ADSORPTION AND RELEASE OF CADMIUM IN SOME SOILS OF EGYPT.

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ABSTRACT

Cadmium adsorption by and release from five soils representing the soils of Egypt; alluvial, calcareous, and sandy soils, were studied. Soil samples collected from the top 20 cm were equilibrated with $CdCl_2$ solutions containing up to 800 μg Cd/ml. The absorbed Cd was subsequently extracted with 1M ammonium acetate (exchangeable-Cd) and 0.125 M copper (II) acetate (complexed-Cd). The adsorption of Cd by soils was highly fitted to Langmuir and Freundlich isotherms with (r^2) values ranging from 0.962 to 0.993 with the former and from 0.945 to 0.987 with the later. The Cd adsorption maxima calculated from Langmuir isotherm ranged from 1.885 to 8.830 mg/g soil (3.35 to 15.71 meq/100g soil) and was significantly correlated with clay + silt content, cation exchange capacity (CEC), and organic matter content (OM). The corresponding values calculated from Freundlich isotherm ranged from 0.18 to 0.88 mg Cd/g soil and were not correlated with any of the studied soil properties. About 80 to 95 percent of adsorbed Cd was retained by cation exchange and complexing sites. Cation exchange sites assumed to play more important role in adsorbing Cd than complexing sites with increasing adsorption, except in calcareous soil, which exhibited a comparable importance of the two kinds of sites. Such results are important to understand the fate of Cd added to the soil through applications of contaminated sludges and wastes and to develop useful environmental guidelines for its potential toxic hazards.
Keywords: Cadmium, Adsorption, Desorption, Heavy metals, Soils of Egypt.

INTRODUCTION

Heavy metal concentrations of soils have received increasing concern because of their harmful effect on the food chain. Among these metals, Cd is the one of the most likely to pose a health problem because under certain circumstances it can accumulate in plants to concentrations that are not toxic to the plant itself but may be harmful to consumers of the plant (King, 1988c).

Cadmium enters the environment as a result of both natural and anthropogenic activities (Naidu *et al.*, 1997). Concentration of Cd released into the soil by pedogenic processes (one of the natural activities) is largely related to the origin and nature of the parent material and in this regard, Levi-Minzi *et al.*(1976) reported that Cd content of common rocks varies between 0.17 mg kg^{-1} in igneous rocks and 2.6 mg kg^{-1} in sedimentary rocks. Anthropogenic sources comprise all contributions made by mankind including industrial processes that contribute to both atmospheric and land depositions, mining and sewage and fertilizer applications. Of the two modes of input, anthropogenic sources are the greater environmental threat as a result of surface inputs to soil making the metal accessible for plant and animal uptake (Naidu *et al.*, 1997). Metals added to soils via sludge application for example, show limited downward movement and remain in the surface 20 cm of soil (Rappaport *et al.,* 1988). The fate of Cd from anthropogenic sources depends

essentially on its retention and mobility in the soil (Naidu *et al.*, 1997) moreover; adsorption of Cd by soil determines its availability to the plant and its movement through the soil (Abd El-Fattah and Wada, 1981).

Uncontaminated soils usually contain less than 1 mg Cd/kg soil, however, a concentration of 1750 mg / kg and of 118300 mg/kg were found in soils around smelter and hazardous waste sites, respectively (Salim *et al.*, 1996). High Cd concentration levels were also reported in polluted soils from Egypt (Elsokkary, 1996; Rabie, Farida *et al.*, 1996; El-Gendi *et al.* 1999; Abou-El Naga *et al.*, 1999; and Badawy and Helal, 2002).

Heavy metals in soils and sediments are generally present in a variety of forms exhibiting different degrees of bioavailability and mobility (Salim et al., 1996). These forms of Cd are (1) adsorptive and exchangeable, (2) bound to carbonate phases, (3) bound to reducible phases (Fe and Mn oxides), (4) bound to organic and sulfides, and (5) detrital or bound to mineral lattice (Forstner, 1985). Navrot *et al.*(1978) found that almost all the adsorbed Cd was exchangeable with 0.5M $CaCl_2$. On the other hand, Soon (1981) found that about 82 to 92 percent of the adsorbed Cd was retained by cation exchange and complexing sites.

The hazardous impact of Cd includes: (1) a decrease in diversity of population of soil flora and fauna, (2) crop yield reductions in sensitive crops (beans, beets, turnip, and corn) at concentrations in solution above 0.30 mg/L and in more tolerant crops(lettuce, tomato, barley and pepper) at concentrations above 4 mg Cd/L (Page *et al.*,1972), (3) blood pressure disorder in animals, and (4) human health problems such as bone disease, lung edema, renal dysfunction, liver damage, anemia, and hypertension (Adams *et al.*, 2004). Therefore, Cd is one of a very small group of metals for which the Food and Agriculture Organization and World Health Organization (1978) have set a limit for the provisional daily intake (70ug Cd/d) by humans (Adams *et al.*, 2004).

For the above-mentioned reasons, Cd adsorption by and release from soils have been the subject of several recent studies (e.g., Levi-Minzi *et al.*, 1976; Navrot *et al.*, 1978; Jarvis *et al.*, 1980; Abd-El-Fattah and Wada, 1981; Aringhieri *et al.*, 1983; King, 1988a&b; Basta and Tabatabai, 1992; Naidu and Harter, 1998; and Hettiarachchi *et al.*, 2003). These studies showed that Cd adsorption and desorption in soils were influenced by soil physical and chemical characteristics such as clay content, CEC, OM, pH, $CaCO_3$, and oxides of Fe, Mn, and Al.

Since there is a dearth of information on Cd adsorption and release in soils of Egypt, the current investigation was initiated (i) to study the adsorption of Cd in five soils representing the arable land in Egypt, (ii) to determine the exchangeability and extractability of the adsorbed Cd.

MATERIALS AND METHODS

Surface soil samples (0-20 cm) were collected from five soils representing the arable soils in Egypt, i.e., alluvial, calcareous, and sandy soils. The samples were air-dried, ground, and passed through a 2-mm sieve. Pertinent soil characteristics are presented in Table (1) along with soil

locations. Particle-size distribution (pipette method), pH (1:2.5 soil:water ratio), organic matter, OM (Walkley-Black method), carbonate content, $CaCO_3$ (Calcimeter method), and cation exchange capacity, CEC (neutral 1M AMOAC) were determined according to Black *et al.*(1965). Initial total Cd concentration was determined by digesting the soil with 4:1 v/v concentrated HCl to HNO_3 following the method of McGrath and Cunliffe (1985) and DTPA-extractable metals by the method presented by Lindsay and Norvell (1978).

Table 1: Sampling location and pertinent characteristics of the investigated soils.

Soil No	Location	`pH	EC, dS/m saturated paste	O M %	CEC me/100g soil	CaCO$_3$ %	DTPA-extractable (mg/kg soil)				Clay %	Silt %
							Fe	Mn	Zn	Cu		
1	Bohairah Governorate site I	8.24	0.90	1.41	46.30	2.31	6.24	5.21	1.92	2.00	56.02	15.52
2	Bohairah Governorate site II	7.74	6.96	1.30	45.00	1.89	8.40	10.00	2.38	2.05	54.73	14.33
3	Sakkarah City, Giza Governorate	7.70	1.40	1.20	39.38	1.80	7.30	8.50	2.00	2.15	31.90	28.50
4	Nubariya, Bohairah Governorate	8.07	2.53	0.52	14.00	28.53	3.15	3.00	1.26	1.64	25.85	22.50
5	Ismailliya Governorate	8.39	0.72	0.37	4.75	2.09	0.93	1.01	0.76	0.51	19.90	5.00

Adsorption isotherms were obtained by batch studies (Naidu and Harter, 1998). Soil samples (1 g each) were shaken for 24 h at 29 $\pm$ 2 'C in separate 50-ml polypropylene tubes with 25 ml solution containing Cd as $CdCl_2$ ranging in concentration from 0 to 0.80 mg/L. The maximum concentration of Cd added was varied depending on the sorption capacity of the studied soils; the samples were centrifuged at 4000 rpm for 15 min and filtered through Whatman 42 filter papers. The concentrations of Cd in the filtrate were determined using an Inductively Coupled Plasma (ICP) emission spectrophotomer Model Ultima 2-Jobin Yuon. The Cd adsorbed by the soil was calculated as the difference between the amount added and the amount remaining in the equilibrium solution.

The extractability of adsorbed Cd was carried out as follows (Soon, 1981): the tube containing the soil and the entrained solution after the adsorption experiment was resuspended in 25 ml of 1M ammonium acetate, pH 7 and shaken for 2 h. The suspension was centrifuged and filtered as above. The extraction was repeated with a second 25 ml of ammonium acetate. The remaining soil was then extracted twice with 25 ml of 0.125 M copper (II) acetate by shaking for 4 h each time. The filtered extracts were analysed for Cd as before. Ammonium acetate and copper acetate have been used by Riffaldi and Levi-Minzi (1975) and Warman and Thomas (1976) to displace exchangeable Cd and Cd held in coordination complexes, respectively (Soon, 1981).Four soils only were used in the extractability experiment and at three initial Cd concentrations, i.e., 0, 5 and 50 μg/ml. All equilibrations were carried out in duplicate.

Data were fitted to the Langmuir and Freundlich equation using linear regression analysis. Simple correlation analysis of the data was carried out following standard methods of statistical analysis (Snedecor and Cochran, 1971).

RESULTS AND DISCUSSION

Adsorption isotherms.

Results obtained for Cd adsorption isotherms are depicted in Fig.(1). Except soil No.5, all the isotherms have the same shape, in which the isotherm rises steeply at the low Cd equilibrium concentrations while at high equilibrium concentrations the curves became convex in relation to the Y axis.
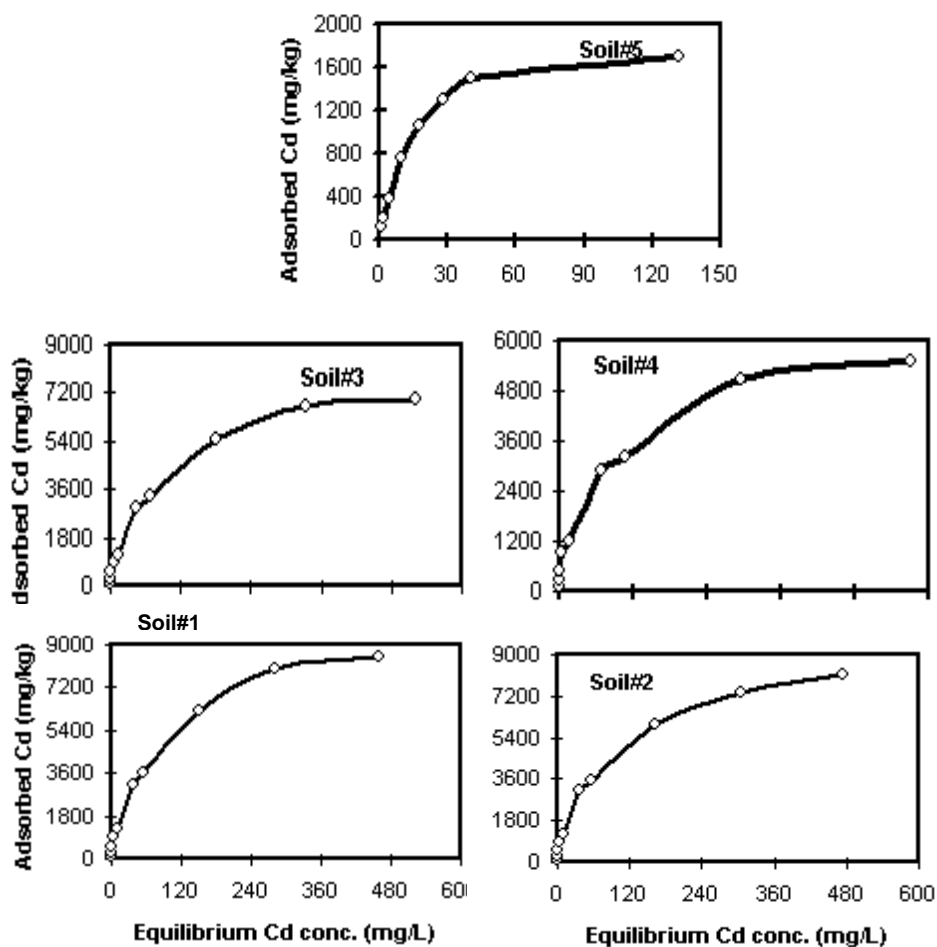

Fig. 1. Cadmium adsorption isotherms for the studied soils. Note the changing scales on the axes of the coordinates.

Similar trends were found for Cd adsorption by soils by Navrot *et al.*(1978) who reported that the first part of the isotherms represents sites with high affinity for Cd as compared to the sites in the second part. This may interpret the different behavior of soil No.5, which contained low contents of clay, OM, and $CaCO_3$ as well as, it may explain the impact of $CaCO_3$ of the calcareous soil (soil No.4), which exhibited almost a similar isotherm to those of the alluvial soils in spite of its relatively low contents of clay and OM (Table 1.).

Over the last 50 or so years numerous soil chemists have attempted to quantify the adsorption processes of various ions with soils by means of models (i.e., Langmuir equation, Freundlich eqution,….etc.). A summary of various models was presented by Travis and Etnier (1981) and Amacher *et al.* (1988).

Numerous researchers have preferred using Langmuir equation because of its usefulness in summarizing a mass of adsorption data into two useful parameters, i.e., maximum adsorption and bonding energy. However, a comprehensive debate has taken place since 1970's about applications and misapplications of the Langmuir equation to soil adsorption phenomena (Veith and Sposito, 1977; Harter and Baker, 1978; Holford, 1978; Harter, 1984 and Schulthess and Dey, 1996). Among the recommendations and conclusions drawn which relates to the present study was "…Where the adsorption maxima for several soils are subsequently correlated with the chemical, and/or physical properties of the soil, the use of such calculated adsorption maxima should not be questioned. Furthermore, it probably makes little difference how the maxima are calculated..". Data of Cd sorbed by unit weight of soil (Q, mg Cd/kg) and equilibrium Cd concentration (C, mg Cd/L) were fitted to linear from of the following adsorption isotherm equations (Fig.2 and 3):

(i) Langmuir equation:

$$C/Q = 1/kb + C/b \quad ,$$

where k (L/mg) is a constant related to energy of adsorption and b (mg Cd/kg) is the maximum adsorption of Cd per unit mass adsorbent, and

(ii) Freundlich equation:

$$Log\ Q = log\ a + n\ Log\ C,$$

where a and n are coefficients, n<1. The coefficient "a" has been reported to be a good index of the relative Cd sorption affinities of soils (Garcia-Miragaya and Page, 1978 and Soon, 1981). Also, a (ug/g) and n (L/kg soil) were reported to be related to maximum adsorption and bonding energy of the adsorbent, respectively (Abd El-Haleem, 2001).

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Fig. 3. Freunlich isotherms fo Cd adsorption by the studied soils. Note
the changing scales on the axes of the coordinates.

The adsorption of Cd by soils obyed the Langmuir adsorption isotherm equation with correlation coefficient of simple determination, r^2, ranging from 0.962 to 0.993 (Table 2).

Concerning the goodness of fit of the data to Freundlich equation, almost similar results to those obtained with Langmuir equation were noticed, with"r^2" values ranging from 0.945 to 0.987 (Table 2). The calculated Cd adsorption maximum from Langmuir equation ranged from 1.885 to 8.830 mg/g (3.35 to 15.71 meq/100g soil) and was slightly more than 70% of the CEC of the sandy and calcareous soils (No.5 and No.4, respectively), while it was about 33% of the alluvial soils' CEC (Table 2). This indicates that Cd ion was not held on the sandy and calcareous soils as strongly as on the alluvial soils, i.e., the exchange cations present in the alluvial soil were more effective in competing with the Cd ion than those present in the sandy or calcareous soil (Cavallaro and McBride,1978). These results are in agreement with those of Levi-Minzi *et al.*(1976), Navrot *et al.*(1978), and Cavallaro and McBride(1978), who found that the retention capacity of Cd was about equal the CEC of the more sandy soils but was only about half the CEC of the more clayey ones. The corresponding values calculated from the Freundlich equation were 0.18 to 0.88 mg Cd/g soil.

Table 2. Coefficients of determination (r^2) for the fit of Cd adsorption data on soils to Langmuir (Lang.) and Freundlich (Freun.) isotherm equations and constants of the tested equations.

Soil No.	r^2		Maxim. adsorption		Bonding energy	
	Lang.	Freun.	Lang.	Freun.	Lang.	Freun.
			b (mgCd/g soil)	a (mgCd/g soil)	k (L/mg Cd)	n (L/kg soil)
1	0.962	0.978	8.83	0.884	29.51	0.354
2	0.969	0.987	8.47	0.539	24.97	0.451
3	0.973	0.969	7.34	0.658	26.41	0.377
4	0.974	0.973	5.65	0.799	49.00	0.323
5	0.993	0.945	1.89	0.179	74.35	0.541

Adsorption Parameters vs. Soil properties.

Correlation coefficients between the parameters (calculated from the studied equations) describing Cd adsorption and some properties of the studied soils are shown in Table (3). Langmuir-maximum adsorption (b) was significantly related to CEC, OM, and clay conten. Inclusion of silt with clay (silt + clay) instead of clay content alone increased the significancy of r (from 0.855, $p < 0.10$ to 0.955, $p < 0.05$). The adsorption maxima, however, was not significantly related to soil pH or $CaCO_3$ content. Several studies on Cd adsorption by soil have reported significant relationships between Cd adsorption and soil pH or CEC. Some studies showed that soil pH was the predominant property that described Cd adsorption (Kuo and Baker,1980). Other studies have shown that CEC was the predominant property affecting Cd adsorption (Singh,1979). Other soil properties that correlated with metal adsorption (e.g., OM and clay content) affect metal adsorption through their CEC property (Basta and Tabatabai,1992; Basta *et al.*,1993).

The bonding energy for the studied soils ranged from 25 L/mg Cdl for the alluvial soil (No.2) to 74 L/mg Cd for the sandy one, soil No.5, (Table 2) and was negatively and significantly correlated with the Cd maximum adsorption of these soils (Table 3). A negative but insignificant correlation was also found in the studies of Cavallaro and McBride(1978) and Salim *et al.*(1996). On the other hand, Basta *et al.* (1992) reported positive and significant correlation between b and k values for Cd adsorption by soils. A very weak correlation (Navrot *et al.*, 1978) or insignificant positive correlation (Levi-Minzi *et al.*, 1976) was also reported. It is worth to note that neither of the two Freundlich equation constants (a and n) was significantly correlated with any of the studied soil properties (Table 3).

Table 3: Simple correlation coefficients between constants of Langmuir(Lang.) and Freundlich (Freun.) adsorption isotherm and some soil properties.

Isotherm	Constant	Soil properties					
		pH	OM	CEC	$CaCO_3$	Clay	Silt +clay
Lang.	b	-0.623	0.925[*]	0.946[*]	-0.155	0.855[†]	0.955[*]
	k	0.763	-0.924[*]	-0.948[*]	0.219	-0.758	-0.952[*]
Freun.	a	-0.263	0.534	0.554	0.303	0.476	0.719
	n	0.284	-0.347	-0.376	-0.284	-0.233	-0.550

†, * = Significant at 0.10 and 0.05 probability levels, respectively.

Release of Cd from soils

The results of exchangeable and complexed Cd from four soils at three initial Cd concentrations; 0, 5 and 50 μg Cd/ml (i.e., 0, 125 and 1500μg/g soil), are shown in Table (4). The exchangeable form was the dominant form of Cd released at 125 or 1500μg Cd rate. The differences in the amounts of exchangeable and complexed Cd at 125μg Cd/g were small for all soils except soil No.5 (sandy soil) and increased with increasing the applied Cd rate. The ratio of exchangeable to complexed Cd increased from 1.37 and 1.57 fold to 2.75 and 3.07 fold for alluvial soils(soils No.1 and 3), respectively, and from 5.63 to 41.22 fold for the sandy soil (soil No.5) with increasing the applied Cd from 125 to 1500μg Cd /g soil. These figures indicate that cation exchange sites assumed to play more important role in adsorbing Cd than complexing sites as adsorption increased. Soon (1981) came to the same conclusion with approximately similar results. On the other hand, no difference was found in the ratio between the two forms (1.23 vs. 1.22 fold) for calcareous soil (soil No.4) with increasing the Cd application rate. Data in Table (4) reveal also that the relative distribution of the adsorbed Cd at the higher two application rates was different from that at the background level (0 μg Cd /g soil). These results may be attributed to the fact that OM, Fe and Mn oxides have a strong affinity for Cd at trace levels (Aualiitia and Pikring, 1987). About 18 or 20% of the adsorbed Cd could not be extracted from alluvial or calcareous soils, respectively, by ammonium and copper acetate solutions, while the corresponding value for sandy soil was only 5% (Fig.4). These results indicate that alluvial and calcareous soil adsorbed more Cd and released less compared with the sandy one. These

results are in agreement with those of Soon (1981), who found that about 82 to 92 percent of the adsorbed Cd was retained by cation exchange and complexing sites.

Table 4. Distribution of adsorbed (Ads.) Cd (μg/g) in soil between exchangeable (Exch.) and complexing (Comp.) sites at three addition levels.

| Soil No. | Added Cd μg / g soil | | | | | | | | |
| | 0 | | | 125 | | | 1500 | | |
	Total	Exch.	Comp.	Ads.[†]	Exch.	Comp.	Ads.[†]	Exch.	Comp.
1	0.54	0.07	0.18	124.95	59.24	43.32	1267.5	718.9	261.1
3	0.63	0.04	0.11	124.48	64.04	41.77	1187.5	696.4	226.7
4	0.99	0.04	0.30	124.80	55.02	44.82	1178.8	486.0	398.1
5	0.25	0.05	0.05	114.03	91.99	16.34	1070.0	940.2	22.8

† Adsorbed Cd calculated as the difference between the added and the remained Cd in equilibrating solutions , the initial adsorbed Cd (at 0 μg/g rate) was not included.
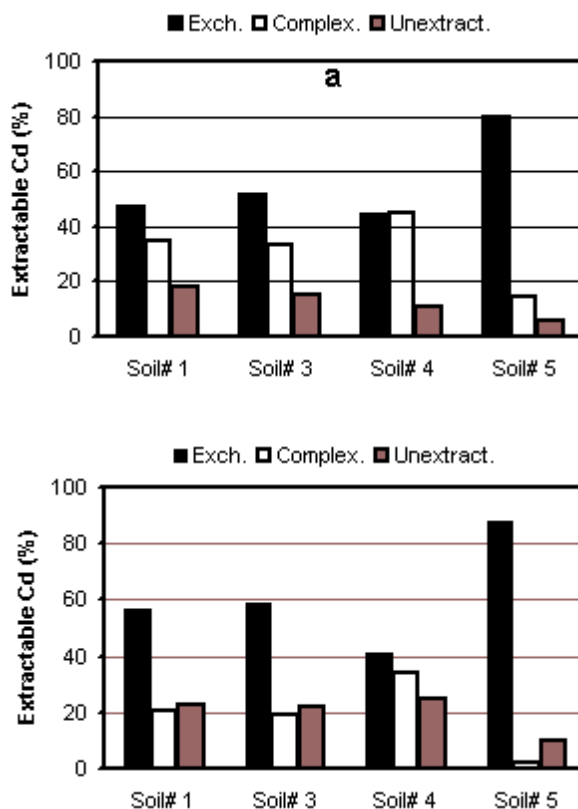

Fig. 4. Average percent distribution of adsorbed Cd in Soil between exchangeable (Exch.), complexed (Complex.), and unextractable (Unextract.) forms at, a, 125 and ,b, 1500 mg Cd/kg soil application rate.

In summary, adsorption of Cd by the three types of soils studied; alluvial, calcareous, and sandy soils, was found to be conformed to the conventional Langmuir equation, as well as, to the Freundlich equation over the concentration range studied. The retention capacity of the studied soils can be ranked as: alluvial > calcareous > sandy soil and best correlated with CEC, "silt + clay", or soil OM content. About 80 to 95% of the adsorbed Cd was subsequently extracted in the exchangeable and complexed forms. The sandy soil released the most and the calcareous one the least. Such results are required in order to understand the fate of Cd and other heavy metals added to the soil through applications of contaminated sludges and wastes and to develop useful environmental guidelines for potential toxic hazards.

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ادمصاص وانطلاق الكادميوم المدمص فى بعض الأراضى المصرية

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أجريت دراسة على ادمصاص وانطلاق الكادميوم المدمص بواسطة خمسة أراضى تمثل الثلاثة أنواع الرئيسية من الاراضى المصرية المنزرعة وهى الرسوبية والجيرية والرملية، وذلك عن طريق عملية الاتزان بين عينات سطحية ممثلة لهذه الاراضى مع محاليل تحتوى على تركيزات تصل الى ٨٠٠ جزء كادميوم فى المليون على صورة كلوريد الكادميوم ($CdCl_2$). تم اختيرت أربعة أراضى عند ثلاث تركيزات لاجراء عملية الاستخلاص بواسطة خلات الامنيوم ١ ع (يمثل الكادميوم المتبادل) ثم بعد ذلك الاستخلاص بواسطة خلات النحاس ٠,١٢٥ ع (يمثل الكادميوم المرتبط بمعقد التربة). أوضحت النتائج أن عملية ادمصاص الكادميوم بالتربة تحت الدراسة تنطبق عليها معادلتى لانجموير و فريندلش فى حدود التركيزات المستخدمة فى الدراسة. وتراوحت السعة العظمى لادمصاص الكادميوم المحسوبة من معادلة لانجموير من ١,٨٨٥ الى ٨,٨٣٠ مجم كادميوم/ جرام تربة (أى ما يعادل ٣,٣٥ الى ١٥,٧١ ميلليمكافئ /١٠٠جم تربة). وكانت هذه القيم ترتبط بعلاقة موجبة ومعنوية ببعض خواص التربة مثل (السلت + الطين)، السعة التبادلية الكاتيونية CEC أو محتوى التربة من المادة العضويةOM وكانت القيم المقابلة المحسوبة من معادلة فريندلش هى ٠,١٨- ٠,٨٨ مجم كادميوم/جرام تربة ولكنها لم ترتبط معنويا بأى من خواص التربة تحت الدراسة . وأوضحت النتائج أيضا أن حوالى٨٠ الى ٩٥ فى المائة من الكادميوم الذى تم ادمصاصة يكون مرتبط على صورة تبادلية exchangeable أو معقدة complexed .

وعملية التبادل الكاتيونى تكون أكثر أهمية من عملية complexation بزيادة كمية الكادميوم المضاف للتربة، فيما عدا التربة الجيرية والتى أظهرت النتائج تساوى أهمية كل من العمليتين السابقتين فى حدود التركيزات المستخدمة فى عملية desorption فى هذه الدراسة.

وهذه المعلومات المتحصل عليها من هذه الدراسة مطلوبة لفهم مصير fate الكادميوم والعناصر الثقيلة الاخرى التى تضاف الى التربة خلال مخلفات المدن والمصانع وغيرها... وذلك من أجل وضع محددات بيئية للآثار السمية المحتملة تحت ظروف الاراضى المصرية.