations in the Bay of Bengal as shown in Figure 1. The first set of samples came from a site 15 km from the south-east coast of

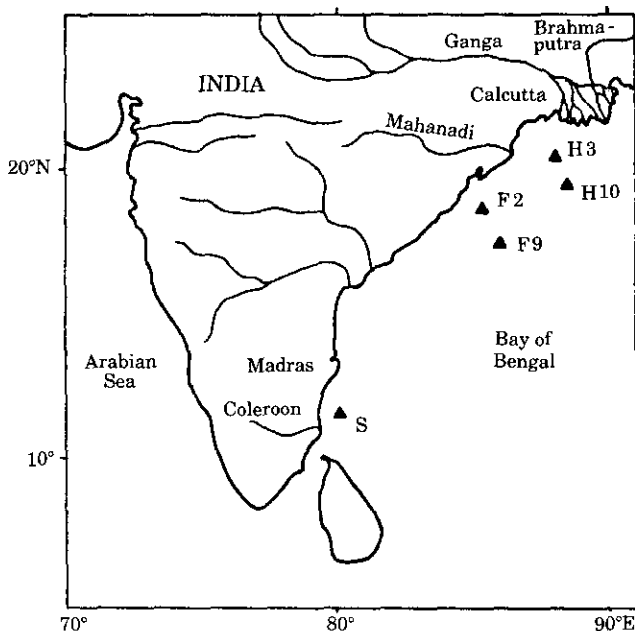


Figure 1. Location of sampling stations (▲) in the Bay of Bengal.

India, in the vicinity of the Coleroon River mouth, in October 1990 on board ORV *Sagar Kanya*. It comprised of zooplankton obtained with a neuston net on 4 different days from the area and a few bottom-feeding fish species such as *Upeneus*, *Nemipterus (Synagris)* and *Sillago*. The samples were obtained towards the end of the south-west monsoon. A second set of zooplankton samples was obtained on cruise 70 of ORV *Sagar Kanya* from the northern Bay of Bengal in December 1991, that is, during the north-east monsoon. All samples were deep-frozen soon after collection and stored at -20°C until analysis.

Three to five specimens of each species of fish were processed and their muscle tissue analysed individually. Other tissues, namely the gills and the liver, were combined and analysed as composite samples of each species. Subsamples were taken from below the lateral line on the left side of the fish, homogenized in the presence of anhydrous sodium sulphate and the resulting dry powder was Soxhlet-extracted with distilled hexane for 8 h. The extract was concentrated to ~ 2 ml, subjected to concentrated H_2SO_4 clean-up, centrifuged and the clear supernatant was analysed on a Perkin-Elmer GC Model 8700 equipped with a ^{63}Ni -ECD (340°C) and an SE-54 capillary column (60 m; $60\text{--}260^{\circ}\text{C}$). In the case of the second set of zooplankton samples an SPB-608 column (30 m; $110\text{--}270^{\circ}\text{C}$) was employed. The various compounds were identified and estimated with reference to authentic compounds obtained from IAEA, Monaco and Supelco, U.S.A.

The detection limits were 0.05 , 0.8 , 0.02 and 0.02 ng g^{-1} for *p,p'*-DDE, *p,p'*-DDT, *p,p'*-DDD and aldrin, respectively. The overall recovery of the analytical procedure was 92–95% and precision 3%.

Results

Table 1 shows the levels of DDT, its metabolites and aldrin in plankton. Total DDT (*t*-DDT; DDT+DDE+DDD) concentrations ranged from 4.0 to 6.2 ng g^{-1} wet weight

TABLE 1. Residue concentrations (ng g^{-1} wet weight) of organochlorine pesticides in zooplankton from the Bay of Bengal

Station No.	Location	Extractable organic matter (%)	DDE	DDD	DDT	t-DDT ^a	Aldrin
S	11°24.6'N, 79°59.9'E	0.54	2.90	1.86	N.D.	4.80	0.78
	11°8.4'N, 79°59.9'E	0.41	1.71	1.02	3.12	5.90	0.36
	11°16.5'N, 80°2.6'E	0.33	1.00	1.02	1.98	4.00	0.42
	11°16.5'N, 79°54.5'E	0.81	2.46	2.28	1.50	6.20	0.78
	Mean	0.52 ± 0.18	2.02 ± 0.72	1.54 ± 0.54	1.65 ± 1.12	5.22 ± 0.88	0.58 ± 0.19
F-2	18°41.6'N, 84°52.1'E	2.05	N.D.	1211.83	N.D.	1211.83	N.D.
F-9	17°42.5'N, 85°45.8'E	0.28	13.21	244.38	52.84	310.23	N.D.
H-3	20°17.3'N, 88°14.9'E	0.87	21.85	830.29	735.61	1587.76	N.D.
H-10	19°44.0'N, 88°29.9'E	0.75	8.14	203.58	57.00	268.73	N.D.
	Mean	0.99 ± 0.65	10.80 ± 7.93	622.52 ± 420.99	211.36 ± 303.51	844.64 ± 571.03	N.D.

^at-DDT=DDT+DDE+DDD.

N.D., Not detected.

(mean $5.22 \pm 0.88 \text{ ng g}^{-1}$) in organisms from the south-east coast. Of this, the parent DDT accounted for a mean value of $1.65 \pm 1.12 \text{ ng g}^{-1}$ in the range of 'not detected' (N.D.) to 3.12 ng g^{-1} , while the metabolites DDE and DDD ranged from 1.0 to 2.9 ng g^{-1} (mean $2.02 \pm 0.72 \text{ ng g}^{-1}$) and 1.02 to 2.28 ng g^{-1} (mean $1.54 \pm 0.54 \text{ ng g}^{-1}$), respectively. Aldrin levels varied between 0.36 and 0.78 ng g^{-1} wet weight (mean $0.58 \pm 0.19 \text{ ng g}^{-1}$).

Samples collected from the northern Bay of Bengal in December 1991 exhibited much higher t-DDT residues amounting to a mean value of 844.64 ng g^{-1} wet weight. Nearly 74% of this was DDD and 25% was DDT while DDE was negligible. Aldrin was not detected.

The range of concentrations of the above compounds in the fish samples is shown in Table 2. The mean t-DDT concentration was $9.95 \pm 6.38 \text{ ng g}^{-1}$ wet weight. The highest value of 19.9 ng g^{-1} was noted in *Nemipterus* sp. and the lowest in *Upeneus* sp. (2.43 ng g^{-1}). Metabolites DDE and DDD ranged, respectively, from 0.94 to 3.93 ng g^{-1} (mean $2.14 \pm 1.12 \text{ ng g}^{-1}$) and 1.15 to 3.88 ng g^{-1} (mean $2.24 \pm 1.01 \text{ ng g}^{-1}$).

Aldrin concentrations in fishes ranged from 0.19 to 0.59 ng g^{-1} (mean $0.35 \pm 0.15 \text{ ng g}^{-1}$).

Discussion

Levels of t-DDT were found to vary widely in zooplankton obtained from different areas of the coastal Bay of Bengal during different seasons. In organisms collected from the northern part of the Bay of Bengal during the north-east monsoon, the levels were much higher than those in organisms from the south-east coast of India. Further, the most predominant metabolite in the northern Bay samples was DDD, accounting for nearly 74% of t-DDT which suggests that these organisms had not been conditioned with the

TABLE 2. Residue concentrations (ng g^{-1} wet weight) of organochlorine pesticides in fish samples from coastal Bay of Bengal

Species (common name)	Tissue and No. analysed	Extractable organic matter (%)	Range Mean SD	DDE	DDD	DDT	t-DDT ^b	Aldrin
<i>Upeneus</i> sp. (goat fish)	M 3	0.47	0.26-1.72	0.04-1.90	N.D.-1.01	1.31-3.23	0.10-0.56	
			0.94	1.15	0.33	2.43	0.32	
<i>Nemipterus japonicus</i> (threadfin bream)	M 5	0.25	± 0.59	± 0.80	± 0.47	± 0.81	± 0.19	
			1.60-7.96	1.72-5.93	6.32-19.34	10.49-30.03	N.D.-1.03	
	3.93	3.88	11.92	19.90	0.59			
	± 2.44	± 1.56	± 4.87	± 7.78	± 0.34			
<i>Syngnatis striatus</i> (threadfin bream)	G ^a 5	2.79	1.15	16.72	23.97	1.08		
	L ^a 5	6.35	31.36	22.46	115.90	4.24		
	M 4	0.78	0.73-2.92	1.55-2.66	1.92-12.79	4.99-18.38	0.15-0.56	
			1.52	2.15	6.51	10.17	0.30	
<i>Sillago sihama</i> (lady fish)	G ^a 4	2.82	± 0.98	± 0.46	± 4.68	± 5.97	± 0.18	
			0.41	1.92	5.31	7.66	0.25	
	L ^a 4	7.67	1.73	3.40	9.08	14.21	0.28	
			1.27-3.82	0.87-3.61	1.76-6.42	3.97-13.88	0.12-0.31	
M 3	0.12	2.17	1.78	3.34	7.30	0.19		
		± 1.17	± 1.29	± 2.18	± 4.65	± 0.08		
L ^a 3	4.98	3.51	6.85	13.87	0.54			

M, Muscle; G, gills; L, liver; N.D., not detected.

^aComposite sample.^bt-DDT=DDT+DDE+DDD.

toxicant earlier (McKinney & Fishbein, 1972). In comparison, the north-eastern Arabian Sea zooplankton value was 189.9 ppb during the south-west monsoon and 87% less during the pre-monsoon period (Shailaja & Nair, 1994). DDT being the predominant component (70–83%) during both the seasons.

In bottom-feeding fish from the coastal Bay of Bengal, the observed levels of t-DDT and aldrin (9.95 and 0.35 ng g⁻¹, respectively) were comparable to the values found in pre-south-west monsoon samples of pelagic fish from the north-eastern Arabian Sea; that is, 10.42 and 0.39 ng g⁻¹, respectively (Shailaja & Nair, 1994). In comparison, the values during the south-west monsoon were 24.67 ng g⁻¹ in pelagic fish from the central part of the eastern Arabian Sea (Shailaja & Sen Gupta, 1989) and 109.9 ng g⁻¹ in those from the north-eastern Arabian Sea (Shailaja & Nair, 1994). Of the different species of fish tested, *Upeneus* sp. had the lowest concentrations of t-DDT, DDE, DDD and DDT. This means that compared to the others, this species has either a higher capacity for biotransformation of the various compounds or that it had not encountered contaminated food in the immediate past which is unlikely because all the fish samples came from the same marine area. Further, in this species the proportions of DDE, DDD and unmetabolized DDT to t-DDT were 39, 47 and 13%, respectively, as compared to 22, 21 and 57% in the other species, confirming a higher metabolic rate in *Upeneus* sp.

On the other hand, the lowest value of aldrin was seen in *Sillago sihama* and not in *Upeneus* sp. making it evident that the pesticide-residue concentrations in different fish species are determined by the biotransformation capacity of the individual species. A similar finding for chlorinated hydrocarbon pollutants in general has been reported by Pyysala *et al.* (1981).

The parent compound, namely, DDT, accounted for 52–70% of t-DDT in different fish tissues, compared to 32% in plankton. Further, in the gills, DDT was significantly higher (85%) than in the plankton. This indicates that these fish had acquired the residues at the bottom probably through ingestion of residue-associated particulates. It is known that DDT is adsorbed to surfaces of suspended particles before it is taken up by living organisms (Harding & Vass, 1979, quoting Sodergren, 1968 and Cox, 1971). For higher organisms such as fish, both contaminated food and contaminated particles are known sources of pollution (Thomann & Connolly, 1984; Opperhuizen & Stokkel, 1988) and particles have been found to contain five to 20 times more chlorinated hydrocarbon residues than underlying sediments (Dawson & Riley, 1977).

The mean t-DDT level in fish muscle was about 90% higher than that in plankton and both DDT and DDD were also significantly higher, individually (234 and 45%, respectively). However, DDE levels were similar (2.14 and 2.02 ng g⁻¹, respectively). DDT and DDD were predominant in the gills too (70 and 25% of t-DDT, respectively), relative to DDE (5%). As DDE is considered to be a tracer of sediment resuspension (Baker *et al.*, 1985) and it formed <5% of total DDT, especially in the gills, it is surmised that the residues were associated with recently-deposited particles rather than with particles resuspended from the sediments. Studies have shown that the enormous amounts of lithogenic material brought into the Bay of Bengal through river outflow lead to a faster settling of the organic macroparticles through the water column with relatively little decomposition (Broecker *et al.*, 1980). Hence, it is likely that DDT associated with suspended particulates is carried down without any significant transformation. As this process is quite rapid, a major part of the DDT entering the southern coastal Bay of Bengal is unavailable, especially to surface organisms. That particulate flux influences the availability of DDT to organisms in the water column is also apparent from the

higher levels of t-DDT found in zooplankton from the northern Bay of Bengal during the north-east monsoon, compared to the levels in zooplankton from the south-east coast (Table 1) because, according to Irtekkot *et al.* (1991), particulate flux in the northern Bay of Bengal during the north-east monsoon is about 35% less than that in the central part of the Bay during the south-west monsoon.

An interesting observation was made by Ramesh *et al.* (1989) regarding the concentrations of t-DDT in air at Porto Novo (11°29'N, 79°46'E) on the south-east coast of India. They found that the concentrations were higher during the period August–January, compared to the rest of the year. Of the various components of t-DDT, p,p'-DDE showed the highest percentage concentration, during the 'dry' season, i.e. January–April; p,p'-DDD showed an increasing trend between June and August and p,p'-DDT from August to December. If the observation of Takeoka *et al.* (1991) that the flux of HCH from the foci of application (agricultural soil) to the air is larger than the flux to the sea (through river discharge) applies to DDT as well, then it would mean that the DDT residues found in the bottom-feeding fish had been brought into the marine environment from air through precipitation and subsequently carried down in association with particulate matter. The fact that higher proportions of p,p'-DDT were found both in the fish (muscle as well as gills, Table 2) and in the air at an observation point close-by (Ramesh *et al.*, 1989) supports this.

As stated earlier, DDD was the major component of t-DDT in zooplankton from the northern Bay of Bengal and at one station (F-2; 18°41.6'N, 84°52.1'E; Figure 1) the entire t-DDT burden (1211.83 ng g⁻¹) was made up of DDD. While it is possible that DDD may undergo further transformation to DDA, an excretable compound, in the higher organisms (Shailaja & Sen Gupta, 1990), it should be a matter of concern that such high concentrations of t-DDT are found in the Mahanadi and Hooghly mouth regions which are known to be rich in penaeid prawn and other commercially important fish resources (NIO, 1986) as these could be affected in the long run.

Aldrin residue levels in different tissues of fish exhibit an interesting relationship with DDT and DDD concentrations; e.g. aldrin:DDT and aldrin:DDD ratios of skeletal muscle, liver and gills were significantly similar, although the levels of the individual compounds varied widely (Table 3). The mean values of the ratios, aldrin:DDT and aldrin:DDD in the three tissues were 0.063 ± 0.002 and 0.16 ± 0.005, respectively. When different species of fish were compared, it was observed that the aldrin:t-DDT ratio was constant at 0.028 ± 0.001 except in the case of *Upeneus* sp. in which it was 0.13, implying that different rates of biotransformation exist for DDT and aldrin in the same species. On the other hand, the aldrin:t-DDT ratio in plankton was much higher than in fish, apparently because of the lower levels of t-DDT in the former resulting from a rapid sedimentation of DDT in association with suspended particulates in the Bay of Bengal. In comparison, the Arabian Sea plankton had a much lower aldrin:t-DDT ratio (0.024 in September and 0.04 in March; Shailaja & Nair, 1994) than the Bay of Bengal plankton (0.11) which is to be expected because of the lower SPM in the Arabian Sea waters.

Recently, t-DDT levels ranging from 'not detected' to 2.38 ng g⁻¹ have been reported in marine fish collected from different landing centres along the Tamil Nadu and Pondicherry coasts on the south-east coast of India (Rajendran *et al.*, 1992). These values are much lower than those observed by us probably because (1) the samples were obtained during the pre-south-west monsoon season and (2) some species of fish such as *Sillago* were analysed at a time (May) when their feeding intensity is known to be low to

TABLE 3. Variation of aldrin with DDT and its metabolites in different fish tissues

Organism	Aldrin:DDT	Aldrin:DDE	Aldrin:DDD	Aldrin:t-DDT ^a
Fish (mean of all species)				
Skeletal muscle	0.063	0.16	0.16	0.035
Gill	0.060	0.85	0.16	0.042
Liver	0.065	0.14	0.17	0.026
<i>Upeneus</i> sp.				
Skeletal muscle	0.97	0.34	0.28	0.13
<i>Nemipterus japonicus</i>				
Skeletal muscle	0.049	0.15	0.15	0.029
Gill	0.064	0.94	0.18	0.045
Liver	0.068	0.13	0.19	0.036
<i>Syngnatis striatus</i>				
Skeletal muscle	0.046	0.19	0.14	0.029
Gill	0.047	0.61	0.13	0.032
Liver	0.031	0.16	0.08	0.019
<i>Sillago sihama</i>				
Skeletal muscle	0.057	0.09	0.11	0.026
Liver	0.079	0.15	0.15	0.039
Plankton	0.35	0.29	0.38	0.11

^at-DDT=DDT+DDE+DDD.

moderate (Bal & Rao, 1984). Thus, the mean t-DDT concentration reported in *Sillago sihama* is $0.06 \pm 0.01 \text{ ng g}^{-1}$, compared to our value of $7.3 \pm 4.65 \text{ ng g}^{-1}$. However, it is intriguing to note that the dominant component of t-DDT was primary DDT in most of the fish species examined.

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