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STUDY OF THE KINETICS DESORPTION OF AGRICULTURAL SULFUR AND SULFUR ASSOCIATED WITH NATURAL GAS UNDER THE INFLUENCE OF DIFFERENT TEMPERATURES AND SOIL TEXTURES

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Abstract:

A laboratory experiment was carried out to study the best incubation temperature (25, 35 and 45 °C) for application in the sulfur kinetic model while treating the soil with two sulfur sources (agricultural sulfur and sulfur associated with natural gas) at a level of 2000 mg S kg^{-1} soil and for incubation periods (15 and 30, 45, 60, 90 and 120 days) in the concentration of dissolved sulfur in the solution of sandy loam and clay loam soils.

The temperature results indicated an increase in soluble sulfur in the agricultural sulfur treatment in both study soils, and the 35 °C treatment gave the highest soluble sulfur compared to the other temperatures.

The studied kinetic model equations First order, Pseudo first order, Second order, Pseudo second order with all their probabilities, Elovich, Intraparticle diffusion and Fractional power succeeded in describing the data on sulfur desorption with time through increasing values of the coefficient of determination (R^2) and decreasing values of the percentage deviation standard (MPSD) and the pseudo-second-order kinetic model can be considered the second possibility (II) to have achieved the highest superiority in describing the data for the reaction in sulfur desorption for sandy loam soil, while the aforementioned kinetic model with its first possibility (I), as well as the Elovich model, gave a close description of the data. Special sulfur desorption (rate of sulfur oxidation) in clay loam soils.

Keywords: Kinetics, Agricultural sulfur, Sulfur associated with natural gas, Influence, Temperatures, Soil textures.

INTRODUCTION:

Sulfur plays a vital role in plant metabolism in addition to being the primary component of important metabolic and structural compounds such as amino acids (cysteine 27%, cystine 26%, and methionine 21%) (Njira and Nabwami, 2015) and Rossini *et al.,* 2018). Sulfur is also one of the components of the tripeptide glutathione (GSH), proteins, fats, vitamins, coenzymes, chlorophyll, and a variety of plant by-products such as glucosinolates in Cruciferae and alliin in Liliaceae (Kopriva *et al.,* 2019). These sulfur-containing compounds function in many biological processes. Such as controlling redox, regulating protein activities, detoxification, and providing plant protection from pests and oxidative stress caused by heavy metals (HARSCO, 2016 and Tabak *et al.,* 2020) Therefore, sulfur plays crucial roles in the catalytic or electrochemical function of biomolecules in cells (Capaldi *et al.,* 2015) .

Sulfur needs to be oxidized to sulfate to be ready for plants by various groups of soil microorganisms, most notably genera of Bacteria *Thiobacillus sp.* and this process is affected by the chemical composition of the fertilizer, the degree of its decomposition, the time of addition, the abundance of microbial groups, and some physical and chemical soil properties such as soil texture, temperature, soil moisture, content and type of organic matter, and the degree of soil interaction, which affects the desorption and adsorption of sulfur in the soil (Degryse *et al.,* 2020 and Kulczycki 2021 and Zhao *et al.,* 2022). Some studies showed an increase in sulfur mineralization in soil at a temperature of 35 °C compared to 20 °C, and the reason for this is attributed to the appropriate conditions for increased hydrolysis (Tanikawa *et al.,* 2014). Sayahi and Souri (2018) found that the amount of sulfate resulting from sulfur oxidation increases with increasing temperatures above 25 °C up to 30 °C.

Many researchers have shown that the use of chemical kinetic inputs can provide a good description of the desorption and adsorption of sulfur by introducing the time factor with the amount of desorption and describing it quantitatively with mathematical equations to make appropriate fertilizer recommendations and managing this element in the soil. Therefore, many attempts have been made to apply mathematical equations to Iraqi soils before Al-Obaidi *et al.,* (2009) and Al-Lami and Al-Khuzai (2017), the mathematical formulas used to describe the desorption of sulfur were divided into two types: The first type is based on the Chemical Kinetics assumption and includes the zero-order, first-order, second-order, and diffusion equations. As for the second type, they are equations. Experimental analysis, which includes the Elovich equation and the power function, and the constants of these equations were used to interpret the amount of sulfur desorption from the soil.

MATERIALS AND METHODS:

Soil samples were collected randomly and in the form of composite samples from the surface layer 0-30 cm at the end of October 2021 from two sites in Basra Governorate, the first from the Barjisiya area (Ts) and the second from the Medina District (Tc). The soil samples were air-dried then ground and passed through a sieve with a diameter of 4 mm to study some physical properties of the soil (Black, 1965) (Table 1) and some chemical properties of the soil (Richards, 1954, Jackson, 1958, Page *et al.,* 1982) (Table 2).

Table (1): Some physical properties of the two study soils

Soil symbol	Field capacity	Sand ratio	Loam ratio	Clay ratio	Soil texture
				%	
Ts	19.60	70.70	21.20	8.10	Sandy loam
Tc	29.7	27.40	34.70	37.9	Clay loam

Table (2): Some chemical properties of the two study soils

Soil symbol	pH	EC	Dissolved ions in soil solution							
			Ca^{+2}	Mg^{+2}	Na^{+}	K^{+}	SO_4^{-2}	Cl^{-}	CO_3^{-}	HCO_3^{-}
		dsm^{-1}	$mmol\ L^{-1}$							
Ts	7.93	2.71	5.62	4.23	4.48	0.59	0.98	18.27	0.00	2.45
Tc	7.79	8.63	8.46	7.79	48.22	3.27	12.42	52.35	0.00	5.19

Soil symbol	CEC	Organic carbon	Organic matter	$CaCO_3$	Ions available in the soil			
					N	P	K	S
	$cmol + Kg^{-1}$	$gm\ Kg^{-1}$ soil			$mg\ Kg^{-1}$ soil			
Ts	7.41	3.54	6.10	169.16	1.12	8.09	39.58	48.85
Tc	23.47	24.71	42.60	445.28	21.62	13.68	197.23	454.58

Used is agricultural sulfur fertilizer produced by the Mishraq Sulfur General Authority, some of whose properties are mentioned in Jassim *et al.* (2021), while some chemical properties of the sulfur associated with natural gas from the (R.S.K.) company that produces it were mentioned, as shown in Table (3):

Table (3): Some primary properties of the sulfur fertilizer used in the study

Source of sulfur	Sulfur %	pH (1:5)	EC (1:5) ds m^{-1}
Agricultural sulfur (Sa)	98.71	3.70	4.40
Sulfur associated with natural gas (Sg)	98.80	3.75	7.34

The laboratory experiment included studying the effect of temperatures of 25, 35 and 45 °C on the concentration of dissolved sulfur in the soil solution by adding a sulfur level of 2000 mg S kg^{-1} soil to test the best temperature and apply it in sulfur kinetic models depending on the source of sulfur (agricultural sulfur Sa and sulfur associated with natural gas Sg). and soil texture (sandy loam Ts and clay loam Tc) with incubation periods of 15, 30, 45, 60, 90 and 120 days while maintaining soil moisture at the limits of field capacity throughout the incubation period. After the end of each incubation period, the dissolved sulfur in the soil solution was estimated to choose the best kinetic equation that describes the desorption of sulfur (Table 4), through the coefficient of determination (R^2) from the linear equation and the percentage of standard deviation Marquardt's Percent Standard (MPSD) according to the following law:

$$\text{MPSD} = 100\sqrt{\frac{1}{N-p}\sum_{i=1}^{N}\left(\frac{q_{ei}^{exp} - q_{ei}^{cal}}{q_{e_i}^{exp}}\right)^2}$$

Nomenclature

q_{ei}^{exp}: Experimental desorption quantity (µg gm^{-1})
q_{ei}^{cal}: Calculated desorption quantity (µg gm^{-1})
N: number of experimental measurements
P: number of parameters in isotherm
q_{e0} :The amount desorption at equilibrium or (t=0) (µg gm^{-1})
q_t : Amount desorption with time
t: time (day)
k_0: Zero order kinetic model constant (µg gm^{-1} d^{-1})
k_1: First order kinetic model constant (1/d)
k_{1p}: Pseudo-first order kinetic model constant (1/d)
k_2: Second order kinetic model constant (gm $µg^{-1}$ d^{-1})
k_{2p} :Pseudo-second order kinetic model constant (gm $µg^{-1}$ d^{-1})
k: Fractional power kinetic model constant (µg gm^{-1} $d^{-\nu}$)
k_p: Intraparticle diffusion kinetic model constant (gm^{-1} $µg^{-1}$ $d^{-0.5}$)
ν: Fractional power kinetic model constant
β : Elovich kinetic model constant (g $µg^{-1}$)
α: Elovich kinetic model constant ($µg^{-1}$ gm^{-1} d^{-1})

The experiments were conducted with three replicates as a factorial experiment and in a completely randomized design (CRD) using analysis of variance through the statistical program SPSS V.23. The averages of the coefficients were compared using the least significant difference (RLSD) at the probability level of 0.05.

Table (4): Kinetic equations (Behnamfard and Salarirad, 2009)

Kinetic models	Equations	Linear expression		Plot	Parameters
Zero-order	$q_t = q_e - k_0 t$		$q_t = q_e - k_0 t$	q_t vs. t	q_e = intercept k_0 = -(slope)
First order	$q_e = q_t \exp(k_1 t)$		$\ln(q_e / q_t) = k_1 t$	$\ln(q_t)$ vs. t	q_e = intercept k_1 = -(slope)
Pseudo-first order	$q_t = q_e [1-\exp(-k_{1p} t)]$		$\ln(q_{e0} - q_t) = \ln q_e - k_{1p} t$	$\ln(q_e - q_t)$ vs. t	q_e = exp(intercept) k_{1p} = (slope)
Second order	$q_t = q_e / (1+q_e k_2 t)$		$q_t^{-1} = qe^{-1} + k_2 t$	q_t^{-1} vs. t	$q_e = (\text{intercept})^{-1}$ k_2 = slope
Pseudo-second order	$q_t = k_{2p} q_e^2 t / (1+q_e k_{2p} t)$	Type I	$t/q_t = 1/k_{2p} q_e^2 + t/qe$	t/q_t vs. t	$q_e = \text{slope}^{-1}$ $k_{2p} = (\text{slope}^2)/\text{intercept}$
		Type II	$1/q_t = (1/k_{2p} q_e^2)(1/t) + (1/q_e)$	$1/q_t$ vs. 1/t	$q_e = \text{intercept}^{-1}$ $k_{2p} = (\text{intercept}^2)/\text{slope}$
		Type III	$q_t = q_e - (1/k_{2p} q_e) q_t/t$	q_t vs. q_t/t	q_e = intercept $k_{2p} = -1/(\text{slope} \times \text{intercept})$
		Type IV	$q_t/t = k_{2p} q_e^2 - k_{2p} q_e q_t$	q_t/t vs. q_t	$q_e = -\text{intercept}/\text{slope}$ $k_{2p} = (\text{slope}^2)/\text{intercept}$
Elovich	$q_t = \beta \ln(\alpha \beta t)$		$q_t = \beta \ln(\alpha \beta) + \beta \ln t$	q_t vs. lnt	β = slope $\alpha = (\text{slope})^{-1} \exp(\text{intercept}/\text{slope})$
Function Power	$q_t = k t^{\nu}$		$\ln q_t = \ln k + \nu \ln t$	$\ln q_t$ vs. lnt	k = exp(intercept) ν = slope
Intraparticle diffusion	$q_t = k_p t^{0.5}$		$q_t = k_p t^{0.5}$	q_t vs. $t^{0.5}$	k_p = slope

RESULTS AND DISCUSSION:

1- The effect of temperature on the rate of dissolved sulfur in sandy loam and clay loam soils:

Figure (1) shows that there are significant differences in the effect of temperature on the average amount of dissolved sulfur in the two study soils. We notice an increase in the concentration of dissolved sulfur at a temperature of 35 °C compared to a temperature of 25 °C, reaching 23.36 and 22.63 mmol S L^{-1}, respectively, in the clay loam soil, and 9.14 and 8.47 mmol S liter-1, respectively, in the sandy loam soil. We also find that the sulfur concentration decreased after that to 21.62 and 7.78 mmol S L^{-1} at a temperature of 45 °C for both soils of the study respectively. The reason for this is that the process of sulfur oxidation is a vital process that depends mainly on the vital activity of organisms' Sulfur-oxidizing microflora and that these organisms have an optimum temperature in which they can operate with maximum effectiveness. Therefore, we find that the temperature of 35 °C represents the appropriate temperature for the activity of these organisms and is a reason behind the increase in oxidation at this temperature (Zhi-Hui *et al.,* 2010 and Kumar *et al.,* 2020) .

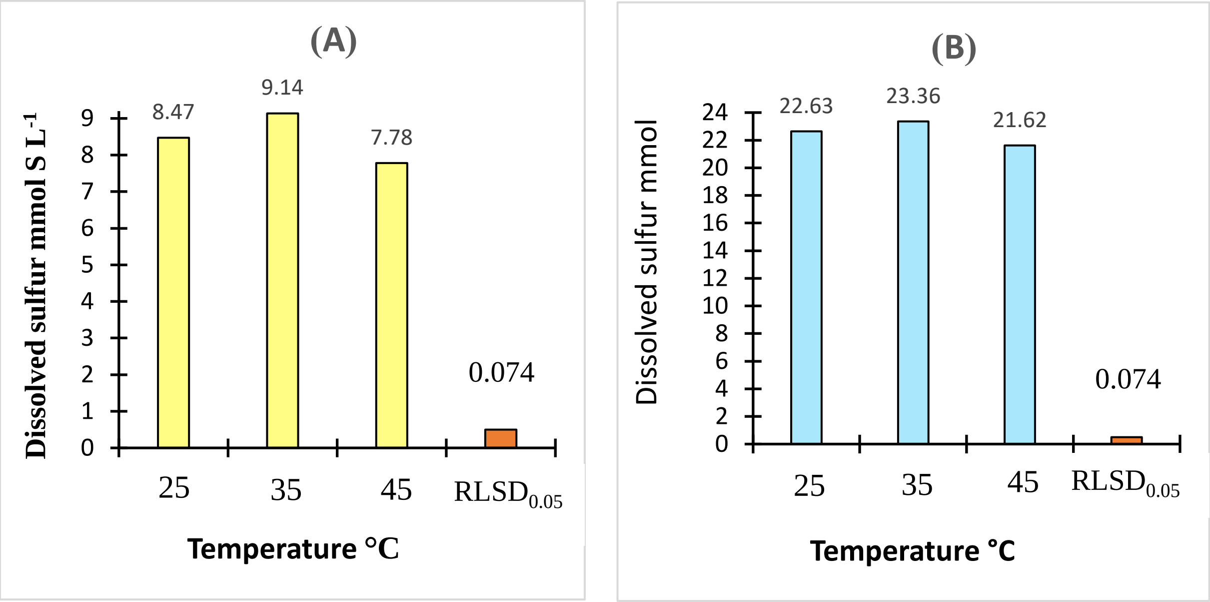


Figure (1): The effect of temperature on the rate of dissolved sulfur in the soil: A) Sandy loam B) Clay loam

2- Study of the kinetics of sulfur desorption from the two sulfur sources in the two soils of the study:

The desorption in Figure 2 (a and b) showed the desorption of sulfur over time depending on the source of sulfur and the soil studied. It was shown that the rate of sulfur desorption varies according to the soil and the source of sulfur. Agricultural sulfur was superior in the amount of desorption compared to the sulfur associated with the gas in both soils, and it was also superior to the clay loam soil, on sandy loam soil with the desorption amount of sulfur. Figure 2 (a and b) also showed an increase in the amount of sulfur desorption over time following the biological oxidation process, but this increase decreased over time as it was characterized by two stages of biological activity and its impact on the desorption of sulfur, where the rate of sulfur desorption generally reached approximately between 90-135%. Of the total amount desorption for the period from 15 - 60 days for the first stage, after that the rate of desorption decreased in both soils, especially the sandy loam soil and for both sources of sulfur, as the remaining amount did not exceed 20% of the total amount for the period from 60 - 120 days, and this reflects the extent of the activity of the oxidation process the vitality of the element sulfur which is an indicator of sulfur oxidation depending on the soil (Anderson, 2020).

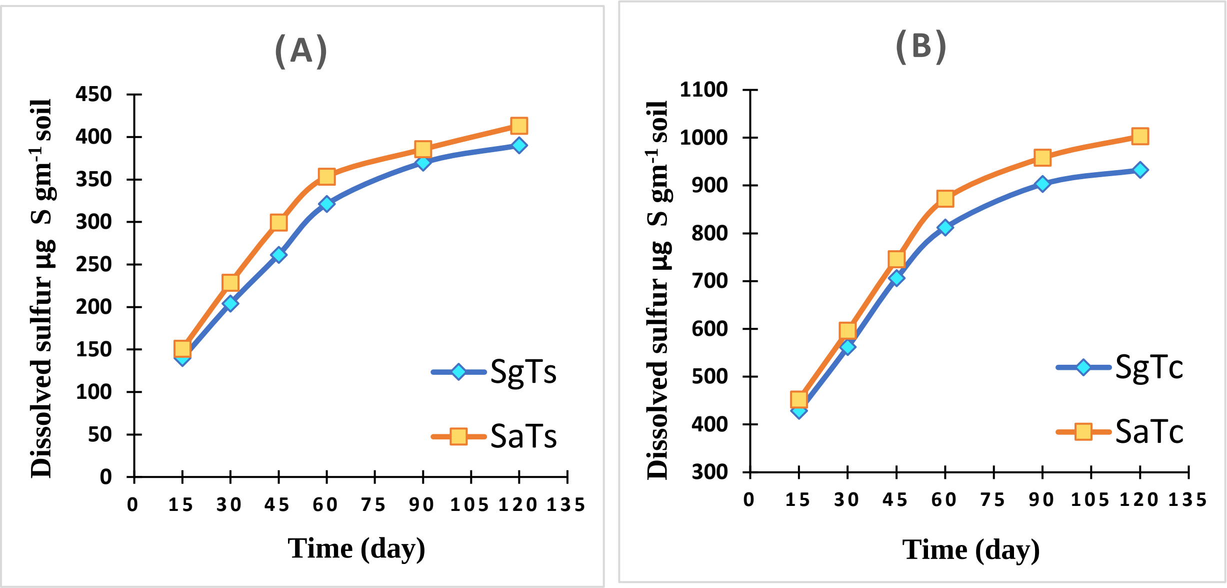


Figure (2): The effect of time on the rate of dissolved sulfur in A) Sandy loam and B) Clay loam

3- Study of the kinetics of sulfur desorption from the two sulfur sources in the two soils of the study:

The relationship between the amount of sulfur desorption over time was subjected to the following kinetic equations: Zero order, First order, Pseudo-First order, Second order, Pseudo-Second order with its four possibilities, Elovich, Intraparticle diffusion and Fractional power to measure the speed of sulfur desorption from the two sulfur sources in the two study soils to understand the mechanism of the sulfur desorption and desorption system over time and to know Which of the kinetic equations referred to above can give the best description of the studied data by knowing the highest coefficient of determination R^2 and the lowest percentage of standard deviation (MPSD) between the actual (experimental) values and the calculated values regarding each equation.

From the desorption of Table 5, it is clear that most of the kinetic equations have succeeded in describing the relationship that controls the desorption of sulfur with time through high values of the coefficient of determination R^2, but some of them also gave high values of the standard error, but the pseudo-second-order kinetic model can be considered in the second possibility (P.S.O. II). excelled in describing the data on the interaction of two sulfur sources (agricultural sulfur and sulfur associated with natural gas) in sandy loam soil over time, as it recorded the highest values for the coefficient of determination (R^2) and the lowest values in the percentage of standard deviation (MPSD) compared to the rest of the kinetic models. Also, an equation for the Pseudo-Second order kinetic model in its first possibility (P.S.O. I) gave the best description of the studied data in terms of an increase in the values (R^2) of the interaction between the two sources of sulfur (Sa and Sg) in the clay loam soil (Tc) with time, which recorded 0.9953 and 0.9955, respectively, compared to the rest of the kinetic models, but gave a slightly higher MPSD value of 4.363 and 4.459, respectively, compared to the Elovich kinetic model, which recorded MPSD values of 4.029 and 3.923, respectively, but this kinetic model (Elovich) at the same time gave values Low in (R^2) 0.9821 and 0.9818, respectively, compared to the kinetic model (P.S.O.). I) as above (Table 5). In this case, it can be said that the equation of the kinetic model of the second order (P.S.O), with its first and second probabilities, gave the best desorption in describing the interaction of sulfur desorption for the two sulfur sources and the two soils of the study with time, and thus the rate of speed of sulfur oxidation (Rate) depends on the square of the reactant concentration. Figures (3-13) show the linear relationships according to the kinetic equations in their various models, which show the accurate description in the kinetic model (P.S.O), whether the first or second possibility referred to previously, of the rate of sulfur desorption speed (rate) for the two sulfur sources in the two study soils over time

Table (5): Values of the coefficient of determination (R^2), percentage of standard deviation (MPSD), equations and kinetic constants in linear form for sulfur kinetic desorption models

Kinetic models	Parameters	SaTs	SgTs	SaTc	SgTc
Zero-order	**Equation**	Y=2.306x+165.050	Y=2.375x+138.560	Y=5.164x+461.060	Y=4.746x+438.970
	R^2	0.8444	0.9057	0.8177	0.8132
	MPSD	17.122	13.433	13.391	13.39
	K_o	-2.306	-2.375	-5.164	-3.864
	q_e	165.050	138.560	461.060	455.240
First-order	**Equation**	Y=0.0084x+5.156	Y=0.0092x+5.030	Y=0.0071x+6.183	Y=0.0070x+6.130
	R^2	0.7602	0.8271	0.8797	0.8741
	MPSD	19.305	17.021	11.241	11.274
	K_1	-0.0084	-0.0092	-0.0071	-0.0058
	q_e	173.469	152.933	484.443	459.436
Pseudo-first order	**Equation**	Y=-0.00135x+7.516	Y=-0.00138x+7.531	Y=-0.0042x+7.354	Y=-0.0037x+7.365
	R^2	0.8561	0.9151	0.9101	0.9012
	MPSD	16.831	12.881	10.027	10.21
	K_{1p}	0.00135	0.00138	0.0042	0.0037
	q_e	1837.204	1864.970	1562.434	1579.715
Second-order	**Equation**	$Y=-3.3*10^{-5}x+0.0057$	$Y=-3.8*10^{-5}x+0.0064$	$Y=-1*10^{-5}x+0.0020$	$Y=-1.1*10^{-5}x+0.0022$
	R^2	0.6628	0.7252	0.7437	0.7409
	MPSD	26.972	25.012	17.474	17.186
	K_2	$-3.3*10^{-5}$	$-3.8*10^{-5}$	$-1*10^{-5}$	$-1.1*10^{-5}$
	q_e	175.500	158.003	494.560	468.384
Pseudo-second order I	**Equation**	Y=0.0019x+0.0691	Y=0.0018x+0.0847	Y=0.0008x+0.0233	Y=0.0009x+0.0242
	R^2	0.9921	0.9900	0.9953	0.9955
	MPSD	4.23	4.241	4.364	4.459
	K_{2p}	$5.224*10^{-5}$	$3.825*10^{-5}$	$2.747*10^{-5}$	$3.347*10^{-5}$
	q_e	526.316	555.556	1250.000	1111.111
Pseudo-second order II	**Equation**	Y=0.0734x+0.0018	Y=0.0804x+0.0019	Y=0.0214x+0.0008	Y=0.0223x+0.0009
	R^2	0.9947	0.9919	0.9786	0.9790
	MPSD	3.658	4.053	5.505	5.193
	K_{2p}	$4.414*10^{-5}$	$4.490*10^{-5}$	$2.991*10^{-5}$	$3.632*10^{-5}$
	q_e	555.556	526.316	1250.000	1111.111
Pseudo-second order III	**Equation**	Y=-40.049x+556.040	Y=-44.496x+537.980	Y=-26.927x+1220.029	Y=-26.080x+1133.534
	R^2	0.9573	0.9531	0.9428	0.9455
	MPSD	6.67	9.766	8.54	8.208
	K_{2p}	$-4.491*10^{-5}$	$-4.178*10^{-5}$	$-3.044*10^{-5}$	$-3.383*10^{-5}$
	q_e	556.040	537.980	1220.029	1133.534
Pseudo-second order IV	**Equation**	Y=-0.0239x+13.561	Y=-0.0214x+11.794	Y=-0.0350x+43.673	Y=-0.0363x+41.952
	R^2	0.9573	0.9531	0.9428	0.9455
	MPSD	10.099	7.607	9.355	9.515
	K_{2p}	$4.212*10^{-5}$	$3.883*10^{-5}$	$2.805*10^{-5}$	$3.141*10^{-5}$
	q_e	567.406	551.122	1247.800	1155.703
Elovich	**Equation**	Y=128.407x-195.167	Y=128.058x-216.175	Y=282.095x-324.468	Y=260.046x-286.020
	R^2	0.9792	0.9849	0.9821	0.9818

	MPSD	4.405	5.274	4.029	3.923
	α	0.036	0.043	0.0011	0.0012
	β	128.407	128.058	282.095	260.046
Intraparticle diffusion	**Equation**	Y=36.462x+34.550	Y=36.973x+8.417	Y=80.901x+174.298	Y=74.470x+174.558
	R^2	0.9303	0.9674	0.9518	0.9487
	MPSD	9.937	6.279	6.194	6.309
	K_p	36.462	36.973	80.901	74.470
Fractional power	**Equation**	Y=0.488x+3.766	Y=0.515x+3.579	Y=0.403x+5.048	Y=0.395x+5.016
	R^2	0.9545	0.9801	0.9743	0.9731
	MPSD	8.075	5.541	4.941	4.979
	v	0.489	0.515	0.403	0.395
	K	43.207	35.838	155.711	150.807

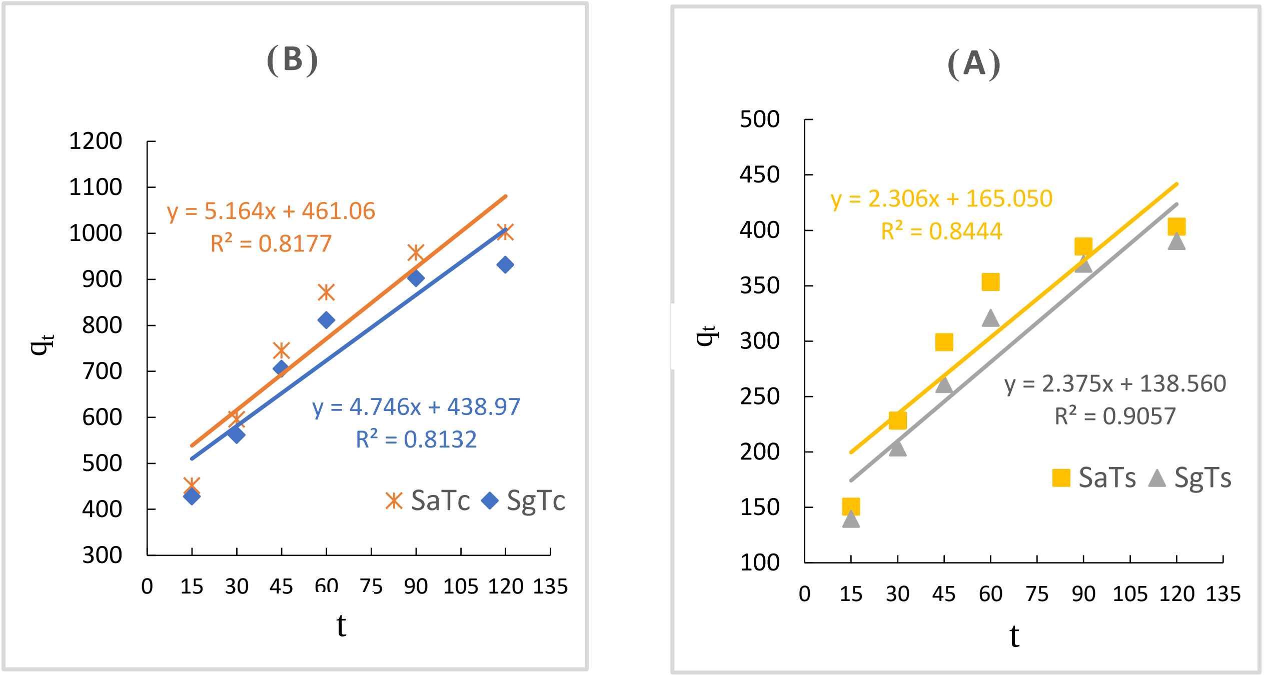


Figure (3): The linear relationship according to the Zero order kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

Figure (4): The linear relationship according to the order kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

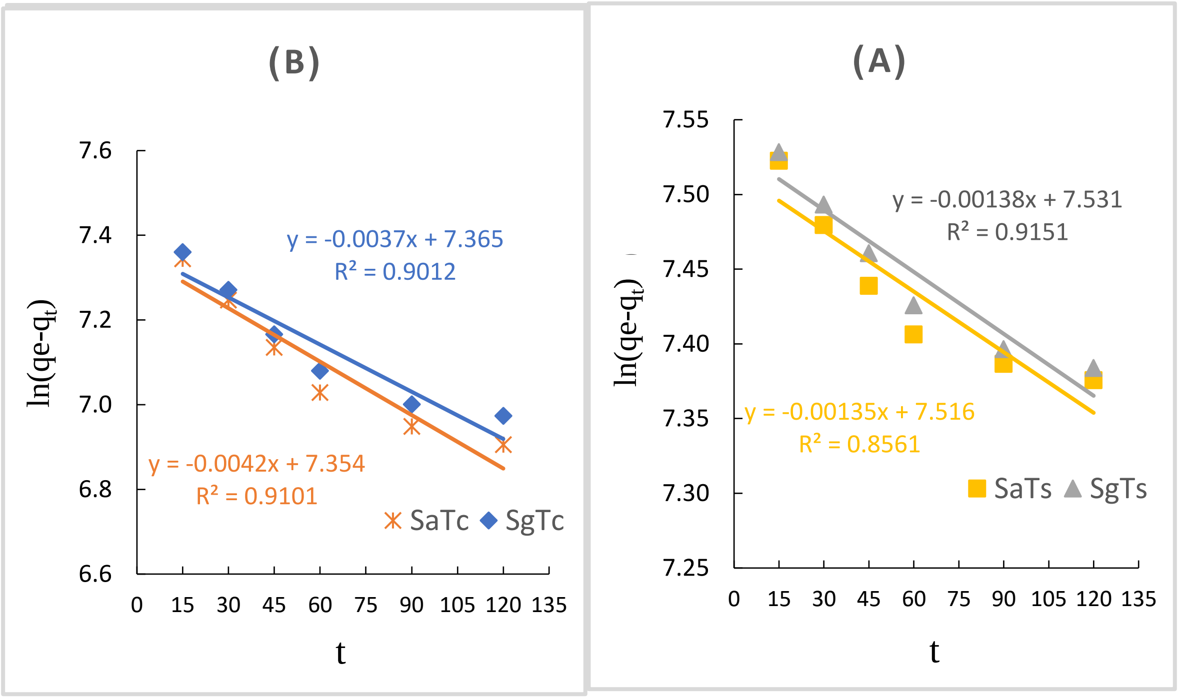


Figure (5): The linear relationship according to the Pseudo-First order kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

Figure (6): The linear relationship according to the Second-order kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

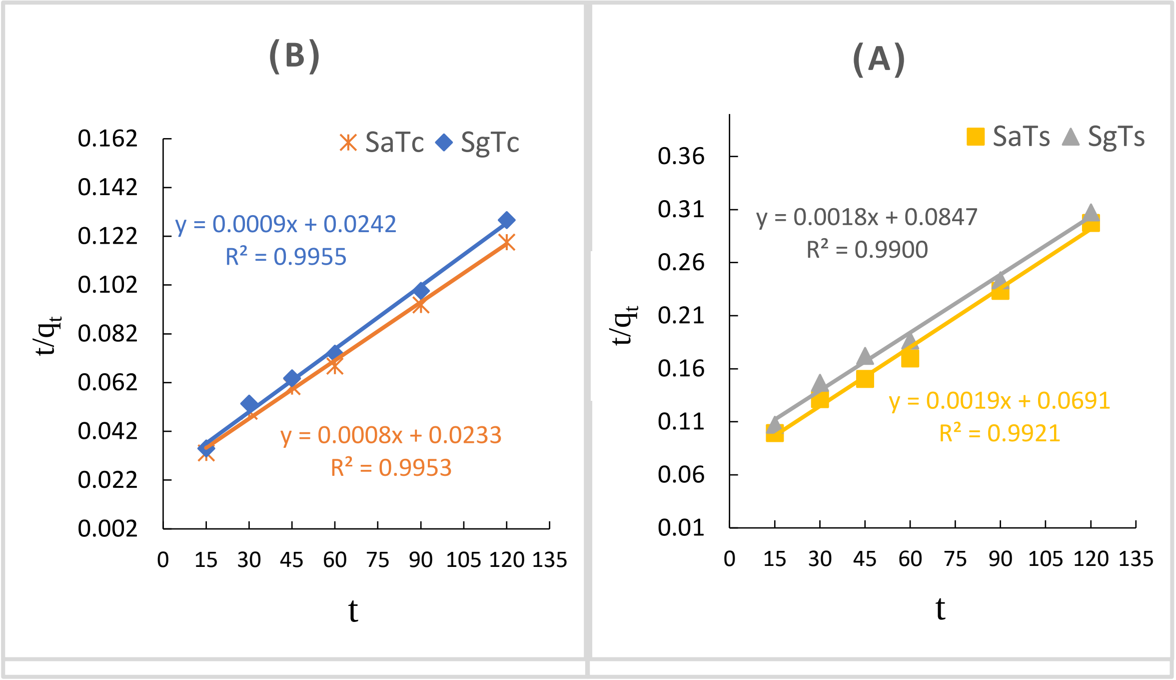
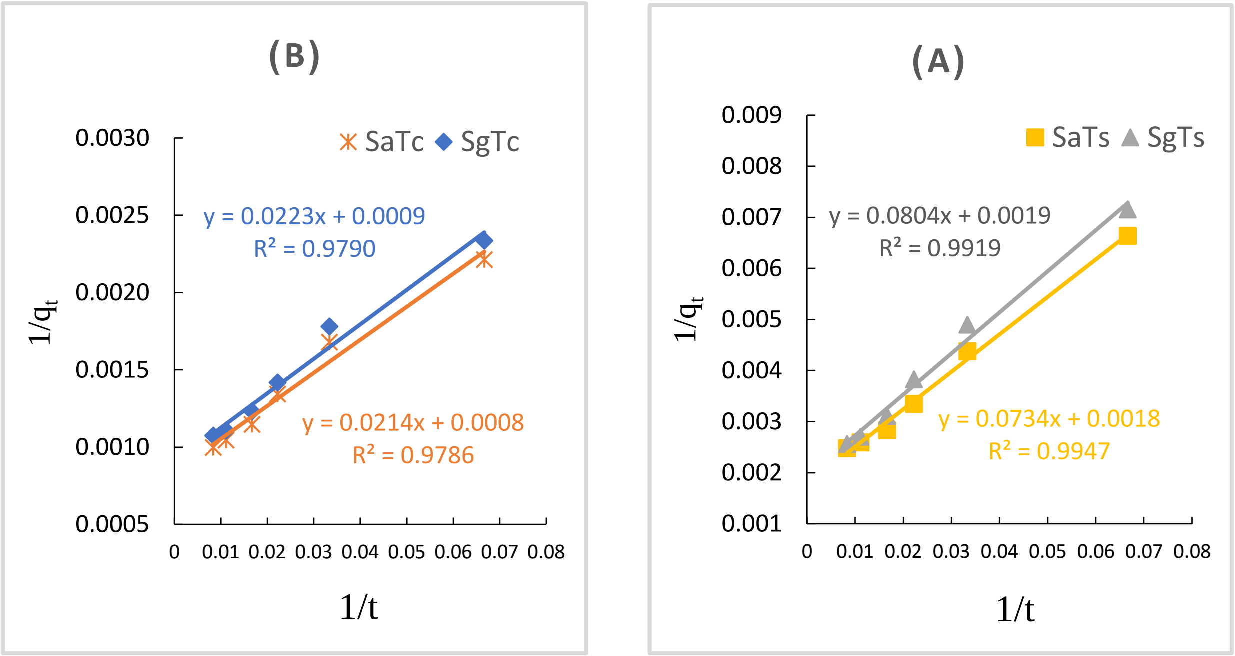
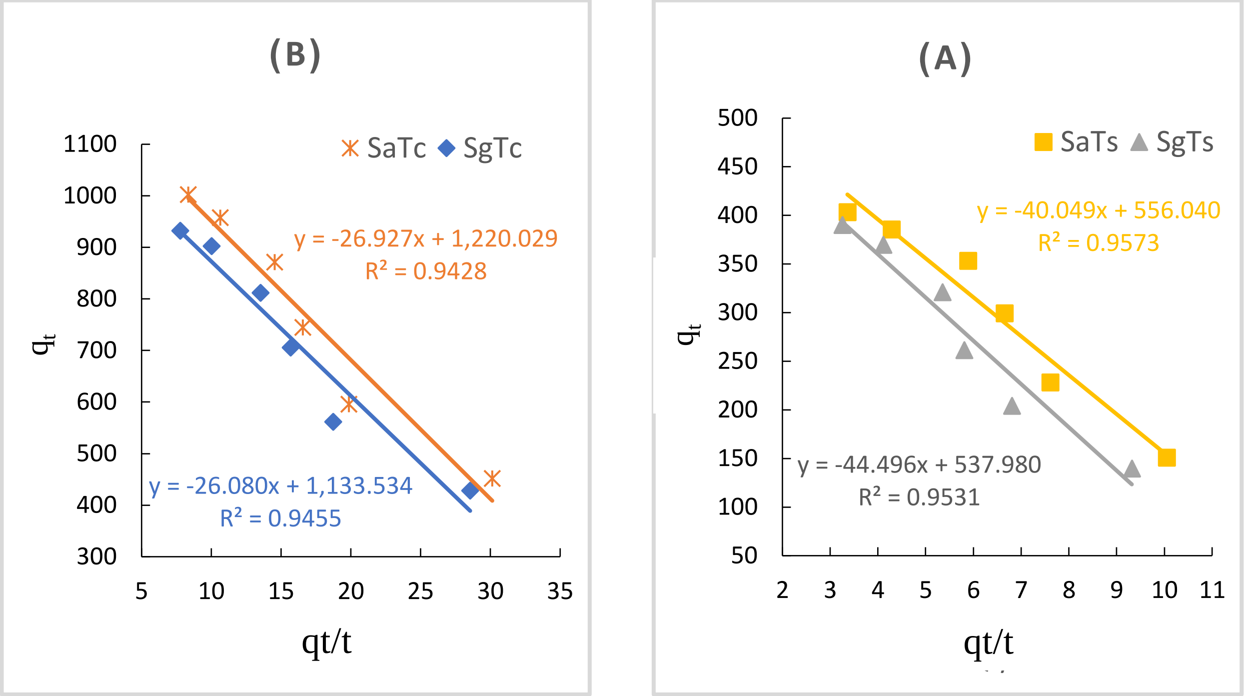


Figure (7): The linear relationship according to the Pseudo-Second order I kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam



kinetic model for the desorption of Figure (8): The linear relationship according to the Pseudo-Second order II agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam



kinetic model for the desorption of Figure (9): The linear relationship according to the Pseudo-Second order III agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

kinetic model for the desorption of Figure (10): The linear relationship according to the Pseudo-Second order IV agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

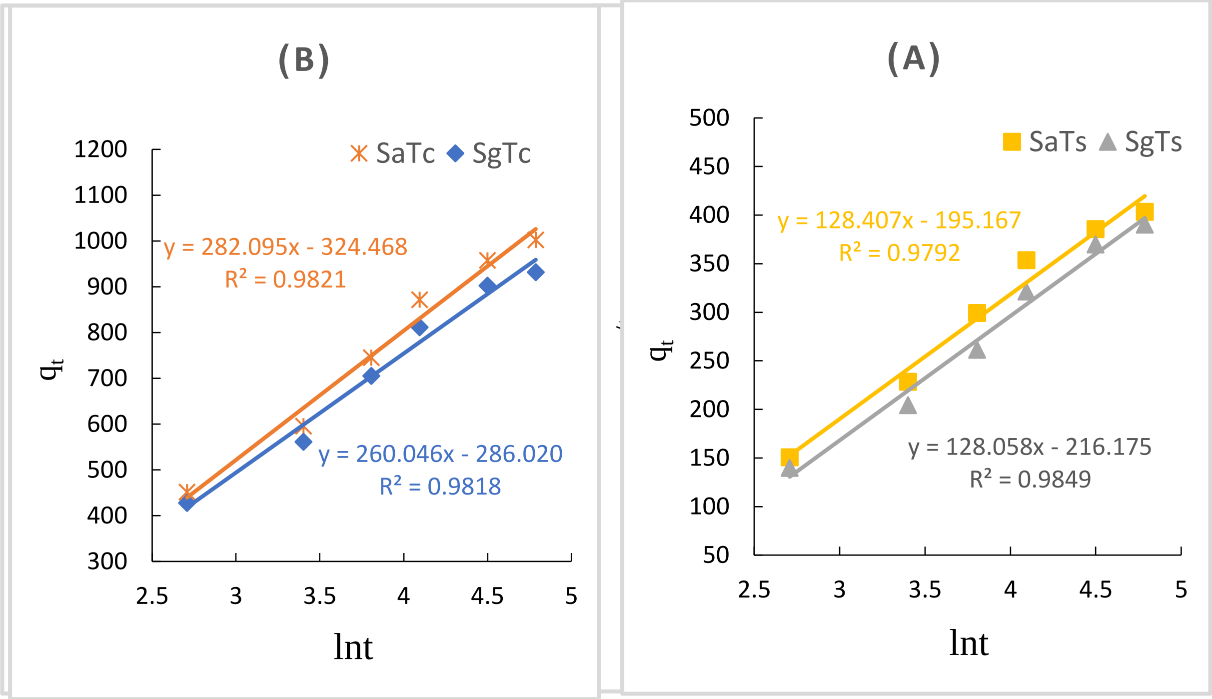


Figure (11): The linear relationship according to the Elovich kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

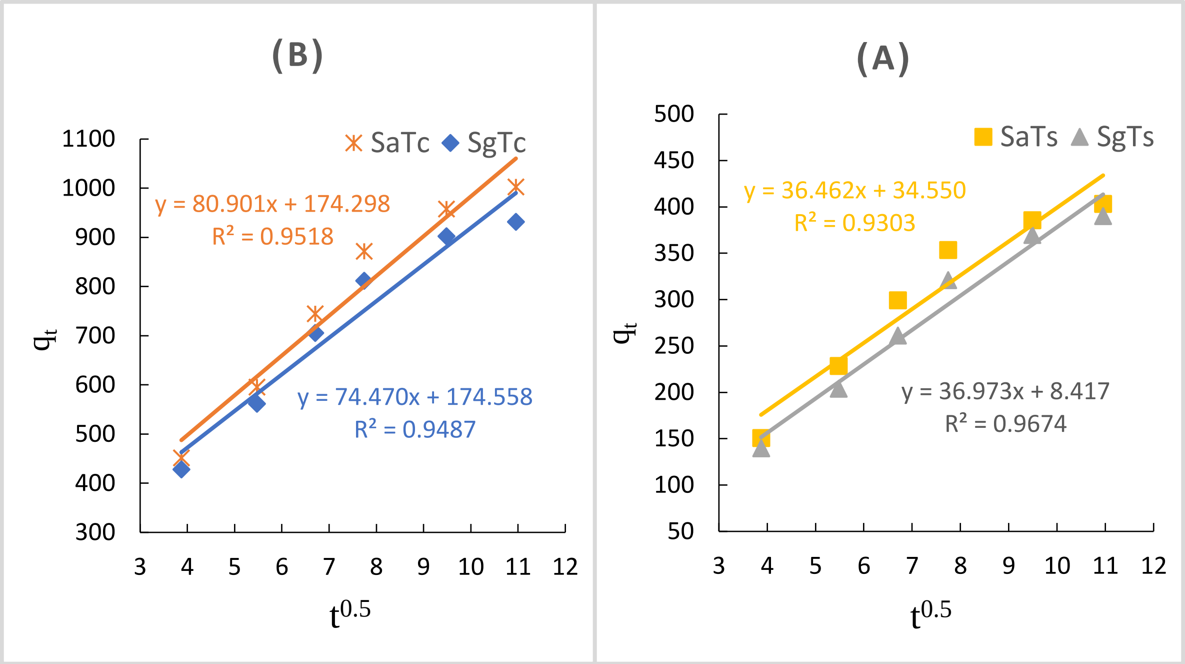


Figure (12): The linear relationship according to the Intraparticle diffusion kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

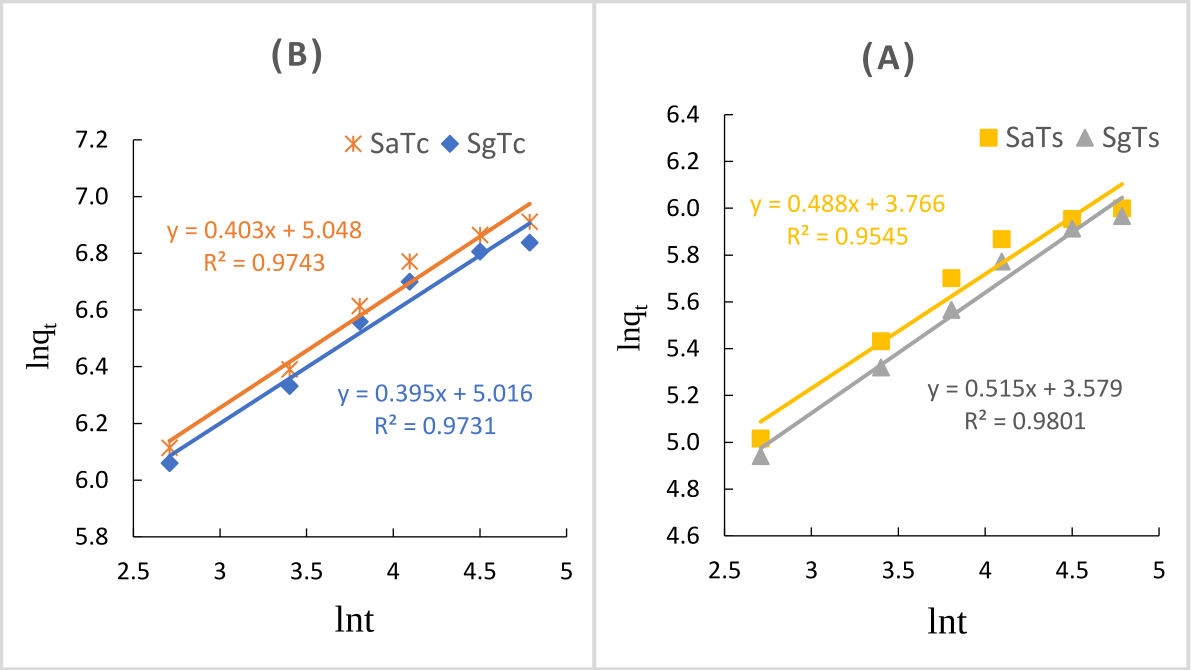


Figure (13): The linear relationship according to the Fractional power kinetic model for the desorption of agricultural sulfur with the sulfur associated with natural gas in the soil texture: A) Sandy loam B) Clay loam

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