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Lindane Pollution Near an Industrial Source in Northeast Spain

L. M. Hernandez, M. A. Fernandez, and Mª. J. Gonzalez

Department of Environmental Contamination, Institute of Organic Chemistry (CSIC), c/ Juan de la Cierva 3, 28006 Madrid, Spain

Since DDT has been legally restricted for use in many countries, lindane, the gamma isomer of hexachlorocyclohexane, (γ-HCH), has become important as a substitute for DDT. Thus, γ-HCH has been used in the control of various wood-inhabiting beetles, in seed treatment, and in pharmaceutical preparations as a scabicide and pediculicide.

Lindane is degraded poorly in the environment: it is hydrolyzed poorly and biodegrades slowly. Lindane is relatively immobile in soil. The bioaccumulation and toxicity to aquatic organisms have been reported in the literature. In the manufacture of lindane, the other HCH isomers are formed as a mixture. During the manufacturing process a mixture of isomers is formed, comprising 65-70% α-HCH, 7-10 β-HCH, 14-15% γ-HCH, 7% δ-HCH, 1-2% ε-HCH, and 1-2% other. Even more dangerous that lindane itself are the waste products from its manufacture (C.I.E.L., 1982).

The town of Sabiñanigo, located in northeast of the Iberian Peninsula, is host to one of only two factories in western Europe that manufacture lindane. HCH waste is being dumped near Sabiñanigo by the chemical company Inquinosa. The Gallego river runs only a few metres away from the main dumping site; apparently, the pesticide waste is leaking into the drinking water supply (Markham, 1989).

The purpose of this investigation are: 1) to determine the levels of HCH isomers in water, soil, vegetation, and invertebrates sampled in five places of the Gallego river; 2) to evaluate biological accumulation of pollutants studied within the food webs; 3) to find out if the residue levels exceeded the limits recommended for HCH in water.

MATERIALS AND METHODS

Location of sampling stations was shown in Figure 1. All stations

Send reprint requests to L.M. Hernández at the above address.

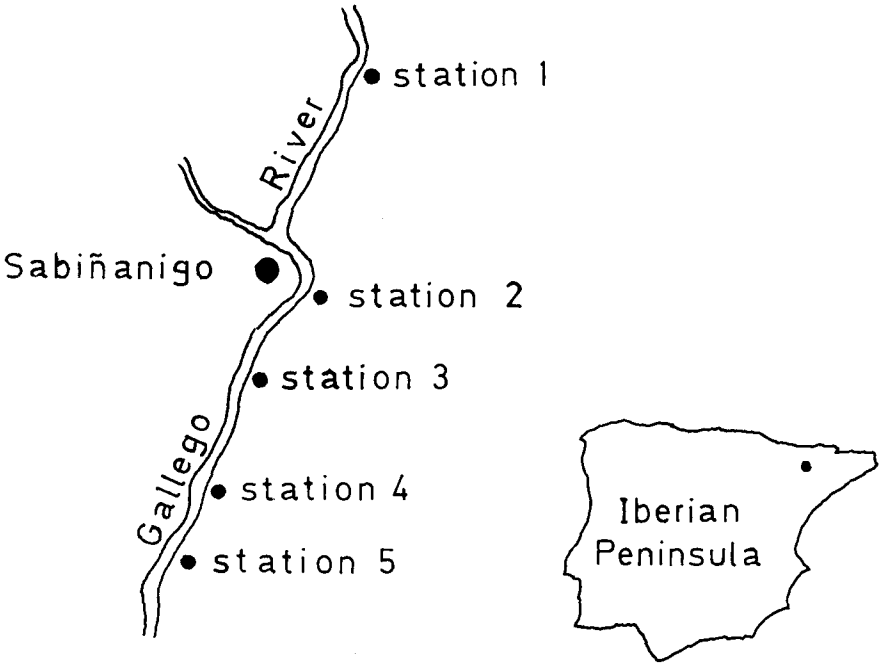

Figure 1. Study area and sampling locations.

were located in the shore of Gallego river. Five stations were selected: station 1 was situated in 6 km upstream from the factory and has been taken as a reference for the "natural pollution" from these pesticides; station 2 was situated beside the factory; stations 3, 4, and 5 were situated in 2, 5, and 7 km downstream from the factory, respectively.

Water samples (three liters at a depth of 25 cm) were collected in glass bottles. Soil samples were collected from the moist shore. Earthworms (Lumbricus terrestris L.) removed from these soils, were placed in petri dishes on moist filter paper three days to void their gut. Plants (Poa sp.) were sampled in the same stations. All samples were collected 30 may 1989. Soil and vegetal samples were dried at 65°C for five days and ground using a stainless steel grinder.

Extraction, purification, and analysis of HCH isomers present in the water samples were carried out according to a method previously described (Rico et al., 1989). The rest of the samples were homogenized, extracted, cleaned, and analyzed following methods already described (Hernández et al., 1988; Hernández et al., 1989). Duplicate were carried out for all samples. Minimum detection limit was 0.01 ppb (μg/kg). Recoveries of HCH isomers ranged from 84-92%, but the residue data in the table were not adjusted on the basis of these recoveries. All the residues are expressed as μg/kg (ppb).

RESULTS AND DISCUSSION

All samples analyzed contained residues of α,β, γ,and δ-HCH (Table 1). In water and vegetal samples α-HCH showed the lowest mean levels, followed by γ,β,and δ-HCH. In soil samples γ-HCH showed the lowest mean levels, followed by α,β,and δ-HCH. In earthworms γ-HCH showed the lowest mean levels, followed by α, δ,and β-HCH. To sum up, mean β and δ-HCH concentrations were higher than mean $\alpha$and γ-HCH concentrations in all samples in this study.

The differences in residue levels among the four isomers examined seem related to differences in some of the physico-chemical properties of the individual HCH isomers. The degradation and distribution in the ecosystem are persistenly affected by these differences. The vapour pressure, especially low in the case of β-HCH, plays an important role. This leads, among other things, to β-HCH being considerably less volatile than α and γ-HCH. On account of the high vapour pressure of α and γ-HCH, these two isomers are readily given up to atmosphere from contaminated surfaces and then distributed widely, whereas β and δ-HCH tend to remain where they were applied. In addition, β-HCH has a very strong affinity for the lipoid structures of plants and animals, whereas γ-HCH exhibits this property to a considerably lower degree (C.I.E.L., 1982).

The data obtained in this study indicate an accumulation of ΣHCH in soils, vegetals and earthworms relative to water concentrations (Table 1). In effect, the average concentration factor, (CF) defined as the ratio of the ppb ΣHCH in soils or vegetals or earthworms to ppb ΣHCH in water, is 128.5, 60.8, and 4604.2, respectively. Likewise, there is bioaccumulation of ΣHCH in earthworms relative to soil (CF = 31.9); however, ΣHCH was not bioaccumulated by plants, only 58% of the ΣHCH concentration in the soil was observed in plants. The different behaviour of the ΣHCH may be due to, a greater bioaccumulation ratio in earthworms and/or to a greater elimination ratio in plants. So then residue levels of the studied organochlorines in the river water may affect aquatic life and lead to their accumulation into the food chain.

In this study, the distance from the lindane manufacturing factory appears to be one of the important factors in determining the levels of HCH isomers residues in samples. It was observed that residues of HCH isomers decreased with increasing distance from the lindane factory.

A Maximum Contaminant Level (MCL) of 4 μg/L for lindane in drinking water was promulgated by the USEPA (1975). The World Health Organization has recommended a drinking water criterion of 3 μg/L (WHO, 1982). The Australian National Health and Medical Research Council (1987) has recommended a MCL of 100 μg/L, whereas Mancy (1972) and Achari et al., (1975) estimated a MCL of 56 and 5 μg/L for lindane, respectively. Worth to be mentioned that residue levels both lindane and ΣHCH isomers, reported

Table 1. Levels of HCH isomers, expressed in ppb, in samples collected in the shore of Gallego river, and concentration factors determined.

Sample	α-HCH	β-HCH	γ-HCH	δ-HCH	ΣHCH	C.F.$_w$	C.F.$_s$
Water 1	0.002	0.003	0.003	0.101	0.109		
Water 2	0.006	0.072	0.064	0.402	0.544		
Water 3	0.016	0.044	0.008	0.392	0.460		
Water 4	0.013	0.013	0.020	0.392	0.438		
Water 5	0.003	0.025	0.006	0.159	0.193		
Soil 1 *	0.93	2.60	0.90	2.61	7.04	64.6	
Soil 2	11.53	45.26	4.60	41.25	102.64	188.6	
Soil 3	4.27	26.77	3.08	28.27	62.39	135.6	
Soil 4	5.38	10.62	4.11	16.03	36.14	82.5	
Soil 5	8.81	7.42	3.44	13.42	33.09	171.4	
Poa 1 *	1.00	1.07	1.63	4.16	7.86	72.1	1.10
Poa 2	1.08	13.63	2.30	17.28	34.29	63.0	0.33
Poa 3	5.57	5.24	5.30	13.96	30.07	65.3	0.48
Poa 4	4.92	2.31	5.00	15.48	27.71	63.2	0.76
Poa 5	0.46	0.14	1.25	6.00	7.85	40.7	0.23
Lumbricus 1	21.32	33.25	11.17	111.81	177.55	1628.8	25.22
Lumbricus 2	838.22	4353.6	1.39	1616.5	6809.7	12517.8	66.37
Lumbricus 3	120.64	1652.3	1.72	1450.1	3224.7	7010.2	51.68
Lumbricus 4	8.82	46.51	3.49	323.30	382.12	872.4	10.57
Lumbricus 5	13.29	122.14	6.71	49.36	191.50	992.2	5.78

C.F.$_w$ = Concentration Factor = Ratio of the ppb HCH in soils or vegetals or earthworms to ppb HCH in water; C.F.$_w$ = Ratio of the ppb HCH in vegetals or earthworms to ppb HCH in soil. * = Dry weight.

in this study are still low compared with the permissible levels for waters.

Our results show that the lindane residue content of the water samples studied (range 3-64 ng/L) are higher than those reported by El-Dib and Badawy (1985) in the Nile river (range 0.1-18.9 ng/L) and by Achari et al. (1975) in Georgetown County (range 0-21.1 ng/L). Likewise, our water samples contain lower concentrations of lindane than those reported by Caceres et al. (1985) in Sao Paulo (Southern Brazil) (range 400-1400 ng/L) and by Ang et al.(1989) in New South Wales (Australia) (range 0-6000 ng/L).

The detrimental concentration of α, β, and δ-HCH in abiotic and biotic substratums is unknown; so then, the significance of our reported α, β, and δ-HCH data, shown in Table 1, is unknown at this time and interpretation of their biological significance

is not possible. C.I.E.L. (1982) stated: "These three substances (α, β, and γ-HCH) differ quite considerably in their resorption kinetics, distribution storage, metabolic conversion and elimination. The real problem is the presence of α and β-HCH, and not the presence of γ-HCH". More integration of field and laboratory research will help determine whether these isomers can pose a serious problem to the flora and fauna.

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