Sorption Kinetics Study of Monocrotophos (MCP) Pesticide in Unsterilized Soil

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Author's contribution

The sole author designed, analyzed and interpreted and prepared the manuscript.

Article Information

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ABSTRACT

Sorption Kinetics of monocrotophos in sandy soil was studied with the aim to establish its fate in the soil. The soil used was characterized with respect to soil pH, organic carbon, moisture, silt, clay and sand content. The sorption kinetic studies were carried out by observing the disappearance of monocrotophos in solution for several days. The kinetic studies showed that sorption of monocrotophos followed first order kinetics and were measured in terms of two processes namely; fast and slow sorption. The amount of pesticide sorbed by soil was found to increase with time and the values showed that the soil has low capacity to sorbe or retain monocrotophos when applied. During this time, rate constant for the disappearance from solution of monocrotophos were found to be 18.2×10^0 s^{-1} and 0.248×10^1 s^{-1} respectively in fast and slow sorption processes. While, the rates for fast sorption and slow sorption were found to be 5.3885 s^{-1} and 0.0315 s^{-1}.

Keywords: Monocrotophos; sorption; kinetic; soil; rate; Freundlich isotherm.

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1. INTRODUCTION

Once pesticides are introduced into the soil environment, whether by direct application, disposal, or a spill, it can be influenced by many processes. They could be translocated, transformed or sorbed. These processes determine the ultimate fate of the pesticide by affecting its persistence and movement in the environment. The optimum life of a pesticides or its persistence is measure by its half-life. Mobility of pesticides is one of the factors that affect the distribution of the pesticides into the soil water phases. The mobility of the pesticide is restricted when associated with solid phase; while on the other hand, the mobility is enhanced when associated with solution [1]. The mobility of pesticides in soil is guided by pH, organic matter, moisture, Clay content and distribution coefficient (K_d) of pesticide. The hazards of pesticides in soil largely depend on their persistence. The longer they persist, the greater are the chances of contamination of soil. Ideally, pesticides should persist only long enough to complete its intended mission and then degrade to harmless products before it is necessary to apply again [2].

In many developing countries, the pesticides use is not being properly regulated leading to residues in food which poses health hazards to the consumers [3]. Therefore, residues of pesticides could affect the ultimate. Many studies reported that the main pesticide residues were organophosphate [4-7]. Organophosphorous pesticides (OPPs) are a group of highly toxic agricultural chemicals widely used in plant protection. Common members of the family are methyl parathion, malathion, dimethoate, phosphamidon, phorate, fenitrothion and monocrotophos [8].

Monocrotophos (Dimethyl (E)-1-methyl-2-(methylcarbamoyl) vinyl phosphate) controls pests on a variety of crops, such as cotton, rice, and sugarcane. It is used to control a wide spectrum of chewing and sucking insects and also mites [8]. Following concern on its toxicity to non-target species especially birds, monocrotophos was voluntarily withdrawn from sale in the US in 1989. It was also banned in Indonesia, Sri Lanka and Philippines while its use is severely restricted in Kuwait and Malaysia [9]. Monocrotophos is still in use in Nigeria and it is usually applied directly on agricultural land primarily to control pests and to improve crop yield with the aim to meet the high demand for food due to the fast growing population [9]. The

long standing practice in which this pesticide was studied intensively on how to be applied to crop and insect pests has been extended to practices such as adsorption, transportation and sorption processes in soil environment in different part of the world. This knowledge is essential to make a better use of the wide variety of chemical compounds that are now available for weed control, and for elucidating possible adverse environmental effects and their control [1]. In Nigeria, there are far-reaching literature on the sub-lethal toxicity of monocrotophos on animals at different concentrations [10,11,9]. However, despite the abundance of literature on monocrotophos in these aforementioned area, there is scantiness of information on the sorption kinetics of this pesticide in soils in Nigeria. Sorption and degradation are the two most important processes affecting the fate and behaviour of pesticides in soil [12]. The degree of sorption from the soil solution on onto the soil phase of the soil may be described by a soil-water partition coefficient (K_D). [12]. Oketunde and Sha'ato [1], have pointed out the critical dependence of degradation kinetics on sorption kinetics, focusing the search for the mechanism of persistence.

One aspect of pesticides interaction with soil that will clearly need attention in this work is sorption rate, since this influence availability of the pesticides to function as such, and to move and degrade. Sorption processes involve an array of phenomena which can alter the distribution of contaminants between and among the constituent phases and interfaces of subsurface system [13].

The objective of this study is to investigate the sorption kinetics of monocrotophos (Dimethyl (E)-1-methyl-2-(methylcarbamoyl)vinyl phosphate) in the soil. Sorption is one of the most important processes in determining the persistence and transport (dynamics) of pesticides in the soil environment [14].

The environmental behavior of monocrotophos can be considered in two ways: mobility and stability (Persistence). Generally, organophophorus pesticides are rapidly degraded with less persistence in soil [15]. Monocrotophos has a half-life of 14-21 days at pH 9 and 25°C, with the rate decreasing at lower pH's and increasing at higher temperatures [16]. Degradation on soil exposed to natural sunlight is rapid (half life less than 7 days) and on dark control samples is slower (half-life approximately

30 days). Monocrotophos is mobile in soil, and although it degrades rapidly it may possess potential for groundwater contamination [16].

2. MATERIALS AND METHODS

2.1 Pesticides and Chemicals

All chemicals used were analytical grade reagents and commercial emulsified monocrotophos which was manufactured by: Sabero Organic Gujarate Ltd India. The concentration of the commercial monocrotophos was 400 g/L and was used as applied by farmers without further purification. Monocrotophos has a molecular weight of 223.2 g /mol and the chemical structure is shown in Fig. 1.
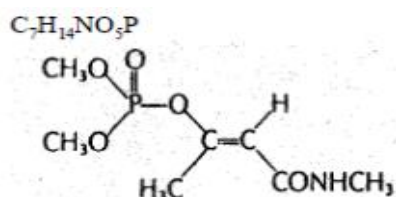

Fig. 1 Chemical structure of MCP

2.2 Soil Sampling and Pre-treatment

Soil samples were collected at depths: 0 - 20 cm from university of Agriculture Makurdi farm Land. They were sieved, free from unwanted debris, made lump free and thoroughly mixed before it was used for the sorption study. The study area is showing in Fig. 2.

2.3 Soil Characterization

The soil was analyzed for pH, percentage clay content, silt content, sand content, moisture content and total organic carbon. The organic matter content of the soil was determined by Walkley and Black modified method [18].

2.4 Preparation of Various Monocrotophos from the Stock

The concentration of the commercial formulation of monocrotophos was 400 g/L, distilled water was used to prepare all aqueous solution. A 1.0×10^{-3} M stock solution of monocrotophos was prepared by diluting $0.285 \ cm^3$ of the commercial formulation to $500 \ cm^3$ with distilled water in a volumetric flask. Separate aliquots of the stock $(0.75 - 4.0 \ cm^3)$ were, then serially diluted to $100 \ cm^3$ with distilled water in volumetric flasks

to furnish various standards $(7.5 \times 10^{-6} - 4.0 \times 10^{-5}$ M) which were used to prepare calibration curves for spectrophotometric determination of monocrotophos in aqueous solutions (Fig. 3). Absorbance of the solutions of the various concentrations was measured at 214nm using UV-Visible spectrophotometer (pharamcia Biotech-ultrospect 2000). The absorbance of the supernatant of centrifuged aliquots of the suspensions was measured using distilled water as reference. To ensure the quality of data obtained, the experiment were carried out in duplicate for each sample and the mean data is presented in Table 1.

Table 1. Monocrotophos concentration (mol/L) and Asorbance

Concentration (mol/dm^3)	Absorbance
8.5 x 10-6	0.176
1.0 x 10-5	0.217
2.0 x 10-5	0.300
3.0 x 10-5	0.487
4.0 x 10-5	0.577

2.5 Sorption Kinetic Studies

To each of the three sample bottles containing 2.5 g of soil, $100 cm^3$ of distilled water was added in order to wet the soil surfaces and stirred for 2 hours and allowed to stand for 30 minutes. The Kinetic run was initiated by the addition of $25 cm^3$ of the standard stock solution $(1 \times 10^{-5}$ M) of monocrotophos into two bottles respectively whereas the third bottle served as the reference. The analysis of the sample commenced ten minutes after addition of monocrotophos $(5.0 cm^3)$ aliquot of the slurry was taken into three 15 cm^3 centrifuge tubes. The tubes were agitated for 3 minutes; the suspension was then centrifuged for a predetermined contact time of 5 minutes without adjusting pH at room temperature. A fixed amount of the supernatant was taken and then filtered using filter paper (Whatmann No. 1). The filtrates were then analyzed in duplicates using UV – Visible Spectrophotometer for quantifying the change of solution concentration with time. The difference between initial and final concentration of monocrotophos was considered as the amount sorbed (Table 3).

2.5 Sorption Kinetics Parameters

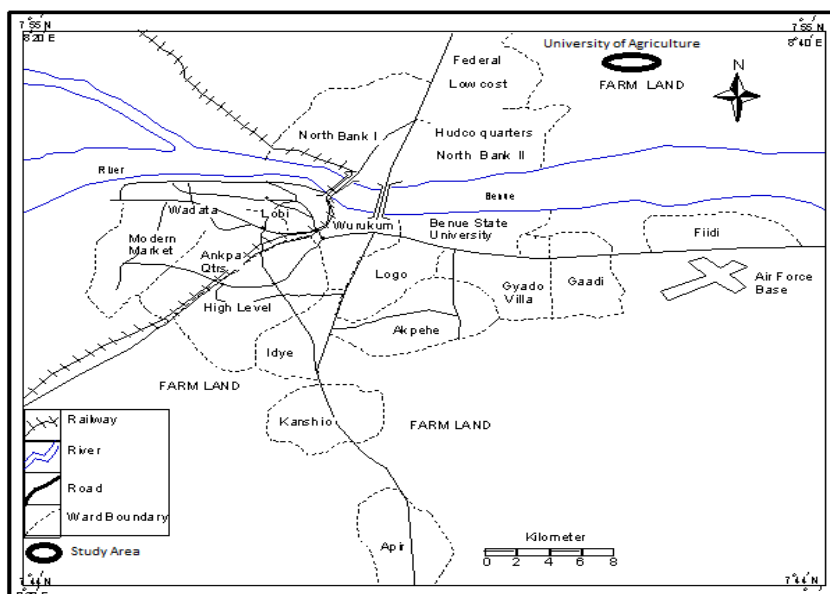The interaction between the pesticide and the soil is represented by the following.

$$P_{ap} + S \rightarrow P.S \qquad (1)$$

Fig. 2. Map of makurdi showing the study area. Source: Adopted from Shabu et al. [17]
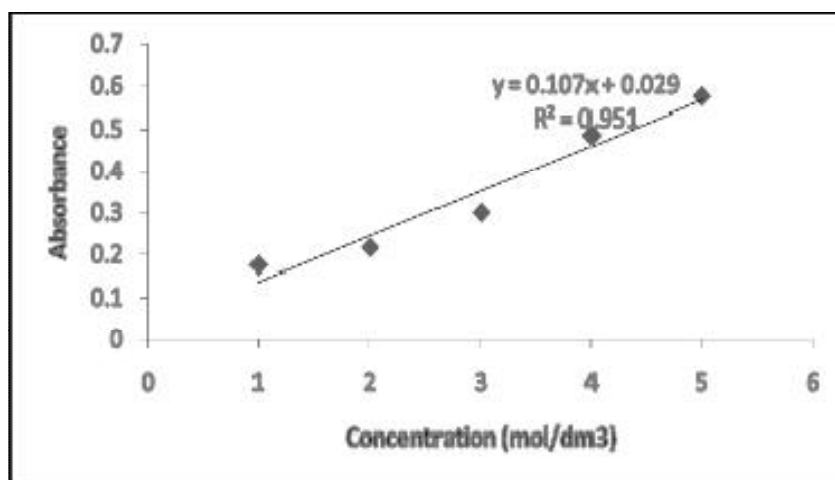

Fig. 3. Calibration curve of monocrotophos at 214 nm

Where "P_{ap}" represents the solution phase concentration of the pesticide, 'S' represents the solution phase concentration of the pesticide bound to the soil surface by reversible labile processes, then

$$X_C = X_{LS} + X_0 \qquad (2)$$

Where, X_c is a measure of the total sorption sites (molg^{-1}) which is called the labile sorption capacity, X_{LS} is the concentration (molg^{-1}) of active sites occupied by pesticides and X_o is the concentration (molg^{-1}) of active sites that are unoccupied. When equilibrium between the soil pesticide and aqueous slurry is reached, the law of mass action may be applied and the weighted average equilibrium constant, K_1 is determined as [1].

$$K_1 = X_{LS} \ /C_P X_0 \qquad (3)$$

Where C_p is the equilibrium concentration of the percentage of the pesticide on the soil solution. The distribution coefficient (K_d) is an empirical constant that described the affinity of the

pesticides molecule for the solid phase relative to the solution.

$$K_D = X_{LS} / C_P \qquad (4)$$

Combing equation (1) (2) and (3) gives equation (4)

$$K_D = K_1 (X_c - X_{LS}) \qquad (5)$$

Equation (4) implies that K_D is not a constant but depends inversely on the surface loading of the pesticide (i.e X_{LS}). The rate of loss of the pesticide from solution to labil surface site can be defined by the formula.

$$-\frac{dC_p}{dt} = K_f C_p X \text{ _____ } K_{des} X_{LS} \qquad (6)$$

Where, K_f is the second order rate constant describing the absorption process. That is, sorption of pesticide to soil surfaces depends on the concentration of the pesticide sites on the soil surface, where a desorption is a first order reaction whose rate constant, K_{des}, depends only on concentration of pesticide molecule on the occupied sites. In case where there is low coverage of sorption sites, then $X_{LS} << X_c$, and therefore, $X_o \approx X_c$ = constant, and the values of $K_{des} K_{LS}$ is very small. Under this condition, equation (6) reduces to equation (7).

$$-\frac{dC_p}{dt} = K_f C_p \qquad (7)$$

K_f is now a psendo-first order rate constant integrating, we have

$$2.3031 log C_p / C_{po} = K'_f \qquad (8)$$

A plot of log (C_{po} – C_{pt}) vs time is expected to be a straight line for the pseudo-first order processes with a slop equal to K'_f /2.303. The pseudo-first order rate constant (K'_f = -2.30 x slop).

3. RESULTS AND DISCUSSION

3.1 Soil Properties

Before the kinetic experiment was carried out, the soil was characterized. Some of the physicochemical properties are shown in Table 2.

The pH range of the soil shows that it is moderately acidic. These properties are likely to influence the fate of monocrotophos in soil to varying extents [19,20]. It usually follows the trend the higher the pH, the higher the base

saturation. It is generally observed that for most soils at a pH of 7 or above, the base saturation approaches 100% [1].

From the texture triangle, the soil was found to be sandy soil, low in clay, silt and organic carbon. The small size fraction that contains clay minerals and organic carbon are those that are particularly active in terms of interaction with the pesticide. The small amount of organic matter and the nature of the clay minerals of this may lead to smaller degree of sorption of the pesticide soil interaction.

The amount of organic carbon content determined by the wet oxidation method is a measure of the total organic carbon of the soil but may be partial due to other component that may be present and was found to be 4.12%. The moisture content was found to be 0.45% after drying at 105°C.

Table 2. Physicochemical properties of soil

Properties	Mean
pH in aqueous $CaCl_2$	5.11
pH in water	6.52
Silt content	5.39%
Clay content	1.96%
Sand content	91.56%
Soil type	Sandy soil
Moisture content	0.40%
Total organic carbon	4.12%

3.2 Sorption and Speciation of Monocrotophos in the Soil – Solution Matrix

The experiment was carried out using slurries maintained over a period of approximately three days from commencement. The method enables separation of the various loci of pesticide molecules in the entire slurry. The effects of the pesticide initial concentration with contact time are shown in Fig. 4. The sorption can be viewed in here as a two phase process, with a rapid initial decrease in concentration of monocrotophos in solution, followed subsequently by a much slower rate. The rapid adsorption occurred at the beginning since there was only physical affinity between the soil and pesticide, and there was a difference in concentration gradient between bulk solution and surface of soil [21], thereby representing a significant motivating force for monocrotophos transfer between the solution and the soil

surface. However, after the initial period, slow sorption may be due to slower diffusion of solute into the interior of the soil. The fig. 4 shows a rapid initial decrease in concentration of the pesticide in solution within the first day and then slowly until no appreciable change in concentration was observed ,while the amount of monocrotophos sorbed by the soil was found to increase with increase in time (Fig. 4).

3.3 Kinetic Evaluation

Several adsorption kinetics models have been developed to understand the adsorption kinetics and rate limiting step [22]. In this work however, Lagergren proposed a pseudo-first order kinetic model was considered to investigate the mechanism of monocrotophos absorption in soil. This is because pseudo first order kinetic model is better for predicting the kinetic process in the experimental conditions than the pseudo second order kinetic model [7,23].

Usually, the pseudo first order kinetic correlation coefficients, r^2 and sorption values are calculated from the plot of log ($C_{p,o} - C_{p,t}$) *vs.* time. Nevertheless, in this work, we a plotted 13 + 1n ($C_{p,o} - C_{p,t}$) against time as shown in Fig. 6 and 7. Rate constant for the disappearance from solution of monocrotophos were found to be 18.2×10^0 s^{-1} and 0.248×10^1 s^{-1} respectively, also values for the fast sorption and slow sorption were also found to be 5.3885s^{-1} and 0.0315s^{-1} respectively (Fig. 5 and 6) .The result revealed that the soil has low capacity to sorb or retain monocrotophos when applied.

Within this period of time for the sorption study, a number of processes occur: the monocrotophos distribute itself between the solution phase and the solid phase, and in the later phase, the amount of pesticide that diffused irreversibly, into soil voids or is otherwise irreversibly bond to the soil surface which is called bound residue (BR). The phenomenon of BR is more associated with soils of high organic carbon, it is expected that the occurrence of bound residues of the pesticide in this soil will be low judging by the amount present in this soil.

3.4 Adsorption Isotherm

Pesticide sorption isotherm was conducted using Freundlich equation in terms of linearized relationship:

$$InQ_e = InK_F + \frac{1}{n}InC_e \qquad (9)$$

Where *Ce* (mg/L) concentration of material remaining in solution after adsorption is completed (mg/l), *qe* (mg/g) is the amount of sorbed pesticide per unit mass of the adsorbent, K_F- Freundlich adsorption coefficient related to adsorption capacity; *1/n* – Freundlich equilibrium constant that characterizes adsorption intensity and energy distribution of adsorption sites [24].

Linear Freundlich Isotherm for the sorption of monocrotophos by soil is presented in Fig. 7. The high correlation coefficient (r^2) of the line was 0.9173 which suggested that the data is fitted well with Freundlich model, an indication that adsorption mechanism was related to non-ideal, reversible and multilayer adsorption with non-uniform distribution of adsorption heat and affinities over the heterogeneous surface [25]. The Freundlich adsorption exponent (1/n = 2.4982) was obtained from slope of this line which were greater than unity (>1.0) indicating a relative increase due to the increasing of initial concentrations [21].
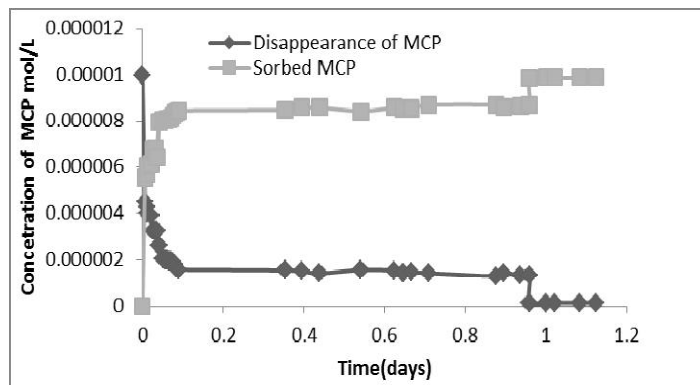

Fig. 4. Effect of contact time, sorbed and initial MCP disappearance in soil
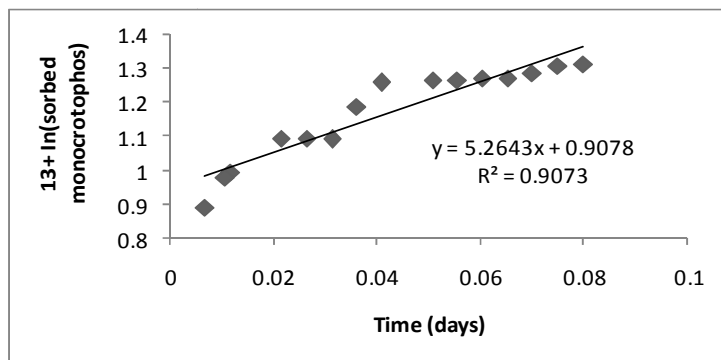
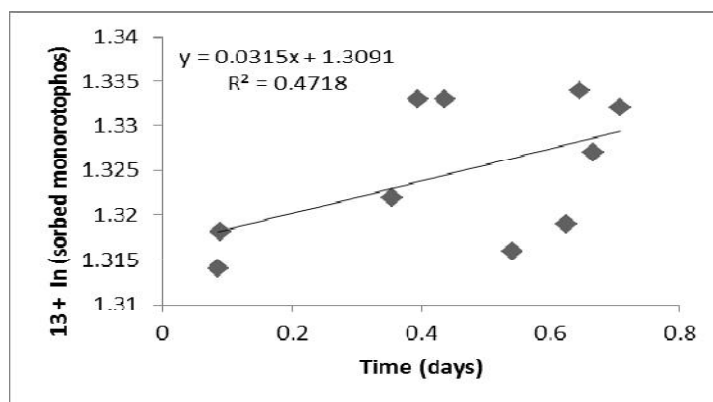**Fig. 5. Sorption kinetics of monocrotophos on soil (Fast)**

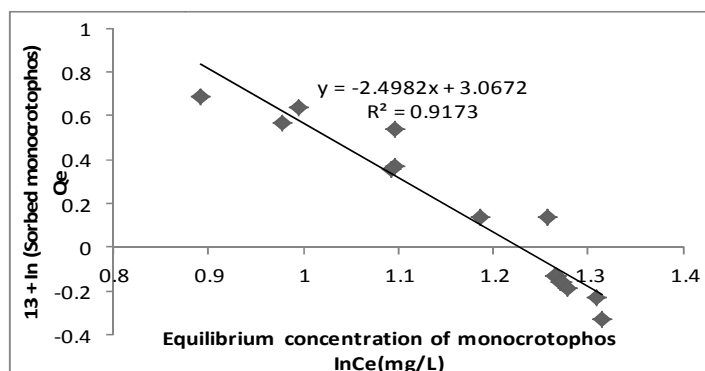**Fig. 6 Sorption Kinetics of monocrotophos on soil (Slow rate)**

Fig. 7. Linear freundlich isotherm for the sorption of monocrotophos by soil

At the beginning of the absorption process, the trend line is almost linear; suggesting that competition for pesticide between soil and aqueous phase was almost the same or increasing at the same rate (at this point Kf could also be named as soil- water partition coefficient K_D). However, as sorption process proceeds with time, the *1/n* value deviated from unity indicating the nonlinear relationship between Qe and Ce with relatively strong adsorption of pesticide on soil as compared to that of water. This is a common feature for adsorption of organic chemicals in soils with low organic matter [18]. Although isotherm does not give confirmations about adsorbate-adsorbent interaction [18,26], but its shape helps in understanding the

Table 3. Sorption in soil

Time (days)	Absorbance	(Mononcrotophos)mol/L at time (t)	Sorbed concentration	Ln(Sorbed Concentration)
0.0000	0.0000	0.00001	0.0000	0.0000
0.0068	0.340	0.0000044819	0.0000055181	-12.1075
0.0118	0.331	0.0000042746	0.0000057254	-12.0049
0.0107	0.315	0.0000039896	0.0000060104	-12.0220
0.0215	0.310	0.0000038860	0.0000061140	-11.9042
0.0264	0.309	0.0000032642	0.0000067358	-11.9042
0.0313	0.253	0.0000032383	0.0000067617	-11.9081
0.0361	0.254	0.0000032383	0.0000064093	-11.8128
0.0410	0.251	0.0000025907	0.0000079534	-11.7419
0.0507	0.209	0.0000020466	0.0000080117	-11.7346
0.0556	0.159	0.0000019883	0.0000080181	-11.7338
0.0604	0.154	0.0000019819	0.0000080570	-11.7290
0.0653	0.155	0.0000019430	0.0000080764	-11.7266
0.0701	0.150	0.0000019236	0.0000081218	-11.7210
0.0750	0.149	0.0000018782	0.0000081995	-11.7114
0.0799	0.145	0.0000018005	0.0000083744	-11.6903
0.0847	0.139	0.0000016256	0.0000084132	-11.6857
0.0896	0.128	0.0000015868	0.0000084456	-11.6819
0.3542	0.123	0.0000015544	0.0000084780	-11.6780
0.3958	0.120	0.0000015220	0.0000085751	-11.6668
0.4375	0.117	0.0000014249	0.0000085751	-11.6668
0.5417	0.127	0.0000015738	0.0000084262	-11.6842
0.6250	0.126	0.0000015415	0.0000085751	-11.6803
0.6458	0.120	0.0000014249	0.0000085233	-11.6666
0.6667	0.118	0.0000014767	0.0000085622	-11.6727
0.7083	0.115	0.0000014378	0.0000087047	-11.6682
0.8750	0.114	0.0000012953	0.0000087047	-11.6517
0.8958	0.111	0.0000014119	0.0000085881	-11.6651
0.9375	0.100	0.0000013601	0.0000086399	-11.6591
0.9583	0.109	0.0000013342	0.0000086658	-11.6561
0.9582	0.105	0.00000014249	0.0000098575	-11.5273
1.000	0.103	0.00000012953	0.0000098705	-11.5260
1.0208	0.011	0.00000012953	0.0000098705	-11.5260
1.0833	0.010	0.00000012953	0.0000098705	-11.5260
1.1250	0.010	0.00000012953	0.0000098705	-11.5260

adsorption mechanism [26]. The Freundlich adsorption exponent values obtained in this work are comparable to those reported by [18,20,27,28]. The extent of adsorption K_F from the intercept of the line was 3.0673 mg/kg.

4. CONCLUSION AND RECOMMENDA-TION

This work was geared towards investigation pesticide – soil interaction. The soil used was characterized with respect to soil pH, organic carbon, moisture content, silt, clay and sand content. The sorption kinetic studies were determined by observing the disappearance of monocrotophos is solution for several days.

The sorption could be described by two distinct kinetic processes, namely, fast sorption (occurs within few hours after spiking the soil) and slow (for the remaining days). The first order plot showed that the concentration of monocrotophos decrease with time (days). Inversely, the amount sorbed was found to increase with increase in time and the values showed that the soil has low capacity to sorbed or retain monocrotophos when applied. This could be probably due to low organic content, abiotic and biotic factors since the soil was not sterilized before used. It was also assumed that the sorption of monocrotophos in the soil could result in low "bound residues" since it is associated with soil reach in organic matter.

In order to have comparable data on the sorption of monocrotophos, more work should be carried out on different classes of pesticide using different agro – environmental zones. This will also enable us to predict their fate in different environment. To avoid microbial degradation, work should be done using sterilized soil. Attempt should also be made to account for hydrolytic degradation since soil surfaces could act as catalyst for pesticide hydrolysis.

COMPETING INTERESTS

Author has declared that no competing interests exist.

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