

Adsorption of Added Cadmium in Relation to Properties of Some Alluvial Soils

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Abstract. In the present work an attempts have been done to study the adsorption capacity of Malaysian soils for cadmium, which consider as an environmental harmful metal. Six soils represent the major soil series used for agricultural production in Malaysia: Typic Sulfaquept (Sedu) Haplic Acrorthox (Segamat) Tropic Fluvaquent (Kundur), Orthoic Tropudult (Durian), Typic Paleudults (Melangit), and Typic Paleudults (Betu Anam) were used. The soils were very acid to slightly alkaline with the lowest pH value at 3.00 (Sedu) and the highest pH 7.3 at Melangit soil. The soils texture varied from silty to clayey. The highest clay content (87%) was found in Segamat series where the lowest (18.9 %) was in Melangit which considered silty soil. The amount of Cd adsorbed by soil depends on their physico-chemical properties. The adsorption data were tested by three different models Langmuir, Freundlich and second order equation. For the Langmuir isotherm r^2 ranged between 0.943 to 0.998 with an average of 0.979, Freundlich isotherm r^2 ranged between 0.519 to 0.991 with an average value of 0.866 where the second degree (r^2) ranged between 0.366 to 0.997 with an average 0.774. This suggests that the Langmuir isotherm provides a good model of the sorption system. The adsorption capacity differs between soils. The highest adsorption capacity was found in Melangit and Kundure ranged between 1800 to 2500 $\mu\text{g}/\text{soil Cd}$. Though Melangit is silty soil but it has high pH and high content of Calcite this suggests the precipitation of Cd and the sorption of heavy metals may be on the edge due to the aluminol sites. The usage of added CdCl_2 as adsorbant metal, CdCl^+ tends to be desorbed from the clay to a larger extent than Cd and hence adsorbed as a monovalent than a divalent cation.

Introduction

Cadmium (Cd) is a naturally occurring, potentially toxic and mobile element present in most soil environments. It is considered an important environmental soil pollutant, as it is a potentially biotoxic heavy metal that is readily absorbed by plants and has potential to enter the human food chain (Chang *et al.*, 1983). Cadmium pollution in soils has been increased during the last decades, mainly due to the large application of farmyard manure, sewage sludges, mining waters or fertilizers obtained from phosphorites, which usually containing a very high Cd content (Mortvedt 1987; Vanni *et al.* 1994).

In contaminated environments Cd can be taken up by animals and humans. It has been reported that chronic exposure of humans through ingestion and/or inhalation to Cd for a longer period of time may cause severe damage to kidney and lungs, and can lead to other pathological symptoms (Nriagu, 1980).

Depending on soil and environmental conditions, Cd added in soil can be retained by both adsorption and/or precipitation reactions. The factors that control desorption and redistribution of soil Cd include Fe oxides content (Davis and Kent, 1990), type and contents of clay minerals (Jobstmann and Singh, 2001), organic matter (Hettiarachchi *et al.*, 2003) and net negative surface charge density (Naidu *et al.*, 1994).

Retention of Cd as well as other heavy metals by soil depends on a complex equilibrium between soluble and adsorbed forms of the metal and reactivity of the metal with soil particles. Swift & McLaren (1991) reported that adsorption and desorption strongly affect the bioavailability of heavy metals and therefore toxicity in soil. Heavy metals in soil exist in soluble and exchangeable forms and as organic and inorganic complexes. This sensitivity is very dependent upon soil type (Tiller 1989). In this work different type of soils have different chemical

properties have been used. Our objectives are:

- (i) To identify the soil which has high capability to adsorbed Cd.
- (ii) To understand adsorption mechanism of these soils using different models.
- (iii) To established correlation between adsorption behavior of these soils and adsorption capacity.

Material and Methods

Soils

Six surface (0-20 cm) soils representative of the major soil series used for agricultural production such as Rubber, oil palm, cocoa and rice which are the most important crops grown mainly in the estate sector were sampled. The soil classification was used based on previous information of the soil map of the area (Table 1). All samples were located using Magellan to specify the latitude (N) and Longitude (E). These soils represent Sedu (Typic Sulfaquept), Segamat (Haplic Acrorthox) Kundor (Tropic Fluvaquent), Durian (Orthoic Tropudult), Melangit (Typic Paleudults) Betu Anam (Typic Paleudults). The soils were selected to be truly representative of the area and to give wide variation in soil properties. They were air dried and ground to pass through a 2 mm sieve.

Table 1. Soil classification and sample sites

Soil	Soil Classification	Latitude (N)	Longitude (E)
Sedu	Typic Sulfaquept	2° 30.238'	102° 48.505'
Segamat	Haplic Acrorthox	1° 56.287'	103° 11.533'
Kundor	Tropic Fluvaquent	1° 56.114'	103° 10.916'
Durian	Orthoic Tropudult	1° 50.443'	103° 07.343'
Melangit	Typic Paleudults	6° 05.287'	103° 20.148'
Betu Anam	Typic Paleudults	6° 31.097'	100° 10.468'

Water soil pH was determined using 1:2.5 soil/water ratio. Organic carbon (OC) by Walkley and Black (1934) method and cation exchange capacity (CEC) were determined by leaching with neutral ammonium acetate solution Piper (1947). Ca-exchange of the soils was determined by 0.025M CaCl_2 Mackay *et al.*, (1986). Mineralogical composition of the soils was determined using X-ray diffraction analysis.

Adsorption measurements

The sorption of Cd was determined by adding 25 mL solution containing each of 0, 5, 10, 20, 40, 60, and 100 mg Cd as cadmium chloride in 0.01M $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ to 1 g soil in 40 ml polypropylene test tube. There were

three replicates for each Cd treatment. The soil-solution was shaken at 240 rpm at room temperature (25°C) for 24 hours and filtered through a Whatman No. 42 filter paper. The concentration of Cd in the filtrate was determined by ICP. The Cd adsorbed was calculated as difference between that in solution initially and that remaining after equilibration. Statistical analysis was run on all treatment.

Adsorption analysis

Two models have been used to study analysis adsorption The Freundlich isotherm model was chosen to estimate the adsorption data according to the following formula.

1) Freundlich

The Freundlich isotherm model was chosen to estimate the adsorption intensity of the sorbent towards the adsorbent. It is an empirical equation employed to describe the isotherm data given by:

$$x/m = AC^b$$

where x/m is the amount of Cd retained by the soil in mg kg^{-1} , C is the concentration of Cd in solution in mg L^{-1} , A is the maximum retention in $\text{m}^3 \text{kg}^{-1}$ and the parameter b is bonding energy. A plot of $\log(x/m)$ versus $\log C$ will yield a straight line of slope b and intercept of $\log A$. The data were fitted to Freundlich equation and regression analysis was used to find the best-fitting straight line. The slope and intercept were used to calculate b and A as well as to test differences among soils for Cd.

2) Langmuir

The Langmuir equation was chosen for the estimation of maximum adsorption capacity corresponding to complete monolayer coverage on the adsorbent surface. The linearized Langmuir equation was used to analyze the data. The equation is given as:

$$C/(x/m) = 1/(k_1 k_2) + C/k_1$$

Where C is the final solution concentration of Cd in mg L^{-1} , x/m is the amount of Cd adsorbed in mg kg^{-1} of soil, k_2 is the adsorption maximum (mg kg^{-1}), and k_1 is a constant related to bonding energy of Cd to soil ($\text{m}^3 \text{kg}^{-1}$).

A plot of $C/(x/m)$ versus C will yield a straight line of slope $1/b$ and intercept of $1/k_1 k_2$. The data were fitted to Langmuir equation and regression analysis was used to find the best-fitting straight line. The slope and intercept were used to calculate k_2 and k_1 as well to test differences among soils for k_1 and k_2 .

Result and Discussions

Characteristics of the soils

Table 2. summarizes the main physicochemical and mineralogical characteristics of the studied soils. The particle-size distribution exhibits a substantial variation in clay and silt contents ranging from 189.0 to 870.0 g kg⁻¹ for clay. Segamat and Kundor Soils showed high amounts of clay 870.0 and 750.0 g kg⁻¹, respectively, whereas Melangit soil has the lowest amount of clay (189.2 g kg⁻¹). The cation exchange capacity (CEC) could be generally determined by the clay content. Therefore, the highest value of CEC (120 Cmol kg⁻¹) was recorded for Kundor soil that was enriched in clay, whereas the lowest one (9.4 Cmol kg⁻¹) was shown for Melangit soil that has the lowest amount of clay. Exchangeable Ca was the dominant cation present on the exchange site of the Kundor soils.

It is clear from Table 2. that most of soils have acidic characteristic where pH values in the range between 3.00 to 7.3. The lowest pH value was at Sedu soil and the highest at Melangit. low organic carbon content shows range between 5.4 to 62 g kg⁻¹ for studied soils. The mineralogy of clay varied. In general, smectite, kaolinite, and mica are the most common clay minerals in study soils consisting of high amount of smectite and low content of kaolinite and mica. The clay fraction of soils Sedu, Kundor and Durian was dominated by smectite with a moderate amount of kaolinite and mica. In contrast, Segamat soil was consisting mainly kaolinite. Melangit soil is dominated with kaolinite and Calcite. However, most of these soils considered high weathered soils have lowest amounts of 2:1 minerals. The clay fraction of soils Durian and Betu Anam consisted mainly of nonphylosilicate minerals such as feldspar. Goethite was detected only in Segamat soils, whereas gibbsite was found in three soil (Sedu, Segamat and Durian). However, quartz formed in very low amounts.

Cadmium Adsorption

Cadmium adsorption varied greatly and depends on the physico-chemical properties of soils used. The Cd adsorption isotherm for all soil samples presented in Figure 1. The shape of Cd (II) was of a “L2” according to Giles and smith, (1974) classification for isotherm which indicates that the data have reached a maximum value, resulting in the presence of the plateau. None of the isotherms display typical Langmuirian shape, with the possible exception of the (Figure 1 e and f). The Figure 1-e & f show a steep increase for the Cd adsorption isotherm after Cd reach 40 µg/ml Cd.

As revealed from the adsorption isotherms, Cd concentration in the equilibrium solutions increases significantly with a little amount of Cd retained by the solid phase. Segamat, Durian, Betu Anam and Sedu showed similar adsorption capacity ranging between 300 to 650 µg/soil Cd. The similarities between these soil can be ascribed to similarities in the CEC, and mineralogy content. However, the highest adsorption capacity was found in Melangit and Kundor ranged between 1800 to 2500 µg/soil Cd. It is expected that the adsorption of Cd on Kundor occur by interexchange with cations saturating the planar positions as well as adsorption to edge sites, because of the high CEC (120 cmol⁻¹kg) and the presence of 2:1 clay minerals the sorption of Cd was high. Hanafi and Sjiola (1998) have shown that soil CEC is the most important factor in controlling the adsorption of Cd in acid tropical soils. Whereas the high Cd adsorption of Melangit could be attributed to the presence of high carbonate in the form of calcite mineral. Undabeytia, *et al.*, 1998; Mahamadi and Zaranyika, 2007 evaluate the effect of excess K, Ca and Mg as interferents during the adsorption of Cd reported that Cd adsorption occurred mainly by physical adsorption and suggests that the Ca ions were more effective in suppressing Cd adsorption.

Table 2. Selected physical, chemical and mineralogical properties of the soils

Soil	Depth cm	Clay g Kg ⁻¹	Texture	pH in H ₂ O	Organic C g Kg ⁻¹	CEC mmol.kg ⁻¹	Exchangeable Ca mmol.kg ⁻¹	Mineralogy of the clay fraction**
Sedu	0-20	490	Si clay	3.0	18.8	38	2	K, Q, Gb, S, M, and J
Segamat	0-20	870	Clay	4.9	21.0	36	11	K, Q, Gb, and Go
Kundor	0-20	750	clay	4.9	45.1	120	77	K, Q, M and S
Durian	0-20	480	Si Clay	4.4	62	31	2	K, Q, Gb, S, M, F, M+M- ML and M+S
Melangit	0-20	189.2	silty	7.3	5.4	9.4	0.18	K, Calcite
Betu Anam	0-20	630	Clay	5.4	15.7	42	21	K, Q, M and F

** k-Kaolinite, Gb- Gibbsite, Go- Goethite, F-Feldspar, M-Mica, ML- Mixed layer, S-Smectite, Q-quartz and J- Jarosite, CEC (Cation Exchange Capacity), OC (Organic Carbon)

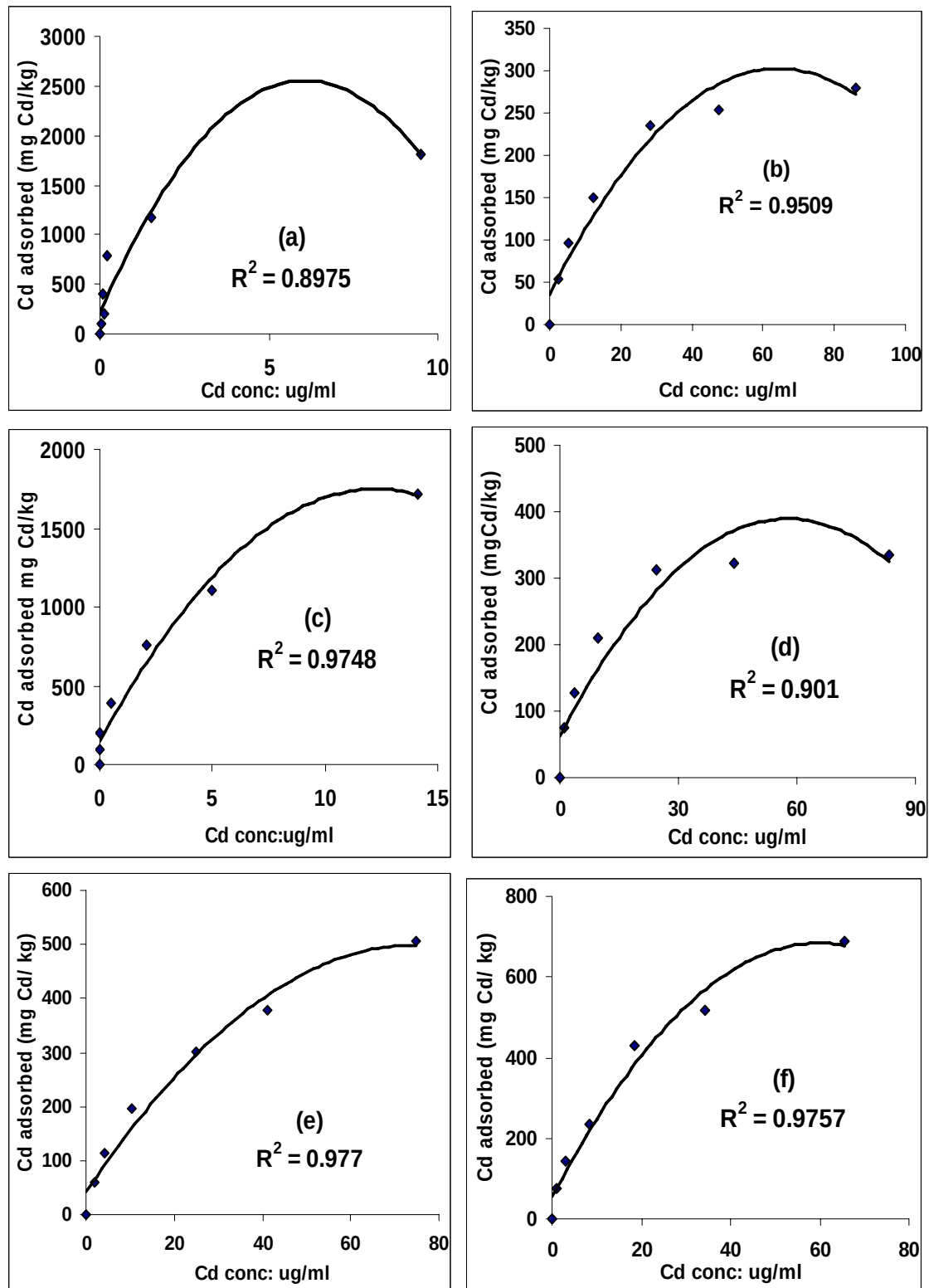


Fig. 1. Shows the adsorption isotherms of adsorbed Cd on soil sampled (a) Melangit (b) Betu Anam (c) Kundur (d) Sedu (e) Durian (f) Segamat.

Moreover, Melangit has high pH and at high pH Stadler and Schindler (1993) suggested that the sorption of heavy metals on the edge sites is mostly due to the aluminol sites.

The mixture of clay minerals containing Segamat (Figure 1-f) soil like goethite-kaolinite surface seems to have a higher affinity for Cd than the other soil (Figure b, d and e). This might be caused by the formation of extra sorption sites at the step sites created by the sorption of goethite particles onto kaolinite.

In general, the high affinity of these soils can be ascribed to more than one reasons: First, the presence of high content of clay fraction. Second, the high CEC of most soils, and lastly the usage of CdCl_2 as adsorbant metal give rise to high absorbance capacity. Undabeytia et. al., (1998) reported that CdCl_2 tends to be desorbed from the clay to a larger extent than Cd. This difference is due to enhanced concentration of the divalent cation at the clay surface.

Hirsch et al., (1989) stated that more Cd was adsorbed as a monovalent than a divalent cation in a suspension containing 50 mM

NaCl; under this condition the solution included several-fold more CdCl_2 than Cd.

Adsorption is the net accumulation of a chemical species at the interface between a solid phase and an aqueous solution phase, leading to a loss from the solution phase (Sposito, 1989). Adsorption reactions can be described by various models. Empirical models provide descriptions of adsorption data without theoretical basis. Examples are the distribution coefficient, the Freundlich adsorption isotherm, and the Langmuir adsorption isotherm and the second order equation. A comparison of the coefficient of regression (r^2) for the three isotherms is shown on Table 3. For the Freundlich isotherm r^2 have a range of 0.519 to 0.991 and an average value of 0.866. The range for r^2 by Langmuir is 0.943 to 0.998 and the average is 0.979. For the second ordered equation isotherm the range of r^2 values is 0.366 to 0.997 and an average value of 0.774. This suggests that the Langmuir isotherm provides a good model of the sorption system. This could be explained by the surface area exposed by soil biomass for the sorption of the cadmium metal ions. The fitting of the data to this model suggests a small surface area for metal adsorption present on the biosorbent, therefore only a monolayer adsorption occurred on its surface in spite of any surface modification. This result was in agreement with Leyva-Ramos et al. (2001), and Igwe. and Augustin (2007).

The bonding energy (k_2 -b) and adsorption

Table 3. Correlation using different equations Longmuir (L) and Freundlich (F) and second order equation

Soils	Isotherm		
	r^2 (F)	r^2 (L)	r^2
Sedu	0.938	0.998	0.845
Segamat	0.991	0.973	0.366
Kundor	0.519	0.943	0.755
Durian	0.988	0.978	0.785
Melangit	0.802	0.993	0.898
Betu Anam	0.957	0.998	0.997

Table 4. Adsorption coefficient ($\log A$ - k_2) and bonding energy (b - k_1) for both Freundlich and Langmuir equations

Soil Name	(Freundlich)		(Langmuir)	
	$\log A$	b	K_2	K_1
Sedu	1.8943	0.3741	0.1823	0.3593
Segamat	1.8867	0.5424	0.0463	0.8233
Kundor	2.6537	0.3636	0.6925	1.7953
Durian	1.6741	0.5657	0.0464	0.6226
Melangit	2.9221	0.4768	1.5573	1.9242
Betu Anam	1.6342	0.4583	0.0850	0.3176

coefficient ($\log A$ - k_1) for both Freundlich and Langmuir are shown in (Table 4). The highest Log A values for Freundlich and Langmuir was 2.9221 $\text{m}^3 \text{kg}^{-1}$ and 1.5573 $\text{m}^3 \text{kg}^{-1}$ for Melangit soil respectively and the lowest was 1.6342 $\text{m}^3 \text{kg}^{-1}$ for Betu Anam soil by Freundlich and 0.04635 for Durian soil by Langmuir. The maximum retention for k_1 and $\log A$ showed high values for Melangit and Kundor soils.

The high $\log A$ value for Melangit soil indicate that soil have high affinity for Cd adsorbed by soil compared with other soils with lower values. Melangit soil considered slightly alkaline $\text{pH} > 7$, and dominated with calcite mineral in the clay fraction which could be an indication of precipitation of Cd as it explained by Ford and Sparks (2000) and Ford et. Al.; (1999). They reported that at higher solute concentrations and $\text{pH} > 6.5$, Cd can be incorporated into neo-formed precipitates, formed at the surfaces of phyllosilicate minerals. These surface precipitates form at solution conditions undersaturated with respect to homogeneous precipitation.

On the other hand the maximum retention of Kundor soil is in agreement with the characteristic of the soil. The fine texture, high exchangeable Ca and high CEC. Therefore, the sorption capacity can be by ionic exchange and adsorption capability.

Conclusion

In this study, batch adsorption experiments were performed to evaluate the cadmium adsorption on six different soils. Based on the results of the adsorption batch Experiments, soils have high affinity for adsorbing Cd. The adsorption mechanism obeys Freundlich and Langmuir equations indicating beneficial adsorption occurring through a monolayer mechanism involving physisorption and/or chemisorption. CdCl increased the Cd precipitation. The adsorption of Cd was well described by the Langmuir models.

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إدمصاص الكاديوم المضاف و علاقته بخصائص بعض الترب الطميية

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كلمات افتتاحية: إدمصاص، كاديوم، لانجمير، فرنديش، الاس الهيدروجيني، الكالسيوم، الترب المروحية الرسوبية.

ملخص البحث. في هذا البحث تمت محاولة لدراسة السعة الإدمصاصية لعنصر الكاديوم على ترب ماليزيا و الذي يعتبر احد ملوثات البيئة السامة. تم اختيار ست ترب تمثل الترب الأساسية التي تستخدم للإنتاج الزراعي و هم هابلك اكروروثك (سيجامات) (Haplic Acrorthox (Segamat) ، تيك سالفاكوبت (سيدو) (Typic Sulfaquept (Sedu) ، تيك فلواكوينت (كندور) (Tropic Fluvaquent (Kundor) ، أورثك تروبادلت (دوريان) (Orthoic Tropudult (Durian) ، تيك باليودالت (ميلاجيت) (Typic Paleudults (Melagit) ، و تيك باليودالت (بيتو أنام) (Typic Paleudults (Betu Anam). و تعتبر هذه الترب حامضية إلى قليلة القلوية تصل أدنى درجة حموضة إلى ٣ في تربة سيدو و أعلى درجة حموضة ٧.٣ في تربة ميلاقنت. و يختلف قوام التربة من سلتى الى طيني. و تصل أعلى كمية طين (٨٧٪) في تربة سيجمات و أقل نسبة طين في تربة ميلاجيت و التي تعتبر سلتية. إن كمية إدمصاص الكاديوم يعتمد على خصائص التربة كيميائيا و فيزيائيا. و لقد تم اختبار نتائج الإدمصاص بواسطة ثلاث نماذج لانجمير، فرنديش و معادلة الدرجة الثانية. و أظهرت النتائج لمعادلة لانجمير أن درجة الميول (R^2) تتراوح بين ٠.٩٤٣ إلى ٠.٩٩٨ مع متوسط يصل إلى ٠.٩٧٩. بينما وجد ان معادلة فرنديش معامل الميول (R^2) يتراوح بين ٠.٥١٩ و ٠.٩٩١ مع قيمة متوسط ٠.٨٦٦. بينما و معادلة الدرجة الثانية (R^2) تتراوح ما بين ٠.٣٦٦ و ٠.٩٩٧ بمتوسط ٠.٧٧٤. و تعتبر معادلة لانجمير أفضل المعادلات التي تمثل عملية الإدمصاص لهذه الترب. إن كمية الإدمصاص تختلف بين الترب فوجد أن تربة ميلاجنت أدمصت اكبر كمية من الكاديوم و ممكن أن يعزى هذا إلى نوعية المعدن السائد في التربة (كالسيت) و إلى ارتفاع درجة الحموضة اللذان بدورهما يحدثا تبادل للكالسيوم الموجود على مواقع التبادل و يترسب الكاديوم. و يعتبر استخدام كلوريد الكاديوم احد الأسباب التي تؤدي إلى إدمصاص الكاديوم كأحادي التكافؤ على سطوح الطين بدلا من ثنائي التكافؤ.

