Arab Gulf J. Scient. Res., Agric. Biol. Sci., **B6** (1), pp. 9-20 (1988)

Zinc Sorption by Some Saudi Calcareous Soils

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ABSTRACT. Zinc sorption was characterized for samples of twenty-two agriculturally important soils in Saudi Arabia. The soil samples had $CaCO_3$ contents ranging from 0.6 - 35.4%, and DTA-extractable Zn contents ranging from 0.19 - 2.42 ug/g. The latter correlated significantly (r=0.469) with the organic matter content of the samples.

Zinc sorption data, obtained by a batch equilibration technique, conformed to both Freundlich and Langmuir equations. The adsorption maxima were very high and ranged from 1260 - 22390 with a mean of 9910 ug/g. Apart from four samples lowest in $CaCO_3$ percentage the adsorption maxima were in excess of the cation exchange capacities (CEC) of the samples. Zinc potentials indicated that, in most cases, Zn did not precipitate as hydroxide or carbonate.

Calcium carbonate percentage and DTPA-extractable Zn originally present in the soil samples accounted for 75% ($r^2 0.001 = 0.652$) and 26% ($r^2 0.05 = 0.423$) of the variability of the adsorption maxima (Sm), respectively. Furthermore, the accountability of $CaCO_3$% was improved to 87% if it and Sm were divided by CEC (*i.e.*, Sm/CEC versus percentage $CaCO_3$/CEC).

Zinc deficiency is of common occurrence in calcareous soils and has been attributed partly to zinc adsorption by carbonates and to its precipitation as zinc hydroxide or carbonate (Thorne *et al.* 1942, Leeper 1952, and Jurinak and Bauer 1956). Layer silicate clays can also adsorb zinc reversibly by cation exchange and irreversibly by lattice penetration (Tiller and Hodgson 1962 and Bingham *et al.* 1964).

Most of the agriculturally important soils of Saudi Arabia are calcareous, sandy and deficient in zinc among other nutrients (Bashour *et al.* 1983). Thus, intensive cropping of these soils will necessitate Zn fertilization for maximizing

their productivity. There is need, therefore, to study zinc sorption by these soils for efficient fertilization because such information is lacking in Saudi Arabia. Furthermore, there is no agreement on the role of $CaCO_3$ in zinc sorption as indicated by previous studies on calcareous and alkaline soil samples of other countries (Udo *et al.* 1970, Singh and Sekhon 1977, Trehan and Sekhon 1977 and Dixit and Dogar 1980). Thus, the main objectives of this study were:

1) to obtain Zn sorption data for important calcareous agricultural soils (Aridisols and Entisols) of Saudi Arabia;
2) to correlate zinc sorption parameters with soil properties, particularly percentage $CaCO_3$, in order to determine whether Zn sorption may be predicted from basic soil properties.

Experimental

Twenty-two surface (0-20 cm) and subsurface (20-40 cm) soil samples were collected by auger, mostly from the same important agricultural areas studied previously by Bashour *et al.* (1983) but not from the exact locations of their studied profiles. In general, the soils belong to the Aridisol and Entisol orders. The samples included in the present study were selected to give a wide range of $CaCO_3$ percentage, from 0.6 to 35.4%. They had no previous history of Zn application. Some of the physicochemical characteristics of these samples are presented in Table 1. Particle-size analysis was carried out by the international pipette method. Soil organic matter was determined by the Walkley and Black method and calcium carbonate equivalent by acid neutralization (Chapman and Pratt 1961). Cation exchange capacity (CEC) was determined by the procedure proposed by Polemio and Rhoades (1977) for calcareous soils. Soil pH was measured electrometrically in a saturated soil paste, and the conductivity of the saturation extract (ECe) was measured by a conductivity bridge (Richards 1954). Zinc was extracted by DTPA (diethylenetriaminepentaacetic acid) and determined by atomic absorption (Lindsay and Norvell 1978).

Zinc sorption data were obtained by a batch equilibration technique. Preliminary studies indicated that a contact time of 24 hr between 1 g soil material and 25 ml of $ZnSO_4$ solution while the suspension was shaken end-over-end by a mechanical shaker at room temperature ($20\pm1°C$) was enough for equilibration. Hence, duplicate 1 g < 2-mm subsamples of each sample were equilibrated with 25 ml of $ZnSO_4$ solutions containing: 5, 20,50, 100, 250 or 500 ug Zn/ml. The suspensions were then centrifuged and the supernatant used for Zn determination by atomic absorption. The determinations were repeated whenever differences between duplications were greater than 5%. The amount of Zn retained was calculated as the difference between initial and final Zn concentrations in solution.

Table 1. Some physico-chemical properties of the soil samples used

Soil sample	Tex-ture	Clay	OM	$CaCO_3$	CEC (meq/ 100g)	SAR	ECe ($mScm^{-1}$)	pH (Paste)	DTPA-extactable Zn (µg/g)
			%						
S_1	cl	34.0	0.62	34.5	13.2	4.7	4.9	7.5	0.72
S_2	ls	12.0	1.24	17.9	7.1	2.8	2.7	7.5	0.50
S_3	scl	24.1	0.76	27.5	7.2	3.1	2.0	7.5	0.22
S_4	sl	12.0	0.33	22.7	8.2	7.1	3.3	7.6	0.22
S_5	ls	9.1	0.55	20.4	4.4	7.2	4.3	7.7	0.19
S_6	ls	6.0	0.94	4.3	8.8	6.6	8.1	7.5	0.40
S_7	sl	4.0	2.40	28.2	16.7	5.5	6.8	7.2	0.42
S_8	ls	10.1	0.51	6.3	11.1	58.6	70.0	7.0	0.25
S_9	cl	30.1	1.14	1.7	18.2	0.7	0.4	7.1	0.33
S_{10}	scl	20.1	4.69	1.4	23.3	0.3	0.7	6.9	0.70
S_{11}	sl	14.1	1.92	1.1	22.1	0.3	0.3	7.3	0.64
S_{12}	s	6.1	2.63	0.6	6.1	1.2	0.8	7.7	2.42
S_{13}	scl	21.0	0.80	15.0	11.1	6.7	6.6	7.4	0.67
S_{14}	scl	23.0	0.88	9.6	10.1	6.3	8.8	7.7	0.47
S_{15}	ls	10.0	0.47	7.4	4.7	2.2	0.7	8.1	0.56
S_{16}	sl	14.1	0.76	8.8	13.3	5.1	7.5	7.5	0.56
S_{17}	scl	15.1	2.26	16.8	23.1	7.6	2.3	7.7	0.82
S_{18}	ls	12.1	2.25	7.0	12.7	4.6	2.5	7.5	0.94
S_{19}	scl	22.1	1.15	8.4	13.3	5.1	4.2	7.4	0.82
S_{20}	sl	18.1	1.64	11.1	12.8	12.6	16.5	7.4	0.67
S_{21}	sl	16.1	0.87	11.9	17.4	1.1	3.0	7.7	0.34
S_{22}	s	8.1	0.62	7.3	5.6	3.4	4.6	7.7	0.48

The following equations were used for evaluating the sorption data:

Freundlich: $\log (x/m) = b \log C + \log a$

Langmuir: $C/(x/m) = \frac{1}{KSm} + \frac{1}{Sm}$

Where: x/m = the amount of Zn sorbed (ug/g), C = the equilibrium Zn concentration (ug/ml), a, b and K are empirical constants and Sm is the adsorption maximum. Correlation and regression analyses were made using standard procedures (Little and Hills 1978).

Results and Discussion

Originally, the soil samples had DTPA-extractable Zn contents ranging from 0.19-2.42 ug/g (Table 1). Most samples had Zn contents below the minimum level (0.8 ug/g) established for similar soils in other countries (Lindsay and Norvell 1978). Extractable Zn correlated significantly at the 5% level (r = 0.469) with organic matter, but not with clay or $CaCO_3$ contents. It seems, therefore, that despite its low level, organic matter plays an important role in Zn reactions in these soils.

Zinc sorption isotherms for loamy sand, sandy loam and sandy clay loam soil samples are presented in Figs. 1, 2 and 3, respectively, for 12 samples containing different $CaCO_3$ levels. The isotherms were nearly linear for samples with a very high percentage of $CaCO_3$ ($\geq$ 17.9%), curvilinear with characteristic plateaus for

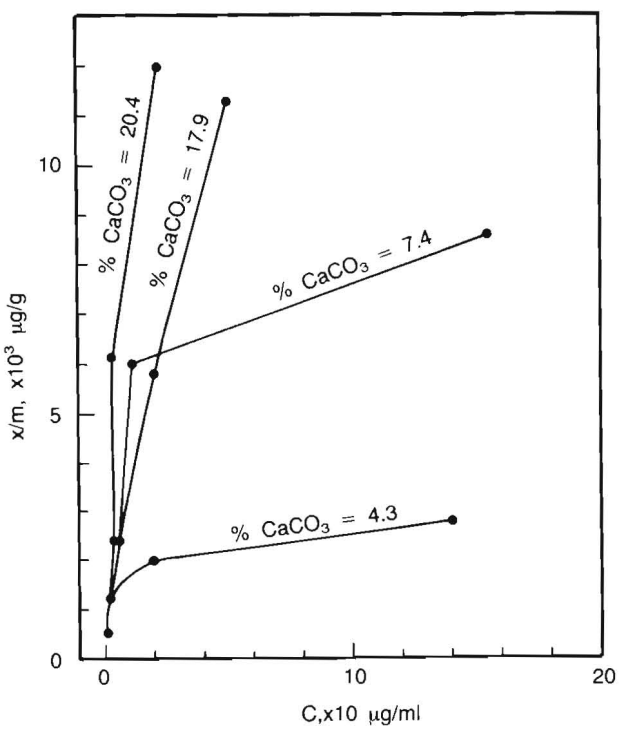

Fig. 1. Zinc sorption isotherms for loamy sand soil samples: S_5, S_2, S_{15} and S_6 containing different $CaCO_3$ levels: 20.4, 17.9, 7.4 and 4.3%, respectively.

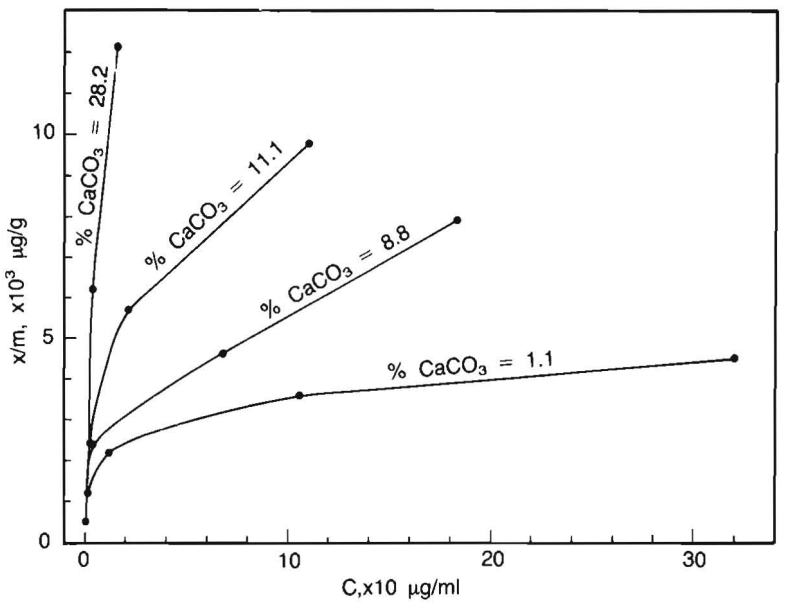

Fig. 2. Zinc sorption isotherms for sandy loam soil samples: S_7, S_{20}, S_{16} and S_{11} containing different $CaCO_3$ levels: 28.2, 11.1, 8.8 and 1.1%, respectively

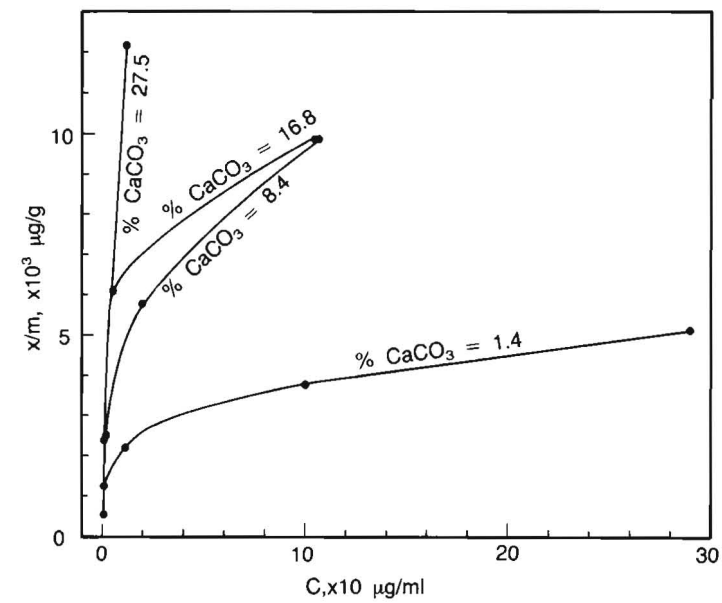

Fig. 3. Zinc sorption isotherms for sandy clay loam soil samples: S_3, S_{17}, S_{19} and S_{10} containing different $CaCO_3$ levels: 27.5, 16.8, 8.4 and 1.4%, respectively.

samples of low $CaCO_3$ levels (< 7%) and assumed in between shapes for the other samples. The effect of $CaCO_3$ percentage on the shape of the isotherms of the remaining ten soil samples was qualitatively similar. The isotherms indicate that at any given equilibrium concentration, Zn sorption increased with increase of $CaCO_3$ percentage. The soil samples completely sorbed the Zn contained in the solutions having the lowest initial concentrations (Ci = 5 ug/ml). At Ci = 20 ug/ml (500 ug/g) - the first experimental points on the sorption isotherms - almost all the Zn was also sorbed by the 22 samples. The amounts sorbed ranged from 480 - 497 ug/g (C.V. = 2%) and constituted about 97 and 99% of the Zn added, respectively. However, at the highest initial Zn concentration (Ci = 500 ug/ml), there were marked variations in the amounts of Zn sorbed by the soil samples. These amounts, for the 22 samples, varied from 1250 - 12212 ug/g (C.V. = 36%) and constituted between 10 and 97% of the Zn added, respectively. These results indicate that the samples have high Zn sorption capacities and that the effect of the soil properties on Zn sorption was most evident at the highest initial concentration. However, at low Zn additions the samples sorbed almost all the Zn and hence the effect of the soil properties was not evident.

The conformity of the sorption isotherm data to linear forms of the Langmuir and Freundlich equations was evaluated by regression analyses. The highly significant correlation coefficients (0.1% level) presented in Table 2 show that, despite the wide range of the initial Zn concentrations, all the sorption data conformed quite well to both equations. Contrary to previous findings, there was practically no need to resolve the sorption isotherms into two or three portions to improve the fit of data, in most cases, to the Langmuir model (Shuman 1975 and Shulka and Mittal 1979). Thus, it seems that on these calcareous soil samples the nature of the Zn-soil interaction was similar over the whole range of the isotherm. For about 8 samples the Freundlich equation yielded higher correlation coefficients (> 1%) than the Langmuir, indicating unlimited sorption for these samples. The sorption data, however, yielded a highly significant (1% level) linear correlation between Freundlich nonexponent constants 'a' and the adsorption maxima, Sm (r = 0.592). Hence, the Langmuir equation was preferred for the forthcoming presentation because it exhibits an adsorption maximum that can be related to soil properties. Though the application of this equation to the data of some of the samples could be an oversimplification, it adequately served the stated objectives of the study.

The adsorption maxima (Sm) obtained were very high ranging from 22390 - 1260 with a mean of 9910 ug/g. This mean value is about 22 times the mean of Sm values reported for eight gleysolic C horizons from Quebec (Karam *et al.* 1983), 22 times the Sm value of B_{2t} horizon of a Rhodic Paleudult from Georgia, USA (Shuman 1975), 6.6 times the mean Sm values of seven Punjab alkaline soils (Singh and Sekhon 1977), and 1.6 times the mean Sm value of 10 Algerian soils (Dixit and

Table 2. Langmuir and Freundlich linear isotherms' constants and coefficients for zinc sorption ($r_{0.001} = 0.654$)

Soil sample	Langmuir constants				Freundlich constants		
	Sm	Sm/CEC	K	r	a	b	r
	$(\mu g/g)\times10^3$		$(ml/g)\times10^3$		$(\mu g/g)\times10^3$	(ml/g)	
S_1	16.28	3.8	115.6	0.656	1.95	0.61	0.938
S_2	14.15	6.1	56.2	0.900	1.06	0.57	0.995
S_3	22.39	9.5	80.9	0.648	1.89	0.66	0.938
S_4	17.18	6.4	98.8	0.849	1.17	0.56	0.686
S_5	16.54	11.5	106.4	0.762	1.89	0.58	0.938
S_6	5.50	1.9	37.1	0.831	1.15	0.25	0.938
S_7	15.30	2.8	260.1	0.894	2.67	0.59	0.980
S_8	10.60	2.9	35.2	0.949	0.85	0.46	0.985
S_9	4.35	0.7	68.7	0.995	0.85	0.28	1.000
S_{10}	5.34	0.7	68.6	0.990	0.82	0.34	0.970
S_{11}	4.57	0.6	97.1	0.995	0.85	0.31	0.970
S_{12}	1.26	0.6	474.3	1.000	0.52	0.17	0.906
S_{13}	8.32	2.3	437.0	0.949	2.12	0.58	1.000
S_{14}	7.09	2.1	99.2	0.854	1.23	0.36	0.970
S_{15}	8.91	5.8	174.6	0.995	1.63	0.36	0.975
S_{16}	7.85	1.8	84.7	0.964	1.51	0.30	0.970
S_{17}	10.32	1.4	227.1	0.995	1.84	0.40	0.843
S_{18}	5.31	1.3	304.1	1.000	1.35	0.27	0.812
S_{19}	10.17	2.3	190.9	0.990	2.00	0.35	0.970
S_{20}	10.11	2.4	161.2	0.990	1.77	0.38	0.995
S_{21}	6.11	1.1	48.7	0.980	0.71	0.39	0.980
S_{22}	10.58	5.8	223.6	1.000	1.81	0.42	0.970

Dogar 1980). The mean bonding energy-related K value of the studied samples was about 0.14 that of the gleysolic C horizons and about 1.5 to 3 times those of the other above-mentioned soils. Thus, it is evident that the studied calcareous samples have very high Zn sorption capacities and moderate bonding energies.

With the exception of the samples with the lowest $CaCO_3$ percentage (S_9, S_{10}, S_{11} and S_{12}), all others had adsorption maxima higher than their CEC'S (Table 2). For one sample (S_5), the adsorption maximum was 11.5 times its CEC. An adsorption maximum in excess of CEC may or may not indicate Zn precipitation depending on the resultant pH and Zn concentration of the equilibrium solutions.

Published research has shown that precipitation may predominate at alkaline pH and high equilibrium zinc concentration (Chester 1965 and Brümmer *et al.* 1983). Because $ZnSO_4$ solutions were used, the equilibrium pH values (pHe) of the suspensions for a given soil are expected to decrease with increasing initial Zn concentration. At a given initial Zn concentration, the differences of pHe among the different soil samples were found to be, relatively, small (Table 3). This may simply be due to their nearly similar initial pH values.

Table 3. Range, mean and coefficient of variation (C.V.) of the equilibrium soil suspensions (pHe) as affected by the initial $ZnSO_4$ concentration used.

Initial zinc concentration	pHe		
	Range	Mean	C.V.
(ug / ml)			%
20	6.9 - 8.4	8.0	6
50	6.1 - 8.1	7.4	8
100	5.9 - 7.5	6.9	7
250	5.7 - 7.0	6.6	6
500	5.4 - 6.7	6.3	6

The pHe decreased as the initial Zn concentrations increased, so that in this experiment low equilibrium Zn concentrations resulted in alkaline media contrary to the high equilibrium concentrations which produced acidic media (Table 3). Because both pHe and the equilibrium Zn concentrations must be taken into consideration when predicting the possibility of precipitation, the solubility product principle was employed for this purpose. Hence, zinc potentials (pZn + 2 pOH) were calculated from equilibrium Zn concentrations (mole/1) and pHe values. At a given initial Zn concentration, zinc potentials exhibited small variations among the different soil samples (Table 4). The data for the individual soil samples (not reported) show that with the exception of S2, S17, S18, and S19 at Ci = 20 ug/ml, all the zinc potential values were higher than those of zinc hydroxide and carbonate (16 - 17), which indicates that most soil samples were undersaturated with respect to these compounds (Jurinak and Bauer 1956). Therefore, in as much as Zn precipitation as hydroxide or carbonate was seemingly not possible for most of the samples, the adsorption maxima in excess of CEC may be due partly to adsorption of Zn as monovalent cations such as $Zn(OH)^+$ along with Zn^{++} (Shulka and Mittal 1979) and partly due to the significant contribution of $CaCO_3$ in Zn sorption.

Table 4. Range, mean and coefficient of variation (C.V.) of the zinc potentials (pZn + 2pOH) in equilibrium solutions corresponding to the initial Zn concentrations

Initial zinc concentration	pZn + 2pOH		
	Range	Mean	C.V.
(μg / ml)			(%)
20	16.3 - 20.0	17.6	5.1
50	17.1 - 20.4	18.2	5.9
100	17.0 - 20.0	18.4	4.5
250	17.3 - 19.4	18.3	3.3
500	17.4 - 19.6	18.2	3.6

The results show that the samples that yielded Sm values lower than their CEC values, contained the lowest $CaCO_3$ percentages, whereas the sample that had an Sm value 11.5 times its CEC had the lowest CEC value. Thus, it seems that $CaCO_3$ contributed significantly to Zn sorption in a manner affected by Soil CEC. This inference was suggested statistically, firstly, by the existance of a highly significant (0.1% level) linear correlation between Sm and $CaCO_3$ percentage ($r = 0.866$) in agreement with previous findings (Udo *et al.* 1970 and Singh and Sekhon 1977), and secondly by the improvement of this correlation when considering Sm/CEC as the dependent and $CaCO_3$ percentage/CEC as the independent variable ($r = 0.934$). This finding indicates that $CaCO_3$ percentage/CEC ratio was a better predictor of Zn sorption than $CaCO_3$ percentage (Sm/CEC = 0.46 + 2.21 $CaCO_3$ percentage/CEC). It further shows that for soils of similar $CaCO_3$ contents, Sm/CEC increased, and Zn precipitation became increasingly important as CEC decreased. Furthermore, zinc precipitation in the samples containing high $CaCO_3$ may be another possibility implying that Zn solubility was governed by factors other than (pZn + 2pOH).

The adsorption maxima did not correlate significantly with organic matter ($r = 0.400$), CEC ($r = 0.332$) or clay content ($r = 0.100$), but correlated significantly (5% level) with DTPA-extractable Zn ($r = 0.510$). It is evident that, contrary to the finding of Trehan and Sekhon (1977), there was a trend for Zn sorption to increase with increase in organic matter (OM). However, the lack of significance may be due to the narrow variation of organic matter in these arid-zone soils. When both $CaCO_3$, and OM percentages were used to predict Zn sorption, a multiple correlation coefficient $R = 0.876$ was obtained. Both $CaCO_3$ and OM percentages accounted for about 77% of the variability of zinc adsorption maxima, whereas percentage $CaCO_3$ alone accounted for about 75% of Zn sorption

variability. Hence, no additional advantage was gained using OM as a second independent variable.

The role of CEC on Zn sorption was overshadowed by the pronounced contribution of $CaCO_3$ percentage. The lack of correlation between Sm and CEC also indicated that Zn sorption in these calcareous soils is not a simple reversible exchange reaction. The adsorption maxima did not relate to ECe or SAR of the soils. It seems that their effects were masked by the dissolution of $CaCO_3$ in the equilibrium solutions.

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(Received 29/12/1986;
in revised form 13/05/1987)

ادمصاص الزنك على بعض الترب الجيرية بالمملكة العربية السعودية

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أجريت الدراسة في اثنتين وعشرين عينة تربة من مناطق مهمة زراعياً في المملكة. تراوحت نسبة كربونات الكالسيوم في عينات الاختبار بين ٦, و٤, ٣٥٪، كما تراوح الزنك المستخلص فيها بين ١٩, ٠ و ٤٢, ٢ ميكروجرام/ جرام. وقد وجد أن الزنك المستخلص في العينات يرتبط معنوياً مع محتوى المادة العضوية فيها (معامل الإرتباط = ٤٦٩, ٠).

طابقت بيانات ادمصاص الزنك في الترب معادلتي كل من فرويندلخ ولانجمير وتراوحت قيم الادمصاص القصوى بين ٢٦, ١ و ٣٩, ٢٢ مليجرام/ جرام وفيما عدا أربع عينات ذات نسبة منخفضة من الجير تجاوزت قيم الادمصاص القصوى السعة التبادلية الكاتيونية للعينات، ولقد دلت حسابات جهد الزنك عدم ترسيبه على هيئة ايدروكسيد أو كربونات.

بينت تحاليل الارتداد أن نسبة كربونات الكالسيوم في العينات والزنك المستخلص فيها يعبران على التوالي عن ٧٥٪ و ٢٦٪ من التغيرات في قيم الادمصاص القصوى، وأن العلاقة بين قيم الادمصاص القصوى ونسبة كربونات الكالسيوم تتحسن كثيراً بقسمة كل منهما على السعة التبادلية الكاتيونية لكل عينة.