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Suitable Models for Describing Sulphate Desorption Kinetics in Selected Bauchi North Soils of Varying Parent Materials in the Nigerian Sudan Savanna

Murabbi Aliyu[1]♥, Abdulkadir M.[2], Azare I.M.[3], Abdu N.[4], Nuhu I. S.[5], Saminu A. I.[1]

[1] Department of Soil Science, Faculty of Agriculture and Agricultural Technology, Abubakar Tafawa Balewa University, Bauchi, Nigeria
[2] Federal College of Education, Gombe, Gombe State, Nigeria
[3] Department of Environmental Science, Federal University, Duste, Jigawa State, Nigeria
[4] Department of Soil Science, Faculty of Agriculture/Institute for Agricultural Research, Ahmadu Bello University, Zaria, Nigeria
[5] Abuja Broadcasting Corporation, Federal Capital Territory, Abuja, Nigeria
♥Corresponding author email: murabbialiyu@gmail.com /amurabbi@atbu.edu.ng

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Abstract. The modelling of sulphate desorption data is critical for a proper S diagnosis and fertilizer formulation to ensure profitable crop production. Five (5) models such as first-order, second-order, Elovich, fractional power, and parabolic diffusion were used to test the best model describing sulphate desorption kinetics in some soils from Bauchi-north, Sudan Savanna, Nigeria. To achieve this, soil samples were collected from three different parent materials namely Basement complex rock and two sedimentary rocks (Kerri-Kerri Formation and Chad Formation). The study showed that the parabolic diffusion and first-order models were found to describe S desorption data satisfactorily, characterized by relatively high R^2 values and lowest S.E values by soil parent materials, respectively. While, the second-order, Elovich and Fractional power equations failed to describe the kinetics desorption of sulphate in all the studied soils, as judged by their high SE values. Therefore, the better fit of S desorption data to the first-order equation is an indication of probable ligand exchange of sulphate ion during the desorption process, and to parabolic diffusion equation suggests that diffusion-controlled phenomena are rate-limiting steps. Based on the findings, we concluded that the rate of sulphate desorption kinetics in these soils is mainly controlled by diffusion-controlled phenomena, which is critical for sulphate-based fertilizer formulations and applications.

Keywords: desorption; kinetics; models; parent material; Sudan savanna; sulphate

INTRODUCTION

The study of desorption kinetics is critical and important in predicting the fate of added S and making proper S fertilizer recommendations to enhance crop production in the era of intensive agricultural practice and climate change. A proper kinetic model is required to assess the rate data to examine the mechanisms of the desorption process, such as mass transfer and chemical reaction. The study of the kinetics of soil chemical processes, even in homogeneous systems, is complex and often difficult to interpret (sparks, 2011).

Scanty information is available in the literature concerning the comparison of different kinetic models for describing sulphate desorption in soils. These kinetic models are designed to give clear information concerning reaction equilibrium with time and to reveal the mechanism involved in the reaction (Abdu, 2006). This research work is intended to solve the problem of S-fertilizer formulation using a kinetic model. In addition, many studies have employed different models to compare the goodness of fit of the models for describing S desorption in the soils (Brajendra *et al*., 2013; Garg *et al*., 2016). The most used models are the First Order, Second-order, Elovich, Fractional power, and parabolic diffusion models. These models can be transformed into linear forms to obtain adjustable parameters by linear regression analysis.

However, the first-order equation has been successfully used to describe S kinetics experimental data by many researchers (Brajendra *et al*., 2013; Farahmand *et al*., 2015 Garg *et al*., 2016). The second-order failed to describe S kinetics desorption data in soils (Brajendra et al., 2013), and also Abdu (2006) noted that for P kinetics desorption data, since most of the soils for both studies had shown inappreciable R^2 values and high SE values. The Elovich

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equation has been successfully adopted for describing the kinetics desorption of S in soils (Garg *et al*., 2016) but has limited utility for describing S adsorption in soils (Farahmand *et al*., 2015). It seems to describe several reaction mechanisms including bulk, surface diffusion, activation, and deactivation of catalytic surfaces (Sparks, 2011). Abdu, (2006) concludes that the fractional power equation is not a suitable model for describing P desorption data in samples taken from different parent materials in Nigeria. The parabolic diffusion model has been utilized by soil chemists in describing the kinetics of ion adsorption and desorption in soils (Agbenin and Raij, 2001; Sparks, 2005).

The parabolic diffusion equation described P kinetics release in soils exceptionally well which confirmed to findings of Abdu (2006). The equation was also found to describe adequately the kinetics of S desorption in soils (Brajendra *et al*., 2013). Sparks, (2011) noted that the determination of rate-limiting steps is another very important aspect of the kinetics study while Agbenin and Raij (2001) and Abdu *et al*. (2008) used this model to conclude that intra-particle diffusion appeared to be the rate-limiting step in phosphorus release kinetics. However, there is little evidence in the literature for a comparison of alternative kinetic models to describe sulphate desorption in Nigerian savanna soils, particularly in the northern Bauchi. Sulphur modelling is an important tool that improves the efficiency of nitrogen (N) use.

Modern fertilizers have a precise amount of NPK with little or no S and this affects soil productivity due to improper fertilizer formulation. Therefore, the soil parent material is an important component that determines the rate at which the sulphate desorption can take place and in turn get assimilated by plants or becomes prone to leaching. The objective of this study was to compare five different kinetic equations for S desorption in the selected northern Bauchi soils from the Nigerian Sudan savanna to find the best model. The modelling of sulphate desorption is thus essential in understanding the chemical behaviour of sulphate ions and in formulating the fertilizer schedule for raising optimum crop production given the low recovery of this ion in most agricultural soils.

METHODS

Site Descriptions

The study was conducted in 2017 in Bauchi, north of the state of Bauchi (Figure 1) and geographically falls within the Nigerian Sudan savanna. Soils in the state are formed from Basement Complex rock (BCR) and the Sedimentary Rocks comprising the Kerri-Kerri formation (KKF) and the Chad formations (CF).

The climate of the study area is typically tropical with distinct rainy and dry seasons. The rainy season starts on average in June-July and ends in November. The dry season lasts from November-May. The rainy season is characterized by high temperature and special variation, averaging about 280 mm per annum. Temperature ranges from 16 to 40°C.

Soil Sampling

Soil samples (0-15 and 15-30cm depths) and replicated twice were collected from 3 different parent materials (Chad Formation (CF), Basement complex rock (BCR), and Kerri-Kerri Formation (KKF)) in Sudan savanna. At each parent material, three (3) locations were selected. A total of 36 composite samples were collected, air dried, gently ground and sieved to remove materials larger than 2mm for laboratory analysis.

The particle size distribution was determined by the Bouyoucos hydrometer method after dispersing with sodium hexametaphosphate (Gee and Bauder, 1986). Using a glass electrode meter, soil pH was determined using 1:1 soil/water and 1:1 soil/0.01 M $CaCl_2$. Organic carbon (OC) was determined by the Walkley-Black dichromate wet oxidation method (Nelson and Sommers, 1982).

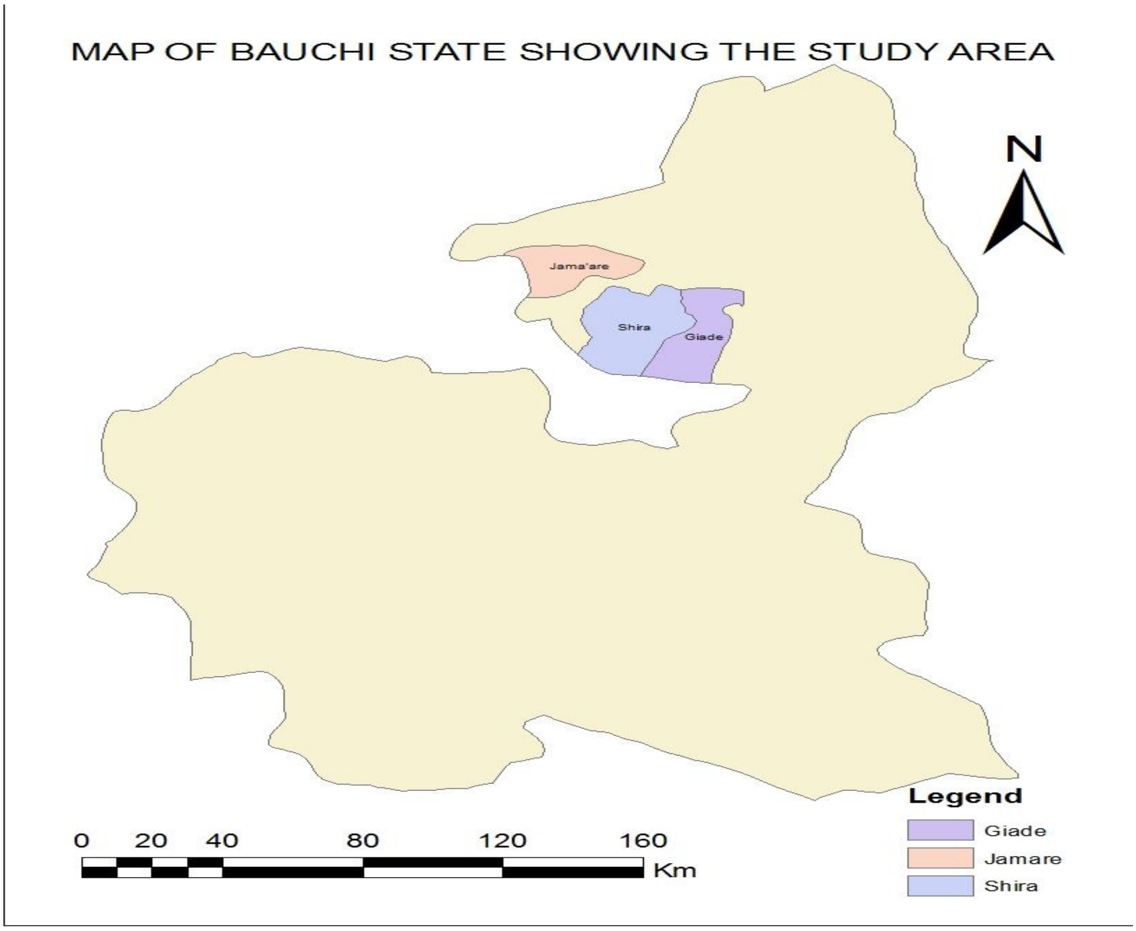

Figure 1. Map of Bauchi state showing study sites

Kinetic Experiments

Kinetics of desorption of added sulphate

For the sorption experiment, 2.5 g of 2 mm sieved soil was placed in separate plastic tubes and 15 ml of 0.01 M NaCl containing 50 mg S g^{-1} (K_2SO_4) was added from a 1000 mg S g^{-1} sulphate stock solution. The soil samples were shaken intermittently for a predetermined equilibrium time of 24 hours. After equilibration, the samples were filtered. The filtrate thus collected was analyzed for SO_4^{2-} by a turbidimetric method by Tabatabai (1982). Adsorbed sulphate was calculated as the difference between added sulphate and sulphate remaining in the solution.

For the desorption experiment, a soil sample with adsorbed S after separating solution sulphate was used for the desorption study. The soil sample was shaken with 15 ml of $Ca(H_2PO_4)_2$ solution containing 500 mg P l^{-1} for 30, 60, 150, 180, and 240 minutes. After shaking, the supernatant was filtered and analyzed for SO_4^{2-} by the turbidimetric method of Tabatabai (1982). The amounts and percentages of sulphate desorbed were calculated concerning the sorbed SO_4^{2-} as follows:

SO_4^{2-} desorbed (mg/g) = SO_4^{2-} recovered (mg/g) – analyzed SO_4^{2-} in solution (mg/g)

Kinetics Modelling of Sulphate Desorption

First-order equation

Lagergren`s first-order kinetic model often describes reactions in the soil mineral/water reactions (Sparks, 2002). The first-order equation is generally expressed as follows:

$$\ln qt = \ln (qo) - (k_1/2.303)\, t \qquad \ldots\ldots\ldots (1)$$

Where In qo and qt are the amounts of sulphate desorbed (mg/g) at equilibrium and at time t (min), respectively, and k_1 is the rate constant (1 /min). Values of qo and k_1 are calculated from the slope and intercept of the plot of lnqt versus t.

Second-order equation

Kinetic rate equation of second order after integration is expressed as:

$$\mathbf{1/q_t = 1/k_2q_e^2 + (1/q_e)\,t} \quad \ldots\ldots\ldots\ldots\ldots(2)$$

Where qe and qt are the amounts of sulphate desorbed (mg/g) at equilibrium and at a time (t) (min), respectively, and k_2 is the equilibrium rate constant (g/mg/min). Values of k_2 and q_e are calculated from the slope and intercept of the plot of $1/q_t$ versus t.

Elovich model

The Elovich equation was originally developed to describe the kinetics of heterogeneous chemisorption of gases on a solid surface (Sparks, 2005). The linear form of the Elovich model is presented by the following equation:

$$\mathbf{q_t = 1/\beta\, In\, (\alpha\beta) + 1/\beta\, In\, t} \quad \ldots\ldots(3)$$

Where q is the amount of sulphate adsorbed/desorbed (mg/g) at a time t (min), α and β are constants. Thus, the constant can be obtained from the slope (1/β) and intercept (1/β) ln (αβ) of the linear plot of q_t versus lnt.

Fractional power model

The linear form of the fractional power model is given in the equation below (Ho and McKay, 2002; Sparks, 2011).

$$\mathbf{q_t = kt^n} \quad \ldots\ldots\ldots\ldots\ldots\ldots(4)$$

$$\mathbf{lnq_t = lnk + nlnt} \quad \ldots\ldots..(5)$$

Where qt is the amount of sulphate desorbed (mg/g) at a time t (min), where k and n are constant with $n < 1$.

Parabolic diffusion model

The parabolic diffusion model often suggests that diffusion-controlled phenomena are rate-limiting (Sparks, 2011). The equation can be expressed in the linear forms given by Sparks (2002):

$$\mathbf{qt = Rt^{1/2} + b} \quad \ldots\ldots\ldots(6)$$

Where qt = quantity of S desorbed (mg/kg) at time t (min); R = an overall diffusion coefficient. Thus, a plot of q_t versus $t^{1/2}$ and b is a constant.

Statistical Analysis

Two-way Analysis of variance (ANOVA) was used to analyze the variation among parent materials. Statistically, significant means were separated by the Least Significant Difference (LSD). All the statistical analyses were carried out at a 95% confidence level using SAS 9.2 (2011).

Fitness of the kinetic models

It is assumed that the model or equation which gives the highest R^2 and lowest standard error of estimates (SE) values is the best model or equation for describing kinetics sulphate of desorption data. The standard error was calculated as follows:

$$SE = \sum [(S_a\text{-}S_b)^2/ (n\text{-}2)]^{0.5}$$

Where S_a and, S_b represent the measured and predicted sulphate desorbed, respectively, and n is the number of measurements.

RESULTS AND DISCUSSION

Soil Physical and Chemical Properties

The physical and chemical properties of soils from three parent materials, namely; Chad formation (CF), Basement complex rock (BCR), and Kerri-Kerri formation (KKF) are shown in Table 1. Sand content was dominant in all three soil parent materials (Table 1). This was also found by many soil scientists (Odunze, 2006; Obi and Akinbola, 2009, Obalum *et al*., 2012; Maniyunda *et al*., 2014; Hassan *et al*., 2016) all for different Nigerian savanna soils. The mean values of sand, silt, and clay fractions in soils from the three-parent materials did not significantly vary ($p > 0.05$) (Table 1). The lack of differences might be attributed to the old nature of tropical soils that are generally highly weathered. Generally, the soils were dominantly sandy loam except in some soils developed from BCR and KKF (Table 1) that were sandy clay loam indicating higher clay contents of the area from the soil samples were obtained.

The soil pH in water was rated acidic in all soil parent materials (Table 1) and is within the range values reported by Raji and Muhammad (2000) for Nigerian savanna soils. The soil pH in $CaCl_2$ was lower

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compared to soil pH in water and the difference between pH in $CaCl_2$ to pH in water was negative and fairly large indicating the dominance of variable charge minerals. The mean values of soil pH in water and $CaCl_2$ were significantly ($p > 0.05$) different among soil parent materials (Table 1). Such differences are expected since the soils developed from different parent materials. This observation corroborates the findings of Jaiyeoba (2006) for soil derived from different parent materials in Nigeria.

The soils are low in organic carbon (Table 1), irrespective of soil parent materials. The mean values of organic carbon in all soils were not significantly different ($p > 0.05$) indicating no influence of parent materials on the organic carbon content of the soils.

Table 1. Selected physical and chemical properties of some soils developed from three different parent materials in Bauchi North, Nigeria

Parameters	CF	BCR	KKF
Sand (g / kg)	778.30a	739.70a	6734.7a
Silt (g / kg)	128.70a	144.40a	141.40a
Clay (g / kg)	98.50a	115.90a	123.90a
Texture (USDA)	SL	SL – SCL	SL – SCL
pH in water	6.54a	6.20b	5.86c
pH in $CaCl_2$	5.48a	5.06b	4.84b
Org. C (g / kg)	5.30a	5.16a	5.37a

CF= Chad Formation, BCR= Basement complex rock, KKF= Kerri-Kerri Formation, SL= Sandy loam, SCL= Sandy clay loam. The data presented are average means. This means that within the same column with the same common lowercase letters, they are not significantly different by 5%

Different Models to Describe Sulphate Desorption Kinetics in Some Soils Derived from Various Parent Materials

The model with appreciable R^2, but higher standard errors (SE) is not a better model to describe experimental data (Abdu, 2006; Rezaei and Naeini, 2009; Brajendra *et al.*, 2013; Garg *et al.*, 2016). The kinetic equations along with values of coefficient of determination (R^2) and standard errors (SE) of soils derived from three different parent materials are presented in Table 2. The ordered or mechanistic models (First-order and second-order) and diffusional models (Elovich, Fractional-power, and parabolic diffusion equations) were used to compare the best fit to the experimental data that satisfactorily describe the sulphate desorption data.

Ordered or Mechanistic models

The first order was the best in all studied soils to describe the reaction rate of S desorption data as evidenced by the overall highest values of R^2 and the lowest values of SE (Table 2). This observation conforms to the findings of previous workers on sulphate desorption data (Abdu *et al.*, 2008; Brajendra *et al.*, 2013; Garg et al., 2016). The goodness

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of fit of the first-order equation on sulphate desorption data from soils derived from the three different parent materials was in the order as follows: CF>BCR>KKF (Figure 1,2,3). The conformity of this experimental data to the first-order equation is an indication of probable ligand exchange of sulphate ions during the desorption process (Abdu *et al*., 2008; Abdu, 2013).

The second-order equation is not suitable to describe S desorption in all the studied soils as can be seen from the large values of SE compared to the first-order equation, the values of R^2, however, are quite high (Table 2). Similar observations were reported by Abdu (2013) for phosphate desorption data, and also by Brajendra *et al*. (2013) and Garg *et al*. (2016) for sulphate release data.

Table 2. Coefficient of determination (R^2) and standard error of estimate (SE) of various kinetic equations for sulphate desorption in some soils developed over different parent materials

Equation	Parent material					
	CF		BCR		KKF	
	R^2	SE	R^2	SE	R^2	SE
First-order	0.9147	0.34	0.9207	0.76	0.976	0.93
Second-order	0.8938	3.53	0.8741	3.46	0.9649	8.28
Elovich	0.7994	2.60	0.7882	2.34	0.8953	1.99
Fractional power	0.7698	3.60	0.7499	3.37	0.8725	3.26
Parabolic diffusion law	0.876	0.00013	0.8678	0.00013	0.954	0.00082

CF= Chad formation, BCR= Basement complex rock, KKF= Kerri-Kerri formation, SE= Standard errors

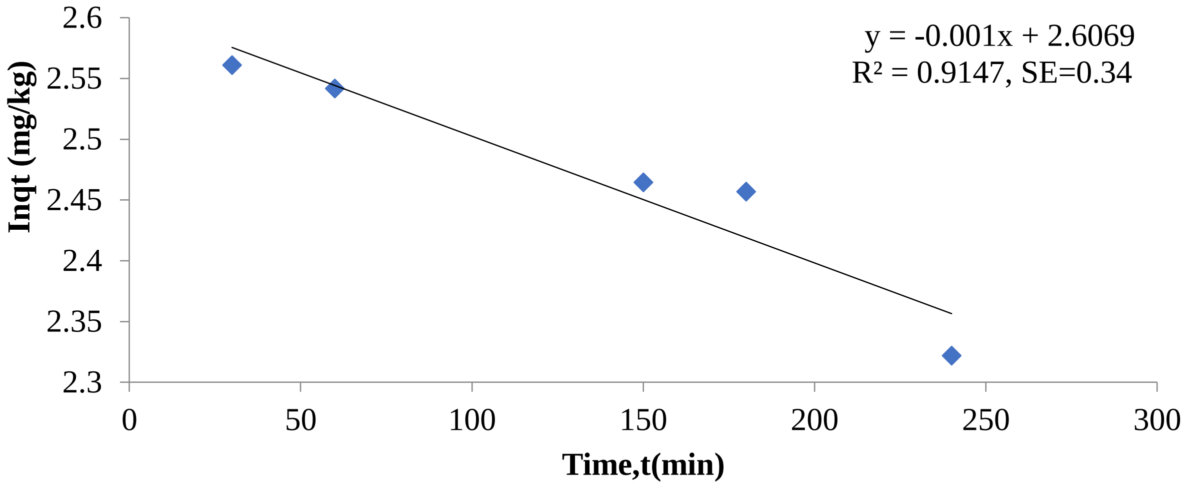

Figure 1. The plot of the first order for describing desorption data from soils developed over the Chad Formation (CF)

Empirical models

The parabolic diffusion, fractional power, and the Elovich models have been regarded as empirical kinetic models (Abdu, 2006). The kinetic of S desorption in all studied soils derived from three different parent materials was also best described by the parabolic diffusion equation (Table 2) as characterized by relatively high R^2 values and low SE values. This observation suggests intra-particle diffusion-controlled exchange

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in all studied soils. This was also found by Garg *et al*. (2016) for sulphate release data from Alfisols soils in India, and Abdu (2006) for phosphate release data in Nigerian savanna soils. The fitness of the parabolic diffusion equation could be arranged in the following order: CF>BCR>KKF (Figure 2.4-2.6).

Both Elovich and fractional-power equations failed to successfully describe S desorption data in this study as can be seen from their large values of SE, though they have appreciable R^2 (Table 2) by the soil of all the different parent materials. This was contrary to the findings of Abdu (2006) who successfully used the Elovich equation to describe phosphate release data in soils from the Nigerian savanna. Recent work by Brajendra *et al*. (2013) on the kinetics of sulphate released by soils showed that Elovich was not suitable to describe S desorption by soils as characterized by the large values of SE.

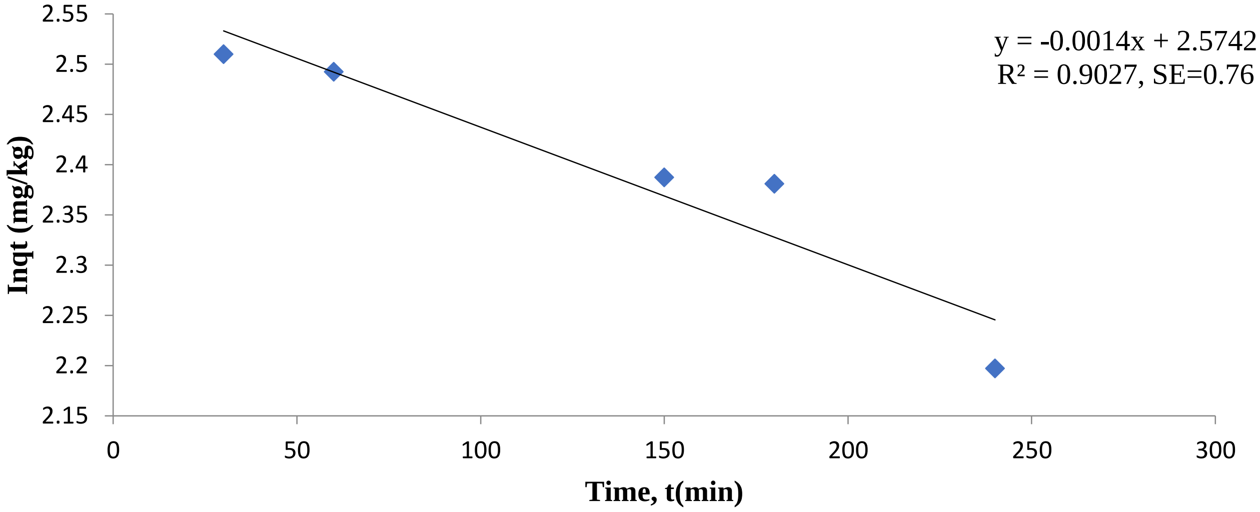

Figure 2. The plot of the first order for describing desorption data by soils Basement complex rock (BCR)

A comparison between best fit kinetic equations (First-order and parabolic diffusion models) by soils that developed over three-parent materials showed that the parabolic diffusion equation could fit the sulphate desorption data better than the first-order equation as can be seen from the lowest values of SE (Table 2). The fitness of sulphate desorption data to the parabolic diffusion model suggests that what governs the kinetics desorption of sulphate by soil was the diffusional process. Similarly, Sparks (2011) noted that the overall kinetic process obeys a diffusion equation since diffusion is the rate-limiting process.

Therefore, many soil chemists speculated that the rate-controlling step in the adsorption or desorption of nutrients was the diffusion of the ions into the subsurface layers of the solid matrix (Sivasubramanian and Talibudeen, 1972; Agbenin and Raij, 2001; Abdu, 2006, 2009; Abdu *et al*., 2008). However, given R^2 and SE of the first-order model, it seems to be good for describing S desorption data.

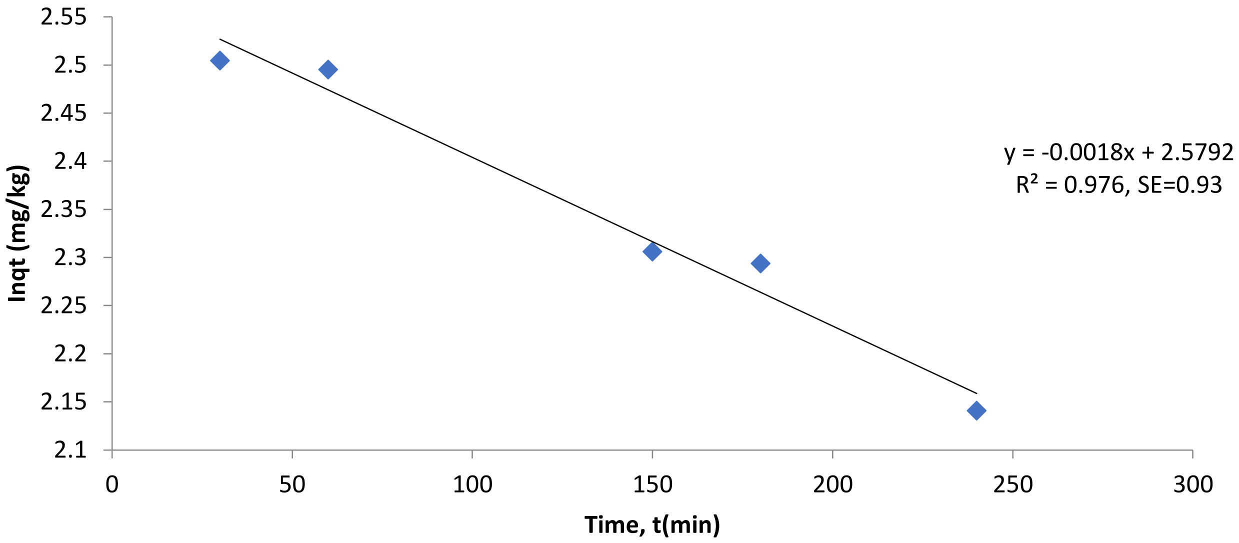

Figure 3. The plot of the first order for describing desorption data by soils Kerri-Kerri Formation (KKF)

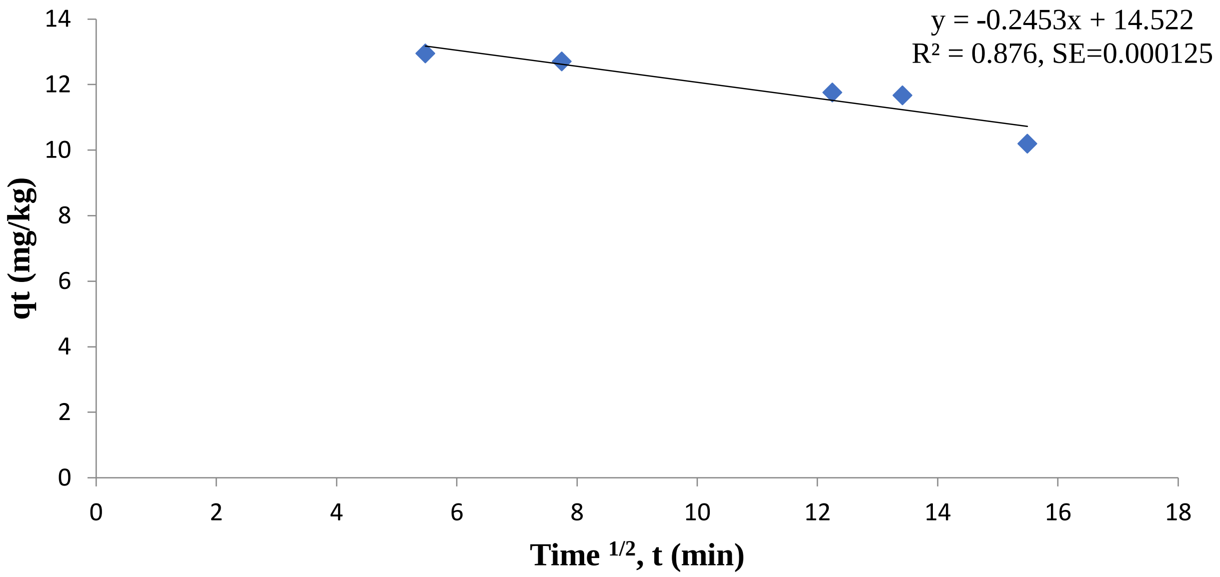

Figure 4. The plot of parabolic-diffusion for describing desorption data by soils Chad Formation (CF)

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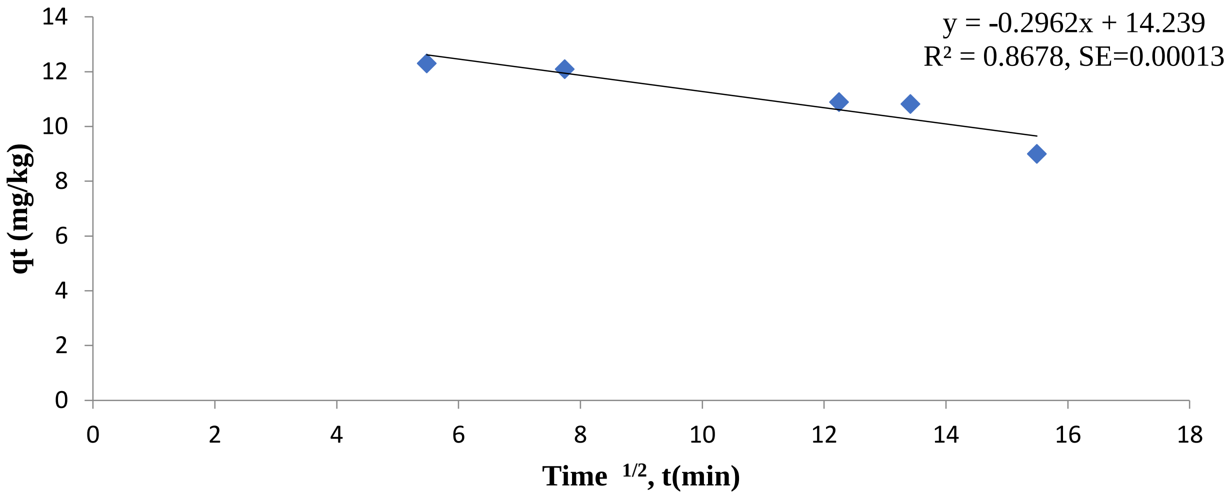

Figure 5. The plot of parabolic-diffusion for describing desorption data by soil Basement complex rock (BCR)

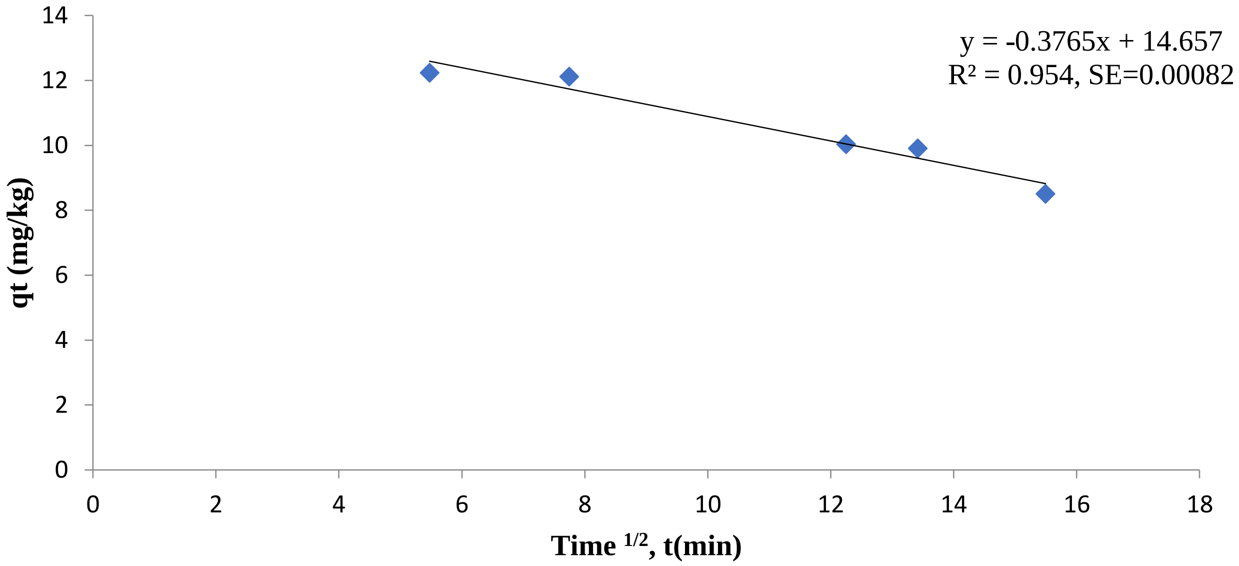

Figure 6. The plot of parabolic-diffusion for describing desorption data by soil Kerri- Kerri Formation (KKF)

CONCLUSION

A variety of kinetic models were tested to compare the best-fitted models to describe S desorption data obtained from soil samples taken in selected Bauchi-north parent materials in the Nigerian Sudan savanna. Parabolic diffusion and first order were found to describe S desorption satisfactorily, as evidenced by their relatively high R^2 values and lowest SE values in all soil parent materials from the northern Bauchi in the Nigerian Sudan savanna. Conformity of S desorption data to the first-order equation is an indication of probable ligand exchange of sulphate ion during the desorption process and the parabolic diffusion equation suggests that diffusion-controlled phenomena are rate-limiting steps. Therefore, the modelling of sulphate is critical in predicting the fate of added S, mobility, and in formulating the fertilizer schedule for raising optimum crop production the because of the low recovery of this ion in most agricultural soils.

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