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ADSORPTION AND DOWNWARD MOVEMENT OF LINDANE IN ORGANIC MATTER RICH SANDY LOAM SOIL OF UTTARANCHAL, INDIA

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ABSTRACT

The effect of soil properties on the adsorption, movement and distribution of Lindane in soils of Uttaranchal, India was studied. The results indicated that soils with different physico-chemical properties have different effects on the adsorption, movement and distribution of pesticide, especially at higher concentration levels. Two composite soil samples were collected that having diversified phycio-chemical properties were analyzed. The Adsorption of Lindane to soils was best found to fit a Freundlich equation and Freundlich's adsorption constant 'K' and '1/n' values were in the range of 0.941 and 0.995 respectively. The highest Kd value (21.24) was received to the Almora soil followed by Udham Singh Nagar soil (20.24) with a high clay content and organic matter content exhibit the high adsorption capacity. In the present study, Lindane downward movement was performed in the packed soil column; the distribution of Lindane was obtained throughout the column and even showed presence in the lower most soil core. The recovered amount of Lindane was detected on Almora (75.38%) followed by Udham Singh Nagar (74.76%) in 0-10cm depth. It was noted that the relatively high organic matter content and clay content of soil restricted the leaching of the soil.

INTRODUCTION

The fate of pesticides in soils is a great environmental concern due to their toxicity, persistence and bioaccumulation. The retention of pesticides in the environment involves complex mechanisms that are influenced by many processes, including volatilization, leaching, adsorption, as well as chemical and biological degradation. Pesticide characteristics (water solubility, tendency to adsorb to the soil and pesticide persistence) and soil characteristics (clay and organic matter) are important in determining the fate of the chemicals in environment (Anonymous, 2009; Pignatello, 1989; Becker *et al*., 2011) and having most important processes affecting the mobility, persistence, bioactivity, toxicity and efficacy of pesticides in the soil environment. Retention is influenced by pesticide adsorption by soil components. The adsorption rate and adsorption capacity of Lindane exhibited highest in organic matter rich soil that retarded the downward movement whereas, least adsorption in sandy soil (Rama Krishna and Philip, 2008). The organic solute diffusion through the soil matrix was strongly affected by the moisture content of the soil (Ryan and Cohen, 1990). The vertical movement of pesticide, soil particles and organic matter with fluid in lower horizons will be very limited, although lateral movement in the soil naturally (Zaman *et al*., 2007). The potential for leaching is the greatest when highly soluble chemicals are used in well-drained sandy soils with low organic matter content (Reddy and Singh, 1993).

The studies pesticide was Lindane or γ-HCH is a broad spectrum insecticide used for control of insect pests in agriculture, vector control in public health programme and also for control of termite. Lindane consisting of almost pure γ-hexachlorocyclohexane (γ-HCH) is a persistent organochlorine insecticide which has been used for decades throughout the world (Li *et al*., 2004). It can retain over a long period of time in soils matrix due to its interaction with organic matter and poor solubility in water. Lindane distribution in soil column is great important pertaining to its influence on crops and ground water contamination. The significant amount of organic matter in examined soils and the chemical characteristics of the pesticides are largely responsible for their adsorption and consequent movement and distribution through soil. Recently, frequent detection of organochlorine insecticides was observed in surface and ground water and different other environmental components in Uttar Pradesh (Nayak *et al*., 1995; Raha *et al*., 1999; Raha *et al*., 2003). With the above context, the aimed at to this study the effect of these soil properties on Lindane adsorption, vertical movement and distribution pattern in organic rich soils of Uttaranchal.

MATERIALS AND METHODS

Area description

The state of Uttaranchal, India lying between 30°20′N 78°04′E latitude which logged with international border Nepal and China alongwith state border includes Uttar Pradesh west to south and Himanchal Pradesh north to west. The total geographical area of the state is 53,566km^2 of which 93% is mountainous and 64% is covered by forest with population density 190/km^2. There are three well defined (Pathak and Sharma, 1985) broad soil groups in Uttaranchal, India viz. (i) Hill soil (ii) Bhabar soil and (iii) Tarai soil respectively. The soils from different physiographic and agro climatic zones of Uttaranchal Pradesh varying widely in their physico-chemical properties were selected for this study.

Soil sampling and analyzed

The studied soils were sampled from different locations in district of Udham Singh Nagar and Almora, Uttaranchal by followed standard procedure and confirmed two composite soil samples from each district separately. Collected soil samples were air-dried at room temperature, powdered, passed through 2 mm sieve, mixed homogeneously and stored at 4°C for analysis. The soil samples were analyzed at various physico-chemical properties by standard procedure described by Jackson, (1973). The soil characterization, summarized in Table 2.

Chemicals description

The analytical grade (98.9%) of Lindane was obtained from Kanoria Chemicals Ltd., Renukut, Uttar Pradesh, India for the study. The chemical properties of Lindane are given in Table 1. Stock solution of Lindane concentration 500mg mL^{-1} was prepared by dissolving requisite quantity in methanol and stored at -20°C in deep freezer.

Adsorption and Movement study

Adsorption experiments were carried out using batch equilibrium technique from the stock solution (500mg mL^{-1}). Bach equilibrium adsorption isotherms were obtained by taking different concentrations each of lindane (250, 500, 750, 1000, 1250, 1500, 2000 and 2500 mg mL^{-1}) in 50 mL conical flasks and the volume of each flask was maintained upto 20mL with distilled water. To these solutions, 1g of each soil was added and the suspension was kept in an incubator for 24h after shaking with period of 3h at mechanical shaker. The incubation period was sufficient to attain equilibrium for the Lindane in soils. The suspensions were then centrifuged at 1500 rpm for 10 minutes and the supernatants were collected. The movement study of pesticides in two different soils was determined by using packed soil column as per method described by (Saeed *et al.,* 1997). Pesticide from soil samples used for adsorption and movement studied were extracted using standard solvent with liquid-liquid partition method and reserved for Lindane analysis.

Lindane analysis

The concentration of Lindane in n-hexane extract was analyzed using Nickel^{63} Electron Capture Detector of Gas Liquid Chromatography; Hewlett Packard 5890 a gas chromatograph equipped with a HP 3392 A integrator. Operating parameters for analysis of Lindane were as follows: 1.8 m ´ 2mm I.D. glass packed with 1.5% OV-17 +1.95% OV-210 of W.H.P. (80-100 mesh) column; oven temp.190°C, injector temp. 210°C, detector temperature 300°C and N_2 as carrier gas; flow rate 70mL min^{-1}. The compounds were identified on the basis of their retention time compared with technical grade reference standard.

Data analysis

The amount of Lindane adsorbed on soil (x/m) was calculated from the difference between the initial and equilibrium concentration (C_e). The data was fit to logarithmic form of the Freundlich equation:

$$\log x/m = \log K + \frac{1}{n} \log C_e$$

Where, log K and 1/n are the constants representing the intercept and slopes of the isotherms respectively where x/m is the concentrations of the adsorbate per unit volume of adsorbent (mg g^{-1}). C_e is the aqueous phase equilibrium concentration in mg mL^{-1}. The distribution coefficient, K_D was calculated as

$$K_D = \Sigma (x/m).C_e/\Sigma(C_e)^2$$

to measure the adsorption extent. The adsorption constants (K) normalized to soil organic carbon content, K_{OC} and soil clay content, K_C. These are important parameters that play a significant role in environmental fate assessment of pesticide chemicals and were evaluated by using the following equation (Swoboda and Thomas, 1968).

$$K_{OC} = \frac{K_D \times 100}{\%\text{ of organic carbon}} \qquad K_C = \frac{K_D \times 100}{\%\text{soil clay}}$$

Multiple regression analysis was done using Microstate package to correlate adsorption with various soil parameters.

RESULTS AND DISCUSSION

Soil analysis

The analyzed physico-chemical properties of soils were presented in Table 2. The values of soil pH varied from 6.3 to 7.6 indicating slightly acidic to neutral in nature, respectively. The content of organic matter and clay on Udham Singh Nagar 36.5 gkg^{-1} and 16.5% and Almora 40.3 gkg^{-1} and 12.5% was observed. However, the average $CaCO_3$ percentage was ranged between 0.05-27.0% and both the sampled soils of Udham Singh Nagar and Almora were classified as sandy loam based on mechanical analysis. The quantity of organic matter and clay attributed a major concern to permit the downward movement of Lindane in the lower depth. Soils of Udham Singh Nagar and Almora reflected high CEC (19.41 and 23.4 [C mol (P^{+}) kg^{-1}]) and surface area due to rich organic matter content which influenced the adsorption pattern of pesticides. Other properties of soils are summarized in Table 2.

Adsorption study of Lindane in Soils

The data on adsorption of Lindane i.e. γ-isomer of HCH in sandy loam soils of Udham Singh Nagar and Almora were presented in Table 3. The adsorption of Lindane in soils was intimated to be in accordance with the Freundlich adsorption isotherm which indicated that the positive correlation between soil parameters viz. organic matter, CEC and surface area of soil where, the adsorption isotherms were drawn between the

Table 1: Properties of the pesticide used in adsorption and mobility study

Pesticide	Chemical Name	Formula	M.W	Ionizability	Solubility K_{sp} (mg/L)	Volatility VP (mm Hg x 10^{-6})	Longevity $T_{½}$ (days)
Lindane.γ-HCH	1,2,3,4,5,6- Hexachlo rocyclohexane(a,a,a, e,e,e confugration of chlorine atom in HCH)	$C_6H_6Cl_6$	290.9	Non-ionic	7.3	95	180

Source: Weber, J.B. (1994)

Table 2: Important physico-chemical properties of soils

Soil Location	Soil Texture	Sand (%)	Silt (%)	Clay (%)	OM (g kg^{-1})	CEC [C mol (P^+) kg^{-1}]	$CaCO_3$ (%)	Surface area (m^2 g^{-1})	EC (d S m^{-1})	pH	Bulk density (Mg/m^3)	Particle density (Mg/m^3)	Water holding capacity (%)
Udham Singh Nagar	Sandy Loam	62.5	21	16.5	36.5	19.4	0.05	279.53	0.06	7.6	1.3	2.25	48.47
Almora	Sandy Loam	62.7	24.8	12.5	40.3	23.4	27	289.47	0.08	6.3	1.15	2.11	54.61

Table 3: Concentration of Lindane following adsorption in soils

Soil	Equilibrium concentration (Ce)and Amount adsorbed (x/m)	Initial Concentration (µg mL^{-1})							
		250	500	750	1000	1250	1500	2000	2500
Udham Singh Nagar	Ce	76.76	154.95	221.16	278.88	347.76	532.74	656.60	973.35
	x/m	173.24	345.05	528.84	721.12	902.12	967.76	1343.40	1526.65
Almora	Ce	57.28	107.94	167.28	228.60	317.80	368.88	497.04	619.56
	x/m	192.72	392.06	582.72	771.40	932.12	1131.12	1502.96	1850.56

a - Concentration in solution following adsorption (µg mL^{-1}); b - Concentration in soil following adsorption (µg g^{-1})

Table 4: Freundlich constants for adsorption of Lindane in soils of Uttaranchal

Constants	K	K_D	K_{OC}	K_C	1/n
Udham SinghNagar	3.460	20.244	954.90	122.69	0.941
Almora	4.543	21.240	907.69	169.92	0.995

amount of Lindane adsorbed (mg g^{-1}) on soils and that in equilibrium suspension (mg mL^{-1}) at a fixed volume of water respectively.

The distribution coefficient, Kd is defined as the ratio of insecticide concentration in the soils and Lindane concentration in solution. The value obtained for the constant, mentioned as given in Table 6, the value of K and Kd considered as a prime approximation in characterizing adsorption capacity of Lindane in both sets of soil. It is observed that high Kd value 20.24 and in both the soils due to significant amount of organic matter that exhibited higher adsorption capacity.

The correlation coefficient between organic matter and Kd and Kc was 0.805 that indicating significant positive correlation between organic matter and Lindane adsorption on examined soils. In general, the adsorption isotherm of soils with varying organic matter content correspondent to the S type (Giles *et al.*, 1960) have observed that low adsorption towards soil affinity at low Lindane concentration. The initial slop of adsorption isotherm, which is a function of soil organic matter content, was noted to be steeper for soils. Besides, the strong binding observed in the present study was attributed to the high clay content with a wide surface area of clay particles facilitates adsorption which is essentially a surface phenomenon (Nyle, 1984).

It is established that the degree of Lindane adsorption on soils of Uttaranchal increased with increasing chemical concentrations which belong to the subclass 'C'-type and the linear relationship being indicative of constant partition of the adsorbate between the solvent and the adsorbent. The correlation coefficient (r) for the adsorption phase ranged from 0.981 to 0.993 indicating the perfect fit of isotherm as per the Freundlich adsorption equation. The linearity of the slope (1/n) worked out in Table 4 with Uttaranchal soils for Lindane, and its range was 0.941 to 0.995. Comparison to both the soils, the higher Lindane adsorption was found in Almora soil (sandy loam) at each concentration might be due to rather high organic matter content than Udham Singh Nagar soil which retained the chemical on soil surface (Table 3). It was revealed that the range of adsorption of Lindane in soils was 60-78% on an average the order of adsorption in studied soils. The affinity of Lindane towards organic matter and clay content of the soils was evaluated by calculating K_{OC} and K_C values which are summarized in Table 4. K_{OC} values of both the soils were much higher than K_C, indicated that contribution of the surface of organic matter in Lindane adsorption was much higher than the clay surface in soils. Similar report was made by (Raj *et al.*, 1999; Kalpana *et al.*, 2002; Kumar and Philip, 2006) the soil separates is primarily responsible for adsorption of pesticides.

Table 5: Movement study of Lindane in soil column

Soil	Depth of soil column (cm)	Lindane mobility in soil column (μ g^{-1})								
		0-2 (cm)	2-4 (cm)	4-6 (cm)	6-8 (cm)	8-10 (cm)	10-12 (cm)	12-14 (cm)	14-16 (cm)	Lindane in Leachate (μ g^{-1})
Udham Singh Nagar	15.5	7.713 (0.187)	6.725 (0.163)	6.084 (0.147)	5.085 -1.123	4.801 (0.116)	2.941 (0.071)	1.698 (0.041)	1.473 (0.035)	4.640 (0.112)
Almora	16	7.175 (0.174)	7.249 (0.175)	6.174 (0.149)	5.25 -0.127	5.216 (0.126)	3.777 (0.091)	2.816 (0.068)	1.240 (0.030)	2.293 (0.055)

Fractions of lindane in parenthesis

Multiple regressions between amount adsorbed (Y) and soil properties provided the equation:

$Y = 1.89 - 0.40 (Clay) + 0.54 (OM) - 0.12 (CEC) + 0.20 (CaCO_3) - 0.20 (Fe_2O_3) - 0.02 (Al_2O_3) - 1.03 (Surface\ area) + 0.42 (EC) + 0.44 (pH) - 0.11 (BD) + 0.80 (PD) - 0.50 (W.H.C.)$

The above equation shows that organic carbon of soil is the most important factor influencing K of Lindane. Therefore, high K value (4.543 and 3.460) in both the tested soil can be explained by the presence of all favorable factors i.e. high organic carbon, surface area and CEC.

Soil organic matter is the principal adsorbent for many non-ionic organic compounds (Mader *et al.*, 1977; Xing and Pignatello, 1996). Lindane is nonionic, non-polar hydrophobic compound and thus, weak attractive interaction such as Vander Waals forces is the dominant forces of attraction in case of the adsorption of Lindane on soil surface and there is reason to believe that the exchange complex of inorganic clays in soils played little role in Lindane adsorption.

Study of vertical movement and distribution of Lindane in soils

Downward movement of pesticide through soil column is important to determine their efficacy as well as their potential for crop damage and environmental pollution (Mersie and Foy, 1986). Lindane is less soluble, its absorption in soil is relatively high that inhibit the possible mobility in soil. In the present study, Lindane movement was performed with soil containing diversifies physic-chemical properties in the packed soil column, the distribution of Lindane was obtained throughout the column and even showed presence in the lower most soil core. Few of Lindane residues were also detected in leachate fractions in tested soil showed in Table 5. Higher chemical was noted within the 10 cm depth (about 75%) of soil layer of the packed column, and remains distributed in lower depth through the soils. The Lindane was detected rather less in leachates.

The lowest recovered amount of Lindane was detected on Almora soil in 0-10cm depth (3.57 %) followed by Udham Singh Nagar soil (3.01%) showed in Table 6. Results revealed lower downward movement of Lindane in both soils and its observed scanty move beyond the 10 cm soil depth possibly due to high adsorption pattern. Leaching behavior of pure grade Lindane were studied in various textural group of soil in pact columns which was drenched with 100 mL of water when Lindane incorporated into the surface of soil. The mobility of Lindane behaved differently in different soils due to significant interaction between soil and elution contents. The relative mobility of pesticides through soil and its longevity are key indicators of the potential for contamination of drainage effluent and ground water by that chemical (Weber and Keller, 1994). The downward movement through soil column studied were normally carried out by applying a pesticide to a soil followed by 50cm of water representing the amount of rainfall needed to produce a crop. In present experiment, vertical leaching loss was carried out of high persistence organochlorine insecticides, viz. Lindane in both the organic rich soils of Uttaranchal, India. Studies showed that the highest concentration of Lindane was found in Udham Sigh Nagar soil with 2cm (7.713 mg^{-1}) and gradually observed reduced trend with lower column. However, the maximum Lindane was found from 0 to 6cm depth (21.673mg^{-1}) in Almora soils presented in Table 5. Similar trends with regard to fraction of Lindane (Table 7 in parentheses) were identified in both types of soils which retained in soil as because less solubility to non-ionic nature of chemicals that have strong affinity towards organics and clay content. Several study of pesticide movement in soil columns have been carried by (Dousset and Mouvet, 1997; Vereecken *et al.*, 1995) that supported good agreement with our findings. Downward movement increases with frequency of irrigation (Frick *et al.*, 1998); increasing temperature accelerates the degradation of pesticides slowing their migration (Singh *et al.*, 2005) and large amount of clay and organic matter in the soil, possibly to diminish the pesticide mobility (Durovic *et al.*, 2009).

Distribution of Lindane in soils affected by properties of pesticides and soils indicated in Table 1 and 2 was studies using two different types of soil considering the textural group sandy loam in tarai and hill region. The distribution coefficient, Kd, for the soil in batch adsorption as well as in columns were calculated. The highest concentration was found at the surface and near surface horizons and gradually decreasing with depth (Table 3). The maximum distribution percent of Lindane was recorded with the Almora soil (94.44%) followed by Udham Singh Nagar soil (88.75%) whereas, less though leachate (5.56%) and (11.25) was obtained showed in Table 6 might be due to high organic matter content (Malik *et al.*, 2009; Singh *et al.*, 2010). Some of the study reported (Priya

Table 6: Percentage distribution of lindane in soil column

Soil	Depth of soil column (cm)	Pesticide	Lindane mobility in soil column (μ g⁻¹)								Lindane in Leachate (μ g⁻¹)
			0-2 (cm)	2-4 (cm)	4-6 (cm)	6-8 (cm)	8-10 (cm)	10-12 (cm)	12-14 (cm)	14-16 (cm)	
Udham Singh Nagar	15.5	Lindane	18.71	16.31	14.76	12.33	11.65	7.13	4.12	3.57	11.25
Almora	16.0	Lindane	17.41	17.59	14.98	12.74	12.66	9.16	6.83	3.01	5.56

and George, 2010) that movement and adsorption of pesticides regulated by both chemical and soil properties. The percent distribution of lindane was maximum associated with upper 0-4cm and gradually lesser with downward upto 16cm depth in both the soils. On the other hand, the vulnerability of ground water for pesticide residues depends upon the ratio of their persistence and residence time in the soil. For these compounds no soil type will guarantee protection against groundwater contamination. Thus, the risk of break through to the ground water of highly persistence chlorinated hydro-carbon viz. γ-HCH or Lindane in future could not be ignored.

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