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Hexachlorocyclohexane Contamination due to Lindane Manufacture at a Site in Spain

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INTRODUCTION

The results reported here detail the levels of hexachlorocyclohexane (HCH) found in environmental samples in the vicinity of the INQUINOSA manufacturing site. The initial pages of the report review some recent literature and give an overview of the toxicological properties of HCH isomers and possible degradation products.

TOXICITY OF LINDANE

Lindane is the name given to the gamma stereoisomer of 1,2,3,4,5,6-hexachlorocyclohexane. This stereoisomer, one of eight well described for the compound, has insecticidal properties and is widely used in agrochemical and pharmaceutical products although its precise molecular mechanism of toxicity is poorly understood (Lopez-Aparicio 1988). EDAP (1986) lists some 75 basic formulations involving lindane in use in European countries which does not include lindane used in wood preserving applications. There is also significant use in developing countries. It has been estimated (see: Kaushik et al. 1987) that DDT and Lindane account for 85% of the pesticides currently used in India. It must be regarded, with other organochlorine compounds, as a ubiquitous global contaminant and has been found in analyses of mosses and lichens from the Antarctic (Bacchi et al. 1986) and from remote northern and arctic areas (Oehme et al. 1985; Oehme & Ottar 1984). The levels recorded are similar (though less variable) to those found in mango leaves in various regions of West Africa (Bacchi et al. 1988).

Lindane has List I (Blacklist) Status in the European Community under Directive 76/464/EEC. It has further been incorporated into the provisional UK "Red List" of substances (DoE 1988) requiring strict limitation on discharges. Hellowell (1988) indicates that no EIFAC criteria have been set for lindane. The US EPA criteria are set at 0.01ug/l for the protection of freshwater aquatic life, 0.004ug/l to protect marine aquatic life and 4.0ug/l in domestic supplies (Train 1979). The World Health Organisation guideline value is 3.0 ug/l for a consumption of two litres per day (WHO 1984). There are a number of household uses of lindane which include seed treatment, insect control in soil, on vegetables, ornamentals and fruit trees. There are also a number of pharmaceutical applications. Lewis et al. (1988) monitored lindane as part of a suite of pesticides in domestic use and found it in samples of indoor air at six of the eight households tested. Kaushik et al. (1987) found lindane in 97.4% of air samples taken in the Delhi area of India with a mean

value of 94 ng per cubic metre of ambient air. Total HCH levels recorded at a mean of 438 ng per cubic metre reflect a level of alpha-HCH of 344 ng/cubic metre on average. The significance of this is discussed further in the section on degradation below.

WHO (1984) details some of the basic health effects of lindane exposure. The oral LD50 for rats is between 125-230 mg/kg body weight. Chronic studies with rats exposed to dietary lindane demonstrated liver cell hypertrophy. At high doses changes were noted in the kidney. Inhalation of lindane by experimental rats resulted, after 180 days, in liver enlargement but no other toxic symptoms. Two years dietary exposure of rats resulted in decreased body weight as compared to controls and disturbance of vitamin C metabolism. Goel et al. (1988) demonstrated the induction of liver enzyme activity as a result of lindane exposure in rats. Payne et al. (1987) review the use of mixed function oxidases in biological monitoring. Cossarini-Dunier et al. (1987) were unable to demonstrate any effect of chronic intoxication upon humoral immune response in carp, although as part of the same study Demael et al. (1987) found important changes in a number of liver and muscle enzymes.

Human toxicity has been reported after prolonged, improper use of a 1% lindane preparation in the treatment of scabies (WHO 1984). Symptoms included irritation of the central nervous system with nausea, vomiting and weak respiration with cyanosis (Blue lips and skin). Production workers exposed to technical HCH exhibited symptoms including headache, vertigo, skin irritation and eye irritation. There was evidence of disturbance of carbohydrate and lipid metabolism and dysfunction of the endocrine system centred around the hypothalamus, pituitary and adrenal glands. Occupational exposure over 11-23 years was associated with toxic hepatitis. The WHO maximum acceptable daily intake for lindane is set at 10ug/kg body weight.

Lindane has also been shown to exert reproductive effects in multi-generation studies of rats. These included an increase in the duration of pregnancy, an increase in stillbirths, delay of sexual maturation in first and second generation animals and paraplegic effects in juveniles. In both rats and rabbits, lindane has been shown to cause post-implantation death of embryos.

The evidence for mutagenic activity is equivocal. Carcinogenic effects have been reported in the livers of mice fed lindane (WHO 1984). Arcos et al (1988) detail the effects of HCH isomers in binary combination with other carcinogenic agents. Haseman et al. (1987) however, record lindane as having no carcinogenic activity. Lopez-Aparicio

(1988) describe changes in phospholipid metabolism in the rat which could be expected to result in cellular dysfunction in target organs. The use of lindane in sheep dip results in long lived residues in the fat of treated sheep (Dagorn et al. 1988).

Lindane is fairly soluble in water as compared to other organochlorine pesticides and this aids in rapid uptake/depuration patterns. Lindane is taken up after oral or dermal exposure and has an estimated weighted bioconcentration factor of 780 as assessed by study of bluegills sunfish. Intraperitoneal doses were consistently found to localise in skin, muscle, fatty tissue and brain. Transplacental movement has also been reported while significant levels have been recorded in human breastmilk in a variety of areas (See eg Karakaya et al. 1987)

In aquatic systems sensitivity of invertebrates tested is only slightly greater than the sensitivity of fish (Train 1979). 96HLC50 values are between 1 and 5 ug/l. Bioaccumulation through an aquatic laboratory food chain was carried out by Hansen (1980) who demonstrated that about 50% of the amount given to the fish was accumulated on a per diem basis and that Daphnia suffered reproductive failure as a result of lindane accumulated from water with a concentration of 7.4ug/l by green algae used as food.

While there is a considerable amount of toxicological data concerning gamma-HCH in the scientific literature, studies of the differential toxicity of the stereoisomers appear to be few. Srinivasan et al. (1988) note minor differences in the toxicological effects of gamma- with respect to beta-HCH. These workers note, however, the remarks of Ulmann (1973) to the effect that the beta-HCH isomer is a very persistent constituent of HCH technical mixes and known to be highly toxic to mammals.

In marine ecosystems appreciable levels of gamma-HCH are found in various environmental compartments. MAFF (1987) describe elevated levels of gamma-HCH at coastal sites on the Irish Sea. The highest concentration in sea water of 26.4 ng/l was recorded at Connah's Quay. This is some six times the US EPA criterion for the protection of marine life. These levels are likely to be transient since they arise from periodic events.

Certainly, the ambient air levels of HCH isomers recorded by Oehme & Ottar (1984) appear to reflect lindane use in spring. Knickmeyer & Steinhart (1988) detail HCH levels in Pagurus spp. (hermit crabs) throughout the North Sea. Gamma-HCH appears to be highest in the early summer, falling somewhat in the winter. In both cases the levels clearly reflect the influence of the major rivers on the continental European coastline. Patterns for alpha-HCH are

less well defined and appear to reflect a degree of conversion from the gamma-isomer. At most stations early alpha:gamma ratios increased in the winter samples. Continuation of this project will be useful in order to establish any trends in levels. The greater magnitude of this ratio in the north west North Sea may indicate that the central area is acting as a sink for these materials.

DEGRADATION

In mammalian systems (WHO 1984), lindane is metabolized to an intermediate hexachlorocyclohexane which upon further degradation yields 2,3,4,5,6-pentachloro-2-cyclohexene-1-ol, two tetrachlorophenols and three trichlorophenols. These are less toxic than the parent compounds. Lindane is slowly degraded by soil micro-organisms and can be isomerised to the alpha and/or delta isomers by micro-organisms and plants. There is evidence that lindane may act synergistically with other pesticides to disrupt normal microbiological processes in the soil (Ray 1984).

There is evidence that gamma-HCH is transformed photochemically to alpha-HCH with a half life of less than a year (Oehme & Ottar 1984). The half life of alpha-HCH is not given. A proportion of alpha and beta HCH isomers were present in technical mixes prepared by the chlorination under light of benzene: about 12% of beta, 12% of gamma and up to 70% of alpha HCH. More recently, gamma-HCH has been separated from the reaction mixture to the extent that pharmaceutical preparations are 99% pure. Agrochemical mixes are significantly richer in gamma-HCH than previously.

The photochemical instability of the gamma isomer and the conversion to the alpha form may largely explain the high alpha : gamma ratios in air reported by Oehme & Ottar (1984) and the predominance of alpha-HCH in rain water samples from Portland, Oregon reported by Ligocki et al. (1985). Similarly, Kaushik et al. (1987) reported a similar preponderance of the alpha isomer in samples from Delhi, India. In aquatic systems the fate of HCH is unclear. Murty (1986) cites evidence that HCH levels in riverwater samples do not decline. In a further study cited by this author, HCH levels in a lake were closely related to the physical phenomena like stratification. There are few data on the fate of HCH in sediments or in sewage treatment works. Schroder (1987) evaluated the fate of gamma-HCH in sewage treatment together with a number of other volatiles and concluded that the bulk would be discharged with the aqueous effluent with a small proportion adsorbing to the sludge. Degradation was not considered an important pathway. A Norwegian study (Carlberg & Boler 1985) showed appreciable levels of HCH

isomers in seals and fish. In general levels were alpha>gamma>beta with alpha levels an order of magnitude greater than gamma.

The predominance of alpha-HCH extends to human dietary intake in food across all age groups considered in the FDA total diet study between 1982 and 1984 (Gunderson 1988). The tabulated data include an FAO/WHO Acceptable Daily Intake (ADI) for gamma-BHC of 10 ug/kg body weight. No ADI values are given for other HCH isomers implying that they are not established. The ADI is the daily intake which is purported to carry with it no appreciable risk over a lifetime consumption. Significantly, for all groups considered, the 2 year age group was subjected to the highest intake of all HCH isomers.

There is the possibility that HCH isomers may be degraded to chlorobenzenes although we are not aware of any evidence for this in the literature. Certainly, it seems likely that chlorobenzenes will be present in the waste materials from lindane production as a result of incomplete conversion of the raw material. The chlorobenzenes are an environmentally difficult group. Hexachlorobenzene is accumulated by organisms (Schuytema et al. 1988) and has been implicated in the perturbation of calcium homeostasis in rats and the demineralization of human bone. It is also thought to be carcinogenic and has been implicated in other metabolic disorders (See: Andrews et al. 1988).

Others of the chlorobenzene group have been shown to undergo reductive dechlorination in anaerobic conditions by bacteria thus allowing the more recalcitrant species such as 1,3,5-Trichlorobenzene to be fully degraded in subsequently applied oxic conditions (Bosma et al. 1988; Haigler et al. 1988). The degradation products of lindane and other HCH isomers could therefore constitute a significant contamination hazard. Barber (1988) notes the threat posed to groundwater by long term persistence of dichlorobenzene.

Given the environmental importance of the HCH isomers, it was decided to investigate the degree of contamination resulting from the INQUINOSA operation.

METHOD AND MATERIALS

Soil and effluent samples were taken in acid washed containers and despatched to the analysing laboratory in the UK. The sediment samples were extracted with acetone which was then filtered, diluted with water and saturated sodium sulphate solution. Extraction with hexane was followed by clean up on a silver nitrate/ alumina column. Analysis was by capillary gas chromatography using an OV 1701 column and an electron capture detector.

Identification was by retention index confirmed by mass spectrometry.

RESULTS

Results of analyses are presented in TABLE I below together with details of the sampling site. Figures are given in mg/kg of sample as received in the case of the sediments and in mg/l in the case of the aqueous samples.

SAMPLE	MATRIX	alpha-HCH	beta-HCH	gamma-HCH
1	sediment	77	66	0.30
2	sediment	0.18	0.27	<0.02
3	aqueous	0.67	2.5	0.74
4	aqueous	0.54	34	<0.02
5	aqueous	0.07	4.6	<0.02

Sample No: 1 was of damp mud from a ditch used to carry drain off from the old Inquinosa dump site at Sabinanigo.

Sample No: 2 was of fine silt from a small stream (Barranco de Bailin) at its point of entry into the Rio Gallego south of Sabinanigo.

Sample No: 3 was an aqueous sample taken from a settling tank for leachate from the dumpsite south of Sabinanigo.

Sample No: 4 was an aqueous sample from a drain taking leachate from the dumpsite at Sabinanigo.

Sample No: 5 was an effluent sample from the INQUINOSA factory direct into the Rio Gallego.

COMMENTS

From these results it is apparent that the manufacture of gamma-HCH and the disposal of unwanted by products has resulted in widespread contamination of the area around the INQUINOSA operation. In the case of sediment in Sample No:1, this should be regarded as hazardous toxic waste and appropriate decontamination operations carried out to minimise the risk of damage to local ecosystems. It is possible that the soils in the area are also contaminated with chlorobenzenes and dioxins and this should be urgently investigated with a view to urgent remedial action if necessary. Sample No: 2, also a sediment shows a lesser degree of contamination with alpha and beta isomers of HCH. Nonetheless these are a cause for grave concern since they are evidence of widespread

contamination of the river system receiving effluent from the manufacturing facility. The levels recorded here are such as to give cause for serious concern about ecosystem health and the consumption of fish from these waters. Analysis of fish tissues should be carried out as a matter of urgency and all fishing prohibited in these waters. Investigation should also be made of the dioxin levels with a view to establishing baseline values in the area prior to the operation of any waste recycling process.

Sample No:3 from the dumpsite effluent settling tank contained effluent grossly unsuitable for discharge to the wider environment. Without suitable criteria set by any regulatory agency it is assumed that the alpha and beta isomers are subject to the same maximum acceptable toxicant concentrations as the gamma isomer. This may be an optimistic assumption. Even so, to dilute 1 litre of this effluent to conform to US EPA criteria of 0.01ug/l would take 391,000 litres i.e. 391 cubic metres of water. The same observations apply to sample number 4 except that in this case 1 litre of effluent would require dilution in 3,454,000 litres or 3,500 cubic metres of water to meet US EPA criteria. Finally, 1 litre of the effluent from the factory itself would require dilution in 467,000 litres of water to meet US EPA criteria. It is likely therefore that during periods of low flow substantial lengths of the river exceed the US EPA criteria. Contamination of the environment on this scale must raise concerns for the health of local inhabitants who are subject to aeolian transport of residue contaminated dusts and who consume foodstuffs irrigated with water from the river on a regular basis. These exposure commitments require urgent evaluation as does that from groundwater abstracted from the area. Drinking water abstraction from the river also requires attention since the possibility exists that some fractions of the population could be approaching the WHO guideline levels set for the gamma isomer in terms of their consumption of other stereoisomers.

The rehabilitation of pesticide contaminated industrial sites is a complex and difficult operation. (See eg: Finnecy & Johnstone 1988; Schmitt & Bjornsen 1988; Exner 1988). Although methods of microbial cleanup have been explored for HCH contaminated sites (Huntjens et al. 1988) these may be isomer specific and are likely to be substrate limited implying that complete decontamination cannot be achieved.

Presently, wastes generated at the INQUINOSA site are landfilled in large plastic containment sacks pending the development of a final disposal method. Quite apart from the mechanical integrity of the sacks causing problems, the possibility that these wastes contain chlorobenzene and benzene residues must be viewed with alarm since they

may well facilitate migration of the wastes through the containment. Such components will almost certainly facilitate the migration of HCH residues through the soil as has been found by Jurgens et al. (1988). This is not a satisfactory storage method given the volume and nature of the wastes being stored. 85% of each production batch in the synthesis of lindane is waste. Additionally landfill operations are subject to volatile emissions even when capped (Karimi et al. 1988). The degree to which materials escape from the present site in this way is unknown but is likely to be significant.

The method of disposal currently proposed for the INQUINOSA site is that employed previously on a German site near Hamburg described by Jurgens et al. (1988). The production of pesticides began in 1951 with the manufacture of hexachlorocyclohexane (HCH) and the separation from this of lindane. The unwanted isomers were stored on the site until 1953 when an "exceptional" recycling process was implemented. Thermal decomposition of the HCH waste rendered a 75% yield of 1,2,4-trichlorobenzene. This allowed the production of 2,5-dichlorophenol and 2,5-dichloro-4-bromophenol. Recycled trichlorobenzene was converted to 1,2,4,5-tetrachlorobenzene which could then be synthesised to 2,4,5-trichlorophenoxyacetic acid (2,4,5-T).

2,4,5-T production at the site stopped in 1983 since prohibitions were made on the transport of the waste which was contaminated with 2,3,7,8-Tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD). Further, in 1984 it was discovered that polychlorinated dioxins were present in the residue from the decomposition of the HCH isomers. Emission limits were imposed by the Hamburg Department of Environmental Protection as follows for 2,3,7,8-TCDD:

2,3,7,8-TCDD contents in residues:	1ug/kg
in air:	1pg/cubic metre
in effluent:	1 ng/l

The plant was unable to meet these limits and was forced to close and is presently undergoing decontamination for industrial development. As of late 1987, no acceptable scheme had been agreed by the authorities with the developers. Soil contamination may be gauged from the published contamination figures derived as part of a graded analytical programme:

Chlorobenzenes, hexachlorocyclohexanes and chlorophenols:

Samples analysed: 2652

in 30 samples (1.1%)	>10,000 mg/kg
in 64 samples (2.4%)	> 1,000 mg/kg <10,000 mg/kg

in 104 samples (3.9%) > 100 mg/kg < 1,000 mg/kg
in 124 samples (4.7%) > 10 mg/kg < 100 mg/kg

The proportion of the remaining 88% contaminated to some degree is not given by Jurgens et al. (1988) but may be substantial. Concentrations of 2,3,7,8-TCDD were found at greater than 100 ppb (ug/kg) in 12 samples and at 1ppb to 100ppb in 132 samples of 254 taken. Levels were generally highest at the surface although the presence of the benzene component is thought to have facilitated migration through the soil to give a level of 98ppb at 5.4 metres depth thus threatening the water table. The highest level recorded was 9mg/kg. Attempts to deal with the HCH residues at the Spanish site using the same methods will result in similar problems of contamination. It seems clear that no environmentally acceptable method currently exists for the disposal of these wastes.

In conclusion, therefore, the manufacture of lindane at the INQUINOSA facility has resulted in widespread contamination of the wider environment and an uncertain exposure commitment of the population served in various ways from the river. The possibility of exposure to chemicals other than HCH isomers such as dioxin compounds or chlorobenzenes also exists. In order to protect both local and wider ecosystems and resources the problem of site rehabilitation should be addressed with urgency. In view of problems experienced with HCH waste recycling in Germany, thermal decomposition treatments should not be used. Further production of lindane and unwanted by-products should be prohibited and efforts concentrated on a clean up.

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