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SEVERITY OF PERSISTENCE AND TOXICITY OF HEXACHLOROCYCLOHEXANE (HCH) TO THE ENVIRONMENT- A CURRENT APPROACH

Sanhita Padhi¹ & Bandita Pati²

Department of Botany and Biotechnology, Ravenshaw University, Cuttack

Abstract

Though the use of Organochlorine pesticides significantly develops the economic status of many developing countries by increasing the crop yield and preventing the vector-borne diseases, but it becomes a serious threat to our environment increasing serious health problems. The persistence of Organochlorine pesticide, Hexachlorocyclohexane (HCH) and its isomers are toxic and cause hepatotoxic, immune-toxic, reproductive, and developmental effects. These become persistent in the soil and environment for a longer period due to their recalcitrant properties and their slow bioconversion into inorganic compounds. Hence, these are biomagnified into the tropic level and cause acute and chronic health problems. Though the use of HCH is banned in developed countries still several developing countries are using lindane (HCH) in domestic and agriculture purposes including India. Different abiotic factors like moisture, pH and Eh, temperature, organic matters affect the persistence of HCH.

Key words: Organochlorine pesticide, vector-borne, Hexachlorocyclohexane (HCH), persistence, recalcitrant, bioconversion, hepatotoxic, immune-toxic, bio-magnification.



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Introduction

Pesticides are chemicals, used mostly in agriculture for protection and higher yield. Among them organochlorine pesticides (OCPs) are most widely used. OCPs are very stable compounds and their half-lives can range from a few months to several years; in some cases decades (Cremlyn, 1991). Organochlorine pesticides (OCPs) have been in wide usage across the world to control agricultural pests and vector- borne diseases (Abhilash and Singh, 2009; Zhang et al., 2011).

OCPs have attracted widespread concern because of their environmental persistence and toxicity (Wang et al., 2016). These are chlorinated hydrocarbons used extensively from the 1940s through the 1960s in agriculture and in mosquito control. These compounds are

typically very persistent in the environment, and are known for accumulating in sediments, plants and animals. Organochlorines have a wide range of both acute and chronic health effects, including cancer, neurological damage, and birth defects. Many organochlorines are also suspected endocrine disruptors. Organochlorines are some of the chemicals found most often in the hundreds of tests of human body tissue: blood, adipose tissue, breast-milk that has been conducted around the world. Because of their chemical structure, organochlorines break down slowly, build up in fatty tissues, and remain in our bodies for a long time.

Many organochlorine compounds have been isolated from natural sources ranging from bacteria to humans. Chlorinated organic compounds are found in nearly every class of biomolecules including alkaloids, terpenes, amino acids, flavonoids, steroids and fatty acids (Grible 1998; Engvild 1986).

Hexachlorocyclohexane (HCH) is an example of OCPs, exists in different isomers like α , β , γ , δ and ϵ , which have been identified in hazardous waste sites (HazDat 2005). Among the HCH isomers, only γ -HCH has insecticidal property and pure γ -HCH is known as Lindane. Lindane and other HCH isomers are toxic and cause hepatotoxic, immune-toxic, reproductive, and developmental effects (USEPA, 2010). Although the productions and application of DDTs and HCHs are banned in developed countries, still several developing countries including India involved in productions and consumptions of DDTs and HCHs for domestic and agricultural purposes (Yadav et al., 2015). Though it is banned in many developing countries, but still these are persistent in soil and aquatic environment due to their less solubility in water.

Persistence of organochlorine pesticides

Persistent organic pollutants (POPs) including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are of significant concern due to their potential toxicity and prevalence in a wide range of environmental media, even at remote geographical locations (Iwata et al., 1994; Kellenborn et al., 1998).

The insecticides are extensively used in agriculture fields for higher yield of crops, which results in the accumulation of these compounds into the soil and environment. In the soil, these compounds are slowly degraded and accumulated. Some of the dead end products of these compounds are getting persistent in the soil and environment. They cannot be further degraded normally.

Persistence of HCH

Persistence of HCH in soils

Studies have shown that HCH is more persistent in temperate soil than in tropical soils. Srivastava and Yadav (1977) have shown that loss of HCH from a tropical soil under maize cover was more rapid than the reported loss from temperate soils. MacRae et al. (1984) reported that soil suspension from tropical soils effected more rapid degradation of HCH isomers than soil suspensions from temperate soils. HCH, applied at 5-30 kg.ha⁻¹ to radish and sugarbeet fields, decreased by 70-90% of the original level in 4 months (Kushwaha et al., 1984). In another study, movement and persistence of lindane was measured in calcareous and sandy loam soil, with and without irrigation. When lindane was applied at 0 to 7.5 and 7.5 to 15cm depth, the insecticide moved downwards with irrigation and measurable amounts were found at 60cm (Stewart and Chisholm, 1971). Meijer et al. (2000) analysed the archived background soils (Broadbalk, 1944-1986) and sludge-amended soils (Luddington, 1968-1990), collected from long-term agricultural experiments in the U.K., for a range of organochlorine pesticides to establish their persistence trends over time. The concentration ranged from 0.1 to 10 ng.g⁻¹ of soil (dry wt.) with □-HCH; dieldrin and p,p¹-DDE consistently having the highest concentrations. They showed that enantio selective microbial degradation was not consistent over time and that the addition of sludge to the soil does not significantly alter the enantiomeric preference of the microbial community. Manz et al. (2001), in studies on persistent organic pollutants from the top soils of eleven agricultural fields of Central Gemany, clearly indicated the deposition of anthropogenic pollutant inputs not only in the past but also today. The reference values exceeded mainly for DDE and lindane. Wang et al. (2016) while studying the levels and patterns of organochlorine pesticides in agricultural soils of a cotton field and other crop fields in eastern central China, the concentrations of HCH was found to be 2.9–56.4 ng g⁻¹, whose compositional study suggested that was mainly due to the use of Lindane.

Persistence of HCH in flooded soil

Raghu and MacRae (1966) provided the first convincing report of anaerobic instability of a pesticide in a flooded soil when they showed rapid degradation (within 90 days) of □-HCH in a Philippine soil upon flooding. A flooded soil planted to rice can be aerobic due to transport of oxygen from foliage to the root region, while an unplanted flooded soil is predominantly anaerobic. Flooding the soil, as in rice cultivation, decreases the persistence of lindane (Raghu and MacRae, 1966). In a subsequent report, MacRae et al., (1967) showed that rapid degradation of other HCH-isomers in flooded soils. They reported

Persistence of HCH in the environment

Extensive studies in temperate countries with aerobic soils have shown that HCH persists in the soil environment for several years. 41% of applied HCH was recovered 11 years after application to a turf soil (applied at 0.25 - 10 lb.acre⁻¹) (Lichtenstein and Polivka, 1959). Stewart and Chisholm (1971) recovered 36, 36, 16 and 12% of α -, β -, \Box - and \Box isomers of HCH from a sandy loam soil after 15 years of HCH application. Lindane was more persistent in a muck soil than in a sandy loam soil. After 3-5 years of application of

lindane to soil, more residues were recovered from the soil amended with lindane at 100 lb.
acre ⁻¹ than 10 lb. acre ⁻¹ . About 7.7% of HCH remained in a sandy loam soil after 15 years
(Chishlom and Macphee, 1972). About 97.5% of HCH was lost from the soil within 100 days
after HCH application in furrows applied at 10kg ha ⁻¹ (Agnihotri et al., 1974). Nash and
Woolson (1967) reported 10% recovery of technical HCH from a sandy loam soil after 14
years; in contrast no lindane was detected 12 years after its application to the surface of Nova
Scotian soils (Stewart and Fox, 1971). Behboodi (1978) determined the residues after 4, 6, 13
and 21 months of application of HCH for upper 15 and 30 cm layers. After 2 years, lindane
decreased from 5ppm to 0.001ppm. Suzuki et al. (1975) studied persistence of HCH isomers
in four types of sandy loam soil for 2.5 years; the most persistent HCH isomer was \Box \Box HCH
followed by \square \square \square \square \square \square and \square \square HCH. About 50% of initial 10ppm HCH application
disappeared within a month and 88% disappeared within 5 months (Pal and Kushwaha,
1977). Similarly \square and \square -isomers of HCH were moderately persistent and degraded to
some extent in 33 days while \square THCH was most stable (Tomizawa and Kazano, 1979). In a
study conducted in Tamil Nadu, more than 95% of the added HCH disappeared in 170-180
days with a half life of 39 days (Jayaraj and Raghupathy, 1986) whereas the reported half life
of HCH was 6 months to 3 years in temperate countries. α -, β -, \square -, and δ -HCH have been
identified in at least 146, 159, 189, and 126, respectively of the 1,662 hazardous waste sites
that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat
2005). □-HCH and other isomers of HCH do not occur naturally in the environment. Most
current releases of □-HCH in the United States are related to its formulation and its use as an
insecticide/acaricide. Historically, the largest source of □-HCH releases to the air resulted
from agricultural application of the pesticide -HCH. Other air releases occurred during the
manufacture of the pesticide. □-HCH can be released to surface water via surface runoff (as
the dissolved chemical or adsorbed to particulates) or via wet deposition of rain and snow
(Tanabe et al. 1982; Wheatley and Hardman 1965). □-HCH can be released to groundwater
via soil leachate. Although available adsorption data indicate that \Box \Box HCH has a low
mobility in soils, the results of monitoring studies suggest that \Box \Box HCH does migrate to
groundwater (Page 1981; Sandhu et al., 1978).

Persistence of different isomers of HCH

Evidence suggested that isomers of HCH are more persistent when applied to the soil in comercial formulations than when applied individually. Stickley (1972) reported that \Box \Box \Box isomer in a crude HCH formulation was more persistent in the soil than lindane. Pure

isomers of \square \square \square \square \square and \square \square HCH did not, however, affect the persistence of pure
$\hfill\Box$ HCH. Tomizwa and Kazano (1979) reported that $\hfill\Box$ -HCH was the most persistent and
bio-concentrated isomer in a rice straw paddy ecosystem. In a long-term study on the
persistence of HCH isomers in field soils, Suzuki et al. (1975) found □-isomers to be most
persistent isomers of HCH. Loss of lindane from soils was attributed not solely to
volatilization, but to degradation as evidenced by the formation of its metabolites □-PCCH
(Claith and Spencer, 1971). Persistence is also dependent on the climate, properties of the
soils, microbial environment, diffusion and method of application, i.e. foliar or soil
incorporation. It is not clear whether rapid loss of pesticides under field conditions in the
tropics or subtropics was due to volatilization of the parent molecules and/or accelerated
chemical or biological degradation under hot and humid conditions of the tropics.
$\hfill\Box$ Pentachlorocyclohexene ($\hfill\Box$ -PCCH) formed by dehydrochlorination, was detected as the
transitory degradation product of $\ \square$ -HCH in moist soils (Yule et al., 1967; Guenzi and
Beard, 1970). Aerobic degradation of soil-applied □-HCH yielded □-PCCH (Yule et al.,
1967) while anaerobic degradataion of \Box -HCH in flooded soil yielded
tetrachlorocyclohexene (□-TCCH) (Sethunathan et al., 1969; Tsukano and Kobayashi, 1972;
Mathur and Saha, 1975; Matsumura et al., 1976; Buerskens et al., 1991; Adhya et al., 1994).
Bachmann et al. (1988a,b) reported the accumulation of □-Pentachlorocyclohexene (□-
PCCH) in trace amounts in an \Box -HCH contaminated soil slurry, which was further degraded
to 1,2,3,5 tetra chlorobenzene, 1,2,3,4 tetra chlorobenzene, 1,2,4, tri chlorobenzene, 1,2 di
chlorobenzene and 1,4 dichlorobenzene. In a recent report by Law et al. (2001), it was
indicated that the concentration of $\Box ext{-HCH}$ was highest in cold, large and oligotrophic lakes
which was attributed to greater inputs from atmospheric deposition and slower loss rates
relative to warmer, temperate lakes. Fisk and Stem et al. (2001) reported that \Box -and \Box -HCH
and lower chlorinated PCB congeners were the most common POPs in a small, herbivorous,
arctic marine zooplankton, Calanus hyperboreus but these POPs do not bio magnify in this
species or likely in other small, herbivorous zooplankton. In situ biodegradation of HCH
isomers is very low (<500mg/kg) in soils (Phillips et al. 2005).

Factors affecting persistence of HCH isomers

Moisture

Moisture regime influences the stability of HCH isomers in soil environment. Jerzy et al. (1971) reported that when soil samples varying in physico-chemical properties were treated with \Box -HCH and maintained at 40% and 95% moisture content for 72 days at 30°C, more rapid degradation occurred at 95% moisture content than at 40% moisture content. A

significant decrease in isomer concentration was observed with higher soil moisture content (Chessel et al., 1988). Hu et al. (2010) reported that the mean concentration of HCH (0.67) in the water shed area was lower than the residual levels of Tibetan plateau. A report by Phillips et al. (2006) showed that Lindane was degraded in soil with both low and high moisture content.

pH and Eh

Chang et al. (1981) reported that increase in pH values in an oxidation pond increased the degradation of HCH. Degradation of \Box \Box \Box \Box \Box \Box \Box \Box HCH was greater in alkaline soil than in acidic soil. Zhang et al. (1982) showed that the rate of degradation of these isomers was relatively low at pH < 8.3 but these compounds were rapidly degraded at pH 10.2. The bioassay test using *Drosophilla melanogaster* showed that the persistence of HCH (at 5 and 10 kg.ha⁻¹ in loam soil with 2.8, 6.0, 9.0 and 12.0 dS.m⁻¹ electrical conductivity) was negatively correlated with electrical conductivity of soil (Varma and Singh, 1974). Sethunathan et al. (1976) reported that after several weeks of flooding when the pH is very low and Eh is positive, the decomposition of \Box and \Box isomers are very low. A report by Phillips et al. (2006) has resulted that Lindane was degraded in soil of near neutral pH. An experiment by Reddy et al. (2011) on clay soil contaminated with lindane and other OCPs reported that, application of voltage gradient across the soil resulted in 28-80% degradation of Lindane by direct electrochemical reductive process at the electrodes.

Temperature

Persistence of HCH and the ratio of \Box - to \Box isomers in residues were higher in an upland soil than in a submerged paddy soil and increased with increasing water content and temperature (Zhang et al., 1988). Lichtenstein and Schulz (1959) reported that temperature and rate of application influence the persistence of HCH in the soil. A report by Phillips et al. (2006) has resulted that Lindane was degraded in soil of moderate temperature.

Organic matter-native or added

Lindane was found to be more persistent in muck soil than in a sandy loam soil. Lichtenstein and Schulz (1959) reported the transformation of Lindane to a non-toxic compound which could be detected by colorimetry but not by bioassay within two weeks after the application to a sandy loam soil. Ponnamperuma (1972) reported that addition of organic matter to a flooded soil increases microbial activity during incubation and lowers the redox potential. Likewise, redox potential dropped more rapidly following flooding in organic matter-rich soils than in soils low in organic matter content. Yoshida and Castro (1970) studied □-HCH degradation in four rice soils (one sandy and three clay soils) of

Phillippines under flooded conditions. Degradation of □-HCH was higher in a sandy loam than in clay soil, at 30°C than at 20°C and was directly related to the native soil organic matter level. Likewise, addition of organic sources such as rice straw (Yoshida and Castro, 1970; Siddaramappa and Sethunathan, 1975; Drego, 1987), green manure of *Gliricidia sepium* and *G. maculata* (Ferreira and Raghu, 1981) or *Astragalus sinious* (Zhang et.al., 1982) further enhanced the degradation of □-HCH in soils. Drego et al. (1990) demonstrated the mineralization of □-HCH in green manure amended soil to ¹⁴CO₂ and other organic volatiles such as benzene.

More rapid degradation of □-HCH in soils rich in organic matter content or in soils with added organic matter under flooded condition has been attributed to the drop in redox potential which favours the anaerobic degradation of \(\subseteq\)-HCH (Siddaramappa and Sethunathan, 1975). Addition of organic substrates such as glutamate and peptone effected rapid degradation of \square -HCH under anaerobic conditions, but not under aerobic conditions (Doelman et al., 1985). Wahid and Sethunathan (1979) reported that soil organic matter is the most important single factor governing sorption-desorption of \Box \Box \Box \Box \Box \Box and \Box -isomers of HCH in aerobic soil and anaerobic (flooded) soils (Wahid and Sethunathan, 1980). Higher the organic matter content greater is the sorption of these isomers in soil. Sorption-desorption of pesticide in a soil would determine its availability for degradation by microorganisms. Wang et al., (2006) reported the loam soils having HCH and other pesticides with different fertilisers like, corn straw, green manure and resulted the decrease in pesticide persistence as pH of the soil was decreased. The persistence of HCH isomers were in the order of □-HCH $> \square$ -HCH is approximately equal to \square -HCH $> \square$ -HCH. Degradation of \square -HCH (Lindane) in surface soils of mollisol amended with farmyard manure and cow-dung slurry was found to be 83% and 91% respectively in comparison to in the un amended soil 79%. Gypsum also degrades Lindane in surface soil, but is less in comparison to organic amendments (Bhatia et al., 2011).

Conclusion

Because of the multiple types and uses of organochlorine pesticides, there are many ways people can be exposed to these. Wind and rain may move pesticides away from where they were used, causing contamination of surface waters, groundwater and/or soil. Using pesticides may increase our exposure to these chemicals, and living or working close to where pesticides are used is also a risk factor. Organochlorine pesticides are not often used in personal products because of their toxicity, but some products, such as lice shampoos, may be a source of exposure. Exposure may also occur through consumption of contaminated foods.

Organochlorine pesticides are carried long distances via atmospheric and oceanic currents from where they are manufactured and used, and build up in the fatty tissues of animals. Many studies have linked organochlorine pesticide exposure with consumption of contaminated animal products, mostly meat, dairy, fish, and marine mammals.

The health effects of organochlorine pesticide exposure depend on the specific pesticide, the level of exposure, the timing of exposure and the individual. Different pesticides result in a range of health symptoms.

The OCPs are regulated by the Environmental Protection Agency (EPA) under the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA). The UN and World Health Organization (WHO) may also influence use of pesticides through treaties and declarations. The Stockholm Convention on Persistent Organic Pollutants, a UN treaty, has established global bans on several organochlorine pesticides including DDT, hexachlorobenzene, pentachlorobenzene, chlordane, dieldrin, endrin, heptachlor, mirex, hexachlorocyclohexane (alpha-HCH, beta-HCH, and gamma-HCH (lindane)), chlordecone. The European Union (EU) recently proposed that endosulfan be included under provisions of the Stockholm Convention as well. Although the U.S. banned the use of lindane for agricultural uses, the Food and Drug Administration still allows use of lindane in pharmaceutical products for the treatment of head lice and scabies.

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