

Organochlorine Insecticide Residues in Water from Five Lakes of Nainital (U. P.), India

V. K. Dua,¹R. Kumari,¹R. K. Johri,²V. P. Ojha,¹R. P. Shukla,³V. P. Sharma⁴

¹Malaria Research Centre (Field Station), Section III, BHEL, Ranipur, Hardwar 249 403, India

²D.A.V. Postgraduate College, Dehradun 248 001, India

³Malaria Research Centre (Field Station), Haldwani, Nainital

⁴Malaria Research Centre, 22 Sham Nath Marg, Delhi 110 054, India

Received: 3 October 1997/Accepted: 7 December 1997

Organochlorine insecticides such as DDT and HCH have been extensively used in public health in India (Gupta 1986). However, due to their chemical nature, they became major environmental pollutions. Several studies were conducted on organochlorine insecticide contamination in lakes and water reservoirs from other part of the world (Kucklick et al, 1994, Tanabe et al. 1983). However few investigations have assessed organochlorine residues in water reservoirs in India i.e. Mahala water reservoir (Bakre et al. 1990), Jamuna water (Agarwal et al. 1986) Ganga water (Nayak et al. 1995; Sinha 1991) and rural ponds water (Dua et al. 1996). All these studies are confined to those area where DDT and HCH were used for the control of vector borne diseases.

Nainital district is situated at an altitude of 1934 m (MSL) at southern extremity of lesser Himalyan zone in Kumaun region of U.P state, India. It is located at 29.23 north latitude and 79.3 east longitude having an area of 6794 km. The terrain is heterogenous consisting of 18% hilly forest, 47% forest, 4% bhabar and 29% terai area with a net work of lakes, rivers, rivulets and canals. The average rain fall ranged from 1275 mm to 3050 mm dependidng on the terrain. Five lakes namely Bhimtal, Sattal, Khurpatal, Naukuchiatal and Nainital are situated in the hilly area of Nainital region. Vital statistics of these lakes are given in Table-I. It may be noted that no insecticide was used in the vicinity of lakes for vector control programme while DDT and HCH were extinsively used in Terai and Bhabar area for malaria control. Water from these lakes was the only source for drinking and domestic use for people residing in these areas. Lakes water was supplied to the community through water pipe after some treatment. Therefore the present study was conducted to monitor the levels of HCH and DDT in the water from these five lakes of Nainital.

MATERIALS AND METHODS

Four 500 ml water samples from each lake were collected in clean 1 litre glass bottles by immersing them about 30 cm below the surface of water during the months of March, July and November, 1994. All samples were brought to the laboratory and stored in the refrigerator.

The extraction of HCH and DDT from water was carried out as reported earlier (Agarwal et al., 1986). Water samples were filtered using Whatman filter paper No. 1. Filter paper was washed with 50 ml distilled water to elute residual pesticide. 250 ml water was extracted three times with 50 ml n-Hexane for 10 minutes in a separating funnel and upper n-Hexane layer was pooled and concentrated to 1 ml using vortex evaporator. The concentrated extract was cleaned with anhydrous sodium sulphate-alumina column eluted with in-haxane-benzene (40 : 60). The eluant was evaporated on a vortex evaporator and kept at 4°C until analysis.

Samples were analysed for organochlorine insecticide residues on Hewlett Packard 5890 gas chromatograph fitted with Ni⁶³ electron capture detector on fused silica capillary TM5column PTE (length 30m, 0.25mm id) Spelco Corp. USA. Nitrogen was used as a carrier gas @ 2ml/min. (split ratio 1 : 100). The injector, oven and detector temperatures were set at 210, 190 and 220° C respectively. All water samples were analysed separately and their mean value was calculated for the determination of insecticide residue in a particular lake. Level of detection for DDT was 0.1ng while for HCH was 0.2 ng. Below this value was termed as N.D (not determined). The correlation between two variables were calculated by Carl Pearson method.

RESULTS AND DISCUSSION

The average percentage recoveries of HCH isomers, DDT and its metabolites in water were more than 85% and recorded values were not corrected for recoveries. Residual levels of HCH and DDT in five lakes under study are given in Table 2 and 3. HCH residues in Khurpatal in the months of March, July and November ranged from 2.408-3.00 ug/L, 4.40-5.349 ug/L and 3.105-3.406 ug/L respectively with the maximum mean HCH level of 4.793 ug/L in July. γ -HCH contributed 57% followed by α -HCH 27% of the total HCH residue of July. In March and November, β -HCH was found in maximum quantity among HCH isomers. No δ -HCH was detected in any sample of lake Khurpatal. DDT residue was also present maximum in July followed by November. pp DDT was major metabolite in July and November while pp DDE was contributed about 60% in March samples. Traces of o-p DDT was also detected in July.

Mean HCH residues in water of March, July and November samples of lake sattal were 3.261, 4.831 and 3.222 ug/L respectively with the maximum concentration in July, γ -HCH contributed maximum among HCH isomers in July while β -HCH was present as major isomers in march and november. DDT contamination was

Table 1. Vital statistics of five lakes used for water sample collection for HCH and DDT contamination

Lake	Height above sea lable a (m)	Length (m)	Breadth (m)	Depth (m)	Area (m ²)	Other details
Sattal	1347	NA ^b	NA	NA	NA	catchment 3.3 km ²
Naukuchiatal	1219	950.7	691.7	40.2	657694	catchment 2.8 km ²
Bhimtal	1372	1700	454	26	772287	catchment 11.9 km ²
Khurpatal	1036	1365	558	38	762768	-
Nainital	1936	1442	462	28	667346	circumference 3492 m

a) m = meter ; b) NA = not available.

also found maximum in July (mean 10.840 ug/L) with pp DDT as major metabolite while pp DDE was present about 43% in March and represented maximum among DDT metabolites. No op DDT was detected in any sample.

The behaviour of HCH and DDT contamination in water of lakes Bhimtal and Naukuchiatal is similar to Khurpatal and Sat tal. Maximum HCH and DDT concentration were observed in July with γ -HCH and pp DDT as major constituents respectively while β -HCH and pp DDE were found maximum in March. Total HCH and DDT contamination were in order of July > November > March. δ -HCH and op DDT were absent in all samples of both lakes. Naukuchiatal had minimum HCH and DDT contamination among all lakes under study.

Mean HCH contamination in water from lake Nainital in July and November, months were similar with slightly higher value in July (3.825 ug/L). Again γ -HCH was found maximum in July and November while β -HCH (75%) represented maximum in March sample. Highest DDT concentration was recorded in lake Nainital with the mean value of 31.336 ug/L in July. One water sample contained as high as 37.165 ug/L DDT contamination. γ -HCH contributed 60% in July and November collection while pp DDE was the major metabolite found in March. The percentage contribution of pp DDT in March, July and November were 42,34 and 16% of the total DDT residues present respectively.

HCH and DDT contamination of varying degree were recorded in all five lakes under study from Dist. Nainital. Maximum concentrations of these residues were found in July while minimum residual levels were recorded in March. Bakre et al.

Table 2. HCH concentrations in water from five lakes of Nainital

Lake	Months	Mean Concentration ug/l			
		α -HCH	β -HCH	γ -HCH	Total HCH
Khurpatal	Mar	0.736 (0.63-0.881)	1.828 (1.778-1.876) ^a	0.105 (0.0-0.340)	2.668 (2.408-3.004)
	Jul	1.342 (1.274-1.506)	0.707 (0.315-0.991)	2.743 (2.135-3.00)	4.793 (4.406-5.349)
	Nov	0.490 (0.403-0.604)	2.600 (2.500-2.667)	0.145 (0.097-0.203)	3.235 (3.105-3.406)
Sattal	Mar	1.217 (1.99-1.237)	1.843 (1.700-1.932)	0.200 (0.187-0.219)	3.261 (3.099-3.369)
	Jul	1.412 (1.382-1.478)	1.167 (1.067-1.280)	2.252 (2.086-2.514)	4.831 (4.631-5.176)
	Nov	0.434 (0.198-0.558)	1.509 (1.002-1.867)	1.279 (1.182-1.371)	3.222 (2.380-3.790)
Bhimtal	Mar	0.488 (0.423-0.542)	2.061 (1.987-2.106)	0.050 (0.0-0.086)	2.598 (2.529-2.691)
	July	0.945 (0.904-0.967)	3.786 (3.20-4.201)	4.081 (3.861-4.372)	8.656 (8.533-8.834)
	Nov	0.641 (0.567-0.738)	2.013 (1.999-2.038)	2.301 (2.049-2.430)	4.955 (4.730-5.167)
Nauku chital	Mar	0.302 (0.295-0.312)	1.001 (0.988-1.013)	0.037 (0.031-0.041)	1.340 (1.337-1.343)
	Jul	0.860 (0.844-0.885)	0.950 (0.911-0.989)	1.311 (1.236-1.396)	3.121 (3.044-3.229)
	Nov	0.145 (0.134-0.154)	1.221 (1.031-1.500)	0.396 (0.333-0.430)	1.749 (1.569-1.987)
Nainital	Mar	0.750 (0.470-0.960)	2.232 (1.860-2.866)	0.211 (0.092-0.370)	2.943 (2.530-3.428)
	Jul	1.012 (0.835-1.270)	1.036 (0.834-1.278)	1.778 (0.983-2.860)	3.8251 (2.818-5.408)
	Nov	0.418 (0.393-0.440)	1.554 (0.918-2.667)	1.914 (1.756-2.070)	3.636 (3.152-4.137)

a = Values in the parentheses denotes range. ND - not detected

(1990) have also found maximum pesticides in water from Mahala reservoir with the onset of rains. This may be attributed to the sub soil movement of water carrying pesticides from the catchment area into the water reservoir. The presence of organochlorine in different lakes may be accounted due to atmospheric transportation of pesticides by evaporation followed by cold condensation mechanism or by misuse of these chemicals in agriculture. It is to point out that

Table 3. DDT concentrations in water from five lakes of Nainital

Lake	Months	Mean concentration (ug/L)				
		pp DDE	op DDT	pp DDT	pp DDD	Total DDT
Khurpatal	Mar	8.917 (8.703-9.206)	(ND)	0.794 (0.784-0.804)	4.827 (4.732-4.889)	14.785 (14.219-15.85)
	Jul	2.735 (ND-3.810)	0.243 (0.201-0.284)	18.581 (17.458-19.753)	2.542 (2.321-2.844)	24.741 (22.567-26.20)
	Nov	4.637 (3.928-5.449)	ND	9.730 (9.333-10.001)	4.327 (3.556-4.889)	18.695 (17.484-20.060)
Sattal	Mar	1.292 (1.053-1.681)	ND	0.681 (0.567-0.870)	1.013 (1.003-1.031)	2.986 (2.628-3.554)
	Jul	1.597 (1.336-1.981)	ND	8.151 (7.954-8.500)	1.084 (1.011-1.131)	10.840 (10.301-11.551)
	Nov	0.824 (ND-1.364)	ND	3.776 (3.493-4.021)	0.496 (0.078-0.786)	5.097 (4.552-5.445)
Bhimtal	Mar	2.674 (2.444-2.870)	ND	0.354 (0.208-0.487)	1.934 (1.872-2.005)	4.961 (4.804-5.083)
	Jul	2.277 (1.140-2.941)	ND	5.718 (4.764-8.204)	1.834 (1.431-2.033)	8.829 (7.937-9.369)
	Nov	0.924 (0.438-1.404)	ND	3.942 (3.883-4.000)	0.709 (0.347-1.033)	5.575 (5.444-5.736)
Nauku-chiatal	Mar	1.154 (0.896-1.422)	ND	0.289 (0.211-0.345)	0.817 (0.637-0.946)	2.261 (2.130-2.366)
	Jul	0.950 (0.900-0.986)	ND	4.330 (4.098-4.845)	0.774 (0.672-0.891)	6.054 (5.852-6.463)
	Nov	0.878 (0.819-0.994)	ND	1.733 (1.600-1.873)	0.847 (0.761-0.900)	3.458 (3.427-3.494)
Nainital	Mar	6.112 (5.481-6.822)	ND	1.913 (0.400-2.183)	5.653 (4.444-7.000)	13.437 (12.892-13.850)
	Jul	1.643 (0.986-2.048)	ND	18.791 (14.815-22.222)	10.902 (9.333-12.991)	31.336 (27.803-37.165)
	Nov	3.481 (3.400-3.557)	ND	8.807 (7.342-9.990)	2.363 (1.690-2.984)	14.652 (13.760-15.080)

ND = not detected a) values in the parenthesis denotes range

HCH and DDT are not used for the control of vector borne diseases near lake areas and their use for agriculture is also banned in India. However, in Terai and Bhabar area of dist. Nainital these insecticides were extensively used for malaria control programme. γ -HCH was recorded maximum in July from all lakes while it was found at minimum levels in March samples. β -HCH was dominated among HCH isomers in March. High level of γ -HCH may be due to continued input of this

Table 4. HCH and DDT residues in tap water from the vicinity of five lakes

Residues	Lakes ^a				
	Kuarpatal	Sattal	Bhimtal	Naukuchiatal	Nainital
α -HCH	1.150	0.943	0.461	0.093	0.206
β -HCH	0.290	1.017	1.077	0.987	1.441
γ -HCH	2.341	0.923	1.092	0.103	0.112
Total HCH	3.782	2.884	2.629	1.183	1.756
pp DDE	3.744	1.484	1.722	1.444	5.126
op DDT	0.225	ND	ND	ND	ND
pp DDT	10.247	7.193	3.721	1.201	7.901
pp DDD	2.798	0.090	0.539	0.104	2.796
Total DDT	17.014	8.767	5.982	2.749	15.822

a = mean of 4 samples ; Concentrations are in ug/L

ND = not detected

isomers. Similarly high value of pp DDT is an indication of recent application while the presence of pp DDE and pp DDD resulted due to degradation process (Bakre et al. 1990).

The contamination of HCH in July ranged from 3.121 to 8.656 ug/L. This range is higher than the EC limit of 3 ug/L established for natural water (Council of the European Communities 1980). Except July samples from Bhimtal lake, all other samples do not exceeded the maximum permissible limit for γ -HCH (4 ug/L) reported by WHO (1984). Mean DDT contamination in all lakes ranged from 6.054 to 3 1.336 ug/L which showed that all lakes water exceeded the maximum permissible limit of DDT (1 ug/L) for drinking water (WHO 1984). Recently Nayak et al. (1995) have found that many water samples from Ganga river exceeded the safe limit while Bakre et al. (1990) reported that almost all samples in Mahala water reservoir were above the maximum permissible limits for DDT and HCH.

Contamination of HCH and DDT was also recorded in tap water used as drinking purposes by the community (Table 4). It is remarkable to note that significant correlation was found between the residual level of tap water and corresponding lake water of HCH ($r = 0.73$; $p < 0.1$) and DDT ($r = 0.93$; $p < 0.001$). All tap water samples exceeded maximum permissible limit for DDT (1 ug/L).

A comparison of HCH residues in different lakes showed that HCH was found maximum in Bhimtal and minimum in Naukuchiatal. The HCH contamination in other three lakes viz, Sattal, Khurpatal and Nainital were approximately same. The residual level of DDT was highest in Nainital lake followed by Khurpatal while it

was found minimum in Naukuchiatal. Low or high contamination of pesticides in these lakes may be due to the location of lakes and the magnitude of agriculture fields near the particular lake command areas associated with the insecticide used. It may be noted that Nainital and Khurpatal lakes are surrounded by small agriculture forms in hills and illegal use of DDT is quite common in agriculture farming thereby resulting very high DDT contamination in these lakes while Naukuchiatal is situated in forest area therefore, DDT contamination was minimum.

Acknowledgements. We thank staff of MRC Haldwani for their help in collection of samples. Thanks are also due to Mr. Rajesh Mittal and S.P. Sethi for technical assistance during sample processing. The authors express appreciation to Mr. R.S. Bhardwaj for typing the manuscript.

REFERENCES

- Agarwal HC, Mittal PK, Menon KB, Pillai MKK (1986) DDT residues in the river Jamuna in Delhi, India. *Wat Air Soil Pollut* 28:89 -104.
- Bakre PP, Mishra V, Bhatnagar P (1990) Organochlorine residues in water from the Mahala water reservoir, Jaipur, India. *Environ Pollut* 63 : 275-281.
- Council of the European Communities (1980) Council directives of 15 July 1980 relating to the quality of water intended for human consumption (80/778/EEC). *Off J Eur Commun L229*: 11-29.
- Dua VK, Kumari Roop , Sharma VP (1996) HCH and DDT contamination of rural ponds of India *Bull Environ Contam Toxicol* 57 : 568 - 574.
- Gupta PK (1986) Pesticides in the Indian Environment. In : Bhatia, B and Varshney, S.K. (eds). Inter print publishers, New Delhi, India, p 12.
- Kucklick JR, Bidleman TF, McConnell LL, Walla MD, Ivanov GP (1994) Organochlorines in water and biota of lake baikal, Siberia. *Environ Sci Technol* 28 : 31 - 37.
- Nayak AK, Raha R, Das AK (1995) Organochlorine pesticide residues in middle stream of the ganga river, India. *Bull Environ Contam Toxicol* 54 : 68 -75.
- Sinha AK (1991) A comprehensive study of Ganga and its dependent. In : Krishnamuru, C.R., Bilgrami, K.S., Das, T.M. and Mathur, R.P. (eds). The Ganga - a scientific study, Northern Book Centre, New Delhi p 125-140.
- Tanabe S, Hidaka H, Tatsukawa R (1983) PCBs and Chlorinated hydrocarbon pesticides in Antarctic atmosphere and hydrosphere. *Chemosphere* 12 : 277 - 288.
- WHO (1984) Guidelines for drinking water quality. II : 190-218.