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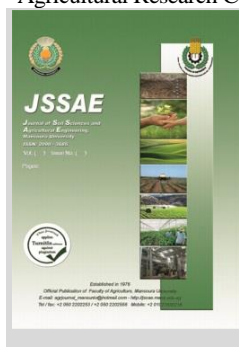## Manganese Release Kinetics in some Calcareous Soils from the Sulaimani Governorate in Iraqi Kurdistan Region

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ABSTRACT

This study was carried out in the Sulaimani Governorate, Iraqi Kurdistan Region, to estimate manganese release kinetics in five calcareous soils including (Sharazor, Qaradagh, Bazian, Mawat, and Surdash). The highest amount of Mn release during 48 hours was least in soil Surdash (64.00 mg kg^{-1}), while Soil Mawat exhibited the highest release (541.82 mg kg^{-1}). The value of first order release rate constant (k_1) from first order differed from 0.076 to 0.102 with a mean of (0.091 h^{-1}). The mean values of the rate constant (a and b) from the power function model were (2.128 mg Mn kg^{-1}(h^{-1})^b and 0.916 mg Mn kg^{-1})$^{-1}$) respectively varied widely with the five soil. The value of second order release constant (k_2) from second order model, varied from (-0.012) to (-0.005) with a mean of (-0.008 mg Mn kg^{-1})$^{-1}$. The first order, power function, and second order were the best-fitted model used to describe Mn release process very well in the investigated soils.

Keywords: Manganese, release, Kinetic, calcareous soils, first order, power function and second order.

INTRODUCTION

The availability of manganese to plants depends on their oxidation state: the modified form of (Mn^{4+}) is not available to plants, while the reduced form (Mn^{2+}) is available to them. Generally, oxidation reactions are primarily biological, however, the reduction can be both biological and chemical in nature (Rengel, 2000). When the growth medium is reduced in oxygen, NO^{3-}, Mn, and Fe act as elective electron acceptors for microbial respiration and are converted to decreasing ionic species. This process increases the solubility and availability of manganese and iron. The most influential soil characteristics that control Mn species in soil solution are redox potential and pH. The oxidation states for Mn are +2 representing MnCO3, +3 representing MnOOH and +4 representing MnO2, respectively, where the amounts of pe + pH are less than 14, 14 to 16.6 and greater than 16.6. (Lindsay, 1991). Ghasemi-Fasaei *et al*., (2012) showed a higher Mn release in clay soils compared to sandy loam soils, this was significantly related to higher initial rates of Mn release in the former than in the latter. Differences in the nature and content of Mn-bearing minerals between soils, according to Krishnamurti and Huang, (1992), result in changes in Mn release characteristics and plant availability. The desorption of sorbed ions from a solid process controls ion availability and leaching into soils. The distribution coefficients, which provide information on the mobility of metals, can be correlated with the absorption and contamination of the plant (Vidal *et al*., 2009). According to Jalali and Moharami, (2013), Fe was released at a faster rate than Mn, which implies that Fe was released more quickly from contaminated soils. In native soils, the Fe and Mn distributions were identical, with Fe-Mn oxides and organic matter (OM) fractions being the most abundant. The key processes that regulate the bioavailability of metals, including Mn, in soils are adsorption, desorption, and precipitation-dissolution (Strawn and Sparks, 2000). According to Graham *et al*., (1991), the availability of Mn for plants is controlled mainly by precipitation and oxidation reactions, and adsorption and desorption processes play a minor role. In a step-by-step regression analysis of the constant values of the best fitted models and the selected soil parameters. GhasemiFasaei, *et al*., (2009) concluded that the quality of the easily reducible Mn oxides (ERMn) was the only soil characteristic capable of predicting any constant value from the best fitted models. According to the equations, the Mn release concentrations increased as the amount of ERMn increased. As a result, ERMn is determined to be the main cause of Mn release in these calcareous soils. Using DTPA as the extraction solution, Bostani and Twofighi, (2010) noted that six mathematical equations (zero order, first order, second order, power function, simplified Elovich, and parabolic diffusion) could be used to explain and quantify manganese. release kinetics in six composite soil samples from Iran. The power function equation, which was used to characterize Mn release kinetics, was the best equation as it had the highest coefficient of determination and the lowest value of the estimation standard among all equations. The kinetics of manganese release in some calcareous soils of the Sulaimani governorate in the Iraqi Kurdistan region was relatively small. As a result, the current investigation began with the following objectives: study the Mn release characteristics of the studied soils & identify the most

effective models for characterizing the kinetics of Mn release in the soils under investigation.

MATERIALS AND METHODS

Soil samples were obtained from the soil surface at a depth of (0-30) cm in some calcareous soils in the Sulaimani governorate in Kurdistan Region of Iraq, including (Sharazor, Qaradagh, Bazian, Mawat, and Surdash). The study area is located between (longitudes 35 ° 15 to 27 N; latitudes 45 07 to 37 E) as shown in the (Fig.1). All five soils were classified as (Argixerolls, Rendolls, Pelloxererts, Rendolls, and Argixerolls) according to the Soil Survey Staff, (2004). Soil samples are air-dried, crushed, and sieved through a 2-mm sieve before analysis and Mn release studies. Some of the basic physicochemical properties of the soil studied include particle size distribution, pH, EC, percentage of organic matter, percentage of calcium carbonate, and CEC determined according to the soil analysis methods as outlined by (Page *et al.*, 1982 and Rayan J. *et al.*, 2001), available Mn content in the soil was determined using the DTPA procedure (Lindsay and Norvell, 1978).
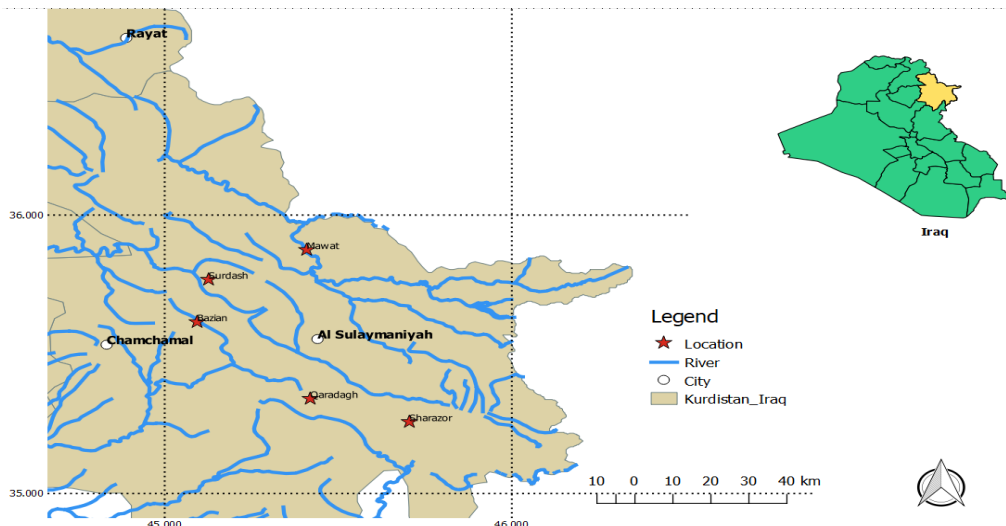

Fig. 1. Soil sampling from studied sites.

Kinetic study procedure:

The DTPA-extracting solution (0.005 *M* diethylene triamine pentaacetic acid, 0.1 *M* triethanolamine, and 0.01 *M* CaCl$_2$) was used to determine the release kinetics at pH=7.3; (Lindsay and Norvell 1978) as an extractant as follows: 50 g of duplicate oven-dried soil were extracted with 100 ml of DTPA-extracting solution at a ratio (m/v of 1:2) of 25 ºC ±1 in a constant temperature shaker for 0.5, 1, 2, 4, 6, 8, 24,48 hours.

To prevent microbiological activity, five drops of toluene have been added. For each shaking period, the soil suspension was filtered instantly through Whatman paper No. 42, then the Mn concentration in a solution was determined using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). Eight models, including: Zero order, First order, Second Order, Third Order, Simple Elovich, Parabolic diffusion, Power function, and Hyperbolic, were used to describe the manganese release in the studied soil (Lopez-Pineiro, and Navarro, 1997) as shown in (Table 1). The model that gave the highest value of the coefficient of determination (R^2) ,and the minimum value of the root mean square error (RMSE) was considered the best model.

Table 1. Eight kinetic models used to describe manganese release in the studied soils.

Models	Equations	Parameters
Zero order	$q_t{}^* = q_0{}^* - k_0 t$	k_0 , zero-order rate constant (mg Mn kg^{-1} h^{-1})
First Order	$\ln q_t = \ln q_0 - k_1 t$	k_1 , first-order rate constant (h^{-1})
Second order	$1/q_t = 1/q_0 + k_2 t$	k_2, second-order rate constant [(mg Mn kg^{-1})$^{-1}$]
Third order	$1/q_t{}^2 = 1/q_0{}^2 - k_3 t$	k_3 , third-order rate constant[(mg Mn kg^{-1})$^{-2}$ h^{-2}]
Simple Elovich	$q_t = 1/\beta s \ln(\alpha s \beta s) + 1/\beta s \ln t$	αs, initial Mn desorption rate constant (mg Mn kg^{-1} h^{-1}) and βs, Mn desorption rate constant [(mg Mn kg^{-1})$^{-1}$]
Parabolic diffusion	$q_t = q_0 - k_p t^{1/2}$	kp, diffusion rate constant [(mg Mn kg^{-1})$^{-0.5}$]
Power function	$\ln q_t = \ln a + b \ln t$	a, initial Mn desorptiorate constant [(mg Mn kg^{-1}(h^{-1})^b] and b , desorption rate coefficient
Hyperbolic	$q_t / t = (q_0 / t \frac{1}{2}) t$	t ½ semi decomposition time

*qt represents the quantity of soil Mn released by DTPA (mg Mn kg^{-1}) after t (h) of extraction, and q$_0$ represents the initial Mn concentration at time = 0.

The determination coefficient and the root mean square error value was calculated according to the following equations:

The coefficient of determination (R^2) = Σ (*q* meas−*q*aepred)^2 /Σ(*q*meas−*q*aepred)^2 + (*q*meas−*q*pred)^2

Root mean square error (RMSE) = {Σ (*q* meas− *q*pred)^2 /n -2}$^{1/2}$

Where **qmeas and qpred , qaepred represent the measured , predicted , and average predicted Mn released, and n is the number of measurements.**

Statistical Analysis

For statistical operations, the XLSTAT software program (Version, 2016) was used.

RESULTS AND DISCUSSION

Physicochemical properties of the soil

Some chemical and physical properties of the soils presented in (Table 2) revealed that the soils differ in their

texture from silty clay to loam, with a range of organic matter, and total calcium carbonate contents ranging from 90 to 25, and 25 to 430 g kg $^{-1}$ respectively. Most of the soil had a neutral reaction (7.48 to 7.90). EC of the soil ranged between 0.40 to 0.90 dS m^{-1}. The CEC values ranged from 26.93 to 41.57 cmolc kg^{-1}, while the extractable Mn by DTPA differed from 2.40 to 7.67 mg kg^{-1}. The texture classes of the studied soils ranged from (loam to Silty clay), indicating that the texture of these

soils ranged from fine to moderately textured soils, and that the soils were slightly to moderately alkaline, with values ranging from (7.48 – 7.9). The electrical conductivity (EC) of the soil samples ranged from 0. 4 to 0.9 dS m^{-1}, indicating that the soils investigated are non-saline. This could be attributed to the relatively higher precipitation and topographic variance of these areas. All soil termed as calcareous soil due to calcium carbonate content was between (25 – 430 gm kg^{-1}).

Table 2. Chemical and physical characteristics of the five soils.

Soil No.	pH	EC (dS m $^{-1}$)	T.CaCO3 (g kg $^{-1}$)	O.M g kg $^{-1}$	Sand g kg $^{-1}$	Silt g kg $^{-1}$	Clay g kg $^{-1}$	Textural class	DTPA Mn mg kg $^{-1}$	CEC cmolc kg $^{-1}$
Sharazor	7.49	0.60	180.00	25.00	37.20	475.10	487.70	Silty clay	2.40	41.15
Qaradagh	7.65	0.70	25.00	17.00	383.50	370.30	246.20	loam	3.46	26.93
Bazian	7.78	0.60	100.00	15.00	55.80	430.50	513.70	Silty clay	7.67	41.57
Mawat	7.48	0.90	25.00	16.00	161.90	434.20	403.90	Silty clay	5.42	39.42
Surdash	7.90	0.40	430.00	9.00	91.60	490.40	418.00	Silty clay	2.91	36.39

The pattern of Mn release

The amount of Mn released in the studied soil as a function of time (0.5 to 48 hours) with DTPA extracting solution is shown in the (Fig.2). The pattern of manganese release models is often characterized by an initial quick reaction, followed by a gradual slowdown until equilibrium was reached after 48 hours. Our findings are similar to those of Bostani and Twofighi, (2010), who observed that Mn release was very rapid in all samples at first (with 25–55 percent of total Mn released during the first 10 hours), then gradually declined until it reached zero in soils after 720 hours.On the other hand, The Kinetics of native Mn release by ammonium bicarbonate diethylenetriaminepentaacetic acid (AB-DTPA) from 10 highly calcareous soils were evaluated for periods ranging from 1 to 1440 minutes by GhasemiFasaei, (2009). The results showed that Mn-release rates were quick at first, then much slower, and that more than 60% of maximum Mn was released from the investigated soils through the first 15 min. They also stated that the pattern of Mn released over time implies that as Mn-release processes continued, Mn-release energies increased. After a 2084-hour kinetic analysis, Jalali and Moharami, (2013) revealed that Fe and Mn concentrations in all fractions decreased, while the Fe-Mn oxides fraction remained the dominant Fe and Mn fraction. During each of the three desorption investigations, the concentration of dissolved manganese content increased initially (within minutes), then declined over the next few hours before reaching a steady value after 4–6 hours (Richard *et al.*, 2013). Similarly, Shi *et al.*, (2014) observed that metal desorption from monodentate sites is fast, but desorption from bidentate sites is slow. In soil Surdash, the largest amount of Mn released for 48 hours was the least (64.00 mg kg $^{-1}$)., on the other hand, Mawat soil had the highest release (541.82 mg kg^{-1}). It could be caused by CEC, organic matter, pH, or clay content. The order of the amount of manganese released into the soil was as follows: Mawat > Sharazoor > Bazian > Qaragh > Surdash. Soil factors such as CEC, organic matter clay content, and pH, play a big role in Mn release patterns. Our findings are consistent with McBride's, (1989) who stated that metal sorption behavior in soil ranges from one soil to another and is impacted by soil properties such as CEC, clay content, pH, and OM. Similarly, Motlagh, (2012) found that the main active

parameters controlling copper release in calcareous soil from northern Iran are CEC and clay content. When anions such as sulfate, carbonate, hydroxide, and phosphate are present. In alkaline soils, precipitation appeared to be the primary metal immobilization process, especially when metal ion concentrations were high (Adriano, 2001).
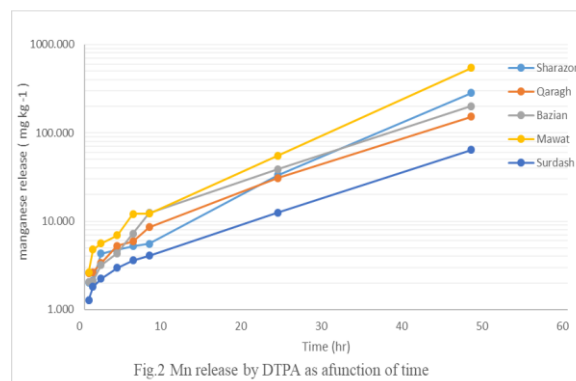

Fig.2 Mn release by DTPA as afunction of time

Mathematical kinetic models used to fit the Mn release data

Eight kinetic models were employed to characterize manganese release in the studied soils by DTPA extracting solution from 0.5 to 48 hours, including zero-order, first-order, second-order, third-order, simple Elovich, parabolic diffusion, power function, and hyperbolic. The model equation, which gave the highest value of determination coefficient (R^2) and the lowest value of root mean square error (RMSE) was considered the best model equation. Due to their high R2 (0.968, 0.850, and 0.566) and low RMSE (0.290, 0. 638, and 0.125), the first order, power function, and second order models given the best fit to Mn release kinetics (Table 3, and 4). While third order, hyperbolic, zero order, parabolic, and simple Elovich could not properly describe Mn release kinetics due to their relatively high values of (RMSE). This is in agreement with the findings of Bostani and Twofighi, (2010), who showed that due to their low standard errors and relatively high correlation coefficients, the first-order, second-order, and parabolic diffusion equations were the best models for predicting Mn release kinetics. While Ghasemi-Fasaei, (2009) showed that two constant rate, simple Elovich, and parabolic diffusion were the best-fitted models for describing Mn release kinetics of agricultural fields of Fars province in southern

Iran. On the other hand, the two best models for describing the desorption Mn data in all soils were two-constant rate and parabolic – diffusion equations Reyhanitabar (2014). Table 4 shows the variable ranges and means of R^2, and RMSE for the studied soils, and eight kinetic models. The average of R^2, RMSE for all the five soil ranked as follows: first order > power function > second order > third order > hyperbolic > zero order > parabolic > simple Elovich.

Table3. The root mean square error (RMSE) and determination coefficient (R^2) of eight kinetic models used to characterize manganese release in the investigated soils.

Eqauation	Sharazor		Qaradagh		Bazyan		Mawat		Surdash	
	RMSE	R^2	RMSE	R^2	RMSE	R^2	RMSE	R^2	RMSE	R^2
Zero order	38.338	0.867	16.354	0.914	21.736	0.913	76.298	0.858	7.078	0.907
First order	0.244	0.982	0.254	0.972	0.434	0.936	0.297	0.974	0.222	0.974
Second order	0.125	0.579	0.096	0.635	0.142	0.521	0.094	0.485	0.168	0.609
Third order	0.086	0.315	0.051	0.407	0.087	0.328	0.046	0.222	0.180	0.327
Simple Elovich	77.795	0.452	39.030	0.512	51.502	0.514	150.800	0.445	16.268	0.509
Parabolic diffusion	56.684	0.709	26.691	0.772	35.254	0.772	110.955	0.699	11.266	0.764
Power function	0.793	0.805	0.585	0.854	0.555	0.896	0.738	0.841	0.519	0.856
Hyperbollic	1.544	0.363	1.555	0.003	1.207	0.209	2.470	0.512	0.779	0.032

Table 4. Range and mean of root mean square error and determination coefficient (R^2) (RMSE) of eight kinetic models were used for describing manganese release by DTPA in the studied soils.

Equation	RMSE		R^2	
	Range	Mean	Range	Mean
Zero order	7.078 - 76.298	31.961	0.858 - 0.914	0.892
First order	0.222 - 0.434	0.290	0.936 - 0.982	0.968
Second order	0.094 - 0.168	0.125	0.485 - 0.635	0.566
Third order	0.046 - 0.18	0.090	0.222 - 0.407	0.320
Simple Elovich	16.268 - 150.8	67.079	0.445 - 0.514	0.486
Parabolic diffusion	11.266 - 110.955	48.170	0.699 - 0.772	0.743
Power Function	0.519 - 0.793	0.638	0.805 - 0.896	0.850
Hyperbolic	0.779 - 2.47	1.511	0.003 - 0.512	0.224

Kinetic Parameters

Table 5 shows the constant values of the best fitted models. Some of these values vary significantly between the investigated soils, possibly due to differences in Mn release rates. The intercept and slope of the linear curves are represented by the constants (q_0, a and $1 / q_0$) and (k_1, b and k_2) of each model. In the first-order model, q_0 represents the intercept, which can be used to indicate the initial release of Mn or the amount of Mn released at time zero, and k_1 represents the slope, which can be used as a constant rate of the index of the first order. release. The value of q_0 ranged between 1,876 and 4,435 with a mean of 3,023 (mg kg-1). The highest value of q_0 recorded in the Mawat soil and the lowest value attributed to the Surdash soil. While k_1 represents the lowest value in Surdash soil attributed to its lower organic matter and cation exchange capacity. On the other hand ,

Table 5. Rate constant values for three kinetics models that best described manganese release by DTPA in the soils investigated.

Soils	First order		Power function				Second order	
	q_0	k_1 (h^{-1})	a (mg Mn kg^{-1}(h^{-1})^b)	B (mg Mn/kg^{-1})$^{-1}$	ab	$1/q_0$	k_2 (mg Mn kg^{-1})$^{-1}$	
Sharazor	2.611	0.100	1.886	0.968	1.826	3.104	-0.008	
Qaradagh	3.186	0.085	2.315	0.850	1.968	3.570	-0.007	
Bazian	3.009	0.093	1.983	0.981	1.945	0.317	-0.008	
Mawat	4.435	0.102	3.051	1.021	3.115	0.197	-0.005	
Surdash	1.876	0.076	1.407	0.762	1.072	0.479	-0.012	
Min	1.876	0.076	1.407	0.762	1.072	0.197	-0.012	
Max	4.435	0.102	3.051	1.021	3.115	3.570	-0.005	
Mean	3.023	0.091	2.128	0.916	1.949	1.533	-0.008	

The value of $1 / q_0$ ranged between 0.197 and 3.570 with a mean of 1.533, while k_2 represents the lowest value in Surdash soil. The lower value of k_1 and k_2 could be attributed to their lower organic matter, this was consistent with the statistical analysis. Curtin, Ryan, and Chaudhary, (1980) have reported similar results reporting that the rate of Mn release was considerably different in the calcareous soils of Lebanon. These differences can be attributed to the difference in the amount of Mn that it supports. minerals in the soils studied (Krishnamurti and Huang, 1988). The value of (a) ranged between 1.407 and 3.051 with a mean of 2.128 and, (b) ranged between 0.762 and 1.021 with a mean of 0.916. The values of the velocity constant (a and b) with the mean are (2.128 and 0.916) from the equation of the power function very different from the five soils (Table 5). The values of (ab) were calculated and listed for the soil studied in (Table 5). According to the statistical study, the Mawat soil had the highest value of constant (ab), equivalent to (3,115), and the Surdash soil had the lowest value, equal to (1,072). This may be due to a higher content of organic matter and CEC in the soil in Mawat soil compared to Surdash soil. The variation in the values of b suggests that the Mn of the soils that provides energy is different. Our results are in agreement with the findings of Dang *et al*., (1994), who believe that an increase in the value of a and / or a decrease in the value of b is an indication of a higher rate of nutrient release.

CONCLUSION

The results of this study showed that the pattern of manganese release models is often characterized by an initial quick reaction, followed by a gradual slowdown until equilibrium was reached after 48 hours. The best model was the first order, power function, and second order based on the highest value of R^2, and the lowest value of RMSE for the description of Mn release from some calcareous soil of Sulaimani governorate, Iraqi Kurdistan Region. For the first order, power function, and second order models, the values of (k1), (a, and b), and k2 were related with some soil characteristics like organic matter and CEC in the studied soils.

ACKNOWLEDGEMENT

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حركية تحرر المنغنيز في بعض الترب الجيرية لمحافظة السليمانية في إقليم كردستان العراق.

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¹قسم الموارد الطبيعة ، كلية علوم الهندسة الزراعية ، جامعة السليمانية ، إقليم كوردستان ـ العراق

²مركز البحوث الزراعية ، السليمانية ، إقليم كوردستان ـ العراق

أجريت هذه الدراسة في محافظة السليمانية، إقليم كردستان العراق، لتقدير حركية تحررالمنغنيز لخمسة أنواع من الترب الجيرية هي (شارزور، قرداغ، بازيان، ماوت، سورداش)، وكانت أكبر كمية تحررللمنجنيز خلال ٤٨ ساعة أقل في سورداش (٦٤,٠٠ مجم كجم ⁻¹)، في حين أظهرت تربة ماوت أعلى تحرر (٥٤١,٨٢ مجم كجم⁻¹). تفاوتت قيمة ثابت معدل التحرر من الدرجة الأولى (k1 من ٠,٠٧٦ إلى ٠,١٠٢ بمتوسط (0.091 h⁻¹) و القيم المتوسطة لثابت المعدل (أ، ب) من نموذج دالة القدرة كانت Mn kg⁻¹ h (2.128 (مجم⁻¹) ، (0.916 ⁻¹ (Mn / kg⁻¹ مجم)) على التوالي تباينت بشكل كبير لترب الخمسة. تراوحت قيمة ثابت معدل التحرر من الدرجة الثانية من نموذج الدرجة الثانية (k2 من (- ٠,٠١٢) إلى (- ٠,٠٠٥) بمتوسط (-٠,٠٠٨ Mn kg⁻¹ مجم)) ، كانت الدرجة الأولى ، دالة القدرة ، والدرجة الثانية هي أفضل النماذج المستخدمة لوصف عملية تحرر المنغنيز بشكل جيد للغاية في الترب المدروسة.