



Impact of Certain Organic Wastes on Some Isotherm Models of Copper and Nickel Adsorption on A Loamy Sand Soil



Atef Abdelaziz Ali Sweed

Department of Soil Science, Faculty of Agriculture, Aswan University, Aswan City, Egypt

A LABORATORY experiment was conducted to study the effect of filter cake and vinasse on copper and nickel adsorption on a loamy sand soil. To attain this aim, five grams of the soil were placed with 0.5 g of filter cake or vinasse in conical flasks. Then, 10 ml of Cu or Ni solutions containing 44 mg Cu or Ni / 10 ml prepared from $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$, respectively, plus 25 ml of distilled water were added to the flasks to give the initial concentration of Cu or Ni of 1257 mgL^{-1} . Two control flasks of the soil plus Cu or Ni and the soil with distilled water were used. The treatments were repeated three times. The soil suspensions in the flasks were shaken at 25°C for five shaking periods of 5, 10, 20, 40 and 60 minutes. The results obtained showed that organic materials resulted in increased efficiency the sorption of Cu and Ni on a loamy sand soil compared to control treatment (without organic materials), and the filter cake was more able to absorb both Cu and Ni compared with vinasse with increased shaking intervals. The highest adsorption amount was 8.53 mg g^{-1} for copper with the non-amended soil at reaction time 60 minute and 4.72 mg g^{-1} for nickel with filter cake + soil treatment at reaction time 20 minute. The results also showed that the Freundlich equation for Cu and Ni absorption was better than the Langmuir equation due to that Langmuir equation is unsuitable of adsorption equations with high concentrations of heavy metals under study.

Keywords: Copper, Nickel, Adsorption, Filter cake, Vinasse and Isotherm models.

Introduction

Organic matter may affect negatively or positively the heavy metal availability as a result of the formation of heavy metal complexes in soils. One of the most important chemical processes which affect the behavior and bioavailability of heavy metals in soils is the adsorption of heavy metals from the liquid phase (soil solution) on the solid phase. This process controls the concentration of metal ions and complexes in the soil solution and thus can exert a major influence on their uptake by plant roots.

The total heavy metal content of the soil is commonly used to indicate the degree of contamination, but the heavy metal concentration in solution mostly determines the actual environmental risk. Also, the key to evaluating the environmental impact of the heavy metals is

the distribution of heavy metals between soil and solute (Temminghoff et al. 1998).

Adsorption of heavy metals in soils involves two mechanisms. One is non-selective adsorption, in which the metallic cations act as counter-ions in the diffuse layer and the other is selective adsorption, in which surface complexes are formed. The importance of each of these mechanisms depends on the metal and the type of soil (Lafuente et al. 2008). The study of adsorption of heavy metals in the soil is an important evaluation tool of the polluting potential of these elements (Dere et al., 2006). To describe the metal adsorption forms in the soil, physicochemistry models are utilized, and several equations are proposed to fit the equilibrium solution data (Moreira and Alleoni, 2010). The most important parameters controlling heavy metal adsorption

* Corresponding author: atefsweed@agr.aswu.edu.eg

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and their distribution between soil and water are solid: solution mass ratio, and contact time (Kent et al., 2000). Several authors have reported that the soil attributes that are more representative in heavy metals adsorption are organic matter (OM), cation exchange capacity (CEC) and Fe and Al oxides (Bradl, 2004; Violante et al., 2010 and Cerqueira et al., 2011).

Nickel is an important heavy metal, and the pollution of nickel has gained importance because of its increased understanding of its persistence and toxicity in ecosystems (Alemayehu and Lennartz, 2010). The normal total range of Ni in soils is 2 – 750 mg kg⁻¹ and the normal range in plants is 0.02 – 5 mg kg⁻¹ (Alloway, 1990). Also, the natural total Cu contents in the soils are between 2 and 250 mg kg⁻¹, on average 30 mg kg⁻¹ (Adriano, 2013). In soil, copper could be found: in the soil solution, adsorbed on exchange sites in the soil, in the oxides, in the crystal lattice of primary and secondary minerals (Selbig et al., 2013) and in soil organic matter and living organisms (Adriano, 2013). Gonçalves et al., (2016) found that the maximum adsorption of Cu on soil (surface layer) was ranged from 825 to 1260 mg kg⁻¹. While Usman, (2008) found that the maximum Cu and Ni adsorbed on six Egyptian soils was ranged from 742 to 1921 mg kg⁻¹ and 80 to 305 mg kg⁻¹ respectively. Isotherm equations as Freundlich and Langmuir equations are used to describe the interference behavior between adsorbent and adsorption surface (Kamari and Wan Nagh, 2009; Jain et al 2004). The major source of heavy metals which entering into the soils are mining and smelting activities and can lead to ecological damages (Li et al., 2006). Copper has recently been widely used in many technical fields, but also in agriculture (fungicides and herbicides). Willful use of copper in agriculture is in inorganic

and organic forms (Pal et al., 2014). Adsorption of copper and nickel in soil depends on the type of soil. Copper has a very high affinity to organic material and the bond between the organic material and copper is much stronger than with other heavy metals (Adriano, 2013).

The aim of the experiment described here was to study the influence of organic wastes from sugar manufacture (filter cake and vinasse) on the adsorption of copper and nickel in loamy sand soil at different reaction time. Also, the effect of organic materials used to restricts contaminants in the soil.

Materials and Methods

A laboratory experiment was conducted in the central laboratory of Soil and Natural Resources Department, Faculty of Agriculture, Aswan University during summer 2019. The used soil in this study was obtained from the cultivated soil near the Nile River in Aswan City at El-Gozaira district (N: 24° 07'5"; E: 32° 53' 50"), Aswan Governorate, Egypt. The soil was analyzed for some physical and chemical properties according to procedure Klute (1986) and Cottenie et al. (1982) and the obtained data are recorded in Table 1.

Two organic amendments were considered in this study i.e. filter cake supplied from a Kom-Ombo Sugar Factory in Kom-Ombo City, Aswan Governorate and vinasse supplied from Egyptian Sugar & Integrated Industries Factors at Hawamdyia, Giza Governorate, Egypt. Some chemical and physical properties of filter cake and vinasse were determined according methods of A.O. A. C. (1995) and the obtained data are recorded in Table 2. Copper and nickel solutions were prepared from copper sulfate (assay 99.8%) and nickel sulfates (assay 99.5%) of Merck Company for Chemicals of Cu and Ni separately.

TABLE 1. Selected properties of the soil used

Parameter	Unit	Value
pH (1:2.5 soil: water susp.)		8.4
EC (1:5 soil: water extract)	dS/m	0.52
OM	%	0.32
Particles size distribution		
Sand		82
Silt	%	6.7
Clay		11.3
Texture according to international classification		Loamy sand
Cation exchange capacity (CEC)	cmol kg ⁻¹	9.2
Cu (1:100 soil: water extract)		0.18
Ni (1:100 soil: water extract)	mg/kg	0.21

TABLE 2. Selected properties of the organic wastes used

Parameter	Unit	Filter cake	Vinasse
pH (1:2.5 organic materials: water)		6.8	4.6
EC (1: 5 organic materials: water)	dS/m	1.8	19.8
OM	%	31.3	4.01
Total C	g kg ⁻¹	181.0	23.3
Total N	g kg ⁻¹	22.0	14.5
C/N ratio		8.2	1.61
Available Cu	mg kg ⁻¹	0.15	0.55
Available Ni	mg kg ⁻¹	0.04	0.46

Portions equivalent to 5 g of sieved soils were placed in a 250 mL conical flask together with either 0.5 g filter cake or vinasse. Another soil portions were left without amending the investigated organic amendments (a control treatment). Twenty five mL of distilled water and 10 mL of copper or nickel solution (4400 mg L⁻¹ from Cu or Ni as initial concentration = 1257 mg L⁻¹) were then added to the contents of the conical flask. Afterwards, the pH of the mixtures was adjusted at 7.0 by 0.01 N of HCl or NaOH, and shook on rotary shaker at room temperature (25 ± 2°C) for 5 time periods of 5, 10, 20, 40 and 60 minutes. The experimental treatments comprised the following: soils and solutions of Cu and Ni with amending organic matter (filter cake or vinasse), soils and solutions of Cu and Ni without amending organic matter. Afterwards, mixtures were quantitatively transferred into 50 ml centrifuge tubes, centrifuged at room temperature (25 ± 2°C) on 3000 rpm and then filtered through Whatman No. 42 filter paper as described by Fontes and Gomes (2003). Each treatment was repeated three times. The filtrates were analyzed for their contents of Ni and Cu using Thermo Scientific, iCAP 7000 Plus Series ICP-OES. The adsorbed amounts of Ni or Cu on each soil (+/- organic amendment) were calculated as the difference between the amounts of Ni and Cu presented initially in the system and the corresponding concentrations at the equilibrium solutions as described by Yassen, and Fakher (2016).

Adsorption data of Ni and Cu by soil and organic matter (filter cake and vinasse) were calculated by estimate of the concentrations of Cu and Ni adsorbed per 5 g soil according to the method by Dada et al. (2012) according to the following equation:

$$C_{ads} = (C_0 - C_{eq}) \dots\dots\dots (1)$$

where:

C_{ads} = the amount of the metal as (mg/5 g of soil) that is retained in the solid phase.

C_0 = the concentration as (mg/L) of heavy metals Cu and Ni in initial solutions.

C_{eq} = the concentration as (mg/L) of heavy metals in solutions after shaking and filtration.

Langmuir and Freundlich isotherm models were applied to describe the adsorption of Cu and Ni ions. The linear formula was used as follows a Langmuir equation. The Langmuir model is as follows:

$$q_e = k_L \cdot C_{eq} \cdot q_{max} / (1 + k_L C_{eq}) \dots\dots\dots (2)$$

Langmuir adsorption parameters were estimated by transforming the Langmuir equation (2) into a linear form.

$$C_{eq} / q_e = 1 / q_{max} \cdot k_L + C_{eq} / q_{max} \dots\dots\dots (3)$$

where:

C_e = the equilibrium concentration of adsorbate (mg /L⁻¹)

q_e = the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g).

q_{max} = maximum monolayer coverage capacity (mg/g)

K_L = Langmuir isotherm constant (L/mg).

It can be represented graphically after draw the relationship between C_e on the X axis and C_e / q_e on the Y axis where that $1 / q_{max}$ represents the slope of the straight line and $1/q_{max} \cdot K_L$ represents the intercept in equation (3). The essential features

of the Langmuir isotherm may be expressed in terms of equilibrium parameter R_L , which is a dimensionless constant referred to the separation factor or equilibrium parameter (Webber and Chakravarti, 1974).

$$R_L = 1/1 + (1 + K_L C_0) \dots \dots \dots (4)$$

where:

C_0 = initial concentration, and K_L = the constant related to the energy of adsorption (Langmuir Constant). R_L value indicates the adsorption nature to be either unfavourable if $R_L > 1$, linear if $R_L = 1$, favourable if $0 < R_L < 1$ and irreversible if $R_L = 0$.

Freundlichequation is also commonly used to describe the adsorption characteristics for the heterogeneous surface (Hutson and Yang, 2000). The adsorption data were fitted to fit the empirical equation proposed by Freundlich:

$$Q_e = K_f C_e^{1/n} \dots \dots \dots (5)$$

where:

K_f = Freundlich isotherm constant (mg/g)
 n = adsorption intensity;
 C_e = the equilibrium concentration of adsorbate (mg/L)
 Q_e = the amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g). Linearizing equation (5) relationship

$$\log q_{eq} = \log K_f + 1/n \log C_{eq} \dots \dots \dots (6)$$

If we draw the relationship between the $\log q_{eq}$ on the Y axis and the $\log C_{eq}$ on the X axis, the output will produce a valid $1/n$ line relative to the capacitance. The equation of the line with the axis ($\log K_f = Y$) is equal to the equilibrium energy. The constant K_f is an approximate indicator of adsorption capacity, while $1/n$ is a function of the strength of adsorption in the adsorption process (Voudrias et al., 2002). If $n = 1$ then the partition between the two phases are independent of the concentration. If value of $1/n$ is below one it indicates a normal adsorption. On the other hand, $1/n$ being above one indicates cooperative adsorption (Mohan and Karthikeyan 1997).

Results and Discussion

The relationship between the amount of adsorbed Cu mg / g on the investigated soil in presence or absence of the organic amendment is illustrated in Figure 1. The highest adsorbed amount of Cu was found in “filter cake + loamy

sand soil” treatment, followed by “vinasse + soil” while the least adsorbed amount was found in the non-amended soil, especially at the short reaction time (5 and 10 min.). On the other hand, Figure 1 show that at reaction time of 60 minutes, the highest amounts of Cu adsorbed were found with the treatment of in the non-amended soil and the lowest values were associated the treatment of soil + vinasse. Johnston and Tombáčz (2002); Abat et al. (2012) and Gonçalves et al. (2016) found that organic matter is mainly responsible for the maximum adsorption of Cu in soils especially with long shaking periods.

Data presented in Figure 2 reveals that that high percentage of Cu sorption reached 93.04 % of the total Cu after 60 minutes on “soil + filter cake” treatment while this percentage was 77.9% in “soil + vinasse” treatment and 96.9% in the non-amended soil. These findings appear the high association between Cu and organic matter reported before that by Al-Janabi and Al-Robiaee (2016).

The Ni sorption was differenced regarding treatments. There are wide differences in adsorbed amounts of Ni from treatment under study (Fig. 3), where the high peak of Ni sorption was found with soil + filter cake treatment at shaking period 20 min. compare with other two treatments and also at different reaction times. There was a difference between Ni adsorbed and Cu adsorbed, where the adsorbed amounts of Ni by the treatment of soil with organic materials were higher than these occurred on the soil+ filter cake treatment and lower than these found with soil+ vinasse treatments. Generally, the obtained data concluded that filter cake application resulted in a more increases in sorption of heavy metals such as Cu and Ni compared with that resulted from vinasse applications or with that soil without organic materials. These results are in similar of studies on adsorption Cu on silt clay loam soil in presence or absence of organic materials (compost) with those reported by Al-Janabi and Al-Robiaee (2016).

In addition data in Figure 3 showed that the Ni sorption curves were oscillating along the curve with increasing shaking periods in the three treatments under study compare with the Cu dsorption with the same treatments. Also, the data in Figure 3 showed that the low effect of reaction time on the adsorbed amounts of Ni compared with that observed with Cu adsorption curve (Fig. 1). These observations were recorded with the studied three treatments.

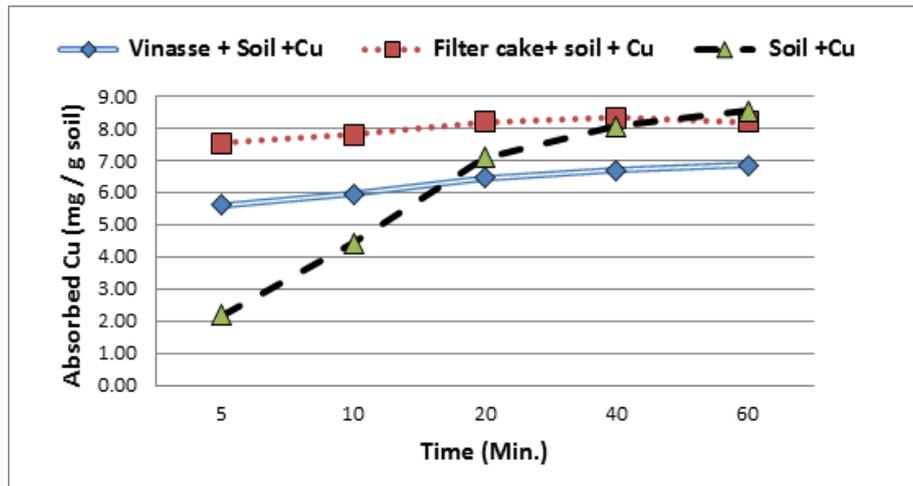


Fig. 1. Effect of filter and vinasse on Cu adsorption (mg/g) on the used soil at different reaction times (minutes).

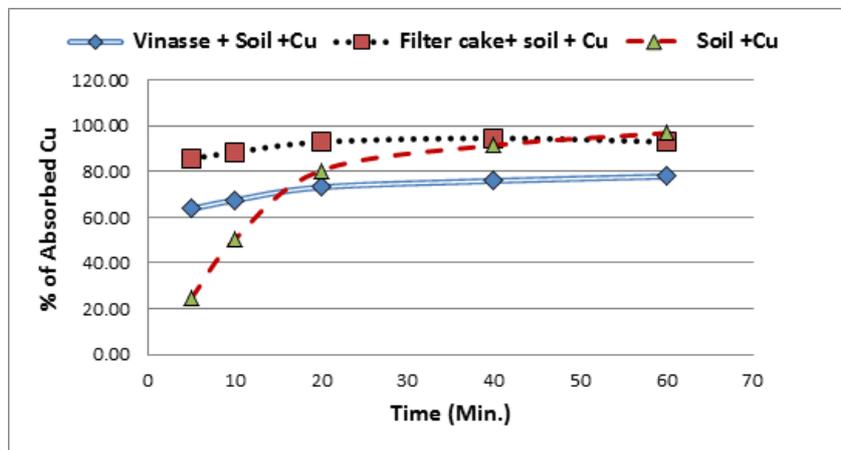


Fig. 2. Adsorbed Cu as a percentage (%) of added Cu on soil and soil-organic matter complexes at different reaction time.

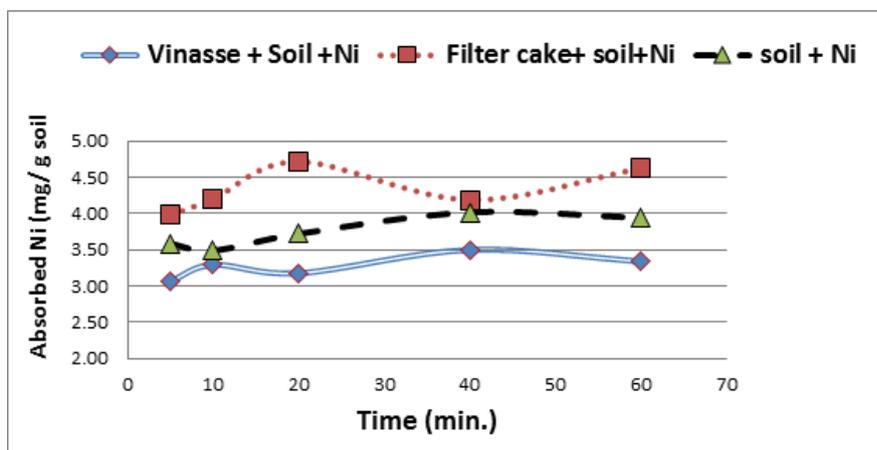


Fig. 3. Effect of filter and vinasse on Ni adsorption (mg/g) on the used soil at different reaction times (minutes).

Data in Figure 4 showed that the high percentage of Ni adsorption on soil and organic materials was 53.59 % of total Ni added at shaking period 20 minutes with the treatment of soil + filter cake but it was 45.63% of total Ni added at shaking period 40 minutes with soil without organic materials treatments of soil + vinasse was 39.74% at 40 min. of shaking period. As shown in Fig (1 to 4), it may be noted that with the same adsorption treatment (materials and shaking period) the adsorption amount of Cu was higher than that of Ni.

To evaluate the sorption mechanism of Cu and Ni on soil with or without the organic materials (filter cake and vinasse), comprehensive studies were conducted on the sorption data using different isotherm models (Langmuir and Freundlich equations). Data presented in Table

3 and Figures 5 and 6 reveal that the dominant mechanisms describing Cu or Ni retention on soil was the adsorption mechanism as the calculated r^2 values were high enough. In this concern, all the " r^2 " values of the Langmuir equation are high significant except for Cu in the in the non-amended control soil. According to the maximum capacity of Cu adsorbed the following sequene was considered: filter cake + soil > vinasse + soil > soil alone. Regarding to the maximum capacity of Ni adsorbed the following order was detected: filter cake + soil > only soil > vinasse + soil. These results agree with Al-Janabi and Al-Robiaee (2016) which pointed out to the positive effect of organic matter on increasing Cu adsorbed on the calcareous soil. Also, these results agree with Ramachandranand D'Souza (2013) which showed adsorption of Ni on entisol in India soils.

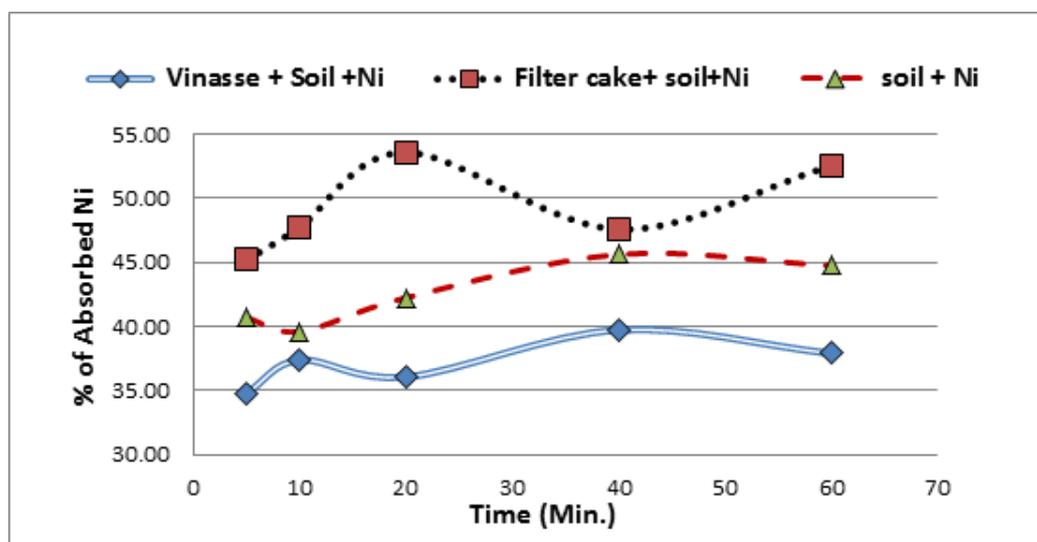


Fig. 4. Adsorbed Ni as a percentage (%) of added Ni on soil and soil-organic matter complexes at different reaction time.

TABLE 3. Langmuir and Freundlich parameters for Cu and Ni adsorption affected by the studied treatments.

Treatment	Langmuir equation parameter				Freundlich equation parameter		
	q_{max} (mg/g)	K_L (L/mg)	R_L	R^2	n (mg/L)	K (mg/g)	R^2
Vinasse + soil +Cu	4.378	-0.0096	-0.100	0.9975	2.474	67.16	0.9925
Filter cake+ soil + Cu	7.139	-0.0930	-0.009	0.9995	9.867	12.84	0.9835
Soil +Cu	2.248	-0.0091	-0.107	0.8952	2.626	42.78	0.7571
Vinasse + soil +Ni	1.218	-0.0020	-1.902	0.9981	0.594	246988.19	0.9988
Filter cake+ soil+Ni	2.155	-0.0031	-0.519	0.9982	0.986	3021.78	0.9984
Soil + Ni	1.597	-0.0024	-0.971	0.9988	0.745	25742.80	0.9991

The data in Table 3 show that Langmuir constants K_L which explain the maximum adsorption (L/mg) of Cu or Ni were negative, leading us to the conclusion that the adsorption behavior of the tested systems does not follow the assumption on which the Langmuir approach is based. Similar results were reported by Kiurski et al. (2011 and 2012)

Freundlich's K values recorded in Table 3 for Cu adsorption data were 67.16, 12.84 and 42.78 for the treatments "soil+ vinasse", "soil + filter" and "soil without organic materials", respectively. On the other hand, the corresponding values for Ni adsorption data were 246988.19, 3021.78 and 25742.80 for the abovementioned treatments, respectively. In general, the values of the Freundlich equation constant (n) of Ni and Cu adsorbed express the absorption capacity.

The results obtained in Table 3 showed that the Ni absorption capacity is higher than the copper absorption capacity in all treatments. Also, results of Freundlich's n values in Table 3 showed that n values for the Cu adsorption data were higher than one 2.474, 9.867 and 2.626 for the treatments of soil+ vinasse, soil + filter and the non-amended control soil, respectively. The obtained results indicate to the adsorption kind is from the cooperative kind which confirmed its from the Langmuir constants. Also, constant's n values of the Ni treatments showed that was natural adsorption because n values are less than one 0.594, 0.986 and 0.745 with these treatments: soil+ vinasse, soil + filter and soil without organic materials, respectively). This was due to the high association between Cu and organic matter. Similar results were reported by Al-Janabi and Al-Robiae (2016).

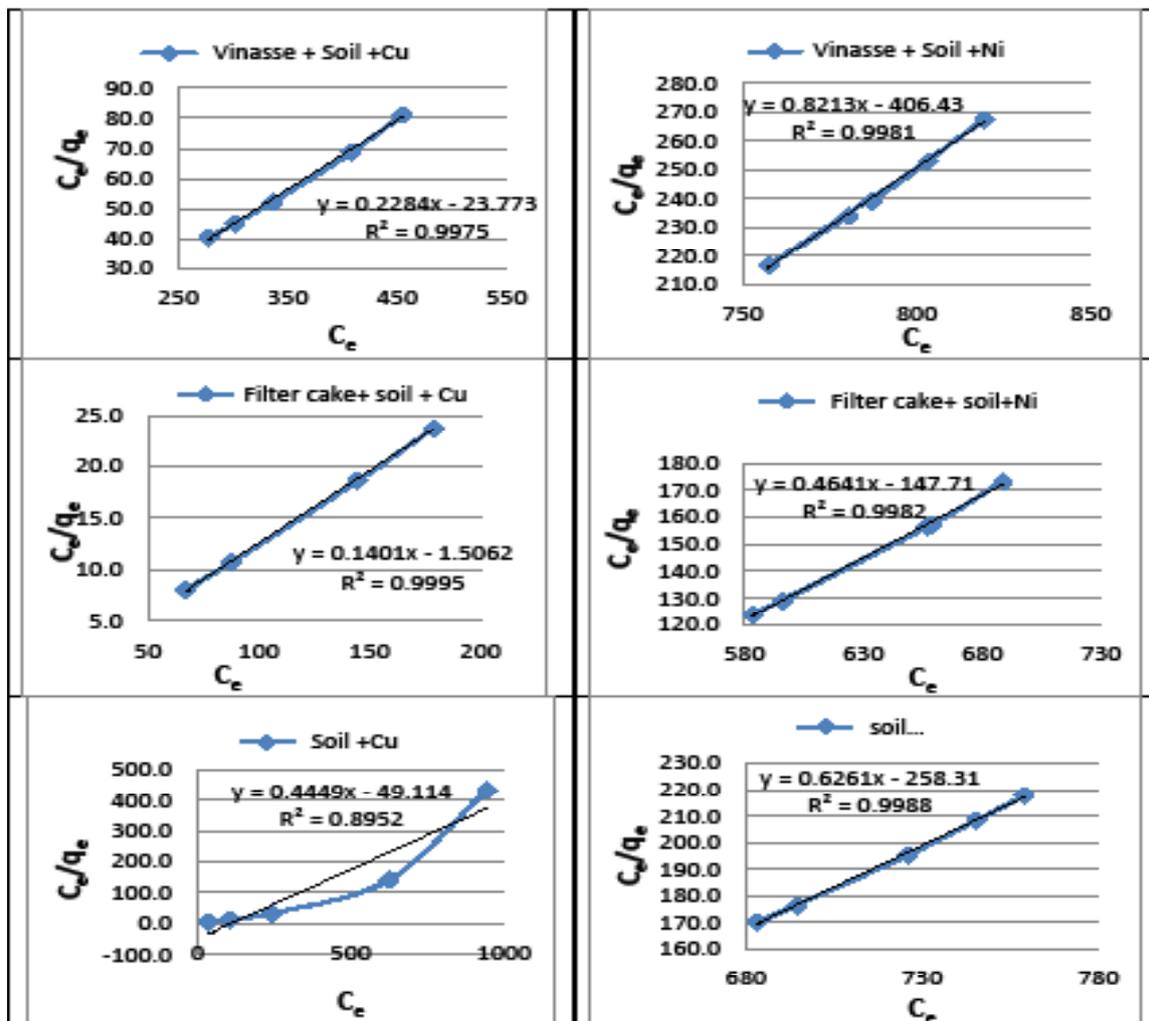


Fig. 5. Linear relationship between C_e and C_e/q_e of Cu and Ni adsorption data applying Langmuir equation.

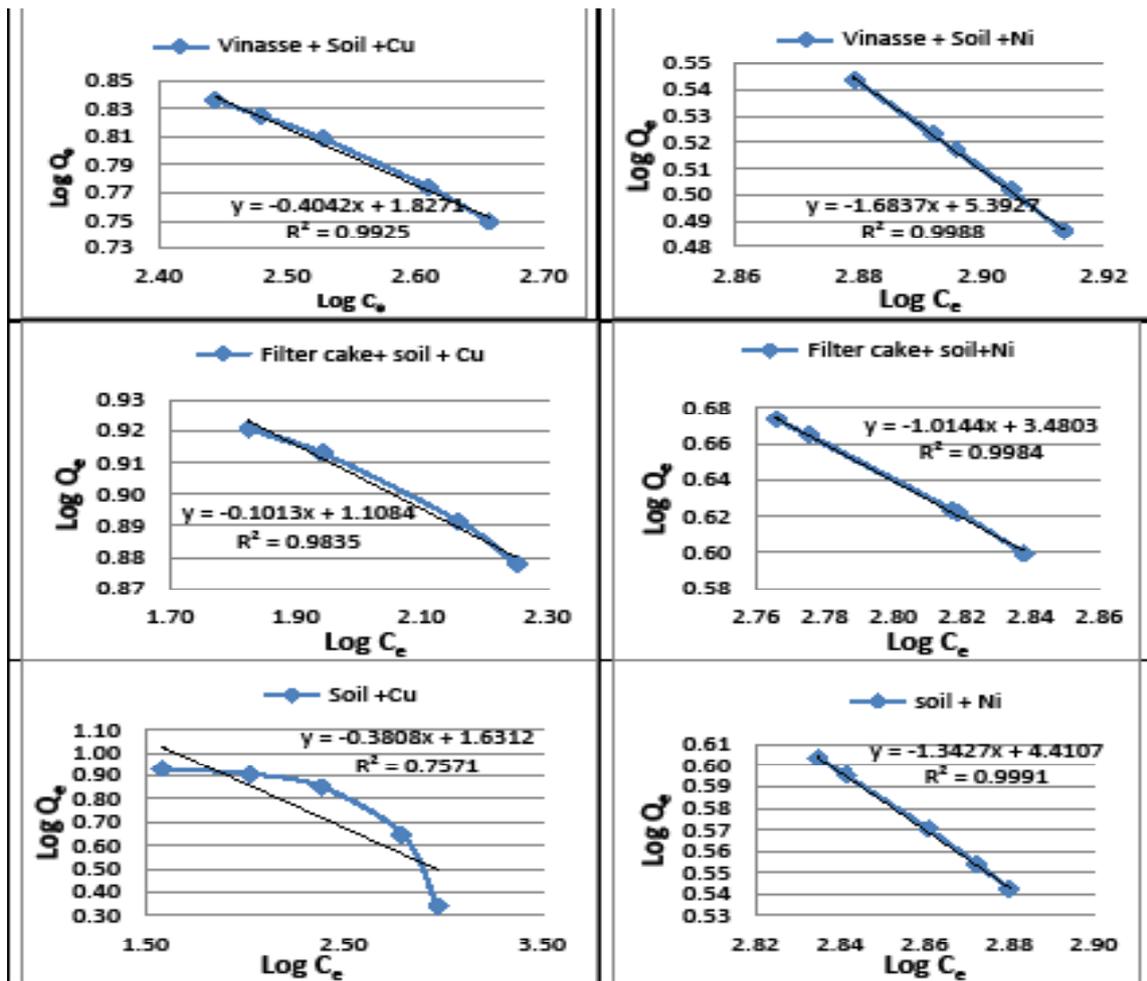


Fig. 6. Linear relationship between Log Ce and Log Qe of Cu and Ni adsorption data Applying Freundlich equation.

Conclusion

It is concluded from this study that the addition of organic matter (filter cake and vinasse) and increasing the reaction time increase Cu and Ni adsorption on the loamy sand soil. Also, the Freundlich model can describe Cu and Ni adsorption better than the Langmuir one. Thus, the addition of filter cake and vinasse protect the plants and groundwater from contamination.

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تأثير بعض المخلفات العضوية على بعض نماذج الأيزثرم لامتماز النحاس والنيكل على تربة لومية رملية

عاطف عبدالعزيز علي سويد

قسم الاراضى - كلية الزراعة - جامعة اسوان - مصر

تم إجراء جربة معملية لدراسة تأثير عجينة المرشحاتوالفيناس على امتزاز النحاس والنيكل على التربة الطميية الرملية. ولتحقيق هذا الهدف ، وضعت 5جم من التربة مع 0.5 جم من عجينة المرشحات أو فيناس في دوارق مخروطية. ثم اضيف 10 مل من محاليل النحاس أو النيكل المحتوية على 44 مجم من النحاس أو النيكل / 10 مل، والتي تم خضرها من $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ و $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ على التوالي . بالإضافة إلى 25 مل من الماء المقطر إلى الدوارق بحيث كان التركيز الابتدائي من النحاس أو النيكل من 1257 مجم/ لتر. تم استخدام اثنين من الدوارق ككنترول بالتربة بالإضافة إلى Cu أو Ni في ماء المقطر. تكررت ثلاث مرات رجعت معلقات التربة في الدوارق عند درجة حرارة 25 مئوية لمدة خمس فترات اهتزاز هم 5 و 10 و 20 و 40 و 60 دقيقة. أظهرت النتائج التي تم الحصول عليها أن المواد العضوية أدت إلى زيادة كفاءة ادمصاص Cu و Ni في تربة طميية رملية مقارنة معاملة الكنترول (تربة بدون مواد عضوية) . وكانت عجينة المرشحات أكثر قدرة على ادمصاص لكلاً من Cu و Ni مقارنة معاليفيناسمع زيادة فترات الهز. كانت أعلى كمية امتزاز تبلغ 8.53 مجم / جم للنحاس مع التربة (بدون مادة عضوية) عند وقت التفاعل 60 دقيقة و 4.72 مجم / جم للنيكل مع عجينة المرشحات + التربة عند زمن التفاعل 20 دقيقة. أظهرت النتائج أيضًا أن معادلة Freundlich لامتماز Cu و Ni كانت أفضل من معادلة Langmuir نظرًا لأن معادلة Langmuir غير مناسبة لمعادلات الامتماز ذات التركيزات العالية للمعادن الثقيلة قيد الدراسة.