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Effect of Temperature and Soil Type on the Adsorption and Desorption Isotherms of Thiamethoxam Using Freundlich Equation

Mohamed R. Fouad

Department of Pesticide Chemistry and Technology, Faculty of Agriculture, Alexandria University, Aflaton St., 21545, El-Shatby, Alexandria, Egypt.

Abstract

The adsorption-desorption isotherms of thiamethoxam in clay loam, clay, sandy loam, sandy clay loam, sand and loamy sand soils at 25&50 ˚C was studied. The amount of thiamethoxam adsorbed and desorbed by soils was significantly influenced by the temperature. Average of adsorbed of thiamethoxam on soils were 13.129, 14.611, 12.305, 6.812 and 6.943 µg g soil^{-1} for clay loam, clay, sandy loam, sandy clay loam and loamy sand soil, respectively. However, the adsorbed amount was reduced to 11.238, 10.450, 7.430, 5.578, and 6.832 µg g soil^{-1} for clay loam, clay, sandy loam, sandy clay loam, sand and loamy sand soils as the temperature was decreased to 25˚C, except for sand soil. The value of K_F in adsorption for clay loam and sandy clay loam soil is greater at 25˚C than that at 50˚C and the opposite in clay, sandy loam, sand and loamy sand soil. Freundlich model was the best fit for thiamethoxam adsorption and desorption on all soils. That adsorption and desorption of thiamethoxam in soils are spontaneous with a high affinity for thiamethoxam.

Keywords: Temperature; Soils; Adsorption; Desorption; Thiamethoxam; Freundlich equation.

1. Introduction

Neonicotinoids are a new generation of insecticides and are the most widely used in the world today [1]. Neonicotinoids are predominantly used for seed treatment for a large variety of crops such as canola, corn, soybean, cotton, rice, sorghum, sugar beets, sweet corn, and wheat. Seed treatment with neonicotinoids can provide excellent protection against a wide range of soil-borne insects as granular products for the control of soil dwelling insect pests, as soil drench around the roots of plants, or in irrigation water for drench and foliar applications [1,2]. It is estimated that more than 90% of the neonicotinoids used in seed dressings enter the soil without being absorbed by the crop [1,3]. Data on adsorption-desorption, degradation, and transport of the neonicotinoids in soils are critical for evaluating the fate and transport of these insecticides in soils and groundwater. Previous studies to obtain these data have been primarily focused on imidacloprid. The data for thiamethoxam are still lacking [1,4]. Thiamethoxam holds registration for 115 crop uses in at least 64 countries [1,5]. It is effective in killing sucking and chewing insects such as aphids, whiteflies, plant hoppers, thrips, and beetles that attack various crops including rice, maize, cotton, vegetables, and mango [1,6]. Thiamethoxam can follow different routes from the soil: retention in organic and/or mineral soil fraction, chemical, photochemical and biological degradation, volatilization, runoff and leaching [7]. The sorption is an important factor regulating pesticides behavior in the environment, being useful information to foresee the contamination potential of surface and ground water so it is important to understand the sorption processes and its relationship with soil parameters. Some pesticide properties, which influence sorption to the soil particles are water solubility, vapor pressure, octanol-water partition coefficient and acid-base ionization constant for ionizable

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*Corresponding author e-mail: mohammed.riad@alexu.edu.eg; mohammed.reid@yahoo.com; (Mohamed R. Fouad).

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compounds. In the soils, mineral composition and soil organic matter are important factors affecting sorption of thiamethoxam [7]. In the Guangzhou section of the Pearl river (China), thiamethoxam, acetamiprid, imidacloprid and clothianidin were detected 100% of the times while thiacloprid was detected with a frequency of 93% with total concentration of the 5 neocotinoids (93 to 321 ng L^{-1}). The equivalent total concentration in soil was (0.40–2.59 ng g^{-1}) with detection frequencies ≥78% [8]. Found that the concentration of neonicotinoids (clothianidin > imidacloprid > thiamethoxam) in arable soils ranged from 0.02 to 13.6 μg kg^{-1} soil, from eighteen sites widely spread out in England [8,9]. Neonicotinoids have been widely detected in the environment with concentrations in the range of parts per billion (ppb)-parts per million (ppm) in soil, parts per trillion-ppb in water and ppb-ppm in plants [10]. Previous studies have reported the presence of neonicotinoids in surface runoff from soils or in groundwater due to leaching [10], with 0.001–225 μg L^{-1} for thiamethoxam in agricultural surface water. Sorption and desorption are basic processes for determining the leaching behavior of thiamethoxam. The sorption of thiamethoxam is governed mainly by soil organic carbon (OC) content and, to a smaller extent, by dissolved OC, soil textural composition and temperature. Adsorption increased with temperature and dissolved OC can compete with thiamethoxam for binding sites on soil OC. Both hydrophobic partition to OC and specific interactions like hydrogen bonds are main driving force for the adsorption of polar pesticides. However, the contributions of these two mechanisms to the sorption of thiamethoxam are not well elucidated. In contrast to sorption, desorption usually does not proceed reversibly, resulting in desorption hysteresis. Desorption hysteresis is a key factor in retarding the leaching and bioavailability of a chemical. However, until now, studies on the desorption of thiamethoxam are fairly scarce [10]. Once in the soil, neonicotinoids may partition between aqueous and solid phases, depending on their properties and those of the soil. The extent of partitioning may have an influence on their bioavailability, mobility, leaching and degradation in soils. Thus, sorption may play a key role in determining the fate, persistence and behavior of applied neonicotinoids in soils [11]. Dissimilar and often conflicting results have been observed, even for one type of neonicotinoid insecticide. This is due to the multiplicity of factors at play and their complex relations to soils and chemical behavior. For instance, while organic matter has been reported to be important in the sorption behavior of a number of neonicotinoids, other reports have found no correlation between neonicotinoid sorption and organic matter content. At the same time, other factors including clay minerals, temperature and pH have been reported as important in the sorption behavior of neonicotinoids. Due to the high water solubility of neonicotinoids, concerns for their potential mobility and leaching into surface and underground water systems have dominated many studies of their sorption behavior. Several studies have reported low sorption coefficients for neonicotinoids in soils, suggesting a high possibility for movement through runoff and leaching into surface and underground water [11]. The global rise in agricultural activity is related to population growth that leads to an increased need for food. Intensive farming predominates in the global agricultural system, aiming to increase productivity and reduce production time. It is necessary to use agricultural inputs to increase soil fertility or combat the spread of pests and diseases in crops. Among the various inputs used in agricultural fields, pesticides have stood out due to their efficiency in controlling weeds, insect infestations, and various pests that inhibit crop development. However, pesticides can cause severe environmental damage [12]. Thiamethoxam was introduced to the agricultural market in the early 2000s and is widely used for crops such as corn and soybeans [13]. The fate of pesticide residues in soils depended largely on the environmental behaviors including leaching, degradation, bioaccumulation, adsorption, and desorption. Among them, adsorption and desorption behaviors played an important role for evaluating the fate and bioavailability of pesticides in soils. Therefore, the adsorption and desorption behaviors of pesticides in soil are receiving an increasing interest [14]. Thiamethoxam is a second-generation neonicotinoid insecticide that is frequently applied to prevent a variety of pests for crops, such as aphids and whiteflies. The residues of thiamethoxam have frequently been detected in agricultural soils [15]. Some studies on the sorption behavior of thiamethoxam have been investigated but most focused on their adsorption behavior. The sorption behaviors of pesticides may be affected by soil physicochemical properties. However, little is available on the sorption behaviors of thiamethoxam and the main influence factors in different soils. Therefore, the adsorption and desorption of thiamethoxam in different soils were investigated to reveal their sorption characteristics and their correlations with soil physicochemical properties. In this study, batch adsorption and desorption experiments of thiamethoxam were conducted in the six different agricultural soils at 25 & 50ºC. The objectives of this study are to determine the adsorption and desorption behaviors of thiamethoxam in different soils and to reveal the correlation between the sorption affinity and soil physicochemical properties. This study will be useful for evaluating the fate of pesticides and soil ecological risks due to the substantial application of pesticides in agricultural soils. The data are important for evaluating the environmental impacts of the use of thiamethoxam and for future field studies.

Experimental

Materials

Thiamethoxam

IUPAC name: 3-[(2-Chloro-1,3-thiazol-5-yl) methyl]-5-methyl-1,3,5-oxadiazinan-4-ylidene nitramide, Trade name: Champ, Chemical class: Neonicotinoid, Molecular formula: $C_8H_{10}ClN_5O_3S$, Molecular weight: 291.71, Activity: Systemic insecticide, Activation: Gets in the way of information transfer between nerve cells by interfering with nicotinic acetylcholine receptors in the central nervous system, Solubility in water: 410 mg L^{-1} (20 °C), Vapor pressure: 6.6x10^{-9} mpa (25 °C), and Rate of application: 20-150 (g.ai ha^{-1}).

Tested soils

The soil samples were collected from the surface layer from different locations in Egypt. The physical and chemical properties were determined at the Department of Soil and Water Sciences, Faculty of Agriculture, University of Alexandria and the data are presented in Tables (1 and 2). Soil samples were air-dried, ground and passed through a 2-mm sieve prior to use [16,17]. The soil texture was determined by the hydrometer method [18]. Soil pH was measured using 0.01 M calcium chloride ($CaCl_2$) in a 1:2 w/w soil: solution slurry. The OM content was determined by dichromate oxidation according to the Walkley-Black method [19].

Table 1

Physical properties of the tested soils

Soil code	Soil type	Particle Size (%)			Texture class
		Clay	Silt	Sand	
A	Alluvial	42	18	40	Clay loam
B		64	24	12	Clay
C	Calcareous	14	11	75	Sandy loam
D		20	13	67	Sandy clay loam
E	Sandy	10	3	87	Sand
F		13	3	84	Loamy Sand

Table 2

Chemical properties of the tested soils

Chemical properties	Alluvial soils		Calcareous soils		Sandy soils	
	A	B	C	D	E	F
EC (ds/m) at 25ºC	1.32	2.06	2.33	5.03	1.18	9.0
Soil pH	8.25	8.22	8.20	8.15	8.51	7.40
Organic matter content (%)	3.31	1.26	1.32	1.54	0.15	0.1
Total carbonate (%)	7.87	15.47	40.09	44.64	4.01	3.76
Soluble cations conc. (meq/L):						
Ca^{++}	3.8	4.0	8.8	18.7	6.0	32.0
Mg^{++}	5.0	3.2	7.0	8.8	2.5	15.0
Na^{+}	9.4	18.1	15.3	22.5	8.3	64.9
K^{+}	0.5	0.4	2.4	0.3	0.6	2.25
Soluble anions conc. (meq/L):						
CO_3^{--}	1.6	0.4	1.6	0.8	0.5	0.0
HCO_3^{-}	2.6	1.5	3.4	4.6	4.0	4.5
Cl^{-}	8.5	16.9	16.5	21.0	7.0	100.0
SO_4^{--}	0.6	1.7	1.8	23.9	6.0	10.0

Methods

Adsorption study

Sorption isotherms by soils were quantified using the batch equilibration technique [20,21]. Experiments were carried out in duplicate with a sorbent mass to thiamethoxam solution ratio of 2:10 for soils. Initial thiamethoxam concentrations of 0.5-50 µg mL^{-1} range were prepared in 0.01 M $CaCl_2$. The thiamethoxam solutions were equilibrated with soil and different in 50-mL polypropylene centrifuge tubes. The tubes were shaken mechanically at 150 rpm at 25&50 °C for a time period to achieve equilibrium based on its kinetic study and centrifuged at 4000 rpm for 15 min. The thiamethoxam concentration in supernatants was determined by spectrophotometer at 256 nm [2]. The amount of pesticide sorbed, Cs, by solid phase after equilibrium was calculated according to [22].

$$q_s = (C_i - C_s) * \frac{V}{M_s}$$

Where q_s is the quantity of pesticide sorbed per mass unit of adsorbent (µg g^{-1}), C_i is the initial concentration of pesticide (µg mL^{-1}), C_e is the equilibrium concentration of the pesticide per mass unit of solution (µg mL^{-1}), V is the volume of added solution (mL) and M_s is the weight of the adsorbent sample (g).

Desorption study

Desorption experiments were conducted immediately after the sorption experiments for all concentrations using parallel system. Following the sorption experiment using a decant refill technique. The background solution was added to each tube for desorption equilibrium step (24 hr). Tubes were shaken to establish a new desorption equilibrium, centrifuged and the liquid phase containing desorbed thiamethoxam was analyzed [4].

Effect of temperature

The tested pesticide adsorption-desorption enthalpy on soils was determined using the batch experiments as described above. The adsorption process was performed at different temperatures (25&50°C). Thermodynamic parameters are calculated from the variation of the thermodynamic equilibrium constant K_o with changes in temperature. Values of K_o are obtained by plotting $\ln(C_s/C_e)$ versus C_s and extrapolating to zero C_s as described by [20]. The standard free energy change (ΔG°) for the interaction was calculated from the relationship;

$$\Delta G^o = -RT \ln K_o$$

Where R is the universal gas constant (8.314 J mol^{-1} K^{-1}), T is temperature in Kelvin. The negative ΔG indicates that the adsorption of thiamethoxam in soil is spontaneous at different temperatures. The standard enthalpy changes (ΔH°) will be calculated from the Van’t Hoff isochore equation:

$$ln\left[\frac{K_{oT2}}{K_{oT1}}\right] = \left[\frac{-\Delta H^o}{R}\right]\left[\frac{1}{T_2} - \frac{1}{T_1}\right]$$

Negative values of the standard enthalpies changes (ΔH°) indicate that pesticide and soil interactions are exothermic and products are energetically stable with high binding of pesticide to soil sites [23].

Freundlich equation

The empirical formula of the Freundlich equation can be written as;

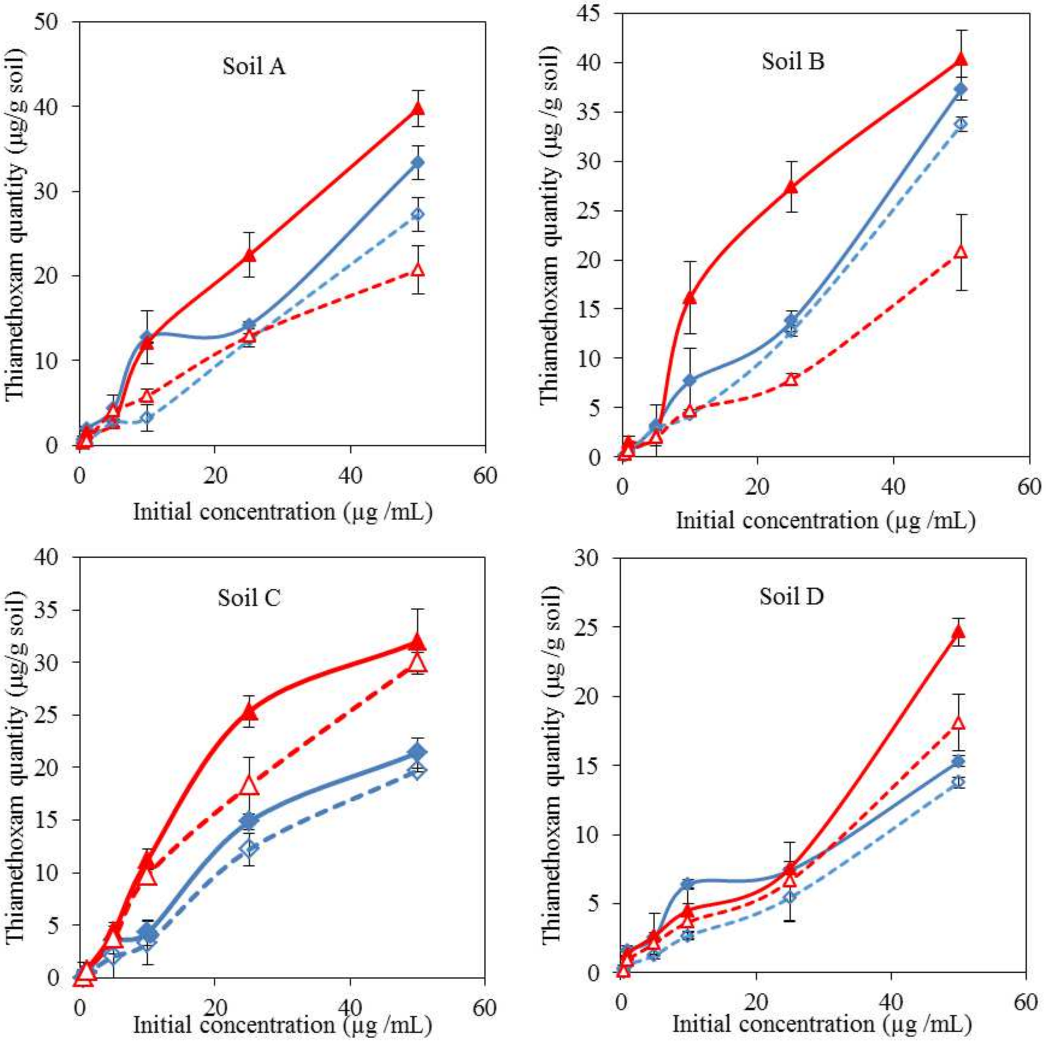
$$log\ q_e = \frac{1}{n}\ logC_e + logK_F$$

Where K_F is a constant indicative of the adsorbent ($mg^{1-(1/n)}$ $L^{-1/n}$ g^{-1}) and 1/n is a constant indicative of the intensity of the adsorption [21,23].

3. Results and discussion

Adsorption-desorption of thiamethoxam in soils at temperature 25&50 °C

Temperature and soil type are an important parameter that can influence the rates and equilibria of different environmental processes [20,23]. Therefore, the adsorption-desorption isotherms of thiamethoxam in clay loam, clay, sandy loam, sandy clay loam, sand and loamy sand soils at 25&50 ˚C was studied. The amount of thiamethoxam adsorbed, desorbed and non-desorbed by the soils (Figures 1 and 2) was significantly influenced by the temperature. At 50 ˚C, the average of the adsorbed of thiamethoxam on soils were 13.129 µg g $soil^{-1}$ for clay loam soil, 14.611 µg g $soil^{-1}$ for clay soil, 12.305 µg g $soil^{-1}$ for sandy loam soil, 6.812 µg g $soil^{-1}$ for sandy clay loam soil, and 6.943 µg g $soil^{-1}$ for loamy sand soil. However, the adsorbed amount was reduced to 11.238, 10.450, 7.430, 5.578, and 6.832 µg g $soil^{-1}$ for clay loam, clay, sandy loam, sandy clay loam, sand and loamy sand soils as the temperature was decreased to 25˚C, except for sand soil. Also, the desorbed amount form soil was decreased from 7.706 to 7.411 µg g soil^{-1} for clay loam soil, 9.041 to 6.027 µg g soil^{-1} for clay soil, 4.689 to 4.250 µg g soil^{-1} for loamy sand soil as the temperature was increased from 25˚C to 50˚C, while the opposite in sandy loam, sandy clay loam and sand soils. The Kd values of adsorption and desorption at 25&50 °C were 0.904, 3.127, 1.049 and 2.995 for clay loam soil, 0.769, 2.732, 1.130 and 2.853 for clay soil, 0.513, 3.117, 0.916 and 3.031 for sandy loam soil, 0.405, 3.305, 0.495 and 3.230 for sandy clay loam soil, 0.494, 2.669, 0.357 and 4.000 for sand soil, and 0.447, 3.102, 0.499 and 3.134 for loamy sand soil. There is a general consensus that the magnitude of Kd values usually indicates the affinity of the compound to the adsorbent matrix [24].



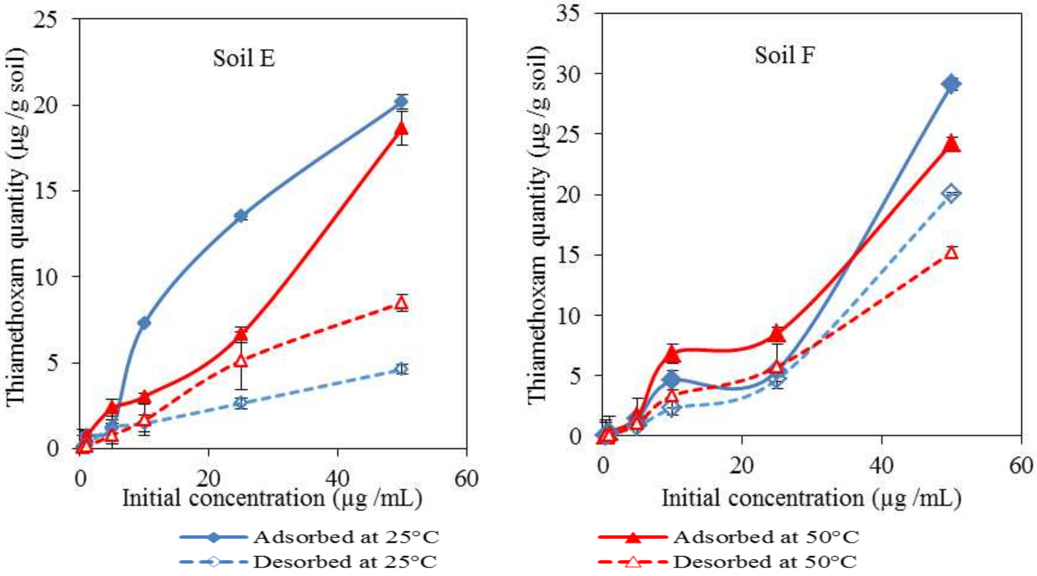


Figure 1: Adsorption-desorption isotherm of thiamethoxam in soils at 25&50 °C

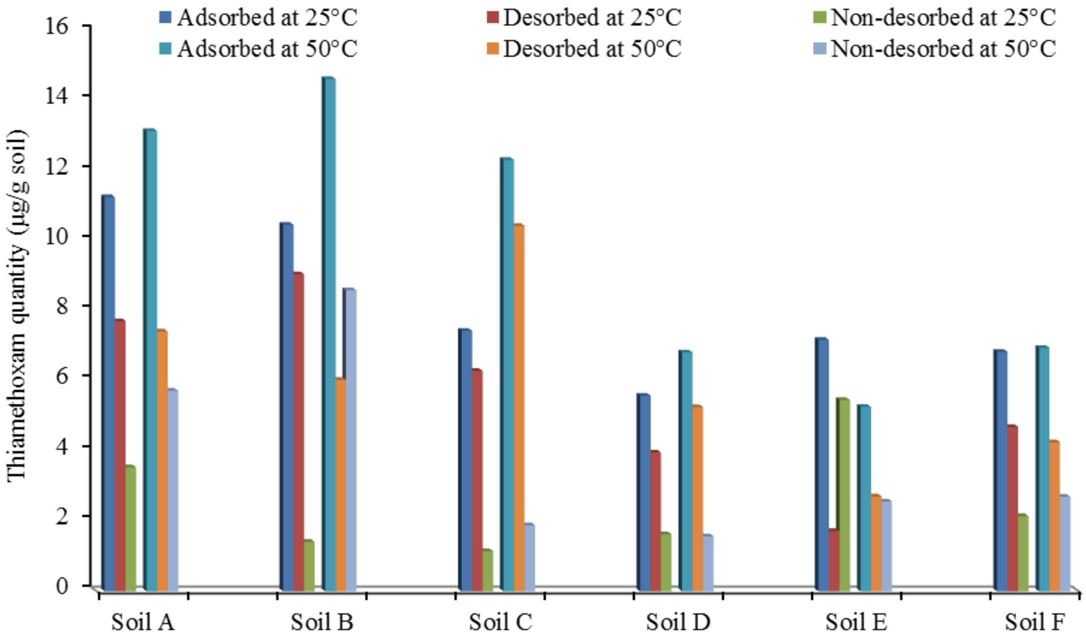
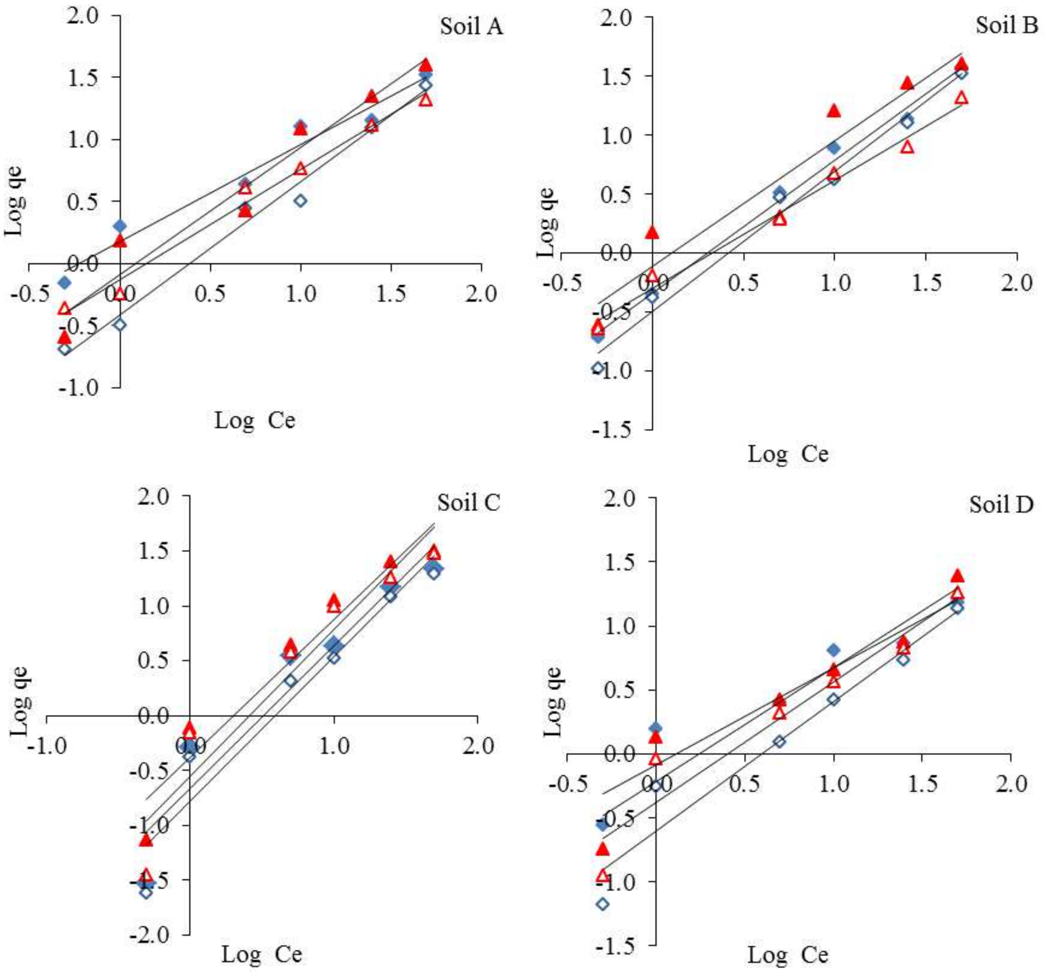


Figure 2. Average of adsorbed, desorbed and non-desorbed tested thiamethoxam (µg/g sorbent) at 25&50 ℃ in soils

Freundlich equation

The data from the adsorption and desorption fate of thiamethoxam in soils at different temperatures corresponded will with the Freundlich isotherm (Figure 3). The values of Freundlich adsorption coefficient (K_F), the Freundlich adsorption exponent (1/n) and correlation coefficient (R^2) for adsorption and desorption of thiamethoxam in soils are presented in Table (3). The value of K_F in adsorption for clay loam and sandy clay loam soil is greater at 25˚C than that at 50˚C and the opposite in desorption, indicate the soil has a higher adsorption capacity at 25°C than that at 50°C. The value of Freundlich adsorption and desorption coefficient K_F for clay, sandy loam, and loamy sand soil are higher at 50˚C than that at 25˚C, indicate that this soils has a higher adsorption capacity for thiamethoxam at 50˚C than at 25˚C. The 1/n values in the case low than unity (1/n < 1), are indicative of adsorption by heterogeneous media where high energy sites are occupied first, followed by adsorption at lower energy sites. Whereas the 1/n values were more than unity (> 1), indicaating relative increased adsorption of insecticide with increasing initial concentration. The correlation coefficient ($R^2 = 0.909\text{-}0.996$), indicating that the Freundlich model was the best fist for thiamethoxam sorption in soils. These results are agreement with those obtained for other pesticides by [20,22,25,26,27,28,29].



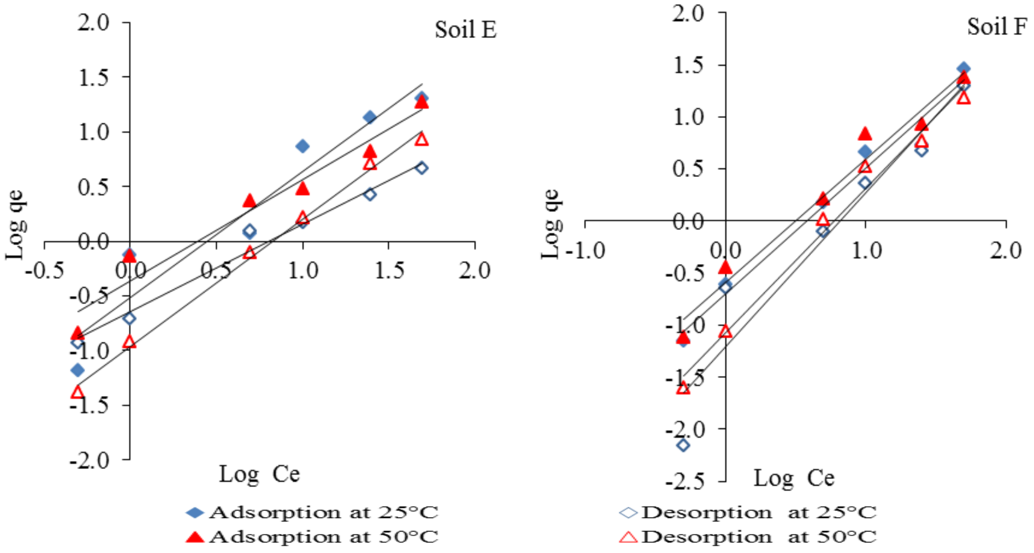


Figure 3. Adsorption-desorption isotherm of thiamethoxamin soils at 25&50 °C fitted in Freundlich equation

Table 3.

Freundlich parameters of adsorption-desorption isotherm of thiamethoxam in soils at 25&50 °C

Soils	K_f		1/n		R^2	
	25°C	50°C	25°C	50°C	25°C	50°C
	Adsorption					
A	1.496	0.807	0.781	1.026	0.965	0.949
B	0.460	0.771	1.127	1.068	0.994	0.921
C	0.215	0.416	1.304	1.258	0.913	0.937
D	0.829	0.615	0.756	0.890	0.911	0.917
E	0.303	0.428	1.153	0.929	0.915	0.956
F	0.202	0.259	1.210	1.186	0.975	0.971
	Desorption					
A	0.380	0.740	1.072	0.884	0.988	0.987
B	0.322	0.501	1.185	0.920	0.987	0.988
C	0.164	0.280	1.319	1.337	0.928	0.909
D	0.253	0.418	1.011	0.944	0.939	0.927
E	0.226	0.109	0.801	1.161	0.978	0.996
F	0.063	0.086	1.481	1.386	0.915	0.984

Thermodynamic parameters of thiamethoxam

The thermodynamic parameters for adsorption and desorption isotherm of thiamethoxam for soils summarized in Table (4). The values of the standard free energy changes (ΔG˚) were negative values. This indicates that the adsorption and desorption of thiamethoxam in soils are spontaneous with a high affinity for thiamethoxam. It also suggests a high persistence and resistance to degradation of thiamethoxam. The same comment was reported by [23]. The standard enthalpy change (ΔH˚) of adsorption in clay, sandy clay loam, sand and loamy sand soil was negative value, indicates the thiamethoxam interaction with clay soil is exothermic and the products are energetically stable with a high binding of thiamethoxam to the soil sites, and except for clay loam and sandy loam soil. Moreover, the standard entropy change (ΔS˚) was negative value in soil for adsorption and desorption isotherms of tested pesticide.

Table 4

Thermodynamic parameters for adsorption and desorption isotherms of thiamethoxam in soils

Thermodynamic parameters	Soil A		Soil B		Soil C		Soil D		Soil E		Soil F	
	25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C	25°C	50°C
	Adsorption											
K_o	3675	400.27	2060.4	2649.8	10414	6378.3	6778.3	7334.9	7932.3	43082	9586.5	9399.9
ΔG°	-20339.2	-16091.4	-18905.5	-21167.2	-22919.8	-23526.1	-21855.9	-23901.4	-22245.4	-28655.8	-22714.7	-24567.5
ΔH°	70972.093		-8053.263		15692.998		-2863.799		-54166.503		-2510.002	
ΔS°	-306.414		-36.417		-129.573		-5527.494		107.118		-67.801	
	Desorption											
K_o	565.98	372.63	11979	21263	22636	15052	1135.3	2238.3	93683	13530	21533	1842.8
ΔG°	-15704.3	-15899.3	-23266.7	-26759.5	-24843.4	-25831.8	-17428.9	-20713.9	-28362.4	-25545.6	-24719.6	-20191.8
ΔH°	13379.405		-18367.898		13061.156		-5527.494		61940.029		6476.285	
ΔS°	-269.546		-40.600		-121.421		-65.132		78.981		-68.289	

4. Conclusion

In general, the adsorption was at 50 °C higher in all soils except sand soil. The desorption was higher at 25 °C in clay loam, clay, and loamy sand soil, while it was higher in sandy loam, sandy clay loam and sand soil at 50 °C. The non-desorbed amount was higher at 50 °C in all soils except for sandy clay loam soil. Adsorption order, clay loam soil > clay soil > sandy loam soil > sand soil > loamy sand soil > sandy clay loam soil at 25 °C; clay soil > clay loam soil > sandy loam soil > loamy sand soil > sandy clay loam soil > sand soil at 50 °C. Freundlich model was the best fit for thiamethoxamin adsorption and desorption in all soils at 25&50 °C.

5. Conflicts of interest

There are no conflicts to declare.

6. Formatting of funding sources

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