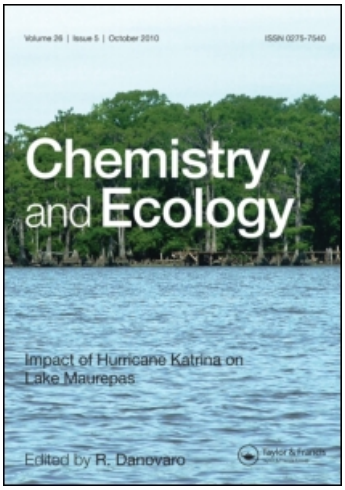

Chemistry and Ecology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713455114

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To cite this Article Rajendran, Ramasamy Babu and Subramanian, Annamalai(1997) 'Pesticide Residues in Water From River Kaveri, South India', Chemistry and Ecology, 13: 4, 223 — 236
To link to this Article: DOI: 10.1080/02757549708035529
URL: http://dx.doi.org/10.1080/02757549708035529

Chemistry and Ecology, 1997, Vol. 13, pp. 223–236
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PESTICIDE RESIDUES IN WATER FROM RIVER KAVERI, SOUTH INDIA

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(Received 10 October 1996; revised 16 December 1996)

The residue levels of presistent chlorinated pesticides such as HCH (hexachlorocyclohexane) isomers and DDT (dichlorodiphenyl trichloroethane) compounds were quantified in water samples collected from the River Kaveri and its distributor River Coleroon in Tamil Nadu, South India. HCH showed higher levels in River Kaveri during premonsoon (July to September) and monsoon (October to December) months, reflecting the HCH usage during that season for paddy crops. But in the case of DDT no clear trend in residue level was observed. The α-HCH was detected as the dominant isomer in all the three sampling sites. Among DDT compounds, p,p′-DDT and p,p′-DDE showed higher percentage of the total. International comparison of residue levels revealed that the present values are comparable to the waters from Asian and South East Asian nations, but lower than some samples from other parts of India. The value of DDT is well below the EEC's maximum acceptable concentration for surface waters and lower than the recommended limit of 2000 ng l^{-1} in USA water for protection of aquatic life (Water Quality Criteria, 1972).

Keywords: Chlorinated pesticides; HCH; DDT; river water

INTRODUCTION

Several hundred pesticides with different physical and chemical properties are used for agricultural and public health purposes. All of these can contaminate eventually natural water bodies (Tanabe *et al.*, 1984; Ramesh *et al.*, 1992; Fingler *et al.*, 1992; Miliadis, 1993). Organochlorine

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pesticides used widely in developing countries are known to resist biodegradation and therefore can be redistributed through the food chain and produce significant contamination at the apex of the food chain.

Rivers and streams are used increasingly as receptacles for the wastes generated on land. River Kaveri (Cauvery) is one of the major riverine systems in South India. The terminus of Kaveri and her discharges are covered by characteristic Coromondel shrub forests and mangroves representing ecosystems which nurture high biological diversity and fecundity. Any man-made input into this system will alter the natural balance of the animal and plant life even if the change may be so small as to be unmeasurable.

No work has been directed towards the contamination of persistent pesticides in water, sediment, flora and fauna of the Kaveri riverine system. The present study aims to identify and measure the concentration of organochlorine insecticides HCH (hexachlorocyclohexane) and DDT (dichlorodiphenyltrichloroethane) in water samples from the tail end region of River Kaveri and its distributor, River Coleroon.

MATERIALS AND METHODS

Sampling Sites

The River Kaveri is the eighth largest river (in terms of discharge) in the Indian subcontinent. It has a drainage area of about 90,000 km^2 covering a distance of 800 km from the Brahmagiri hills (12°25′N; 75°34′E) in the Sahayadri range in the Western Ghats until the river mouth at the Bay of Bengal. It is one of the most intensively utilised river basins in India. Since most of the water is withdrawn for irrigation, several distributaries form a network of a broad (about 100 km) and shallow delta at the river mouth. Coleroon is one of the main distributors of the River Kaveri covering a distance of about 200 km. It acts as an overflowing river. The Kaveri riverine system comprises Kaveri, Coleroon and other small distributors. Most of the area in Tamil Nadu state and especially the Kaveri basin gets substantial rain and the river witnesses a good river flow during the north east monsoon (October to December), although water flow during summer (April to June) is normally minimal.

Water samples were collected monthly at two locations in the Kaveri (Manikaranai-11°06′N; 79°42′E (site I) and Melaiyur-11°07′N; 79°45′E (site II)) and one location on the Coleroon river (Kollidam-11°21′N; 79°50′E (site III)) (Fig. 1) for the period of 18 months during October 1990 to March 1992.

Extraction and Cleanup

All the solvents used in residue analysis were of analytical grade and thoroughly distilled in all glass apparatus to avoid contamination. After thorough washing, the glassware was air-dried and wrapped with pre-cleaned (with Acetone) aluminium foil and baked in a hot air oven at 150°C overnight to remove traces of organic contaminants and finally rinsed with *n*-hexane before use.

Pesticides were extracted from the water according to the method described by Ramesh *et al.*, (1990). About 20 litres of water was collected in a polyethylene carboy and passed through pre-washed and dried Amberlite XAD-2 resin packed in a glass column of 10 mm i.d and about 100 mm length, and stored at 4°C until analysis. The organochlorine insecticides adsorbed to the Amberlite XAD-2 resin

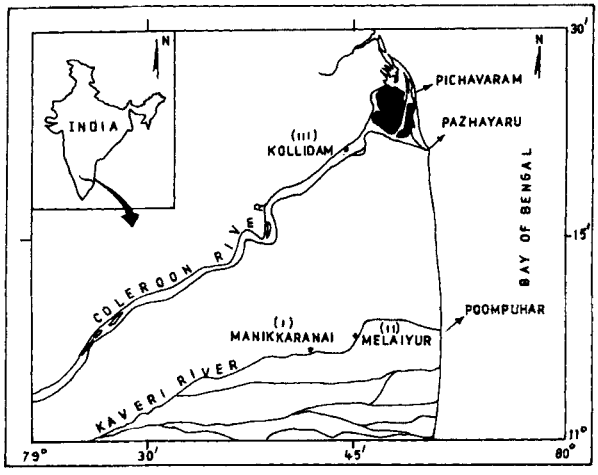

FIGURE 1 Map showing the sampling stations in Kaveri and Coleroon Rivers.

were eluted with 150 ml of ethanol into a separatory funnel containing 100 ml hexane and 750 ml hexane washed water and thoroughly shaken for 10 minutes. After partitioning, the hexane layer was collected and concentrated to 5 ml in a Kuderna-Danish (KD) concentrator. The concentrate was cleaned up with 5% fuming sulphuric acid in concentrated sulphuric acid, and then with hexane washed water.

Identification and Quantification

Identification and quantification of HCH and DDT were carried out in a Gas Chromatograph (GC), Hewlett Packard 5890-series II equipped with ^{63}Ni Electron Capture Detector (ECD). A glass column (2 mm i.d) of 6 ft. length packed with 3% OV-25 WHP 100/200 was used for all determinations. The oven temperature was 191°C held for 12 minutes and programmed at a rate of 5°C min^{-1} to 216°C and then held for 20 min. Nitrogen gas at a rate of 30 ml min^{-1} was used as the carrier gas. Injector and detector temperatures were kept at 225°C and 250°C respectively. They were quantified as individually resolved peaks based on retention times, in comparison with the corresponding peak height of standards (Ultra Scientific Co., USA). Peaks were integrated by a Hewlett Packard 3392 integrator.

To assure the accuracy and precision of the method, recovery studies were made by fortifying the sample with a known quantity of pesticide mixture of p, p′-DDE, p, p′-DDT and α-HCH. Recoveries ranged from 85 to 92 per cent and the results were not corrected for recovery percentage.

RESULTS

The variation in residual concentrations (range and mean) of HCH isomers (α, β and γ) and DDT compounds (p, p′-DDE, p, p′-DDD, o, p′-DDT and p, p′-DDT) in water samples collected from Kaveri and Coleroon rivers are given in Table I.

The concentration of α-HCH ranged from 1.9 to 101.3 ng l^{-1} at site I (Manikaranai), 10.2 to 83.2 ng l^{-1} at site II (Melaiyur) and 1.4 to 19.1 ng l^{-1} at site III (Kollidam). β-isomers were varied from 7.15 to 79.3, 9.15 to 67.1 and 0.7 to 11.4 ng l^{-1} at site I, II and III respectively.

TABLE I Seasonal variation of HCH and DDT residues (range and mean) levels ($ng\,l^{-1}$) in the water of Rivers Kaveri and Coleroon during 1990–'92

Season/Site		*HCH*			*ΣHCH*	*p,p'-DDE*	*p,p'-DDD*	*o,p'-DDT*	*p,p'-DDT*	*ΣDDT*
		α	*β*	*γ*						
Premonsoon	I	47.4–101.3 (74.3)	27.9–33.6 (30.8)	21.1–62 (41.6)	137.3–156 (146.7)	0.21–0.85 (0.53)	0.11–0.3 (0.21)	0.35–1.01 (0.68)	0.58–0.95 (0.77)	1.25–3.11 (2.18)
	II	32.3–83.2 (57.8)	26.1–29.7 (27.9)	63.1–69 (66.1)	121.4–182 (151.7)	0.11–1.82 (0.97)	0.04–0.5 (0.27)	0.27–0.75 (0.51)	0.38–1.1 (0.74)	0.8–4.17 (2.49)
	III	1.4–3.9 (2.71)	2.38–2.9 (2.71)	0.41–4.9 (2.55)	4.7–10.1 (7.97)	0.36–1.28 (0.81)	0.08–0.21 (0.13)	0.38–0.73 (0.58)	0.92–0.99 (0.95)	1.94–3.01 (2.47)
Monsoon	I	2.13–61.2 (41.4)	11.1–79.3 (31.3)	8.05–63 (22.2)	40.4–131 (94.8)	0.12–1.4 (0.87)	0.06–0.51 (0.23)	0.33–0.88 (0.64)	0.35–1.21 (0.78)	2.05–2.99 (2.51)
	II	22.4–53.1 (35.9)	12.2–67.1 (29.4)	7.75–73 (30.0)	69.2–115 (95.2)	0.11–1.52 (0.59)	ND–0.45 (0.27)	0.26–0.63 (0.39)	0.38–0.98 (0.55)	0.75–3.27 (1.80)
	III	1.9–13.1 (7.97)	0.7–7.96 (2.89)	0.35–9.2 (3.49)	3.2–24.8 (14.4)	0.16–1.01 (0.42)	0.08–0.38 (0.2)	0.21–0.85 (0.52)	0.44–0.96 (0.62)	0.97–2.14 (1.76)

TABLE I (Continued).

Season/Site		*HCH*			*ΣHCH*	*p,p′-DDE*	*p,p′-DDD*	*o,p′-DDT*	*p,p′-DDT*	*ΣDDT*
		α	β	γ						
Post monsoon	I	14.7–43.3	7.15–39.4	5.8–17.3	30.3–79	0.36–1.82	0.11–0.5	0.36–0.86	0.55–1.25	2.06–3.48
		(26.1)	(19.1)	(16.3)	(53.9)	(0.92)	(0.25)	(0.63)	(0.84)	(2.63)
	II	10.2–63.3	10.3–41.1	2.2–27.1	23.7–106	0.12–1.4	ND–0.25	0.15–0.78	0.37–0.57	1.05–2.78
		(23.9)	(25.5)	(9.05)	(58.4)	(0.67)	(0.11)	(0.39)	(0.45)	(1.63)
	III	9.2–19.1	4.6–11.4	0.1–4.2	17.6–30.2	0.12–1.4	ND–0.22	0.26–1.01	0.42–0.93	0.87–2.61
		(12.9)	(7.52)	(2.73)	(23.2)	(0.56)	(0.1)	(0.57)	(0.65)	(1.88)
Summer	I	1.9–2.55	7.3–10.2	0.9–1.4	11.3–13	0.43–0.63	0.29–0.39	0.12–0.48	0.21–1.02	1.05–2.52
		(2.23)	(8.75)	(1.15)	(12.2)	(0.53)	(0.34)	(0.3)	(0.62)	(1.79)
	II	19.3–26.4	9.15–9.7	3.45–5.8	32.4–41.3	0.16–0.75	0.25–0.6	0.6–0.75	0.35–0.95	1.95–2.46
		(22.9)	(9.43)	(4.63)	(36.9)	(0.46)	(0.43)	(0.68)	(0.65)	(2.21)
	III	2.45–4.1	1.95–7.7	0.2–1.3	7.35–11.3	0.11–0.96	0.17–0.26	0.35–0.78	0.29–1.06	0.94–3.05
		(3.22)	(5.32)	(0.87)	(9.42)	(0.44)	(0.23)	(0.5)	(0.71)	(1.88)

ND- Not Detected
ΣHCH- Sum of α, β and γ-isomers
ΣDDT- Sum of p,p′-DDE, p,p′-DDD, o,p′-DDT and p,p′-DDT

The respective concentration of γ-isomer at site I, II and III were ranged from 0.9 to 63, 2.2 to 73 and 0.1 to 9.2 $ng\,l^{-1}$.

The levels of DDT metabolite p,p'-DDE varied from 0.12 to 1.82, 0.11 to 1.82 and 0.11 to 1.4 $ng\,l^{-1}$ at site I, II and III respectively. Another metabolite, p,p'-DDD, was found from 0.06 to 0.51, ND to 0.6 and ND to 0.38 $ng\,l^{-1}$ at site I, II and III respectively. The concentration of parent compounds, o,p'-DDT and p,p'-DDT, ranged from 0.12 to 1.01 and 0.21 to 1.25, 0.15 to 0.78 and 0.35 to 1.1, and 0.21 to 1.01 and 0.29 to 1.06 $ng\,l^{-1}$ at site I, II and III respectively. In all the three sites ΣHCH concentration was more (3.2–182 $ng\,l^{-1}$) than ΣDDT concentration (0.75–4.17 $ng\,l^{-1}$).

DISCUSSION

A clear seasonal variation in the distribution of HCH was observed. HCH was found more during premonsoon (July to September) followed by monsoon (October to December), post monsoon (January to March) and summer (April to June) seasons. Sites I and II at Kaveri river had more or less similar residual distribution pattern and higher concentrations of both HCH and DDT than that of Site III (i.e., Coleroon river).

The occurrence of the high concentration of HCH compounds at sites I and II during premonsoon and monsoon months, matching a wet season reflecting the use of HCH (10% BHC dust) to rice crops in that cropping season in the entire deltaic region. Drainage of agricultural wastewater would be the important source of HCH residues in Kaveri river as reported elsewhere (Bevenue *et al.*, 1972; Rajendran, 1984; Karthikeyen, 1988).

Li (1975) also reported that agricultural run-off from fields and grazing lands could be considered as an important route of pesticide transport to the nearby water bodies. Moreover, the concentration of pesticides increased with the sub-soil movement of water carrying pesticides from the catchment area into water bodies. In addition, rainfall may be also a possible reason for higher concentration during wet season. The concentration observed in water also depends on enrichment and dilution phenomena caused by rainfall (Polemio *et al.*, 1983).

The Kaveri river is bordered on both sides by paddy fields and the river water is much used for irrigation purposes and further acts as a disposal point for agricultural channels, so the chances of contamination of this river with pesticides (HCH) used is more than that of its distributor in the Coleroon. The low residue levels of HCH recorded at site III (Coleroon River) even during premonsoon and monsoon months (the cropping season in this region) could be due to the absence of local source with land runoff leaching from nearby fields to Coleroon River as suggested by Rajendran (1984).

These observations are quite agreeable with the reports of El-Dib and Badawy (1985), who have reported increased residue levels (BHC 46 $ng\,l^{-1}$) at Al-Mansora, one of the most productive agricultural area in the Nile delta, reflecting the extensive usage and leaching of insecticides. Kilikidis *et al.* (1992) confirmed the increase in the concentration of organochlorine pesticides in the waters of Strimon river of Northern Greece during the period of increased agricultural activity. Miliadis (1994) observed maximum concentration of lindane in Iliki Lake in Greece and further explained that the maxima indicate the agricultural origin of the lake's pollution. Tan and Vijaletchumy (1994) recorded high levels of DDT (ND-190 $ng\,l^{-1}$), HCH (ND-320 $ng\,l^{-1}$) and endosulfan (ND-310 ngl^{-1}) in the waters of rivers flowing through major rice growing areas than from the rivers along the east coast of Peninsular Malaysia. These pesticides used in agriculture have been a contributing factor for higher levels recorded.

Among the HCH isomers, the concentration of α-isomer was greater followed by β and γ at sites I and III, but at site II the α-HCH was followed by γ and β respectively. Usually the γ-isomer is greater (i.e. 14%) in technical grade following α-HCH. However, the β-HCH was greater at sites I and II, which may be due to isomerization of α to β in the environment (water) as suggested by Hayes (1982). Moreover, β-isomer is more persistent than α and γ-HCH. The mean percentage composition of all the three isomers and its order of distribution is shown in Figure 2.

The ratio of α to γ-HCH indicate the time spent by HCH isomers in the environment as well as their transportability (Falandysz *et al.*, 1994). In the present study higher α/γ ratio found (Tab. II) during the summer season than in the other seasons in all the three sites, and indicate the photochemical transformation of γ-HCH as pointed out

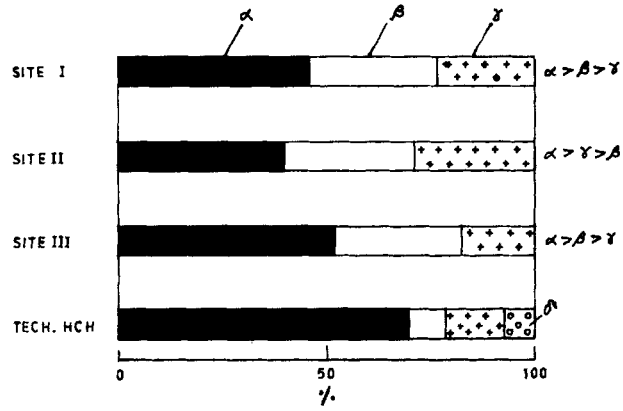

FIGURE 2 Mean percentage composition of HCH isomers.

TABLE II Mean concentration ratio of pesticide residues

Compound	*Premonsoon*			*Monsoon*			*Post Monsoon*			*Summer*		
Site	*I*	*II*	*III*	*I*	*II*	*III*	*I*	*II*	*III*	*I*	*II*	*III*
α/γ-HCH	1.79	0.87	1.06	1.86	1.2	2.28	1.6	2.64	4.7	1.94	4.95	3.7
p,p′-DDE/ΣDDT	0.24	0.4	0.33	0.35	0.33	0.24	0.35	0.41	0.3	0.3	0.21	0.23
p,p′-DDT/ΣDDT	0.37	0.3	0.39	0.31	0.31	0.35	0.32	0.28	0.35	0.35	0.29	0.38

by Pacyna and Oehme (1988) and also the isomerization of γ-HCH to α-HCH (Benezet and Matsumura, 1973).

DDT and its metabolites does not show any such clear difference between stations and between seasons. Relatively low concentrations of DDT and high concentrations of HCH recorded in the present investigation could be attributed to the quantities of these pesticides used in public health activities and agriculture. Similar observations were also made by Iwata *et al.* (1994) in Chao Pharya river, Thailand, Pentai Jeram river of Malaysia, Ciliwung river of Indonesia and Yamuna river of India, and Hernandez *et al.* (1992) in Guadalquivir river of Spain (Tab. III). The government of India has severely restricted the use of DDT except for malaria control. HCH was also banned for use in vegetables, fruits, oilseed crops, but is still heavily used in controlling pests of cereal crops and commercial crops.

Comparing the values of DDT compounds in different seasons, showed that the mean concentration of p,p′-DDD was more during

TABLE III Range/Mean concentration of ΣHCH and ΣDDT in water samples from various rivers in the world

Name	*Survey Year*	*Concentration ($ng\,l^{-1}$)*		*References*
		ΣHCH	*ΣDDT*	
1. Nile river, Egypt	1982	0.1–18.9*	8.6–58.8	El-Dib & Badawy (1985)
2. Bengawan Solo river, Indonesia	1984	0.94	1.2	Hillebrand *et al.* (1989)
3. Kali Porong river, Indonesia	1984	1.0	1.4	Hillebrand *et al.* (1989)
4. Kupa rivers, Croatia	1988–'89	<1–20	1–6	Fingler *et al.* (1992)
5. Major rivers of Malaysia	1990–'91	ND-320	ND-190	Tan & Vijayaletchumy (1992)
6. Guadalquivir river, Spain	1990	9–137	1–33	Hernandez *et al.* (1992)
7. Chao Pharya river, Thailand	1990	18	0.34	Iwata *et al.* (1994)
8. Pentai Jeram, Malaysia	1991	1900	1.7	Iwata *et al.* (1994)
9. Ciliwung river, Indonesia	1991	3.1–22	0.19–0.27	Iwata *et al.* (1994)
10. Ilike lake, Greece	1991–'92	15*	NA	Miliadis (1994)
India				
1. Hoogly river, Calcutta	1982–'84	1–400	2–560	Thakker (1986)
2. Yamuna river, Delhi	1984	NA	2100–34000	Subba Rao *et al.* (1986)
3. Ganga river	NA	100–22000	NA	Halder *et al.* (1990)
4. Vellar river, Tamil Nadu	1987–'89	26–3900	0.051–4.8	Ramesh *et al.* (1990)
5. Yamuna river, Delhi	1989	660	120	Iwata *et al.* (1994)
6. Kaveri river, Tamil Nadu	1990–'92	79.3	2.16	Present study
7. Coleroon river, Tamil Nadu	1990–'92	15.4	1.93	Present study

*- Lindane
NA- Date Not available
ND- Not detected

summer than in other seasons at all the sites, indicating the photochemical degradation of parent compound into DDD during summer months. Figure 3 shows the mean percentage composition of DDT compounds, where p,p′-DDE was predominant at site I and II but at site III the parent compound, p,p′-DDT, was greater. Another metabolite, p,p′-DDD, was low. In general high values of p,p′-DDE and p,p′-DDT in the present investigation indicate their longer residence time in the environment.

The mean ratio between α and γ-HCH and DDT and its metabolites are given in Table II. At both sites in River Kaveri, the p,p′-DDE/ΣDDT ratio was greater during most of the year, inferring that the degradation of DDT to DDE in the environment is greater or the addition of DDE residue through river runoff or the addition of airborne residues through precipitation. At site III (Coleroon River) the p,p′-DDE/ΣDDT ratio was less than p,p′-DDT/ΣDDT throughout the study period. The water control constructed for irrigation purposes in Kaveri River resulted in poor dispersal of pesticides released to the river. Such poor water exchange resulted in the degradation of DDT to its metabolite DDE which may be another reason for high p,p′-DDE/ΣDDT ratio, even though at Coleroon River there is no such water control from its starting point at Mokkombu. So, the pesticides reaching Coleroon River are flushed away to the sea at a faster rate than at River Kaveri where the pesticides stay for a longer duration and are metabolised.

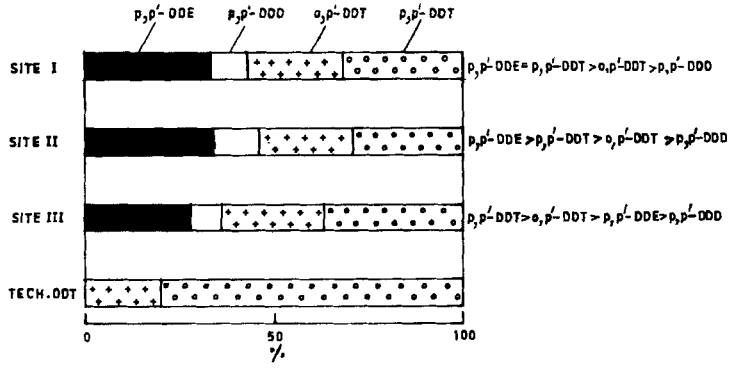

FIGURE 3 Mean percentage composition of DDT compounds.

The levels of DDT recorded are comparable with Bengawan Solo and Kali Porong Rivers of Indonesia (Hillebrand *et al.*, 1989); Kupa River of Croatia (Fingler *et al.*, 1992); Chao Pharya River (Thailand), Pentai Jeram River (Malaysia), Cliwung River (Indonesia), and Ulsoor Lake ($0.87\,ng\,l^{-1}$), Mandovi River ($3.1\,ng\,l^{-1}$), Hoogly River ($1.5\,ng\,l^{-1}$) and Vellar River ($0.87\,ng\,l^{-1}$) of India (Iwata *et al.*, 1994), but quite lower than some other rivers (Hoogly and Yamuna) of India (Tab. III).

The concentrations of DDT residues found in water samples from Hoogly and Yamuna Rivers in 1982–84 were more (Thakker, 1986; Subba Rao *et al.*, 1986) when compared the concentrations recorded in 1989 (Iwata *et al.*, 1994) (Tab. III). It shows clearly the phasing out of DDT in agriculture in India and minimum use in public health programmes.

CONCLUSION

The quantified residue levels (HCH to 79.3 $ng\,l^{-1}$ and DDT to 2.16 $ng\,l^{-1}$) in the present study are well below the EEC's maximum acceptable concentration of 1000–3000 $ng\,l^{-1}$ in surface water for pesticides (E.C. Council Directive, 1980). The concentration of p,p′-DDT (0.29–1.25 $ng\,l^{-1}$) was lower than its recommended limit of 2000 $ng\,l^{-1}$ in USA water for protection of aquatic life (Water Quality Criteria, 1972). The maximum gamma-HCH concentration ($73\,ng\,l^{-1}$) observed is still below the ecologically acceptable level ($100\,ng\,l^{-1}$) in fresh waters prescribed by UNEP (1989). The residue levels of HCH and DDT quantified here is lower even though large quantities are used. The pesticide consumption in the lower reaches of Kaveri River is rather higher ($45\,kg\,ha^{-1}$) than in the upper reaches $38.2\,kg\,ha^{-1}$ (Agnigothrudu, 1985). Takeoka *et al.* (1991) pointed out that 99.6% of the applied insecticide (HCH) in the Vellar watershed, India (coastal area) were removed/transported to air and only 0.4% is drained to the estuary. Moreover, about 75% of the flux to the estuary is again removed to the air, and finally 0.1% is drained ultimately to the sea. Further, they stated that HCH flux to the coastal water bodies through water discharge is less significant. Its residence time in the aquatic environment is quite short whereas transfer to the atmosphere

is large in the tropical areas. In conclusion, the prevailing climatic conditions in the study area may be considered as playing a major role in transport of the residue to more distant places. Further intensive monitoring study on pesticide residues in terrestrial, aquatic and atmospheric media is needed to find out the pesticide flux data through rivers into the adjacent seas.

Acknowledgement

The authors thank the University authorities and the Department of Ocean Development, New Delhi for the facilities provided. One of the authors (R.B.) wishes to thank the Ministry of Environment and Forests, New Delhi for the award of fellowship.

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