

On chlorinated hydrocarbons in the Indian Ocean

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Abstract - This paper reviews the data available on the distribution of organochlorine compounds such as DDT and its metabolites, aldrin, dieldrin, endrin, isomers of HCH and the PCBs in the sediment, water, zooplankton, fish and seals from the Indian Ocean. Concentration of total DDT (t-DDT; DDT + DDE + DDD) in fish from the western Arabian Sea ranged from 2 - 11 ng.g⁻¹ (wet weight) while endrin and dieldrin were in the range of 0 - 45 ng.g⁻¹ and 0 - 5 ng.g⁻¹, respectively. On the other hand, the concentration of t-DDT in the eastern Arabian Sea (along the west coast of India) was in the range of 7.38 - 179.1 ng.g⁻¹ in the sediments while it was 15.8 - 444.0 ng.l⁻¹ in water. The concentrations of aldrin, dieldrin and HCH were, in the sediments, 0.95 - 35.7, 0 - 0.88 and 0.44 - 17.9 ng.g⁻¹, respectively, in water, 1 - 10, 2 - 50 and 0.2 - 9.0 ng.l⁻¹, respectively. The level of t-DDT in zooplankton was significantly high (50 - 1630 ng.g⁻¹). However, the highest concentrations of t-DDT were observed in the sediments of the Bay of Bengal (range 20 - 780 ng.g⁻¹). In the same region, concentrations of aldrin, dieldrin and HCH were 20 - 530, 0 - 510 and 10 - 210 ng.g⁻¹, respectively. These high concentrations are due to larger anthropogenic inputs from the east coast of India. The metabolite DDE constituted the major fraction of t-DDT in the fishes from the Arabian Sea suggesting that this region is not subject to high inputs of DDT.

As regards the other regions of the Indian Ocean, very high concentrations of t-DDT (5.5 - 12053 ng.g⁻¹), HCH (0.3 - 6813.0 ng.g⁻¹) and PCBs (2.1 - 3876 ng.g⁻¹) have been reported in the various tissues of the Australian fur seals while in the fish from the South African coast t-DDT and PCBs were in the range of 1 - 1240 and 1 - 310 ng.g⁻¹, respectively. In the southern Indian Ocean (in the Antarctic region) the levels of all the chlorinated hydrocarbon compounds were low viz., t-DDT 0.005 - 0.025, HCH 0.27 - 0.80 and PCBs 0.04 - 0.08 ng.l⁻¹.

INTRODUCTION

Chlorinated hydrocarbons are well known for their high persistence, toxicity and accumulative characteristics. These compounds have a high lipid/water partition coefficient which leads to their accumulation in organisms and biomagnification (RISEBROUGH *et al.*, 1967). The ubiquity of such toxic compounds as environmental pollutants has been well documented for all the major terrestrial, fresh water and marine environments. While the use of these compounds has been banned or restricted in most of the industrialised countries, they are still used extensively in the developing countries. In fact, the use of organochlorine pesticides such as DDT and HCH is considered to be inevitable for agricultural and public health in order to sustain food supply and to prevent the propagation of malaria (GOLDBERG, 1976). On an average 55,000 tonnes of pesticides are used in India every year for agricultural development and vector control programme (SEN GUPTA and QASIM, 1985). The world pesticide consumption was reported to be around 4571 thousand tonnes during the year 1980 and it is projected to go up to 6445 thousand tonnes by the year 1995 with an annual growth rate of 2.3 % (BHAT, 1985). Such an alarming trend of pesticide consumption causes a major concern about global pollution.

Studies on the distribution of chlorinated hydrocarbons in the marine environment have been carried out by several scientists from all over the world. However, most of the studies are confined to the northern hemisphere and little information is available on their distribution in the Indian Ocean (TANABE and TATSUKAWA, 1980; TANABE

et al., 1982a,b ; TANABE *et al.*, 1983 ; KUREISHY *et al.*, 1978 ; SARKAR and SEN GUPTA, 1987, 1988a,b, 1989, 1990 ; KANNAN and SEN GUPTA, 1987; SHAILAJA and SEN GUPTA, 1989 ; DOUABUL *et al.*, 1987 ; DE KOCK and BOSHOFF, 1987 ; DE KOCK, 1985 ; DE KOCK and LORD, 1986 ; SMILLIE and WAID, 1987 ; and RICHARDSON and WAID, 1983).

This paper consolidates and reviews the data available so far on the levels of organochlorine residues in the biotic as well as the abiotic segments of the Indian Ocean.

MATERIALS AND METHODS

Data base

Several groups of anthropogenic organochlorine compounds have been identified and quantified from the Indian Ocean. These include DDT, cyclodiene, hexachlorocyclohexane (HCH) and industrial compounds such as polychlorinated biphenyls (Table 1).

Table 1. Organochlorine compounds detected in the marine environment

p,p'-DDT	2,2-bis-(p-Chlorophenyl)-1,1,1 trichloroethane
o,p'-DDT	2-(o-Chlorophenyl)-2-(p-chlorophenyl)-1,1,1-trichloroethane
p,p'-DDE	2-(o-Chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethylene
o,p'-DDE	2-(o-Chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethylene
p,p'-DDD	2,2-bis-(p-Chlorophenyl)-1,1-dichloroethane
o,p'-DDD	2-(o-Chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethane
HCH	α , β , γ , - isomers of 1,2,3,4,5,6-hexachlorocyclohexane
Aldrin	1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene
Dieldrin	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4-endo-exo-5,8-dimethanonaphthalene
Endrin	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo-endo-5,8-dimethanonaphthalene
PCBs	Congeners of polychlorinated biphenyls

The documented levels of chlorinated hydrocarbons reported here were determined from different types of samples such as sediment, water and marine organisms including seals. The analyses were conducted with the help of a gas chromatograph coupled with a Ni⁶³ electron capture detector and in some cases with a GC - MS, particularly for the analysis of PCBs. Fig. 1 distinguishes the various regions of the Indian Ocean from where the data have been gathered and the types of samples investigated from each region. The presence of organochlorine pesticide residues has been reported in various samples namely, fish, zooplankton, sediment and water from the Arabian Sea (SARKAR and SEN GUPTA, 1987,1989, 1990; SHAILAJA and SEN GUPTA, 1989; KANNAN and SEN GUPTA, 1987 and DOUABUL *et al.*, 1987), as also in the sediments and zooplankton from the Bay of Bengal (SARKAR and SEN GUPTA, 1988a,b ; 1990).

Some data on the concentration of chlorinated hydrocarbons in the fishes from African coastal waters are available (DE KOCK and BOSHOFF, 1987; DE KOCK and RANDALL, 1984 ; DE KOCK, 1985 ; DE KOCK and LORD, 1986).

Table 2. Mean concentrations of different chlorinated hydrocarbons in the Indian Ocean (ppb)

Region ^b	Sample ^c	Chlorinated Hydrocarbons ^a							Source
		HCH	Aldrin	Dieldrin	Endrin	t-DDT	PCBs		
I	Fish (13)	N.D.	N.D.	N.D.	2.5 (0-5)	5.9 (2-11)	N.D.	Douabul et al, 1987	
II	Fish (48)	N.D.	N.D.	N.D.	N.D.	59.17 (8.1-204)	N.D.	Shailaja and Sen Gupta (1989)	
III	Zooplankton (33)	N.D.	N.D.	N.D.	N.D.	500 (50-1630)	N.D.	Kannan & Sen Gupta, 1987; Sarkar et al. (1990)	
IV	Water (6)	2.72 (0.26-9.4)	4.43 (1.42-9.8)	20.75 (2.1-50.9)	N.D.	115.2 (15.81-444)	N.D.	Sarkar and Sen Gupta (1987)	
V	Sediment (42)	5.03 (0.44-17.9)	8.72 (0.95-35.7)	0.88 (0-0.88)	N.D.	48.31 (7-179.1)	N.D.	Sarkar and Sen Gupta (1989)	
VI	Sediment (87)	54 (5.0-200)	136 (15-390)	185.5 (50-500)	N.D.	165 (20-790)	N.D.	Sarkar and Sen Gupta (1988 a,b)	
VII	Fish (57)	N.D.	N.D.	N.D.	N.D.	110 (0-1200)	95	De Kock and Boshoff (1987)	
VIII	Water (6)	0.48 (0.21-0.93)	N.D.	N.D.	N.D.	0.002 (0.001-0.002)	54	Tanabe et al. (1982)	
IX	Seal muscle (11)	0.26 (0.1-0.6)	N.D.	N.D.	N.D.	21.1 (5.5-42.1)	9.02 (2.1-40.6)	Smillie and Waid (1987)	
X	Water (25)	1.66 (0.27-7.3)	N.D.	N.D.	N.D.	0.03 (0.005-0.12)	0.098 (0.051-0.25)	Tanabe et al. (1983), Sarkar & Sen Gupta (1989)	

N.D. - Not detected, a - Values within parentheses indicate the conc. range,

b - Regions of investigations in the Indian Ocean : I-W.Arabian Sea; II-NE Arabian Sea;

III-Central Arabian Sea; IV- SE Arabian Sea; V-E.Arabian Sea; VI-NE Indian Ocean (Bay of Bengal);

VII-W.Indian Ocean (South African Coast); VIII-SW. Indian Ocean.

c- Values within parentheses indicate the number of samples analysed.

Table 3. Mean concentrations of DDT isomers and metabolites in the Indian Ocean

Region ^b	Sample ^c	DDT-Isomers and Metabolites ^a								Source
		pp'-DDT	op'-DDT	pp'-DDE	op'-DDE	pp'-DDD	op'-DDD	pp'-DDD	op'-DDD	
I	Fish (13)	3.18 (1-13)	2.25 (0-5)	1.92 (1-5)	2.00 (0.47)	N.D.	N.D.	N.D.	N.D.	Douabul et al. 1987
	Fish (13)	N.D.	N.D.	50.48 (8.7-171.3)	N.D.	37.10 (0-162.5)	N.D.	N.D.	N.D.	Shailaja and Sen Gupta (1989)
III	Zooplankton (33)	52.50 (0-900)	16.40 (0-2000)	23.00 (0-330)	N.D.	77.53 (0-1630)	N.D.	N.D.	N.D.	Kannan & Sen Gupta, 1987; Sarkar et al (1990)
	Water (6)	23.78 (2-56)	25.10 (0-251)	29.19 (2.52-20.3)	29.11 (4.3-64.5)	N.D.	N.D.	137 (0-137)	N.D.	Sarkar and Sen Gupta (1987)
V	Sediment (42)	8.78 (0.28-4.63)	2.30 (2.2-2.4)	14.99 (2.38-47.6)	55.76 (3.3-159)	17.60 (0-17.6)	N.D.	N.D.	N.D.	Sarkar and Sen Gupta (1989)
	Sediment (87)	85.7 (10-200)	28.00 (20-40)	57.77 (10-340)	69.23 (20-170)	107.50 (20-300)	150 (40-350)	N.D.	N.D.	Sarkar and Sen Gupta (1988 a,b)
VII	Fish (57)	25.00 (0-180)	N.D.	N.D.	N.D.	85.00 (0-170)	N.D.	N.D.	N.D.	De Kock and Boshoff (1987)
	Water (6)	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	Tanabe et al, (1982)
IX	Seal muscle (11)	1.97 (0.3-3.3)	N.D.	17.60 (4.2-41.4)	N.D.	1.47 (0.1-2.3)	N.D.	N.D.	N.D.	Smillie and Waid (1987)
	Water (25)	0.03 (0.005-0.12)	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	Tanabe et al. (1983), Sarkar & Sen Gupta (1989)

N.D. - Not detected, a - Values within parentheses indicate the range, b - Regions investigated in the Indian Ocean

I - W. Arabian Sea; II - NE Arabian Sea; III - Central Arabian Sea; IV - SE Arabian Sea; V - E. Arabian Sea; VI - NE Indian Ocean (Bay of Bengal); VII - W. Indian Ocean (South African Coast); VIII - SW. Indian Ocean; IX - E. Indian Ocean (Australian coast); X - E. Indian Ocean. c - Values within parentheses indicate the number of samples analysed.

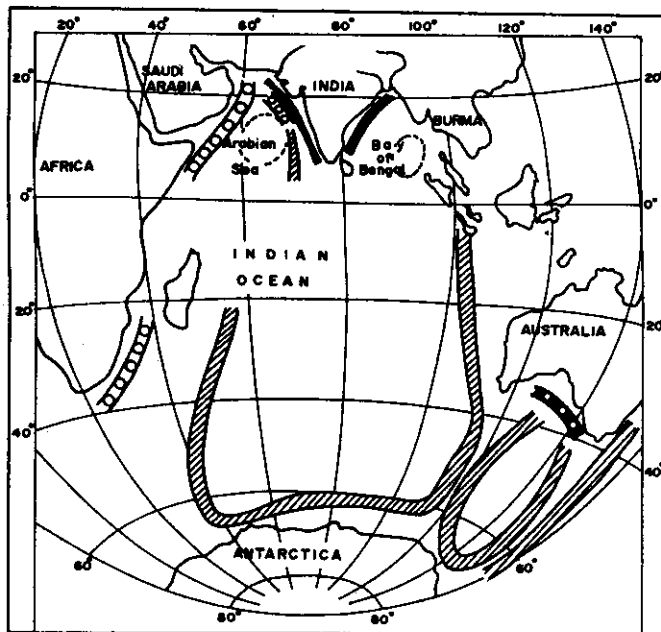


Fig. 1. Regions of investigation in the Indian Ocean. Organochlorines in sediments; PCBs, DDTs, HCHs in water; Organochlorines in fishes; Organochlorines in seals; Organochlorines in zooplankton.

There are some reports on the chlorinated hydrocarbons in the seals and shellfish from the Australian coastal region (SMILLIE and WAID, 1987 ; RICHARDSON and WAID, 1983).

Considerable data are available on the residue levels of various chlorinated hydrocarbons in water in the southern Indian Ocean (TANABE and TATSUKAWA, 1980; TANABE *et al.*, 1982a,b ; TANABE *et al.*, 1983).

The mean concentration levels of various chlorinated hydrocarbons and DDT metabolites in different regions of the Indian Ocean are reproduced in Tables 2 and 3, respectively. The variation of their mean concentration levels are shown by histograms (Fig. 2 and 3) in order to illustrate the extent of pollution in different regions.

DISCUSSION

DDT

Total DDT (*p,p'*-DDT + *o,p'*-DDT + *p,p'*-DDE + *o,p'*-DDE + *p,p'*-DDD + *o,p'*-DDD) was detected in almost all the regions of the Indian Ocean under investigation. Concentration of *t*-DDT was significantly high in the zooplankton (50 - 1630 ng.g⁻¹) from the central Arabian Sea (KANNAN and SEN GUPTA, 1987) whereas those in sediment from the Bay of Bengal and southeastern Arabian Sea were in the range of 20 - 790 and 7 - 179.1 ng.g⁻¹, respectively (SARKAR and SEN GUPTA, 1987, 1988a,b). However,

it was very low in the surface water from the southern Indian Ocean, particularly the Antarctic region (0.21 - 0.93 ng.g⁻¹; TANABE *et al.*, 1982a,b; 1983). The distribution of t-DDT in the Indian Ocean is exhibited by the contour diagram (Fig.4). A clear picture of the mean concentration levels of t-DDT in different regions of the Indian Ocean is reproduced by a three-dimensional diagram (Fig.5). It clearly illustrates the extent of pollution in the Indian Ocean. Fig. 3 shows the variation of the composition of the different isomers and metabolites of DDT available in different regions of the Indian Ocean. The most striking feature is that almost all the isomers and metabolites of DDT were present in the northern Indian Ocean and among them p,p'-DDT and p,p'-DDE were found to be dominant. o,p'-DDD was sparingly detected in a few sediment and water samples from the Bay of Bengal and the Arabian Sea. p,p'-DDD could be identified in fish, zooplankton, seals and sediments from different regions of the Indian Ocean. Another interesting point is that the sediment along the east coast of India contained an elevated concentration of t-DDT (20 - 790 ng.g⁻¹). This is due to the influx of huge

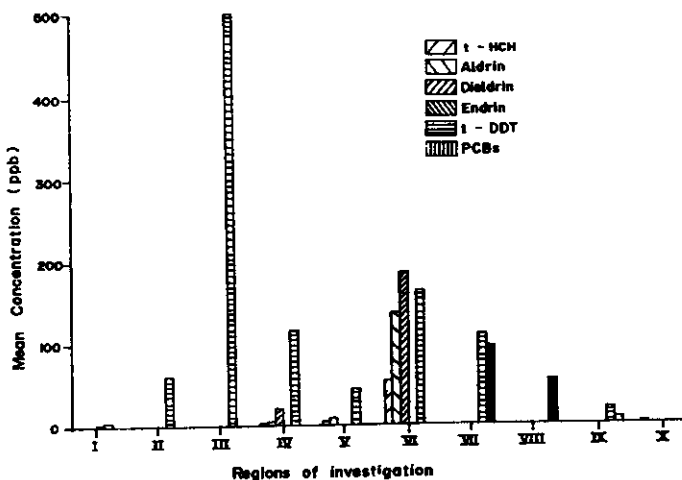


Fig.2. Variation of the mean concentration of chlorinated hydrocarbons in the Indian Ocean.

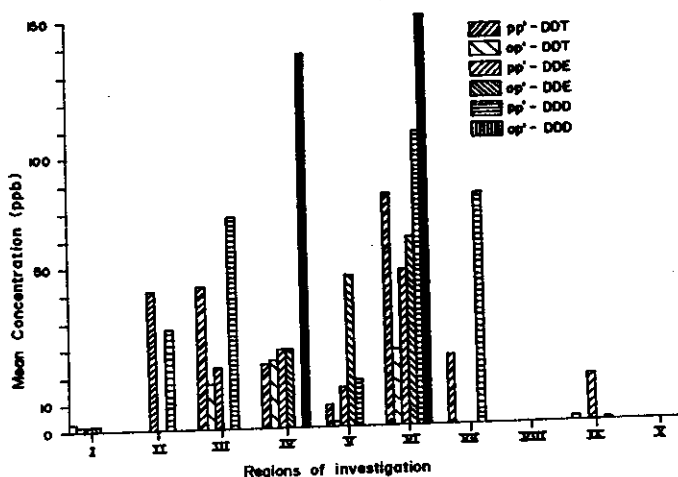


Fig.3. Variation of the mean concentration levels of the isomers and metabolites of DDT in the Indian Ocean.

amounts of pesticide residues into the Bay of Bengal through runoff from the eastward flowing perennial rivers. Besides, the sediment samples were mostly sandy-clay type which adsorb pesticides to a great extent (PICER *et al.*, 1977). On the contrary, the sediments from the west coast of India were found to contain relatively less amount of t-DDT (7.38-179.1 ng.g⁻¹). It was observed that there is a good correlation ($r = 0.85$) between the DDE with respect to the total DDT concentration in the sediment from both the coasts of India. This indicates that the degradation of DDT to its metabolite DDE occurs in the marine sediments. Such a degradation of DDT to DDE may be attributed to the biochemical effects of the benthic organisms and physico-chemical characteristics of the sediments.

It is reported that only two compounds of the DDT family, p,p'-DDT and p,p'-DDD were detected in the fishes from the South African coastal waters (DE KOCK and BOSHOFF, 1987). This could possibly be due to the metabolic transformation of p,p'-DDT

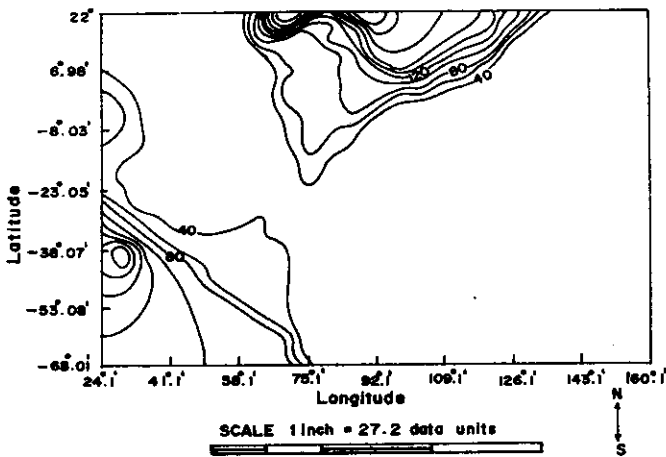


Fig.4. Distribution of total-DDT in the Indian Ocean.

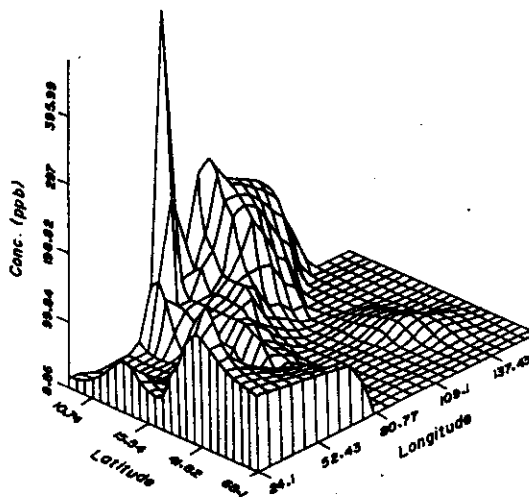


Fig.5. Total - DDT profile in the Indian Ocean.

to p,p'-DDD in the physiological system of various species of fishes available in that region. Among them, *Lichia amia* and *Mugil capensis* were found to accumulate considerable amounts of DDT (10 - 910ng. g⁻¹) in their body tissues.

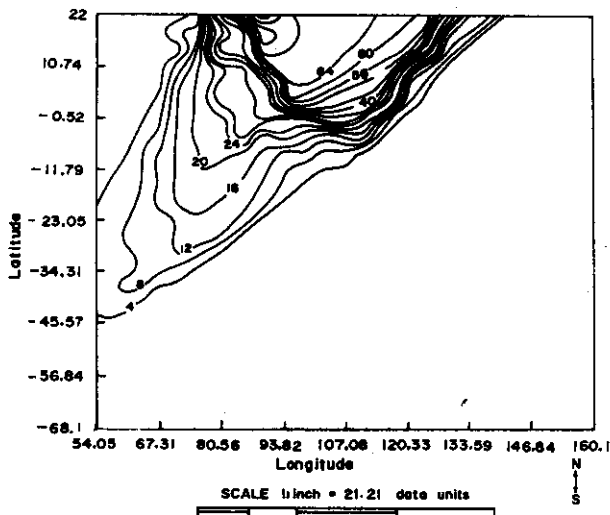


Fig.6. Distribution of total-HCH in the Indian Ocean.

A wide range of variation in the concentration levels of DDT was observed in muscle (5.5 - 42.1 ng.g⁻¹), liver (0.3 - 77.7 ng. g⁻¹), testes (0.4 - 31.4 ng.g⁻¹) and blubber (232 - 6831 ng. g⁻¹) of the Australian fur seals. The observed variation in the DDT concentration in different tissues of the seals may be due to the relative sensitivity, sustaining capacity of each of the individual species of the seals (both male and female) and the partition coefficients of the pesticides with respect to the lipid contents of various tissues and water. However, it was observed that the DDT levels in the blubber of male seals increase with age (ADDISON and SMITH, 1974) reaching a steady state in later life.

The total DDT content of the fishes from the Arabian Sea showed a greater accumulation of pesticide residues in the liver than in the muscle. Concentration of DDT in the surface water from the eastern Indian Ocean was in the range of 0.005 - 0.12 ng. l⁻¹. Such a low concentration of DDT in water clearly indicates its low solubility. However, the concentration of DDT was significantly high in coastal waters (15.81 - 444ng. g⁻¹) from the eastern Arabian Sea (SARKAR and SEN GUPTA, 1989).

The observed differences in the concentration of DDT in water and sediment from different regions of the Indian Ocean may be attributed to the influence of various physico-chemical characteristics of the ocean as is evident from the multiple regression analysis of such parameters as pH, salinity, total organic carbon, clay minerals, Fe, Co, Ni, Mn, Zn, Cu etc., on the degradation of DDT to DDE (SARKAR, unpublished) in the marine environment.

HCH

HCH was detected mostly in the Indian coastal waters and in the southern Indian Ocean (SARKAR and SEN GUPTA, 1987; 1988a,b; 1989; TANABE *et al.*, 1982; 1983; 1985). The distribution of total HCH in the Indian Ocean is depicted by the contour diagram (Fig.6). HCH was present in considerable amounts in the Indian coastal

sediment ($0.44 - 200 \text{ ng.g}^{-1}$) whereas it was in the range of $0.26 - 9.40 \text{ ng.l}^{-1}$ and $0.1 - 93.0 \text{ ng.l}^{-1}$ in the waters of the Arabian Sea and the southern Indian Ocean, respectively. HCH was rarely detected in the fishes from the eastern Arabian Sea (SHAILAJA and SEN GUPTA, 1989). However, there are some reports of its presence in different tissues of the fur seals from the Australian coastal water. It was detected mostly in the muscles and to some extent in the liver, blubber and testes of the male species of the seals (SMILLIE and WAID, 1987).

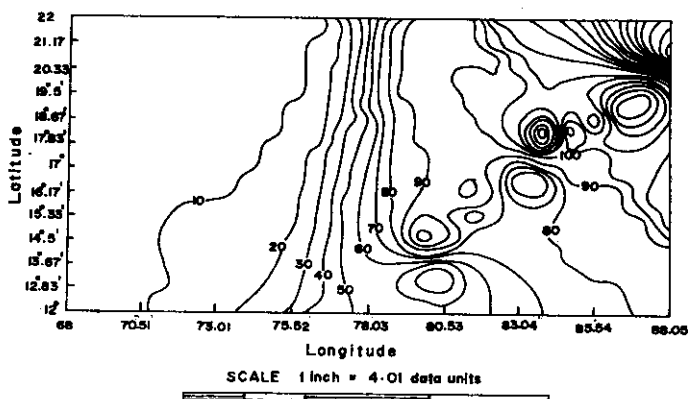


Fig.7. Distribution of Aldrin in the Indian Ocean.

HCH detected in different regions of the Indian Ocean was composed mainly of three components namely, α , β , γ isomers of which α and γ - isomers contributed around 45 - 48% whereas β - isomer 5 - 10% of the total HCH concentration. Extensive use of lindane (γ - HCH) as well as technical HCH in the tropics and southern hemisphere accounts for the high proportion of α and γ - isomers in the Indian Ocean.

Aldrin

It was one of the most stable cyclodiene organochlorine compounds detected in the Indian Ocean particularly in the sediment and water from the Indian coastal region. Distribution pattern of Aldrin in the Indian Ocean is presented by the contour diagram in Fig. 7. It was observed that the concentration of Aldrin in the sediments in the Bay of Bengal ($15 - 390 \text{ ng.g}^{-1}$; SARKAR and SEN GUPTA, 1988a,b) was higher than that in the Arabian Sea ($0.95 - 35.7 \text{ ng.g}^{-1}$; SARKAR and SEN GUPTA, 1989). It was low in water ($1.42 - 9.8 \text{ ng.l}^{-1}$) from the west coast of India. Aldrin was also detected in fishes and zooplankton from the Bay of Bengal (SHAILAJA - personal communication).

Dieldrin

This highly toxic cyclodiene chlorinated compound was detected in rather low concentration in the Indian Ocean particularly in the Indian coastal region. It was found to be abundant in the sediment from the Bay of Bengal ($50 - 500 \text{ ng.g}^{-1}$; SARKAR and SEN GUPTA, 1988a,b) but scanty in the eastern Arabian Sea ($0 - 0.88 \text{ ng.g}^{-1}$; SARKAR and SEN GUPTA, 1987). It was, however, present in considerable amounts in water from the southeastern Arabian Sea ($2.1 - 50.9 \text{ ng.l}^{-1}$; SARKAR and SEN GUPTA, 1989). Such a sharp variation of the concentration of Dieldrin in the sediment and water could probably be due to a faster rate of degradation of pesticide in the sediment phase than in the water (PATIL *et al.*, 1972). Also, normally the

degradation potential varies in different locations depending on the environmental conditions.

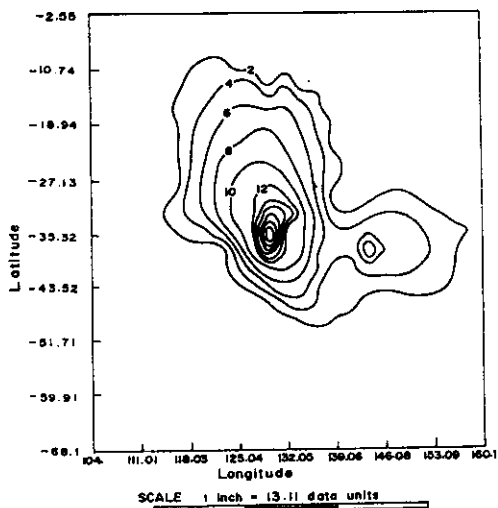


Fig.8. Distribution of PCBs in the Indian Ocean.

Endrin

Endrin was rarely detected in the marine environment. It was reported to be in the range of 0 - 2.5 ng. g⁻¹ in fishes from the northwestern Gulf region (DOUABUL *et al.*, 1987). Such rarity of this compound in the marine environment may be attributed to the extent of its use and unstable nature, being isomerized into the more stable keto form as follows:

PCBs

Polychlorinated biphenyls is a generic name of a distinct class of chemical compounds. There are altogether 209 congeners of PCBs. In fact, it has now become a ubiquitous pollutant due to its high persistence and bioaccumulative nature. Extensive work has been carried out all over the world on its abundance in the environment. However, as regards its presence in the Indian Ocean, it was detected mostly in the southern Indian Ocean. A complete picture of its distribution in the Indian Ocean is shown in the contour diagram (Fig.8) . Of the 209 congeners of PCBs , only a few could be detected in most of the samples from the Indian Ocean. It was observed that the components of Arochlor 1254 and Kanechlor were more prevalent than the other isomers and congeners of PCBs in the Indian Ocean. In fact, they were found to be abundant in the fishes in the South African coastal waters (1 - 310 ng.g⁻¹; DE KOCK and BOSHOFF, 1987) and in different tissues of fur seals from the Australian coastal water (SMILLIE and WAID, 1987). The concentration of PCBs is significantly high in the blubber (53.4 - 3836 ng. g⁻¹) and liver (44 - 114 ng. g⁻¹) but relatively less in the testes (2.1 - 55 ng. g⁻¹) and muscle (35 - 72 ng.g⁻¹) of the seals. It was found to be in the range of 35 - 72 ng. l⁻¹ in water from the eastern Indian Ocean (TANABE *et al.*, 1982a,b). PCBs were detected even in the Antarctic water (0.057 - 0.25 ng.l⁻¹; TANABE *et al.*, 1982a,b; 1983). The above findings relating to its occurrence in different regions of the Indian Ocean clearly indicate the ubiquity of such substances in the environment.

Acknowledgement - The authors are highly thankful to Dr. B.N. DESAI, Director, N.I.O., for encouragement and keen interest in this work and to Dr. M.S. SHAILAJA for all help.

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