

# Equations Adsorption Isotherms for Biuret on Soils, Paper and Cortex Plant Application of the Freundlich, Langmuir, Temkin, Elovich, Flory-Huggins, Halsey, and Harkins-Jura

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**Abstract:** Adsorption is considered to be one of the most effective technologies widely used in global environmental protection areas. Modeling of experimental adsorption isotherm data is an essential way for predicting the mechanisms of adsorption, which will lead to an improvement in the area of adsorption science. In this paper, we employed seven isotherm models, namely: Freundlich, Langmuir, Elovich, Temkin, Harkins-Jura, Halsey, and Flory-Huggins to correlate seven sets of experimental adsorption isotherm data, which were obtained by batch tests in laboratory. In order to determine the best fit isotherm model, the correlation coefficient ( $R^2$ ) for each parameter was used to evaluate the data. The kinetics study for adsorption processes were found to verify the first order rate law. Thermodynamic parameters ( $\Delta G^\circ$ ,  $\Delta H^\circ$  and  $\Delta S^\circ$ ) were also calculated, indicating the endothermic processes.

**Keywords:** biuret, adsorption isotherm, thermodynamic parameters.

## 1. INTRODUCTION

The main advantages of adsorption process are low operating cost, easily available sorbent sources, minimization of the volume of sludge to be disposed of, recovery of the metal from the adsorbents and regeneration of the adsorbents for reuse [1] and is recognized as the most efficient, promising and widely used fundamental approach in wastewater treatment processes [2]. Adsorption is well-established technique for the removal of low concentrations of organic pollutants from large volumes of potable water, process effluents, wastewater, and aqueous solutions. A number of natural and synthetic adsorbents have been studied by various researchers for the removal of heavy metal ions [1]. The performance of an adsorbent can be studied by adsorption isotherm data, which can be obtained by a series of experimental tests in labs. Modeling the adsorption isotherm data is an essential way for predicting and comparing the adsorption performance, which is critical for optimization of the adsorption mechanism pathways, expression of the adsorbents capacities, and effective design of the adsorption systems [3].

Biuret ( $C_2H_5N_3O_2$ ) is a known contaminant of urea fertilizers. Biuret is formed from the combining of two molecules of urea with one molecule of ammonia released during the pilling or granulation of crystalline urea when it is heated near its melting point ( $132.8^\circ C$ ). Pure biuret contains ( $g\ kg^{-1}$ ) C 233, N 408, O 300, and H 49. It has been shown that biuret is both stimulatory and inhibitory to plant growth. In agriculture, high concentrations of pure biuret or biuret in urea fertilizer can be toxic to crops whether applied to soil or foliage. Biuret might stimulate growth at low concentration through acting as a plant growth regulator and/or soil microbial activity regulator, at higher concentrations as a slow release N fertilizer and be inhibitory and ultimately toxic at even higher concentrations. However, little information is available on the mechanisms of stimulation of tree growth by biuret. We hypothesized that stimulation by biuret of tree growth could be a direct metabolically regulated plant response or the results of enhancing soil nutrients, especially N availability through microbially mediated soil processes, or both. These potential properties of biuret could make it an attractive alternative N fertilizer to traditional N [4].

Soil is a heterogeneous mixture of several components, many of which are organic and inorganic compounds of varying compositions and surface activities [5].

The soil properties on which the biuret-exposed crop is grown are important in determining potential toxicity. Biuret is not retained in soil and is easily leached. Plants are generally less sensitive to biuret when it is applied to soils containing appreciable amounts of clay or organic matter, or of low pH. The soil properties on which the biuret-exposed crop is grown are important in determining potential toxicity. [6].

## 2. SOILS

### 2.1. Materials and Methods

Fresh soil samples were taken from four soil samples, Collected from four main agricultural fields, representing a range of physico-chemical properties, and two samples represent paper of plant, and cortex of plant. Subsamples of homogenized soils were analyzed for moisture content, organic matter content, particle size distribution, texture, pH, loss on ignition and exchangeable basic cations. The details were characterized in one of our previous articles [7]

### 2.2. Adsorption Experiments

Adsorption of the biuret from aqueous solution was determined at temperature ( $25 \pm 1$  °C) employing a standard batch equilibrium method. An aqueous stock solution of biuret of  $100 \text{ mgL}^{-1}$  was prepared by diluting  $1 \mu\text{l}$  in 100 ml de-ionized water [8]. The stock and working solution were stored. Duplicate air-dried soil samples were equilibrated with different biuret concentrations (5, 10, 15, and  $20 \mu\text{g ml}^{-1}$ ) at the soil solution ratio 1:10. The samples plus blanks (no biuret) and control (no soil) were thermostated and placed in shaker for 0.5, 1, 2, 3, 3, 4, 6 and 24h. The tubes were centrifuged for 20 min. at 3000 rpm. One ml of the clear supernatant was removed and analyzed for the biuret concentration. Biuret identification was done by UV, at wavelength 280 nm. To study the effect of temperature, the same experiments were done at temperatures ( $25, 35, 45 \pm 1$  °C) employing a standard batch equilibrium method [9].

## 3. RESULTS AND DISCUSSION

To have an insight into the adsorption behavior of biuret on the soils, paper and cortex plant samples, and to gain the optimal fitting of theoretical model, the experimental data from batch experiment were analyzed using Distribution Coefficient and seven parameter isotherm equations, Freundlich, Langmuir, Temkin, Elovich, Flory-Huggins, Halsey, and Harkins-Jura, in which linear regression analysis was used to evaluate whether the theoretical models have better or worse fit for the experimental data [10]. The respective parameters of these isotherm models have been enumerated in (Table 1, and 2).

### 3.1. Linear Adsorption Coefficient (Distribution Coefficient)

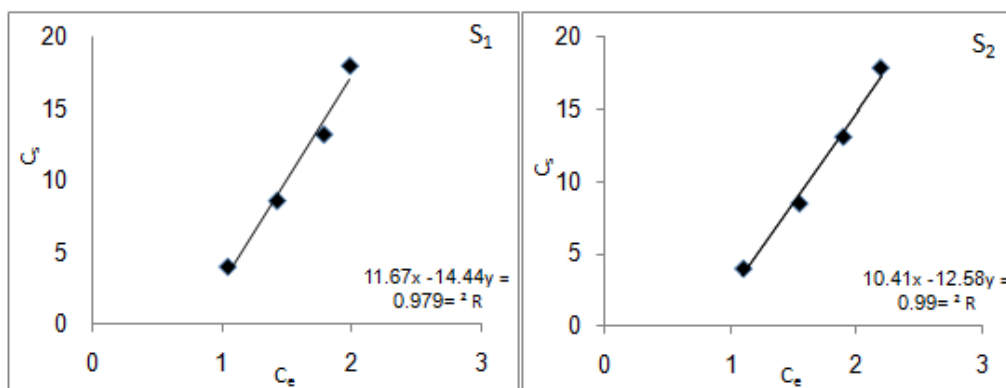
The distribution coefficient ( $K_d$ ) was calculated using the equation (1) [11-12].

$$C_s = K_d \cdot C_e \quad (1)$$

The distribution coefficient ( $K_d$ ) was calculated by taking the ratio of adsorption concentration in soil ( $C_s$ ) and equilibrium concentration in solution ( $C_e$ ). The results were summarized in (Table 1) and Figure (1).

**Table 1.** Adsorption distribution coefficients for biuret adsorption on soils, paper and cortex plant

Parameters	S <sub>1</sub>	S <sub>2</sub>	S <sub>3</sub>	S <sub>4</sub>	Paper	cortex
$K_d$	14.446	12.581	10.905	21.691	14.666	32.752
$R^2$	0.9799	0.9900	0.9438	0.9636	0.9816	0.9205



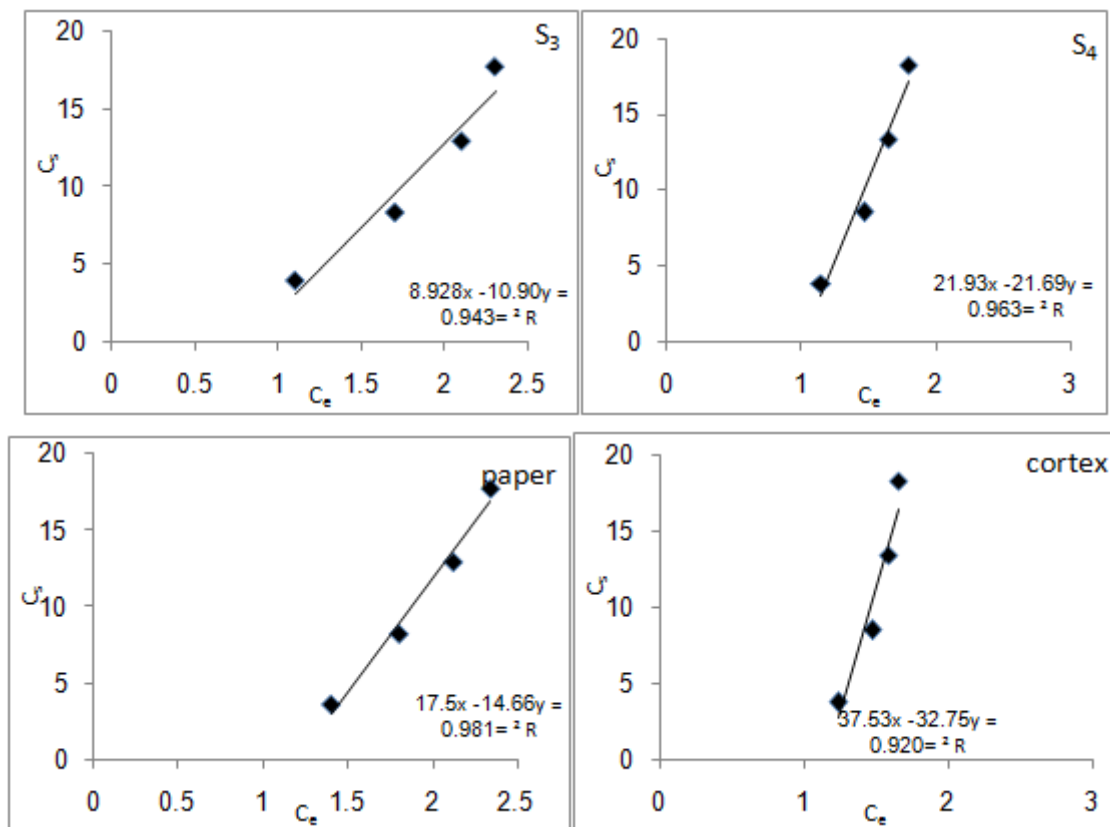


Figure 1. Distribution coefficients for biuret on soils, paper of plant, and cortex of plant

### 3.2. Adsorption Isotherms

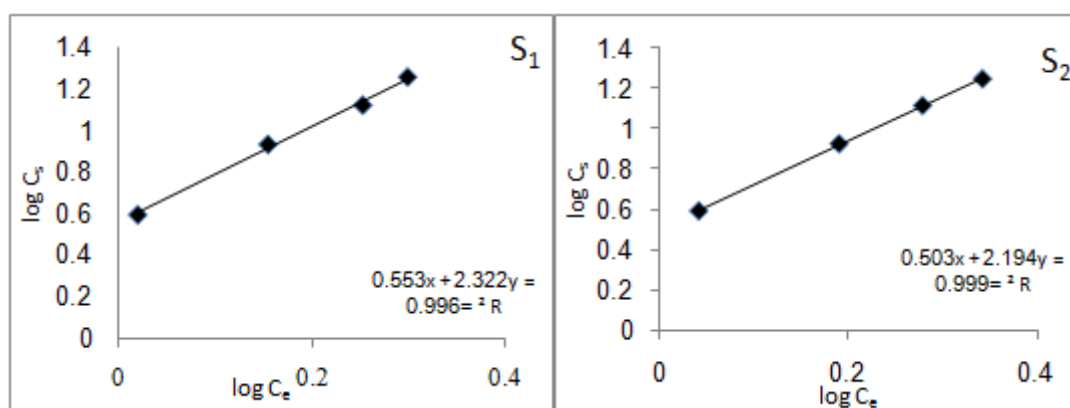
#### 3.2.1. Freundlich Adsorption Isotherm

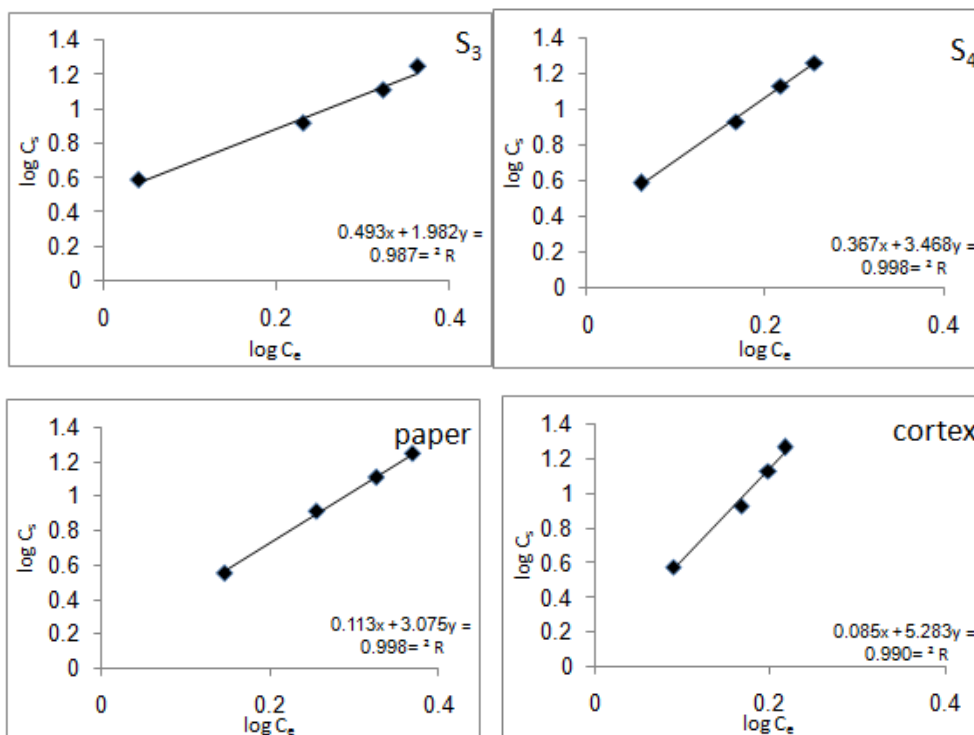
Adsorption isotherm parameters were calculated using the linearized form of Freundlich Equation (2) [13]

$$\log C_s = \log K_F + 1/n \log C_e \quad (2)$$

$C_s$  and  $C_e$  were defined previously,  $K_F$  is Freundlich adsorption coefficients, and  $n$  is a linearity factor, and it is also known as adsorption intensity,  $1/n$  is the slope and  $\log K_F$  is the intercept of the straight line resulting from the plot of  $\log C_s$  versus  $\log C_e$ . The values of  $K_F$  and  $n$  calculated from this regression equation showed that Freundlich adsorption model effectively describes isotherms for biuret in all cases. The results were summarized in (Table 2).

The plotting according to Freundlich isotherm model investigate the homogenous linearly of adsorbent with sorbent with  $R^2 = 0.9873-0.9997$  Figure (2) that is very close to survey results.





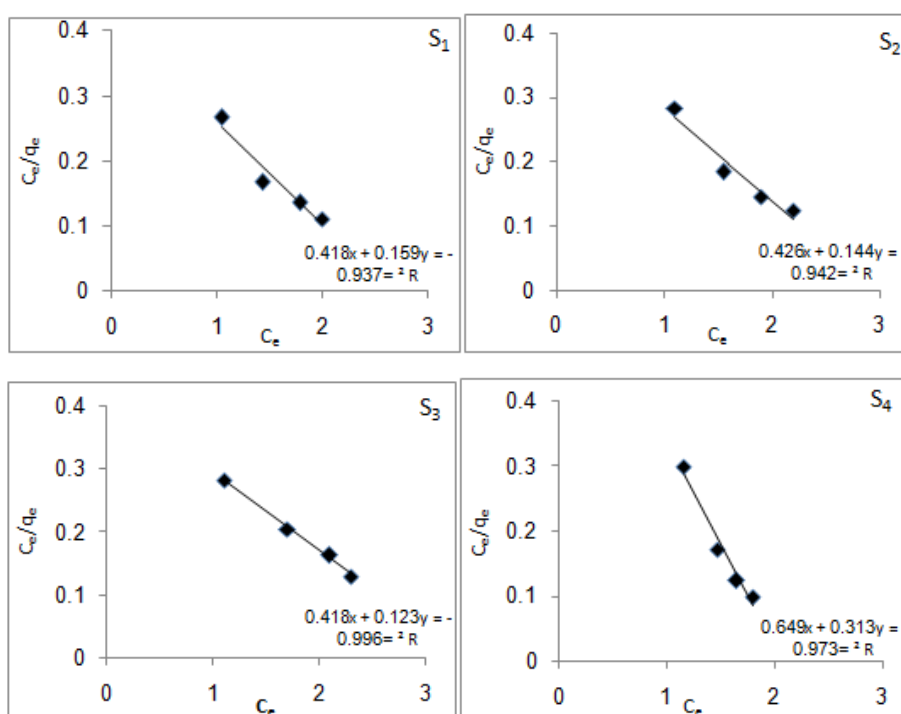
**Figure 2.** Freundlich isotherm of biuret on soils, paper of plant, and cortex of plant

### 3.2.2. Langmuir Adsorption Isotherm

The Langmuir adsorption model is the most common model used to quantify the amount of adsorbate on an adsorbent as a function of partial pressure or concentration at a given temperature. This equation expressed by relation.

$$\frac{C_e}{q_e} = \frac{1}{C_m} + \frac{1}{C_m K_l C_e} \quad (3)$$

in this equation,  $q_e$  ( $\text{mg} \cdot \text{g}^{-1}$ ) is the solution adsorbed by the surface and  $q_e$  is equilibrium constant of adsorption and  $C_m$  is the capacity of adsorption in saturated single layer and  $C_e$  ( $\text{mg} \cdot \text{l}^{-1}$ ) is solution in equilibrium state Figure(3), value  $R^2=0.9378-0.9968$ , and its calculated parameters can be seen in (Table 2) [14].



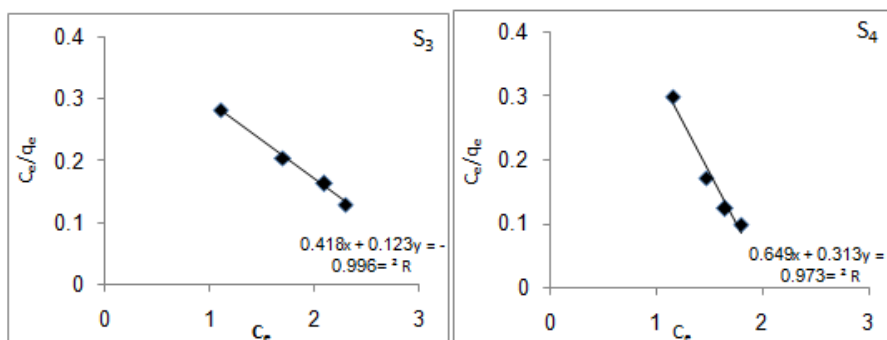


Figure3. Langmuir isotherm of biuret on soils, paper of plant, and cortex of plant

### 3.2.3. Elovich Model

The equation defining the Elovich [15] model is based on a kinetic principle assuming that the adsorption sites increase exponentially with adsorption, which implies a multilayer adsorption. It is expressed by the relation:

$$\frac{q_e}{q_m} = K_E C_e \exp\left(-\frac{q_e}{q_m}\right) \quad (4)$$

where  $K_E$  is the Elovich equilibrium constant ( $L \text{ mg}^{-1}$ )  $q_m$  is the Elovich maximum adsorption capacity ( $\text{mg g}^{-1}$ ). If the adsorption obeys Elovich equation, Elovich maximum adsorption capacity [16] and Elovich constant can be calculated from the slopes and the intercepts of the plot  $\ln(q_e/C_e)$  versus  $q_e$  (Table 2), coefficient of determination ( $R^2=0.9476-0.9885$  Figure (4))

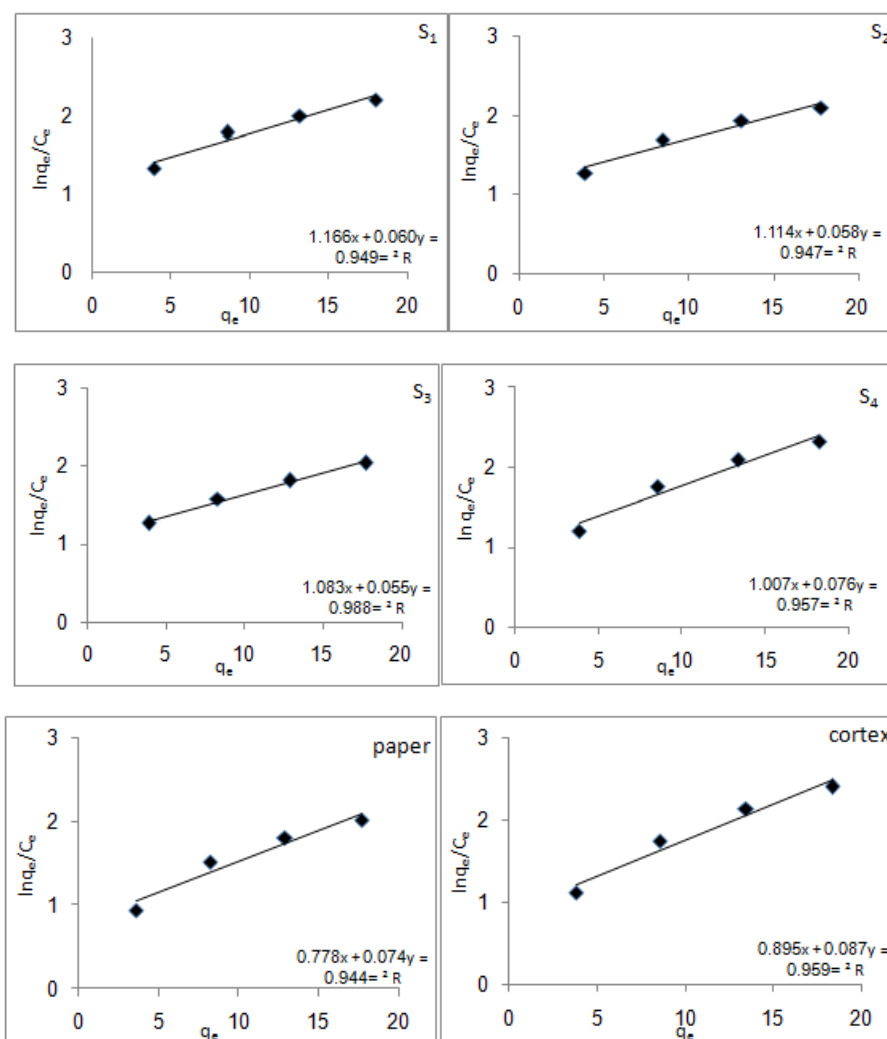


Figure4. Elovich isotherm of biuret on soils, paper of plant, and cortex of plant

### 3.2.4. Temkin Model

The Temkin isotherm model assumes that the adsorption energy decreases linearly with the surface coverage due to adsorbent–adsorbate interactions. The linear form of Temkin isotherm model is given by the equation: [17].

$$q_e = B \ln K_T + B \ln C_e \quad (5)$$

Where A and B are the Temkin isotherm constant (L/g) and heat of sorption (J/mol) respectively. R is the gas constant (J/mol/K), b is the Temkin isotherm constant linked to the energy parameter B, as shown in equation:

$$b = RT/B \quad (6)$$

T is the absolute temperature in kelvin Figure (5) and its calculated parameters are given in (Table 2)[14].

The linear form of this model is shown in Figure (5). As described in Equation (5),  $K_T(1.0475-1.2588 \text{ L}\cdot\text{g}^{-1})$  is the equilibrium binding constant, which indicates the maximum bonding energy;  $B_T(17.102-45.968 \text{ KJ/mol})$  is the constant related to the heat of adsorption, these constants are calculated from the plot  $\ln C_e$  and  $q_e$  and tabulated in (Table 2).

However, the Temkin equation is good for predicting the equilibrium in the gas phase (when arrangement in a tightly packed structure with the same orientation is not required); conversely, the liquid-phase adsorption isotherms (complex adsorption systems) are usually not suitable to be represented [15, 18], based on the coefficient of determination ( $R^2 0.8953-0.9603$ Figure (5)).

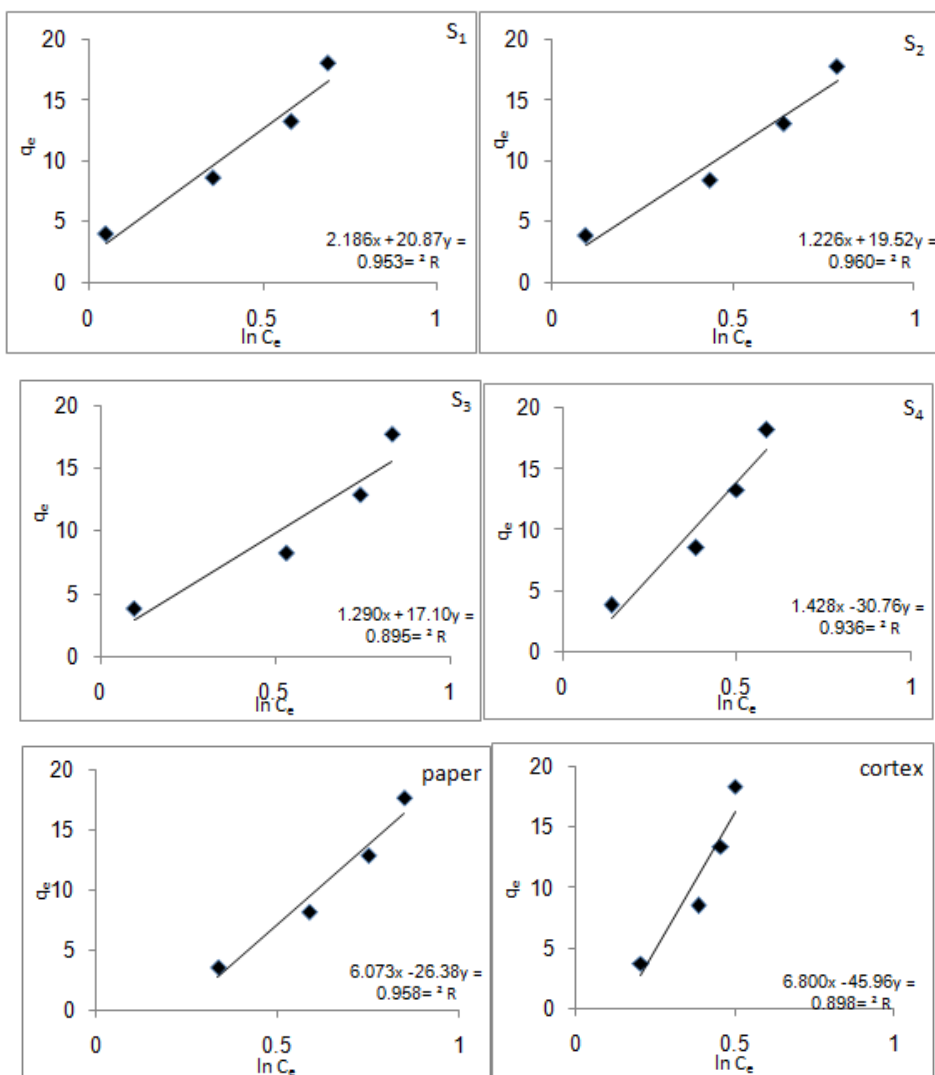


Figure 5. Temkin isotherm of biuret on soils, paper of plant, and cortex of plant

### 3.2.5. Harkins-Jura Isotherm

This isotherm model can be discussed according to the [19] equation 7:

$$\frac{1}{q_e^2} = \frac{B}{A} - \frac{1}{A} \log C_e \quad (7)$$

By inspecting the gained Figure (6) it is obvious that correlation coefficient due to this does not give a good coincidence with experimental data [19].

The Harkins-Jura isotherm model for biuret adsorption onto soil, paper and cortex plant samples are presented in Figure 6 and the relevant isotherm parameters (Table 2) show that the values of  $R^2$  are located in the range of 0.8744-0.9476, which indicate a better fits to the biuret adsorption onto  $S_3$  sample. This result reveals that biuret adsorption onto paper of plant and  $S_3$  samples are in support of the multilayer adsorption rule [20-21]. Values of constants B and A ( $\text{mg} \cdot \text{g}^{-1}$ ) of H-J isotherm model (Equation (7)) were obtained from plot  $1/q_e^2$  vs.  $\log C_e$ . Value B 0.2105-0.3505, where value A (1.8181-5.0000).

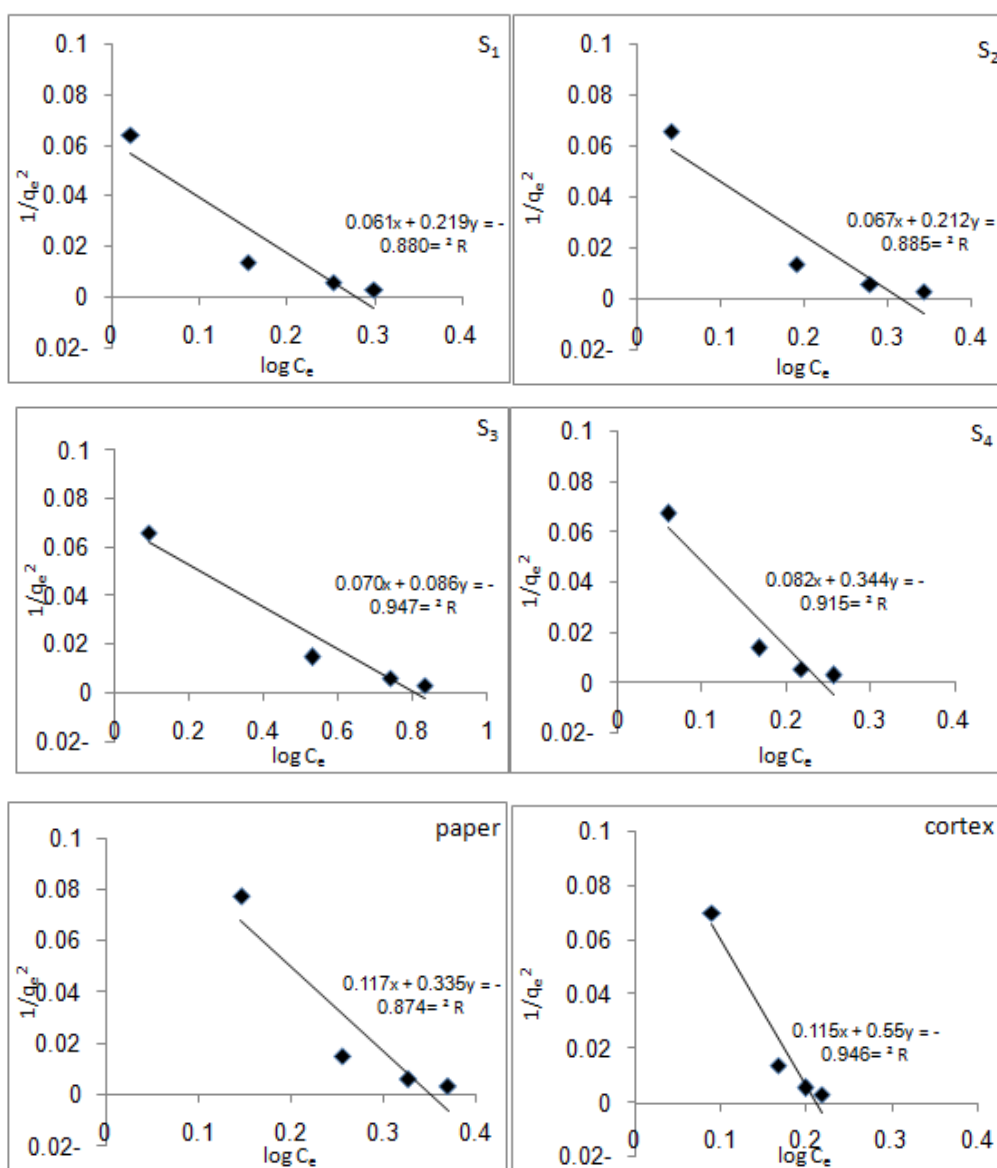


Figure 6. Harkins-Jura isotherm of biuret on soils paper of plant, and cortex of plant

### 3.2.6. Halsey Adsorption Isotherm

The Halsey adsorption isotherm can be given as [22]

$$\ln q_e = \frac{1}{n} \ln K - \frac{1}{n} \ln C_e \quad (8)$$

This equation is suitable for multilayer adsorption and the fitting of the experimental data to this equation attests the heteroporous nature of the adsorbent. The Halsey isotherm parameters are obtained from the plot of  $\ln q_e$  versus  $\ln C_e$  [22]. Figure (7) reflected the relatively better agreement to the adsorption data with  $R^2$ , value of  $R^2$  0.9873-0.9997, value of  $K_H$  1.0378-1.7741, and value of  $n$  0.18925-0.5044.

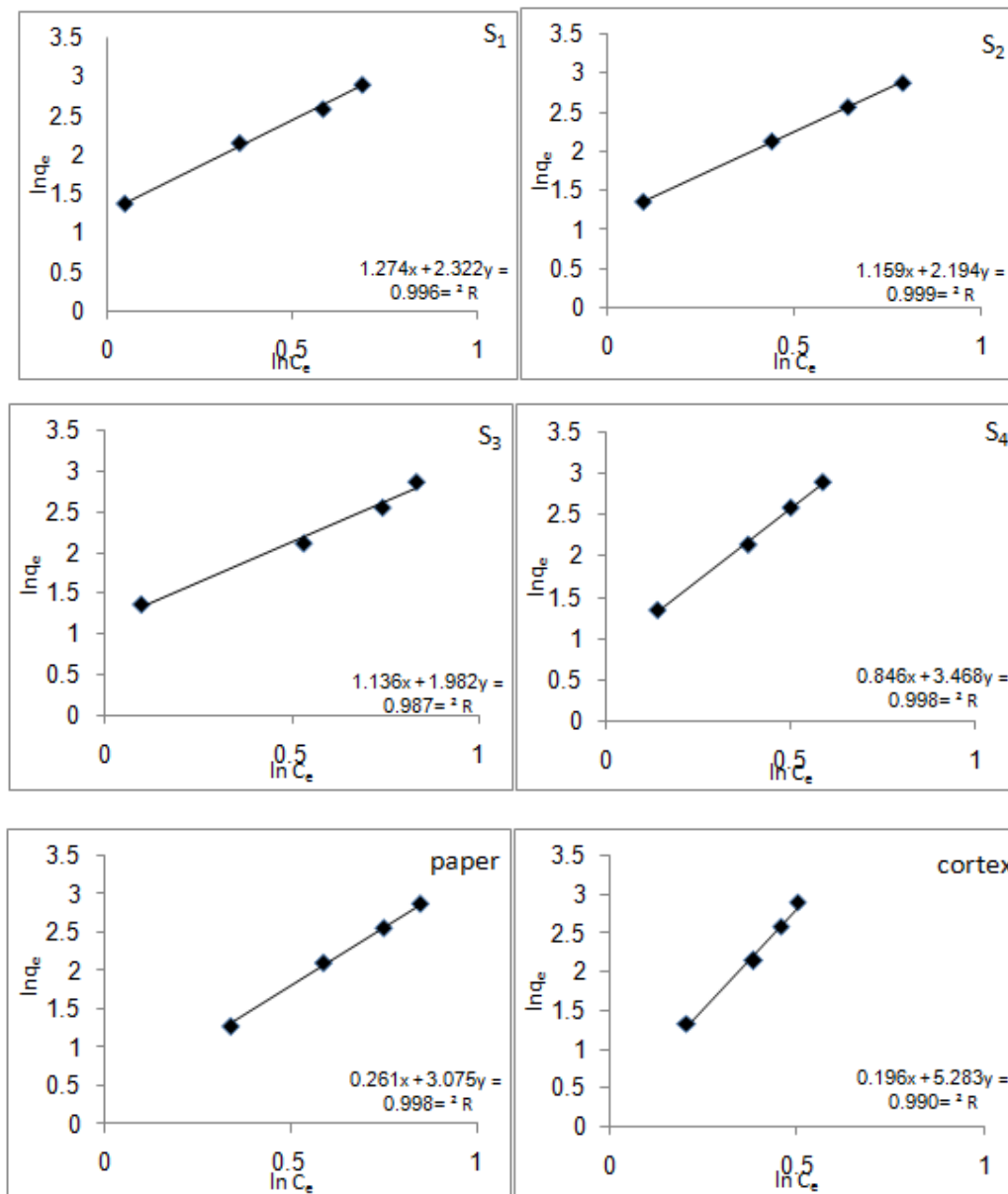


Figure 7. Halsey isotherm of biuret on soils, paper of plant, and cortex of plant

### 3.2.7. Flory-Huggins Adsorption Isotherm

The Flory-Huggins model is represented by the following equation:

$$\ln \frac{\theta}{C_0} = \ln K_{FH} + n \ln(1 - \theta) \quad (9)$$

Where  $\theta$  is the degree of surface coverage,  $n$  is the number of metal ions occupying sorption sites,  $K_{FH}$  is the equilibrium constant of adsorption and  $C_0$  is equilibrium biuret concentration. A plot of  $\log \theta / C_0$  against  $\log (1 - \theta)$  yielding a straight line was made to confirm the model. Calculated parameters can be seen in (Table 2).

The  $\theta$  in equation 10 was calculated from the relation below [23]:

$$\theta = 1 - \frac{C_e}{C_0} \quad (10)$$



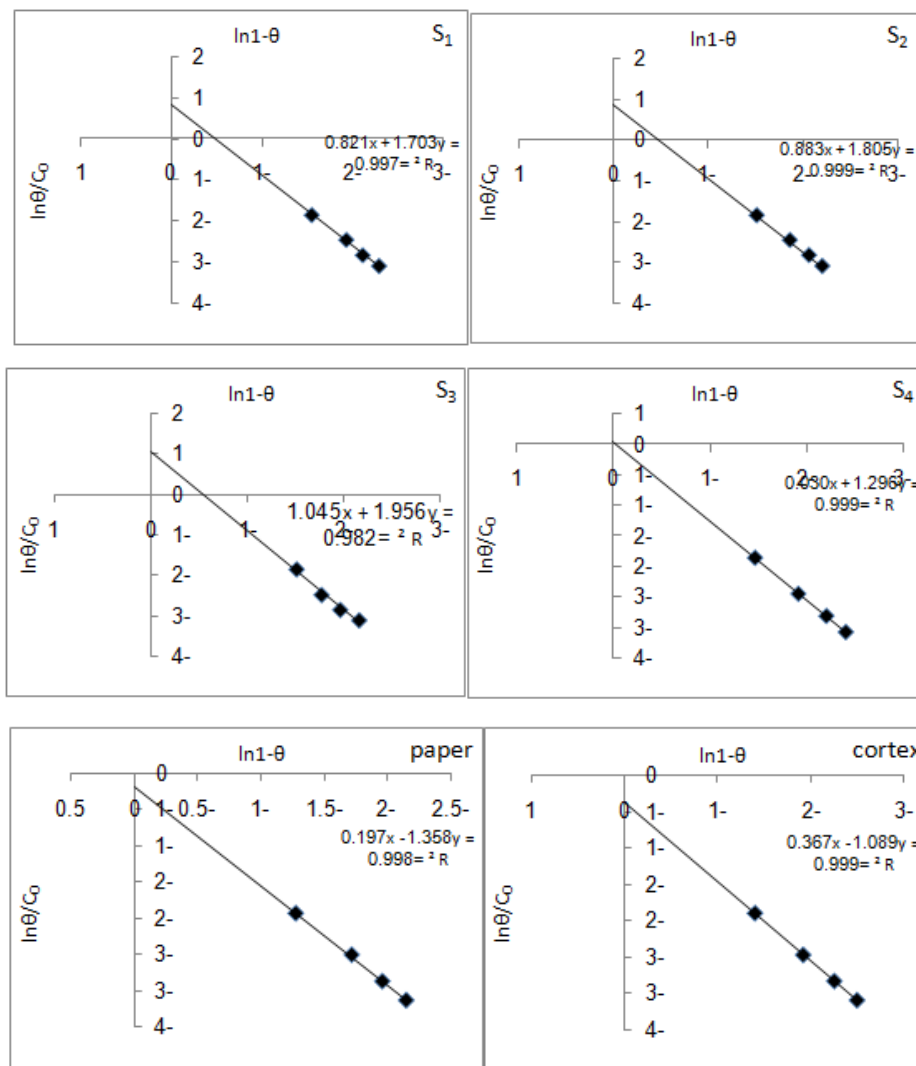


Figure 8. Flory-Huggins isotherm of biuret on soils, paper of plant, and cortex of plant

Table 2. Values of isotherm constants for the adsorption of biuret onto soils, paper of plant, and cortex of plant

models	parameters	S <sub>1</sub>	S <sub>2</sub>	S <sub>3</sub>	S <sub>4</sub>	paper	cortex
Frundlich	K <sub>F</sub>	1.7396	1.6546	1.6380	1.4445	1.1200	1.0889
	n	0.4305	0.4557	0.5044	0.2882	0.3251	0.1892
	R <sup>2</sup>	0.9966	0.9997	0.9873	0.9984	0.9988	0.9901
Langmuir	K <sub>l</sub>	0.3802	0.3375	0.2964	0.4830	0.3632	0.5582
	C <sub>m</sub>	6.2774	6.9444	8.0710	3.1867	3.6995	1.7497
	R <sup>2</sup>	0.9378	0.9429	0.9968	0.9738	0.9369	0.9925
Harkin-Jura	A	4.5662	10.8577	11.5207	6.6934	6.8728	4.1858
	B	0.2808	0.7318	0.8076	0.5535	0.8054	0.4847
	R <sup>2</sup>	0.8806	0.8856	0.9476	0.9156	0.8744	0.9460
Halsey	K <sub>H</sub>	1.7312	1.6963	1.7741	1.2764	1.0886	1.0378
	n	0.4305	0.4557	0.5044	0.2882	0.3251	0.1892
	R <sup>2</sup>	0.9966	0.9997	0.9873	0.9984	0.9988	0.9901
Elovich	K <sub>E</sub>	0.1945	0.1780	0.1637	0.2083	0.1541	0.2136
	q <sub>m</sub>	16.501	17.123	18.0504	13.140	14.134	11.467
	R <sup>2</sup>	0.9491	0.9476	0.9885	0.9573	0.9446	0.9595
Temkin	K <sub>T</sub>	1.1104	1.0648	1.0783	1.0475	1.25887	1.1594
	B <sub>T</sub>	20.870	19.525	17.102	30.766	26.3810	45.968
	R <sup>2</sup>	0.9532	0.9603	0.8953	0.9361	0.9580	0.8988
Flory-Huggins	K <sub>FH</sub>	2.2745	2.4186	2.8442	1.0313	1.2188	1.4441
	n	1.7036	1.8058	1.9562	1.2960	1.3580	1.0892
	R <sup>2</sup>	0.9971	0.9996	0.9821	0.9999	0.9989	0.9999

### 3.3. Thermodynamic Study

To investigate the thermodynamic nature of soils for biuret, different standard parameters  $\Delta G^\circ$  (Gibbs free energy),  $\Delta H^\circ$  (enthalpy) and  $\Delta S^\circ$  (entropy) have been calculated using the equations

$$\Delta H^\circ - T\Delta S^\circ = -RT \ln K_c \quad (11)$$

$$R \ln K_c = \Delta S^\circ - \frac{\Delta H^\circ}{T} \quad (12)$$

Where  $K_c$  is the distribution coefficient constant for adsorption,  $R$  is the gas constant (KJ/mol/K) and  $T$  is the absolute temperature (Kelvin). Based on the Van't Hoff plot of  $\ln K_c$  versus  $1/T$ , the values of  $\Delta H^\circ$  and  $\Delta S^\circ$  were determined from the slope and intercept respectively Figure (9) [24].

The determination of the thermodynamic parameters ( $\Delta H^\circ$ ,  $\Delta S^\circ$  and  $\Delta G^\circ$ ) for biuret as given in (Table 3) can provide mechanism insights into biuret adsorption onto soils, paper and cortex plant. The values of the standard enthalpy change ( $\Delta H^\circ$ ) were positive in the (soils, paper and cortex plant), indicating that it is an endothermic process for biuret adsorption onto soils.

The values of the Gibbs free energy change ( $\Delta G^\circ$ ) were all negative at two temperatures studied herein as expected for a spontaneous process under our experimental conditions. The higher the reaction temperature, the more negative the value of  $\Delta G^\circ$ , indicating that the adsorption reaction is more favorable at elevated temperatures [25].

However, the values of the standard entropy change ( $\Delta S^\circ$ ) in soils were all positive for biuret sorption onto soils, which indicates that during the whole adsorption process, some structural changes occurs on soils surface and thus leading to an increase in the disorderness at the soil- water interface. In addition, whether or not a surface adsorption reaction is an associative or dissociative mechanism, strongly depends on the value of  $\Delta S^\circ$ . When the value of  $\Delta S^\circ$  is higher than  $-10 \text{ J/K mol}^{-1}$ , a dissociative mechanism controls adsorption [25].

The higher values obtained for  $\Delta S^\circ$  in agricultural soil than in undisturbed soil confirmed that agricultural soil has comparatively low sorption capacity for biuret which leads to less  $k_c$  [26].

**Table 3.** Values of ( $\Delta G^\circ$ ,  $\Delta H^\circ$  and  $\Delta S^\circ$ ) for the adsorption of biuret onto soils, paper of plant, and cortex of plant

Sample	T1	T2	T3	$\Delta H^\circ$ kJ/mol	$\Delta S^\circ$ J/K mol	$R^2$
	$\Delta G^\circ$ KJ/mol	$\Delta G^\circ$ KJ/mol	$\Delta G^\circ$ KJ/mol			
S <sub>1</sub>	-232.428	-349.159	-452.183	3041.3	-10.988	0.9987
S <sub>2</sub>	-261.082	-235.255	-690.531	6221.1	-21.472	0.7050
S <sub>3</sub>	-292.328	-600.487	-671.568	5321.7	-18.962	0.8847
S <sub>4</sub>	-174.915	-747.740	-809.579	9201.2	-31.733	0.8223
paper	-243.438	-299.239	-240.191	310.98	0.16230	0.8024
cortex	-140.156	-156.935	-94.9740	826.83	-2.2591	0.8969

## 4. CONCLUSIONS

The results derived from this work indicate that the thermodynamic parameters are related to both the nature of sorbate and the nature of solid particles. The thermodynamic analysis of biuret adsorption indicates that the

surface reaction of biuret adsorption onto soils is an endothermic and spontaneous process. Results of thermodynamic studies revealed that biuret sorption reaction in soils, paper and cortex were less susceptible to biuret toxicity due to obtaining high  $k_d$  values at high temperature (45°C). But at ambient temperature (25°C), agricultural soils were more prone to biuret toxicity than undisturbed soil indicating that such soils will pose more problems of biuret contamination and its toxicity to the plants. Thus soil properties, nature of pollutant and soil environment particularly temperature needs to be considered in the assessment of soil quality. The negative values obtained for  $\Delta S^\circ$  in agricultural soil also confirmed that agricultural soil has high sorption capacity due to high degree of order at solid-solution interface during the sorption of biuret. The findings in this study are quite important to understand the physicochemical behavior of the interested radionuclides in the natural environment. The experimental adsorption data was fitted according to the following order: Freundlich = Halsey > Flory-Huggins > Langmuir > Elovich > Temkin > Harkin-Jura isotherm models.

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