# Chapter Twenty-Eight

# OCCURRENCE AND DISTRIBUTION OF PERSISTENT CHLORINATED HYDROCARBONS IN THE SEAS AROUND INDIA

## A. SARKAR

Chemical Oceanography Division National Institute of Oceanography Dona Paula, Goa - 403004, India

### INTRODUCTION

Organochlorine compounds are well known for their high persistence, toxicity, low water solubility and lipophilic characteristics. Because of such lipophilic nature they are being accumulated in the biological organisms as well as human tissues. It has thus become a global concern due to widespread occurrence of several organochlorine pesticides such as DDTs, HCHs, aldrin, dieldrin, heptachlor, hepox, etc. and various congeners of PCBs in the environment! Potential sources from which they are likely to reach the marine environment are rivers, the atmosphere, direct dumping into the sea and shipping activities. Upon entering the sea, a variety of interactions takes place with the compound. It involves many transition between different compartments such as water, suspended matter, sediment and organisms. Several groups of anthropogenic organochlorine compounds have been detected and reported frequently in the marine environment. These are DDT, cyclodiene, HCH insecticides, industrial compounds such as polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB), etc. Although most of the industrialized countries have banned or restricted the use of persistent chlorinated hydrocarbons because of their accumulative characteristics and harmful biological

effects, they are in use extensively in the developing countries especially in the Southern hemisphere and tropical regions.

The use of chlorinated hydrocarbon pesticides such as DDT and HCH (BHC) is considered to be unavoidable in the developing countries for agriculture and public health in order to sustain food supply and to prevent the propagation of malaria.2 In India around 75000 metric tons (MT) of technical grade pesticides are being used to cover 182.5 million hectares of cultivated lands.3 The present demand for technical grade pesticide may go up to 144000 MT by the end of this century. Approximately 10000 MT of DDT and 7000 MT of BHC are being used for control of vectors of diseases as part of its vital public health programs for malaria, filarias, Kalazars, etc.3

As far as the occurrence and distribution of PCB in the environment are concerned it has now become an ubiquitous substance. PCBs is a generic name of many isomers and congeners with different numbers of chlorine atoms substituted in biphenyl rings. This group of chemicals was first synthesized by Schmidt & Schulz<sup>4</sup> (1881) before the turn of the century and has been produced commercially since 1929. Because of their large production and worldwide use, PCBs have already extended their boundaries of distribution over the global environment, as is evidenced by their detection in the Arctic and Antarctic atmosphere, hydrosphere and biosphere. 5.6 The persistent and bioaccumulative nature of PCBs have been recognized particularly in aquatic ecosystems, where the stepwise accumulation in higherranking predators of the foodchain is rather common.7-9 PCBs currently present in the global environment were estimated to be about 370 x 103 tons. Of this, almost the entire amount, approximately 360 x 103 tons are retained in coastal sediments and open ocean water. Particularly the oceanic water contains the major portion of PCB residues, over 60% of the world environmental PCB load, clearly indicating that the open ocean water serves as a vast reservoir and final sink of PCBs. It has been estimated that about 780 x 103 tons of PCBs are still in use in the older electrical equipment and other products and deposited in landfills and dumps or in storage. The concern for PCB pollution in aquatic organisms has also been emphasized as the primary route of exposure to humans and domestic animals due to consumption of fish, etc., from contaminated waters!0-13

The cumulative effects of the residues of various organochlorine pesticides and PCBs over time on our coastal environment can be expected to be considerable in view of the fact that 25% of India's population lives in the coastal areas and marine fish is one of the major foods consumed by them. Uptake and accumulation of highly persistent organochlorine compounds by sediments, micro-organisms and fish lead to build up of such compounds in the foodchain thereby posing a serious threat to human health. This chapter deals with an exclusive evaluation of occurrence and distribution of various organochlorine compounds in different compartments of the seas around India extending up to the Antarctic Ocean.

### MATERIALS AND METHODS

Marine environmental samples were collected from different compartments of

the seas around India. The regions of investigation in different parts of the Indian Ocean are depicted in Figure 1. Water samples were collected by an *in-situ* sampler. It was deployed in different locations with the help of a self lubricating hydrographic winch PVC coated wire on board, ORV-SAGAR KANYA. The Sea-Star® in-situ sampler is an instrument designed to pump large volumes of water slowly (50 -200 ml/min) through an extraction column which concentrates chlorinated hydrocarbons from water. It provides an accurate measurement of the volume of water extracted. The extraction column is a teflon cylinder (37 x 2.5 cm o.d) containing Amberlite XAD-2 resin. Prior to deployment of the instrument in water, the retention capacity of each of the columns for different organochlorine compounds and their recovery were determined.<sup>14</sup> It was observed that the retention capacity of the XAD-2 column for chlorinated hydrocarbon averaged 95 + 12% indicating average column efficiency of 94  $\pm$  14%. Organochlorine compounds were eluted from the columns by 250 ml distilled methanol followed by 300 ml dichloromethane. Volumes

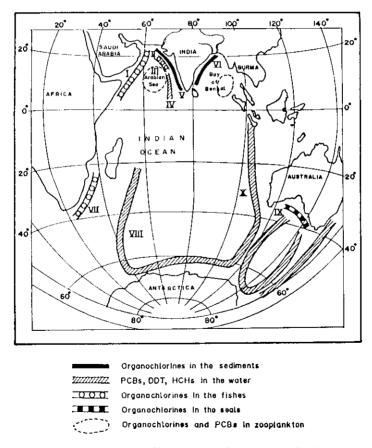


FIGURE 1. Map showing the regions of investigation of the residues of various organochlorine compounds in the Indian Ocean.

of eluates were reduced to 1 ml by evaporation on a Kuderna Danish (KD) evaporator and the aliquots were then treated with activated copper powder to remove sulfer. It was then passed through deactivated (10%) alumina followed by deactivated (5%) silica following the micro-column technique:5

Sediment samples were collected in aluminum foil (precleaned with distilled hexane) from different locations of the seas around India (Figure 1) with the help of Van veen grab on board ORV-SAGAR KANYA and RV GAVESHANI. Immediately after collection, sediments were preserved in deep freezer to avoid bio-degradation.

After thawing 100-150 g (wet wt.) sediments were first treated with 5 ml 1M solution of ammonium chloride and shaken thoroughly for 4 hours to facilitate the desorption of the pollutants adsorbed on the sedimentary particles. In order to determine the water content of each of the sediments a small quantity of sediment from each of the samples was dried in the oven at 120°C. Chlorinated hydrocarbons were extracted from the sediment by shaking the moistened sediment first with 150 ml acetone (nanograde) for about 8 hours followed by post extraction with 150 ml hexane (nanograde) and acetone (nanograde) (1:1) for 1 hour. The combined extracts were then transfered to hexane and the volume is reduced to 1 ml by evaporation in a KD evaporator. Just before evaporation, 1 ml iso-octane (nanograde) was added to each of the containers having the extract to prevent them from complete dryness. The aliquots were purified following the steps as described above.

Zooplankton samples were collected in aluminum foil (precleaned with distilled hexane) using Neuston net (200 u mesh size) on board ORV-SAGAR KANYA and RV GAVESHANI from different locations of the seas around India (Figure 1). Immediately after collection, the samples were preserved in the deep freezer. Organochlorine compounds were extracted from the zooplanktons by soxhlet extraction with distilled hexane (nanograde) for about 8 hours. The volume of the extracts was then reduced to 1 ml by evaporation in a KD evaporator followed by purification as described above.

Various species of fishes were collected with the help of fishing nets on board from different places of the seas around India (Figure 1) and preserved in the deep freeze. Organochlorine compounds were extracted from the fishes following the solvent extraction method and cleaned up according to the procedure described above.

### CHEMICAL ANALYSIS

The determination of chlorinated hydrocarbons was performed on a gas chromatograph (Perkin Elmer 8700 and 3920B) coupled with electron capture detector (Ni<sup>63</sup>). A capillary column (SE-54) (30 m in length, 0.25 mm i.d.) was used for the analysis. The temperature of the oven was programmed as follows: initial temp. 110°C for 2 min., raised to 180°C at the rate of 10°C per min. and held for 8 min., raised to 220°C at the rate of 4°C per min. and held for 5 min., finally raised to 270°C at the rate of 4°C per min. and held for 15 min. The equilibration time was 2 min. The temperature of the detector and the injection port was fixed at 340°C and 200°C respectively. In order to determine the percentage of recovery

of various organochlorine compounds from the marine environmental samples, 100 µL of an internal standard (chlorobiphenyl congener, IUPAC-88) was spiked into each of the sample and was found to be 99.9%. The reproducibility of the chromatogram was checked by comparing the response of the internal standard spiked (100 µL of 0.1 ppm IU 88) into 0.9 ml standard solution of PCBs mixtures containing various congeners. For accurate determination of organochlorine compounds in each of the samples, PCBs fractions were pre-separated from organochlorine pesticides by silica (5% deactivated) column chromatography following the micro-column technique!5 Organochlorine pesticides and various congeners of PCBs were identified and quantified with reference to the standard solution of pesticides and individual congeners of PCBs obtained from Supelco, Switzerland and Promochem, Germany, respectively.

### RESULTS AND DISCUSSION

Chlorinated hydrocarbons identified and quantified in the seas around India are the compounds of the DDT family, cyclodienes, hexachlorocyclohexane and industrial compounds such as polchlorinated biphenyls (Table 1). The residue levels of various organochlorine pesticides and PCBs were determined in different compartments of the Indian Ocean. Figure 1 clearly shows the types of organochlorine compounds detected in different regions of the Indian Ocean. The compounds of the DDT family were detected in all the compartments of the ocean whereas HCH isomers in water, sediment and seals only. Aldrin and Dieldrin were detected in water and sediment from the Arabian Sea and the Bay of Bengal. Endrin is sparingly detected in various species of fishes16 and sediments17-19 only. PCBs were found to be present in considerable amounts in the fishes, water, seals, sediments and zooplankton. Figure 2 clearly shows the variation of the mean concentration levels

TABLE 1 Organochlorine Compounds Detected in the Seas Around India

India	
-pp'-DDT	2,2-bis-(p-chlorophenyl)-1,1,1-trichloroethane
op'-DDT	2-(o-chlorophenyl)-2(p-chlorophenyl)-1,1,1-trichloroethane
pp'-DDE	2-(o-chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethylene
op'-DDE	2-(o-chlorophenyl)-2-(p-chlrophenyl)-1,1-dichloroethylene
pp'-DDD	2,2-bis-(p-chlrophenyl)-1,1-dichloroethane
op'-DDD	2-(o-chlorophenyl)-2-(p-chlorophenyl)-1,1-dichloroethane
HCH	A, B, Y-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
<b>PCBs</b>	Congeners of polychlorinated biphenyls

of various chlorinated hydrocarbons in the Indian Ocean. Figure 3 exhibits a clear picture of the variation in the mean concentration levels of the isomers and metabolites of DDT in the Indian Ocean. A detailed discussion on the distribution of chlorinated hydrocarbons in different compartment of the ocean is given below.

### **SEDIMENT**

The distribution of various organochlorine pesticides in the sediment along the east and the west coasts of India is clearly delineated in the map (Figure 4). The most interesting finding is the trends in distribution pattern of t-DDT, aldrin, dieldrin and HCHs in the sediments!<sup>7-19</sup> The sediment of the east coast of India was found to contain relatively higher levels of organochlorine pesticides than those in the west coast (Figure 4). Such a significant variation in the distribution pattern of pesticides may be attributed to the influx of huge amounts of pesticides in contaminated water through the runoff from the rivers on the east coast of India into the ocean. There are several other factors for which the coastal sediments are being

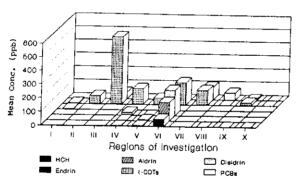


FIGURE 2. Mean concentration levels of Organochlorines in the Indian Ocean.

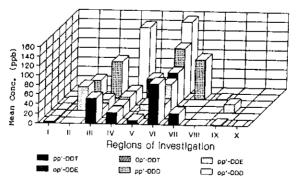


FIGURE 3. Mean concentration levels of metabolites of DDT in the Indian Ocean.

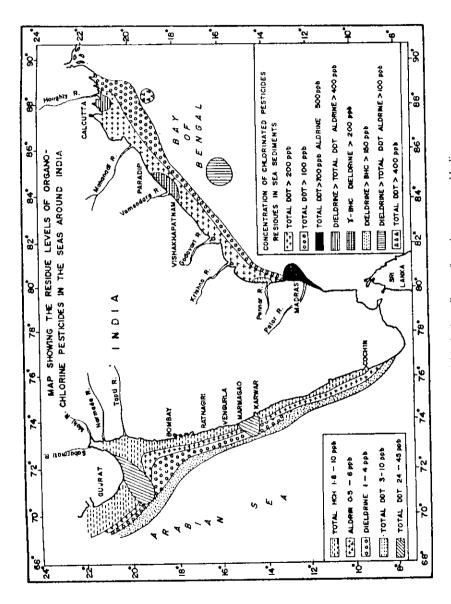


FIGURE 4. Distribution of organochlorine pesticides in the sediments from the seas around India.

contaminated with pesticides. The most striking point of observation is that there are a number of major rivers, namely, Hooghly, Mahandi, Vamsodhara, Godavari, Krishna, Pennar, Palar, etc. carrying a large volume of water into the Bay of Bengal. On the other hand, there are a few rivers, namely, Sabarmati, Mahi, Narmada and Tapti along the Gujarat coast of India. Although there are a few more small rivers falling on the west coast of India, they do not contribute as much into the seas as far as the contamination by pesticides is concerned. It was observed that the concentration levels of various organochlorine pesticides were quite high near the mouth of the rivers along the coasts and relatively less off the coasts. The most contaminated site is identified near the mouth of the Palar river where the concentration of t-DDT and aldrin were around 700 ppb and 500 ppb respectively. Dieldrin was identified discretely in a few samples collected from the east coast of India as indicated in the map (Figure 4). It has been reported that on an average 1800 MT of DDT, 270 MT aldrin, and 12000 MT of BHC were consumed every year in India.3 Such a hugh amount of consumption of organochlorine pesticides is most likely to contribute a considerable amount of their residues in the marine environment. Among the isomers and metabolites of DDT in the sediment, pp'-DDE and op'-DDE were found to be highly dominant over the other. In fact, there is a good correlation (0.83) between the percentage composition of DDE with respect to t-DDT in the sediment along the coasts of India.

However, a very recent study on the occurrence and distribution of the residue levels of various organochlorine pesticides in a few sediment samples along the east coast of India indicated that the concentration of these compounds were quite low (HCH, 0.008-0.02 ppb, aldrin, 0.008-0.13 ppb, dieldrin, 0.017-0.15 ppb, endrin, 0.046-0.490 ppb, and t-DDT, 0.10-0.97 ppb). Such remarkably low levels of the residue of organochlorine pesticides in the sediment may be attributed to characteristics of the sediments (i.e. the texture of the sediment), clay mineral composition, retaining capacity of various organochlorine compounds, diffusivity (rate of diffusion of contaminants into the underlying layers), bioturbation, bioirrigation, population and growth of benthic organisms, etc. The texture of the sediment collected recently were mostly composed of sands and silts with a very low percentage of clay minerals whereas sediments collected during the period 1986-87 were found to be composed of more than 50% clay minerals. Thus it can be clearly understood that the retaining capacity of the sediment for organochlorine pesticides was enhanced due to higher percentage composition of clay minerals in the sediment. Above all, an awareness of environmental pollution among the people might have drastically reduced the load of input of contaminants in the coastal water. Besides, the fate and persistence of various pesticides are greatly influenced by various physio-chemical characteristics (pH, salinity, total organic carbo, metal ion concentration, clay mineral concentration, etc.) of the sediments.<sup>20</sup>

A few congeners of PCBs namely, CB-14, CB-16, CB-29, CB-31, CB-28, CB-66, CB-99, CB-101, CB-119, CB-153 and CB-138 were detected in the sediment along the east coast of India.<sup>21</sup> The variation of their concentration levels were shown in Figures 5 & 6. Of all these congeners, CB-14, CB-28, CB-138, CB-153, and CB-119 were found to be highly prominent. The presence of PCBs in the sediment along the east coast of India can be attributed to industrial discharge along the east coast of India.

### WATER

lsomers of HCH, aldrin, dieldrin, DDT and PCBs were detected in water of different regions of the Indian Ocean16 (Figure 1). Of all these organochlorine pesticides, total-DDT was found to be present in considerable amounts (15.81 - 444 ng/L). The most striking point of observation is that the concentration of t-DDT is very low in the open ocean water of the Indian Ocean. The variation in the concentration levels of various organochlorine compounds in water of different regions is shown in Figure 2. Such a remarkable variation in the concentration levels of t-DDT in the marine environment can be envisaged from the fact that huge amounts of pesticides in contaminated water are being discharged into the coastal regions regularly whereas the presence of pesticides in the open ocean water is mainly due to atmospheric transport of pollutants. 22,23 The concentration of HCHs was found to be quite low in water compared to those in the sediment which may be due to the hydrophobic characteristics of organochlorine compounds as well as dispersion of the pollutants in the open ocean.

In regards to the concentration levels of PCBs, they were present relatively in higher

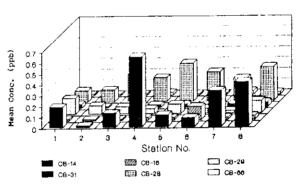


FIGURE 5. Concentration levels of PCBs in sediments along the east coast of India.

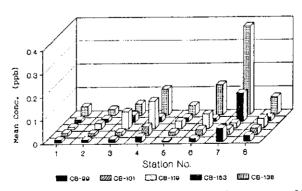


FIGURE 6. Concentration levels of PCBs in sediments along the east coast of India.

concentrations in the south west Indian Ocean than the eastern Indian Ocean. 22-25 It may be due to larger input of these compounds from the African coast.

### ZOOPLANKTON

Among the organochlorine pesticides, compounds of DDT family were found to be abundant in the zooplankton from the north eastern Arabian Sea<sup>26-30</sup> The concentration of t-DDT in zooplankton was significantly high (mean conc. 500 ppb) in this region. This can be attributed to the input of huge amounts of DDT into the coastal water through agricultural drainage from the adjoining areas and river runoff. It is apparent that the biomass of zooplankton are quite high in the estuarine and coastal water and various depth strata will each be subjected to a different level of organochlorine contamination. The availability of organochlorine compounds in zooplankton is dependent on quality, quantity and size of competing living and inert particles because organochlorines are very hydrophobic and adsorb readily to other surfaces.31-33 Moreover, particulate matter high in organic contents accumulates and retains a greater concentration of organochlorines than inorganic particles.32-36 Organochlorines are also incoporated into collidal micelles believed to be formed by humic substances in the presence of ionic species<sup>37-40</sup> Humic substances may comprise as much as 90% of the dissolved organic matter in the seawater.41-42 It clearly indicated that most organochlorines are probably bound to particulates or associated with the micellar structures present in seawater. Thus, the high concentration levels of DDT found in the zooplankton might be due to its associations with various other organisms being collected along with the zooplankton.

Concentration levels of various congeners of PCBs (17.62 - 105.7 ppb), were determined in the zooplankton from the Arabian Sea29 Altogether, 31 congeners of PCBs were detected. However, only 17 congeners of PCBs, namely, 2 isomers of trichlorobiphenyl [CB-26, CB-28 (1.57-9.54 ppb)], 5 isomers of tetrachlorobiphenyl [CB-42, CB-44, CB-49, and CB-60 and CB-70 (0.55-6.85 ppb)], 6 isomers of pentachlorobiphenyl [CB-97, CB-99, CB-101, CB-105, CB-116 and CB-118 (0.55-11.28 ppb)] and 4 isomers of hexachlorobiphenyl [CB-138, CB-141, CB-149, and CB-153 (0-6.83 ppb)] were found to be present in almost all the samples. The other congeners of PCBs, namely, CB-4, CB-11, CB-14, CB-15, CB-16, CB-26, CB-31, CB-61, CB-65, CB-128, CB-151, CB-181, CB-183 and CB-198 were sparingly detected in a few samples. The distribution pattern in the concentration levels of the individual congeners of PCBs in the zooplankton showed that they were in abundance in the coastal region and steadily decreasing off the coast.29 Such a variation in their concentration levels may be due to industrial discharge into the ocean from the adjoining areas. The decreasing trend in the distribution pattern may be attributed to the dispersion of the pollutants due to the dynamic situation of the ocean. The concentration levels of various congeners of PCBs were in the order, CB-101 > CB-99 > CB-28 > CB-52 > CB-149 > CB-42 > CB-118 > CB-60 > CB-44 >CB-138>CB-105>CB-116>CB-49> CB-97>CB-141.

### FISH

Isomers of HCHs and DDTs were detected in various species of fish collected from different regions of the seas around India 16,30 (Figure 1). However, the concentrations of the isomers of HCHs were too low to be quantified. The concentration of total-DDT was found to be in the range of 8.1-204 ng/g (dry wt.) and the mean concentration level was 59.17. It was found to be fairly high in the fish collected from the South African coast43 probably due to extensive use of DDT in the coastal regions. Only 23 species of fish were analyzed from the Arabian Sea (Shailaja and Sen Gupta, 1989) and it was observed that the fish from the estuarine region contained the highest amount of t-DDT residues burden (344.7 ppb) followed by coastal variety. Among the metabolites of DDT, DDE constitutes a major proportion. Apart from these organochlorine pesticides, PCBs were also detected in the fish from the South African coast<sup>43</sup> (Figure 2). The mean concentration level of PCBs was found to be 95 ppb. This clearly shows the extent of contamination of the South African coastal water.

### SEALS

Organochlorine pesticides and PCBs were also detected in the seals, Arctocephalus pusillus doriferus from the Australian coastal water44 (Figure 1). Organochlorine compounds were determined in different organs of the seals. All of the seals contained PCBs, DDT, DDE and some DDD; some of them contained trace amounts of Hexachlorobenzene (Y-HCB) and HCH particularly in the blubber. In all samples the pp'-DDT were present in high proportions compared to the op'-DDT. An extensive study on the evaluation of the distribution of PCBs and organochlorine pesticides within animals clearly indicated that PCB and DDT levels in the blubber of male seals increased with age45 but reaches a plateau value in later life. The mean concentration levels of various compounds in seal were, HCH, 0.26 ng/g; t-DDT 21.1 ng/g and PCBs, 9.02 ng/g (Figure 2).

### CONCLUSION

The residue levels of various persistent chlorinated hydrocarbon pollutants such as DDTs, HCHs, aldrin, dieldrin, endrin, and PCBs were determined in different compartments (water, sediment and marine organisms) of the seas around India. The trends in distribution patterns of various organochlorine pesticides and PCBs in the seas around India indicated that the east coast of India is much more contaminated than the west coast because of the input of huge amount of pesticides and other organochlorine compounds into the ocean through the river runoff, industrial discharge, as well as agricultural drainage. The compounds of the DDT family were detected in almost all the compartments of the Indian Ocean. Concentrations levels of t-DDT were significantly high in zooplankton and various species

of fish. Among the isomers and metabolites of DDT, DDE constitutes a major portion of total-DDT. The most striking point of observation is that the concentration levels of various organochlorine pollutants were quite high in the sediments near the mouth of the rivers on the east coast of India. HCH, aldrin and dieldrin were not found to be present in the marine organisms. However, endrin was sparingly detected in a few samples of fish. PCBs were detected in sediments and zooplanktons from the Indian coastal water and in various species of fish, seals and water from the southern Indian Ocean.

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