Kinetics of Zinc Sorption by Some Calcareous Soils of Iran

A. Reyhanitabar[1*], M. M. Ardalan[2], N. Karimian[3], G. R. Savaghebi[2], and R. J. Gilkes[4]

ABSTRACT

The kinetics of Zn sorption by ten calcareous soils of Iran were measured and fitted to several equations (zero-, first-, second-, third-order, parabolic diffusion, simple Elovich, and exponential equations). Two initial concentrations of zinc i.e. 20 and 80 mg Zn l^{-1} were used and observations were carried out for 100 hours. The time required to reach the near equilibrium state was about 24 hours, by which time most of the Zn in solution had been sorbed. Zero-order, first-, second-, and third-order equations did not adequately describe zinc sorption kinetics. Both simple Elovich and exponential equations did adequately describe the data. Regression analysis indicated that the total $CaCO_3$, active $CaCO_3$, clay content, and specific surface area were closely related to the coefficients of these two equations. The magnitude coefficients of the equations were positively related to these soil properties. Thus, Zn sorption kinetics can be predicted from data collected during routine soil evaluation.

Keywords: Aridisols, Carbonate, Entisols, Kinetic models, Soils of Iran, Zinc sorption.

INTRODUCTION

Most studies on zinc sorption and desorption by soils have been based on equilibrium conditions. However, due to slow chemical reactions, plant uptake, fertilizer addition, and other factors, agricultural soils are rarely in an equilibrium state with regard to Zn sorption and release. Considerable research work including detailed modelling of kinetic equations has been reported for the kinetics of sorption of phosphorus and potassium by soils. There are few comparable descriptions of the kinetics of Zn sorption by soils (Sparks *et al.,* 1980; Dang *et al.*, 1994; Taylor *et al,.* 1995) and there are none for Iranian soils. The kinetic equations used in the studies of sorption of ions by soils are diverse (Table 1) and include: zero-, first-, second- and third-order kinetics, parabolic diffusion, Elovich and exponential equations (Griffin and Jurinak,1973; Kuo and Lotse, 1974; Chien and Claytone, 1980; Aharoni and Suzin, 1982; Sharply, 1983; Sparks, 1989; Aharoni *et al.,* 1991; Dang *et al.,* 1994; Sparks, 1995; Taylor *et al.,* 1995). There has been no evaluation of the most appropriate equation for describing Zn sorption by calcareous Entisols and Aridisols such as those that occur in Iran. In this region the quantity and also the quality of clay and calcium carbonate has a major influence on Zn sorption in soils, whereas organic matter shows little effect, probably due to the low OM content of these soils and not to the OM itself (Reyhanitabar *et al.,* 2007).

The objectives of this study were to compare the suitability of different kinetic equations to describe Zn sorption by some calcareous soils of Iran and to investigate the

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[1] Department of Soil Science, Faculty of Agriculture, University of Tabriz, Tabriz, Islamic Republic of Iran.

[*] Corresponding author, e-mail: areyhani@tabrizu.ac.ir

[2] Department of Soil Science, Faculty of Soil and Water , University College of Agriculture and Natural Resources, University of Tehran, Karaj, Islamic Republic of Iran.

[3] Department of Soil Science, Faculty of Agriculture, Shiraz University, Shiraz, Islamic Republic of Iran.

[4] School of Earth and Environment, Faculty of Natural and Agricultural Science, University of Western Australia, Crawley, WA 6009, Australia.

Table 1. Kinetic models used in this study.

Model	Equation	Parameters
Zero-order reaction	$q_t = q_0 - k_0 t$ [a]	k_0, Zero-order rate constant (mg Zn kg^{-1} min^{-1})
First-order reaction	$\ln q_t = \ln q_0 - k_1 t$	k_1, first-order rate constant (min^{-1})
Second-order reaction	$1/q_t = 1/q_0 - k_2 t$	k_2, second-order rate constant[(mg Zn kg^{-1})$^{-1}$]
Third-order reaction	$1/q_t^{\,2} = 1/q_0^2 - k_3 t$	k_3, third-order rate constant [(mg Zn kg^{-1})$^{-2}$ min^{-2}]
Parabolic	$q_t = q_0 + k_p t^{0.5}$	k_p, diffusion rate constant
Exponential	$q_t = a\, t^{\mathbf{b}}$	**a**, Zn sorption magnitude constant [(mg Zn kg^{-1}(min^{-1})^b] and **b** sorption rate constant
Simple Elovich	$q_t = 1/\beta_s \ln(\alpha_s \beta_s) + 1/\beta_s \ln t$	α_s, initial Zn sorption constant (mg Zn kg^{-1} min^{-1}) and, β_s, Zn sorption rate constant [(mg Zn kg^{-1})$^{-1}$]

[a] q_0 and q_t are the amount of Zn sorption (mg Zn kg^{-1}) at time zero and t, respectively.

relationships between the parameters of the various equations and the physical and chemical properties of these soils. Results may be used in identifying the optimum desorption times for these complex calcareous soils where soil solution Zn is controlled by diverse sets of sorption/desorption and crystallisation/dissolution processes (Udo *et al*., 1970; Uygur and Rimmer, 2000).

MATERIALS AND METHODS

Surface soil composite samples (0-30 cm) were collected from 10 locations in the central parts of Iran (Ghazvin Plain, Tehran Province, and Qom Province), representing 10 soil series in two taxonomic orders, namely, Entisols and Aridisols. The samples were air dried, passed through a 2-mm sieve, and used for the determination of their physical and chemical properties. The following analytical procedures were employed: clay by the hydrometer method (Day, 1965); pH of 1: 4 soil to 0.01 M CaCl$_2$ suspension by glass electrode; organic carbon (OC) by the Walkley-Black method (Allison, 1965), cation exchange capacity (CEC) by replacing exchangeable cations with sodium in sodium acetate (NaOAc) solution, removal of excess NaOAc by washing with alcohol, exchanging Na by ammonium acetate

(NH$_4$OAc) and determining the Na concentration in the extract by flame photometry (Chapman, 1965); and total CaCO$_3$ by acid (HCl) titration (Allison and Moodie, 1965). Because the total carbonate of soil (CCE) determined by acid digest is a poor measure of carbonate reactivity, the active CaCO$_3$ equivalent (ACCE), which is fine particle-size calcite, was determined by the NH$_4$-oxalate method (Drouineau, 1942). Specific surface area (SSA) was measured using the N$_2$-BET method (Brunauer *et al*., 1938) with a Micrometric Gemini III 2375 surface area analyzer. Available zinc was measured with Diethylenetriaminepent-aacetic acid (Lindsay and Norvell, 1978). The properties of the soil samples are listed in Table 2. Clay contents range from 106 to 410 g kg^{-1}, pH (CaCl2) from 7.3 to 7.8, organic carbon from 6.3 to 15.5 g kg^{-1}, and SSA from 10 to 39 m² g^{-1}. Calcium carbonate equivalent ranged from 38.0 to 228.0 g kg^{-1} and active CaCO$_3$ equivalent ranged from 13.4 to 99.8 g kg^{-1}. DTPA extractable Zn values ranged from 0.7 to 4.40 mg kg^{-1}. X-ray diffraction (XRD) analysis of random powders identified abundant quartz and showed that calcite was the sole carbonate mineral. X-ray diffraction patterns of the clay fraction identified illite, chlorite, and smectite as the major clay minerals in all of the soil samples (data are not shown).

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Table 2. Measured characteristics of the samples of 10 calcareous Aridisols and Entisols used in this study.

Soil No	Soil Series	pH	CEC (cmol$_c$ kg^{-1})	Cla	CCE	ACCE	OC	SSA (m² g^{-1})	Zn [a] (mg kg^{-1})
				(------------g kg^{-1}--------------)					
1	Kooskak	7.63	16.4	325	80	38.2	8.3	26	2.3
2	Saeed Abad	7.70	12.4	106	46	19.2	9.3	10	2.2
3	Ahmadabad[b]	7.72	19.2	226	72	27.2	8.3	20	4.4
4	Kord Amir	7.80	16.3	308	136	62.6	11.2	19	2.3
5	Chitgar	7.30	24.6	188	98	13.4	9.2	16	7.0
6	Firoozkooh	7.77	20.2	307	228	99.8	15.5	39	2.6
7	Yaftabad	7.66	14.8	165	38	27.3	7.3	13	2.3
8	Damavand	7.60	19.2	410	94	40.8	9.2	39	1.9
9	Roodhen	7.51	32.0	134	194	58.5	6.3	12	1.2
10	Karaj	7.66	17.0	142	108	34.7	6.4	21	0.7
	Minimum	7.30	12.4	106	38	13.4	6.3	10	0.7
	Maximum	7.80	32	410	228	99.8	15.5	39	7
	Mean	7.64	19.2	231	109	42.2	9.1	21.5	2.7
	St. deviation	0.1	5.0	100.0	60.1	25.5	2.6	10.4	1.8

[a] DTPA-extractable Zn, [b] Soil taxonomy is Aridisols and remainder are Entisols.

Kinetics Experiment

Sorption kinetics was studied using duplicate 1 g soil samples in 50 ml centrifuge tubes. Twenty millilitres of 20 mg Zn l^{-1} and 80 mg Zn l^{-1} solutions of ZnSO$_4$ in 0.01M CaCl$_2$ were added to the soil samples. The suspensions were shaken for 0.5, 1, 3, 6, 9, 12, 24, 36, 48, 60 and 100 hours at 25°C in a constant temperature chamber. At the end of the shaking period, the suspension was filtered, and the filtrate was analyzed for Zn by ICP-MS (Inductively Coupled Plasma Mass Spectroscopy) using a Perkin-Elmer Elan instrument. Differences in the measurements data between duplicate samples were generally small; therefore, average values were considered henceforth. The kinetic equation parameters and physicochemical properties of the soils were set as the dependent and the independent variables, respectively, for statistical analysis of bivariate and multivariate relationships.

RESULTS AND DISCUSSION

Zinc sorption characteristics of the studied soils have been previously reported by authors (Reyhanitabar *et al*., 2010). In spite of the fact that precipitation of zinc is probable in alkaline condition of calcareous soils, based on our previous study, zinc does not necessarily precipitate in the studied soils. A variable pressure scanning electron microscopy (VPSEM) showed that zinc was uniformly distributed in the soils matrix up to an initial concentration of 500 mg Zn l^{-1} to the limit of the spatial resolution of this technique**.** There were no local concentrations of zinc or spatial associations of other elements with zinc and there was little indication of zinc precipitation (Reyhanitabar *et al.,* 2010).

Plots of zinc sorption versus time for three soils i.e. soil 1, 5, and 7, and the two initial Zn concentrations are shown in Figure 1 and are representative of the data for all soils. Zinc sorption increased rapidly up to about 24 hours (1,440 minutes) beyond which a near steady-state condition was attained where most added Zn (380,385 and 400 at C1 and 1,300, 1,450 and 1,600 mg Zn kg^{-1} soil at C2) had been sorbed. As shown in Figure 1, for both initial concentrations (20 and 80 mg Zn l^{-1}), the sorption rate and amount of Zn sorbed after 6,000 minutes were different for the three soils, presumably

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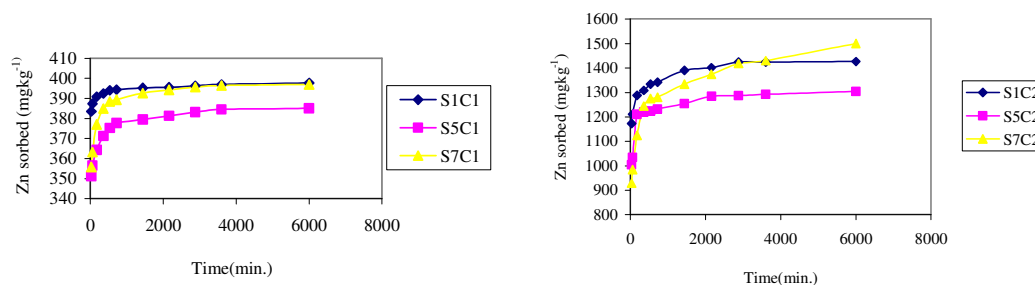
Figure 1. Zinc sorption with time for three soils and two initial concentrations of zinc (C1 is 20 and C2 is 80 mg l^{-1}).

reflecting differences in soil properties. The zero-, first-, second- and third-order equations, the parabolic diffusion, Elovich and exponential equations were tested for their suitability to describe the kinetics of Zn sorption. Sorption kinetics were poorly described by the zero-, first-, second-, third and parabolic equations as indicated by low values of R^2 (Table 3). Several equations that provided a poor description of the data exhibited a systematic departure of data points from the fitted curves (e.g. parabolic diffusion equation, Figure 2, A). High values for the coefficient of determination (R^2) indicated that the best models for describing the data for all soils were the exponential and the simple Elovich equations (Table 3, Figure2 B and C). Dang *et al.* (1994) and Taylor *et al.* (1995) obtained similar results for Zn sorption by Ultisols and Vertisols. According to Aharoni and Ungarish (1976), when sorption data are plotted as reciprocal rate, $Z= (dq/dt)^{-1}$ versus time, a straight line is produced, if sorption follows Elovich

kinetics. Plots of Z against time (e.g. soil 1, C2, Figure 3) are linear with $R^2 = 1$ for the present data, indicating the applicability of Elovich kinetics. The amounts of Zn sorbed by these soils for times up to 100h were predicted using the fitted simple Elovich and exponential equations (Figure 4 A and B). The excellent agreement between the predicted and the measured sorbed Zn for both soils confirms that the simple Elovich and the exponential equations accurately describe Zn sorption by these soils, although there are small systematic departures of the data points from the regression lines for both equations. Values of simple Elovich parameters for the soils are shown in Table 4, because of high numerical values of α_s, its value is shown as *ln* α_s. The Elovich equation reduces to a simplified version for the circumstances where α_s β_s *t>1*. The minimum observed value of α_s for an initial concentration of 80 mg Zn l^{-1} was 2,980 and the product α_s β_s was 25.03 for the first minute, therefore, the simplified form was

Table 3. Range and mean of values of the coefficient of determination (R²) for the seven kinetics equations for two initial concentration of Zn.
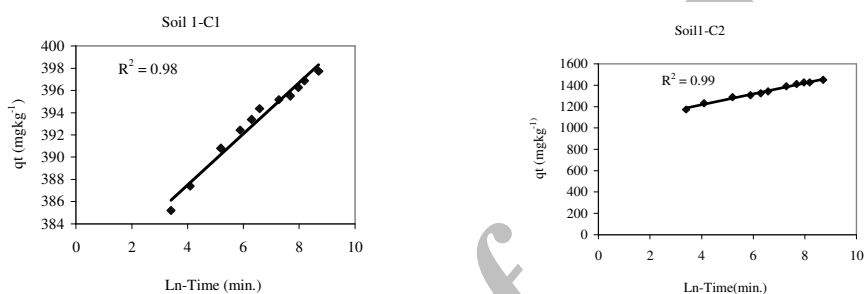
Equation	R²(C1 [a])		R²(C2 [b])	
	Range	Mean	Range	Mean
Simple Elovich	90-98	94	95-99	98
Exponential	90-98	93	94-99	97
Parabolic diffusion	62-82	70	71-85	80
Zero-order reaction	40-60	48	48-67	58
First-order reaction	40-59	47	46-63	55
Second-order reaction	39-58	46	44-60	52
Third-order reaction	39-57	46	42-58	48

[a] 20 mg Zn kg^{-1}, [b] 80 mg Zn kg^{-1}, as initial concentrations of Zn.
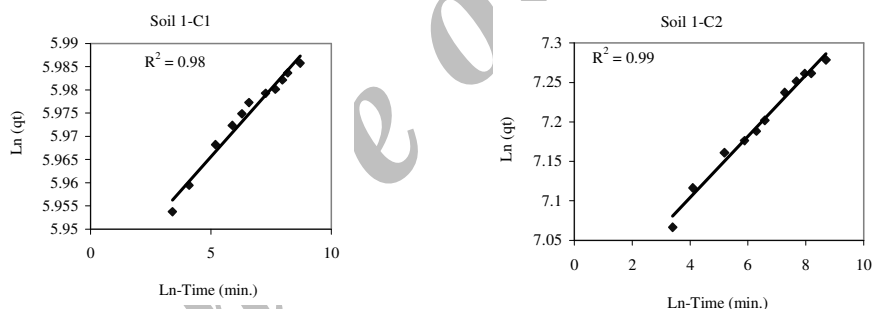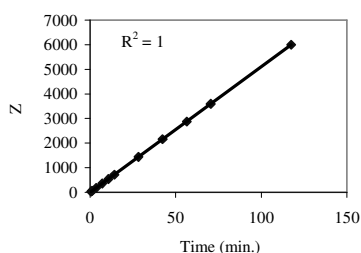
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(A)
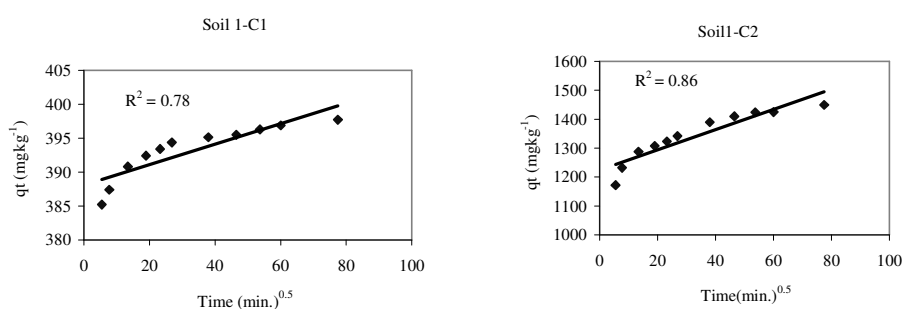
(B)

(C)

Figure 2. Zn sorption data for soil number 1, (A) fit of parabolic diffusion equation, (B) fit of simple Elovich equation, (C) fit of exponential equation. q_t is the amount of Zn sorption (mg Zn kg^{-1}) at time t.

Figure 3. Plot of Z (inverse sorption rate) versus time for soil number 1.

adopted. The constants α_s and β_s can be used to compare Zn sorption kinetics of soils under the same experimental conditions. The slope of the plots for the simple Elovich equation changes with solution concentration and solution to soil ratio (Sharpley, 1983) and, consequently, can not be directly compared under different experimental conditions (Sparks, 1995). For example, in the present research the values of α_s and β_s for 20 mg Zn/l

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Table 4. Values of exponential and Elovich equation coefficients for Zn sorption by Iranian soils. Unit of "a" coefficient is (mg Zn kg^{-1}(min^{-1})^{b} and b is dimensionless and for αs and βs units are (mg Zn kg^{-1} min^{-1}) and (mg Zn kg$^{-1)}$ $^{-1}$, respectively.

Soil series	Initial concentration	Exponential equation		Simple Elovich equation	
		a	**b**	*ln α$_s$*	*β$_s$*
Kooskak	C1 [a]	378	0.0059	165	0.43
	C2 [b]	1041	0.0388	23	0.019
SaeedAbad	C1	295	0.037	24	0.074
	C2	576	0.107	8	0.0084
AhmadAbad	C1	351	0.015	60	0.17
	C2	788	0.078	11	0.010
KordAmir	C1	380	0.0053	183	0.48
	C2	1011	0.049	18	0.015
Chetigar	C1	331	0.018	50	0.15
	C2	867	0.050	18	0.017
Firoozkooh	C1	391	0.0023	427	1.09
	C2	1284	0.024	39	0.028
Yaftabad	C1	342	0.018	50	0.14
	C2	699	0.089	10	0.0094
Damavand	C1	383	0.0048	203	0.52
	C2	1175	0.036	25	0.019
Roodhen	C1	374	0.0082	117	0.31
	C2	1100	0.044	20	0.015
Karaj	C1	343	0.017	53	0.15
	C2	801	0.073	12	0.011

[a] 20 mg Zn kg^{-1}, [b] 80 mg Zn kg^{-1}, as initial concentrations of Zn.
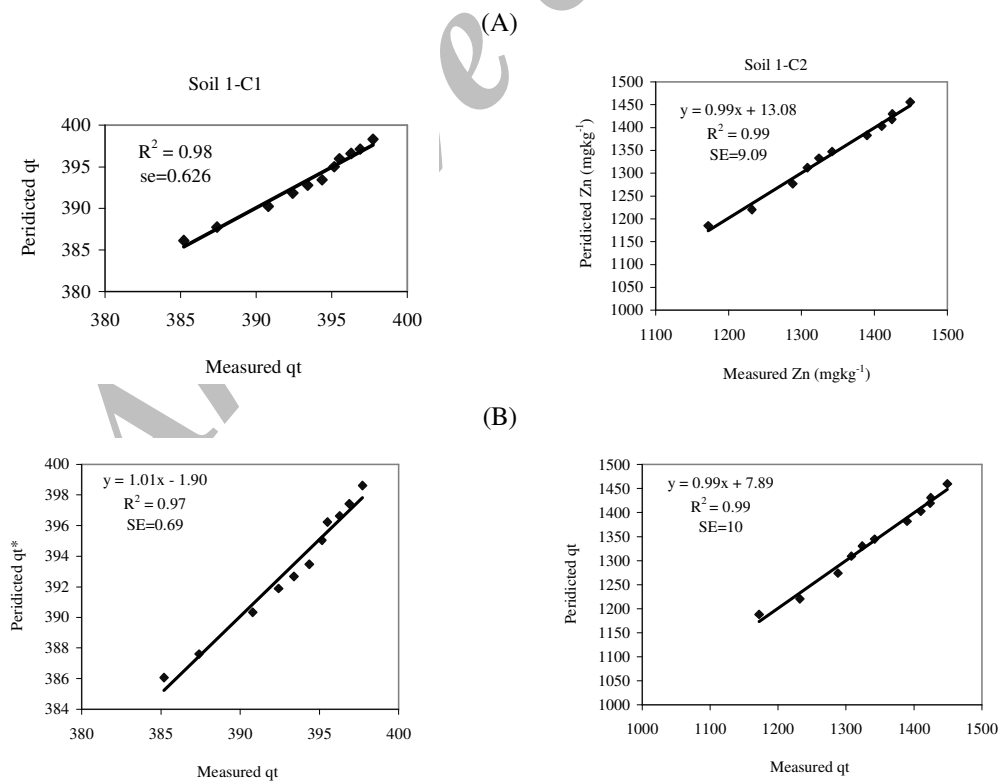
Figure 4. Relationship between predicted and measured values of zinc sorbed for soil number 1 for the two initial concentrations of Zn (C1= 20 mg l^{-1}, C2= 80 mg l^{-1}). The predicted values were calculated from: (A) the simple Elovich equation. (B) the exponential equation. q$_t$ is the amount of Zn sorption (mg Zn kg^{-1}) at time t.

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initial concentration are much higher than the values for 80 mg Zn l^{-1}. Some researchers have suggested that multiple linear segments in simple Elovich plots could indicate a change from one type of binding site to another (Atkinson *et al.,* 1970). However, such mechanistic suggestions may not be correct (Sparks, 1989) and, for these Iranian soils, there is not an evident separation of the simple Elovich plots into two discrete linear segments (Figure 2B). Values of the *a,* and *b* coefficients for the exponential equation are shown in Table 4 and both coefficients differ greatly between soils. The values of **a** for 20 mg Zn l^{-1} initial concentration are lower than for 80 mg Zn l^{-1} initial concentration. Constant *a* is equal to the Zn sorbed *qt* when *t= 1s*. This constant is a measure of the capacity of the soil to sorb Zn, which, at constant pH, reflects soil properties such as specific surface area, mineralogy, and organic matter content (Udo *et al.,* 1970). Constant *b* is a measure of the rate of sorption of Zn by surface sites and

reflects the strength of bonds between Zn ions and the surface of soil particles, although some workers consider that it may also reflect internal diffusion within soil particles (Barrow, 1986). Both *a* and *b* coefficients vary greatly between soils reflecting the substantial differences in soil properties.

Effect of Soil Properties on Zinc Sorption

The values of the coefficients of the exponential and simple Elovich equations were related to soil chemical and physical properties by statistical analysis (Table 5). There are similar bivariate relationships for the two initial Zn concentrations, except for the OC content of the samples. For the exponential function, clay has positive linear relationships with *a* and *b* and both specific surface area and calcium carbonate equivalent are positively related to *b*. Active

Table 5. Correlation coefficients (r) for linear relationships between soil properties and constants of the two kinetic models that best described Zn sorption.

Soil properties	Initial concentration	Exponential equation		Simple Elovich equation	
		a	*b*	*Ln α$_s$*	*β$_s$*
Clay	C1	0.73*	0.70*	0.64*	0.64*
	C2	0.66*	0.65*	0.63*	0.63*
Silt	C1	0.47	-0.48	0.44	0.44
	C2	0.43	-0.42	0.43	0.43
SSA	C1	0.68*	-0.67	0.80**	0.80**
	C2	0.75*	-0.69*	0.80**	0.77**
OC	C1	0.35	-0.34	0.81**	0.81**
	C2	0.49	-0.44	0.69*	0.69*
CEC	C1	0.33	0.37	0.11	0.1
	C2	0.46	-0.5	0.32	0.32
pH	C1	0.22	-0.11	0.38	0.38
	C2	0.04	0.14	0.07	-0.03
CCE	C1	0.64*	-0.63*	0.70*	0.74*
	C2	0.78*	-0.72*	0.76**	0.72*
ACCE	C1	0.77*	-0.72*	0.90**	0.90**
	C2	0.75*	-0.69*	0.81**	0.74*
DTPA-Zn	C1	-0.3	-0.18	-0.21	-0.21
	C2	-0.2	0.008	0.08	0.03
Olsen-P	C1	0.61	-0.6	0.61	0.62
	C2	0.51	-0.54	0.54	0.56

* and ** are significant at 0.05 and 0.01 levels of probability, respectively.

calcium carbonate shows a strong positive relationships with *a* and a negative relationship with *b*. Active calcium carbonate had the strongest overall relationships with both *a* and *b*. Clay, calcium carbonate equivalent, active carbonate, and specific surface area are positively correlated with *ln α_s* and *β_s* parameters of the simple Elovich equation which is consistent with the relationships observed for the coefficients of the exponential function. However organic carbon is also strongly, positively related to the simple Elovich coefficients, which was not the case for the exponential coefficients (Table 5). Again, ctive calcium carbonate has the closest relationships with both coefficients (β_s and ln α_s). The finding that clay and organic carbon content have strong relationships with *β_s* and *ln α_s* is similar to that reported by Taylor *et al.* (1995) for Zn sorption by soil but these authors reported that CEC was also strongly correlated with *α_s* and *β_s*. This was not observed in our study. The present research indicates that it is active calcium carbonate that is generally most closely related to the coefficients of the equations, but these statistical associations can not be assumed to indicate that Zn is mostly sorbed by fine carbonate. In particular, several authors have concluded that, at adequate Zn concentrations in soil solution, various Zn hydroxide and carbonate minerals may precipitate (Udo *et al.,* 1970; Uygur and Rimmer, 2000).

Step-wise multiple regression equations (Table 6) greatly increased the accuracy of the prediction of equation coefficients relative to the simple bivariate relationships considered in Table 5. The exponential equation *b* coefficient for the higher initial Zn concentration is very accurately predicted ($r^2= 0.98$) by an equation containing clay, pH and active calcium carbonate. Stepwise regression showed that SSA, ACCE and CCE are the soil characteristics that are most predictive of the coefficients of the simple Elovich equation.

CONCLUSION

Statistical analysis has indicated that clay, SSA, ACCE and CCE may play important roles in determining the amount and rate of adsorption of Zn by calcareous soils of Iran. However, these statistical associations do not provide direct evidence of the Zn sorption processes in these soils. It might be inferred from these findings that the agronomic effectiveness of Zn fertilizer will decrease as elevated levels of clay and carbonate increase the amount of Zn sorbed by the soil. Furthermore, as these two basic soil properties are commonly determined in soil and land capability surveys, this information might be used as a basis for developing recommendations for application rates of Zn fertilizers.

ACKNOWLEDGEMENT

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Table 6. Step-wise multiple regression equations relating soil properties and the rate constants for exponential and simple Elovich equations for Zn sorption at two initial concentrations (C1 and C2)**.**

Rate constants	Step-wise regression equation	R^2
b-C2-exponential	-0.83-0.0011 clay+0.13pH+0.011ACCE	0.98**
ln (α_s)-C1-Elovich	-87.2+3.11 ACCE+5.15 SSA	0.96**
ln (α_s)-C2-Elovich	-10.2+0.41 SSA+1.24 CCE	0.99**
β_s-C1-Elovich	-0.203+0.0081 ACCE+0.013 SSA	0.96**

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سینتیک جذب روی توسط برخی خاکهای آهکی ایران

ع. ریحانی تبار، م. م. اردلان، ن. کریمیان و غ. ر. ثواقبی، ر. ج. گیلکس

چکیده

سینتیک جذب روی در ده خاک آهکی از ایران اندازه گیری و به معادلات گوناگون سینتیکی برازش داده شدند (معادلات سینتیکی درجه صفر،اول،دوم،سوم،پارابولیک پخشیدگی، الوویچ ساده شده و معادله تابع توانی). دو غلظت اولیه روی یعنی ۲۰ و ۸۰ میلی گرم بر لیتر مورد استفاده قرار گرفته و مشاهدات در طول ۱۰۰ ساعت انجام گرفت.زمان مورد نیاز برای رسیدن به شبه تعادل ۲۴ ساعت به دست آمد که در آن بیشتر روی محلول توسط خاکها جذب می شوند.معادلات درجه صفر،اول، دوم، و درجه سوم نتوانستند سینتیک جذب روی را توصیف کنند ولی معادلات الوویچ ساده شده و تابع توانی بخوبی داده ها را توصیف کردند. آنالیز رگرسیونی نشان داد که کربنات کلسیم کل،کربنات کلسیم فعال،مقدار رس و سطح ویژه خاکها بطور نزدیکی به ضرایب این دو معادله مربوط میشوند.ضرایب این معادلات بطور مثبت و معنی دار به خواص خاکی ارتباط پیدا می کنند.بنابراین سینتیک جذب روی را می توان از روی داده هایی که بطور معمول در ارزیابی خاکها اندازه می گیرند به دست آورد.

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