

Study of phosphorus adsorption behaviour of soils of the Qazvin plain, NW Iran

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Abstract

Received: 2023-07-19

Accepted: 2024-02-04

Published online: 2024-02-04

Associated editor: A. Horel

Keywords:

Adsorption
Alkaline soils
Fertilizer
Isotherm
Kinetics
Phosphorus
Saline soils

Information of phosphorus adsorption characteristics in soils is of great importance for determining and applying proper management strategies in order to improve crop productivity and reduce environmental damages. A comprehensive equilibrium and kinetic adsorption study was designed to evaluate the adsorption behavior of phosphorus and the effect of soil properties on phosphorus retention in four different agricultural soils of Qazvin plain. The results showed that the Langmuir isotherm ($r^2 = 0.998$, $s = 0.007$), and Kuo and Lotse equation ($r^2 = 0.946$, $s = 0.009$) had the highest accuracy in equilibrium and kinetic modeling of phosphorus adsorption onto the soil surface. The results of linear regression analysis showed that the maximum adsorption capacity (q_m) was positively correlated with clay and calcium carbonate contents and negatively correlated with the percentage of organic matter ($p < 0.05$). Additionally, the effects of three distinct parameters including salinity (0 to 30 ppt), temperature (12 to 38°C), and pH (2.5 to 11.5) on phosphorus adsorption were found to be significant ($p < 0.05$; $p < 0.01$). According to these results, by changing the temperature from 12 to 25°C and 25 to 38°C, the maximum adsorption capacity increased in the range of 1.4–1.9 times. A rapid 82% increase in the adsorptive removal of phosphorus was observed at pH 5.36 to 7.5. However, with increasing salinity, the amount of adsorption decreased gradually and reached a plateau of about 0.189 mg g⁻¹ over 10 ppt salinity. Furthermore, the results of thermodynamic test of the investigated soils showed that the process was endothermic and spontaneous.

1. Introduction

Phosphorus is considered one of the most important soil nutrients for plant growth and development (e.g. Raghothama and Karthikeyan, 2005). In agricultural areas, high amounts of phosphate fertilizers are applied every year in order to increase the physiological efficiency of crops, and a part of the added phosphorus is not used by the plants due to phosphorus fixation or sorption (e.g. Narsian and Patel, 2000; Idris and Ahmed, 2012). Following the accumulation of phosphorus and the saturation of binding sites on soil particles, the ability of soil to retain phosphorus decreases and the possibility of phosphorus losses through runoff or drainage water increases, which can lead to eutrophication of aquatic environments (e.g. Sims et al., 1998; Smith et al., 1999; Allen et al., 2006; Torma et al., 2020).

Comprehending adsorption-desorption process, its associated mechanisms, as well as models describing it, are essential for understanding phosphorus transport from high-P soils to plants, groundwater, or surface waters. Phosphorus adsorption and desorption characteristics have been analyzed in several studies using different equilibrium isotherms such as Langmuir

and Freundlich equations (e.g. Xia and Wang, 2009; Wolde and Haile, 2015; Bai et al., 2017; Yang et al., 2019), and kinetic models such as the pseudo-first-order, and pseudo-second-order models (e.g. Hamdi et al., 2014; Zafar et al., 2016). The information obtained from the adsorption/desorption models can help develop effective fertilizer use strategies based on phosphorus availability and mobility in the soils and subsequently optimize crops yield while reducing environmental hazards (e.g. Maguire et al., 2001; Zhang et al., 2005).

Phosphorus adsorption and its bioavailability in soils are controlled by clay, Al and Fe oxides, exchangeable Ca content, soil texture, organic matter, pH, hydraulic conductivity, degree of phosphorus saturation and ionic strength of soil solution, as well as by environmental factors such as salinity and temperature (Bubba et al., 2003; Moazed et al., 2010; Debicka et al., 2015; Bai et al., 2017). Management practices (including different agricultural and tillage systems) also affect the amount of phosphorus retention in the soil by changing properties such as pH, calcium carbonate and organic matter content (e.g. Debicka et al., 2015; Fink et al., 2016; Zafar et al., 2016; Nobile et al., 2018). Despite advances in research about phosphorus dynamics and adsorption in the soils with dif-

ferent physicochemical properties (Cheung and Venkitachalam, 2006; Santos et al., 2011; Zhang et al., 2015; Guedes et al., 2016), the role of soil properties in phosphorus adsorption behaviour of soils is less studied (Thomas et al., 2015; Wolde and Haile, 2015; Antoniadis et al., 2016; Campos et al., 2016; Nobile et al., 2018), and even less information is available for saline soils (Hussain et al., 2003; Zhang and Huang, 2011; Bai et al., 2015). Furthermore, there are still many uncertainties regarding how the phosphorus adsorption process is influenced by the changes made in soil properties due to soil management practices.

None of the previous studies has addressed phosphorus behavior in soils of different areas of Qazvin plain. Qazvin plain is one of the most important plains of Iran in terms of cultivating various crops and using phosphate fertilizers in this plain is common among farmers. Therefore, the objectives of this research were to (1) assess equilibrium isotherms of phosphorus adsorption including the linear, Langmuir, Freundlich and Van Huay equations in four agricultural soil samples from this region; (2) study the correlation between the maximum adsorption capacity and soil properties including the content of clay, calcium carbonate and organic matter; (3) examine the changes in phosphorus adsorption over time using kinetic models including the pseudo-first-order, pseudo-second-order, Kuo and Lotse (1974), Panda et al. (1978) and Barrow and Shaw (1975) equations; (4) investigate the effect of salinity, pH, and temperature on phosphorus adsorption.

2. Materials and methods

2.1. Study area

The soil samples were collected from four villages of Mehdi Abad (36°09' N 49°52' E), Kamal Abad (36°08' N 50°03' E), Zaferan (36°06' N 50°06' E), and Koochar (36°07' N 50°04' E)

Table 1
Region climatology and soil taxonomy of the studied soils

Location/Soil	USDA soil taxonomy		Region climatology		
	Order	Suborder	Climate	mean annual temperature	mean annual precipitation
Zaferan (Z)	Aridisol	Xeric Haplocambid	Cold semi-arid	13.5°C	256.5 mm
Koochar (K)	Aridisol	Xeric Haplocambid			
Mehdi Abad (MA)	Aridisol	Xeric Haplocalcid			
Kamal Abad (KA)	Aridisol	Xeric Haplocalcid			

Table 2
Physical and chemical parameters of the studied soils (Z, K, MA, KA)

Location/Soil	ECe (dS m ⁻¹)	pH	CaCO ₃ (%)	Organic matter (%)	Total P (mg kg ⁻¹)	DTPA extracted Fe (g kg ⁻¹)	Sand (%)	Silt (%)	Clay (%)	Soil texture
Zaferan (Z)	0.77	7.18	19.00	0.54	735.0	5.6	28	35	37	Clay loam
Koochar (K)	0.65	7.88	13.22	1.00	807.8	5.2	40	29	31	Clay loam
Mehdi Abad (MA)	0.66	8.22	12.56	0.83	871.0	8.0	32	27	41	Clay
Kamal Abad (KA)	0.85	8.17	14.69	0.80	688.4	6.2	44	24	32	Clay loam

located in the north of Qazvin plain for phosphorus adsorption studies. All the samples were taken from fine textured or heavy agricultural soils (with long-term use of phosphate fertilizers in agricultural production), where phosphorus adsorption onto soil particles is visible. The studied soils belong to the Quaternary period with high level piedmont fan and valley terrace deposits. The information related to the classification of the studied soils (taken from the GIS-based soil maps of the study area prepared by Soil and Water Research Institute) and the region climatology (Saghafian and Sanginabadi, 2020) is given in Table 1.

2.2. Soil collection and analysis

In the late spring of 2018, soil samples were collected from the four sites (0–30 cm depth) and were air-dried, crushed and sieved (<2 mm) to determine the physicochemical properties and to conduct adsorption experiments. The available iron was extracted by DTPA solution and measured by atomic absorption spectroscopy (Shimadzu AA-6200) (Lindsay and Norvell, 1978). The total phosphorus content of the soil samples was measured by the nitric acid and perchloric acid digestion methods using a spectrophotometer (Shimadzu UV-1700) at a wavelength of 470 nm and Ammonium Molybdate-Vanadate solution as a reagent according to Murphy and Riley (1962) and Kuo (1996). Soil organic matter content (organic carbon) was determined based on Walkley-Black wet oxidation method (Walkley and Black, 1934). Soil calcium carbonate percentage was also determined by the titration method (Loeppert and Sparks, 1996). The results of soil analysis are presented in Table 2.

2.3. Equilibrium adsorption studies

A standard method recommended by SERA-IEG 17 was used to perform equilibrium experiments (Nair et al., 1984).

One gram of dried and crushed soil (from each of the samples including Z, K, MA and KA) passing through a 2-mm sieve was weighed and placed in 60 mL containers. Solutions with different phosphorus concentrations of 0, 5, 10, 15, 20, 30, and 80 mg l⁻¹ were prepared by adding certain amounts of potassium monophosphate, KH₂PO₄ (Merck) in 0.01 M Calcium Chloride, CaCl₂ (Merck) solution. 25 ml of the solution was then added to each soil sample to give a 1:25 ratio of soil to the solution. Three drops of chloroform were added to each container containing soil and solution to prevent microbial activity. Soil suspension samples were placed in a shaker at 250 rpm and 25°C for 24 hours. After this time, the suspensions were allowed to settle for one hour and the samples were filtered through a Whatman No. 42 filter paper. The concentration of phosphorus in the transparent solutions was then measured by the vanadate molybdate method using a spectrophotometer (SpectroDirect/Lovibond) at a wavelength of 470 nm (Pierzynski, 2000). The amount of adsorbed phosphorus in each soil was determined by equation 1 (Wolde and Haile, 2015).

$$q_e = \frac{(C_0 - C_e)\forall}{w} \quad (1)$$

Where q_e is the amount of phosphorus absorbed by the soil (mg g⁻¹), C_0 is the initial concentration of phosphorus (mg l⁻¹), C_e is the concentration at equilibrium (mg l⁻¹), \forall is the volume of solution (l) and w is the weight of soil (g).

2.4. Equilibrium isotherms

Linear (Moazed et al., 2010), Langmuir (Langmuir, 1918), Freundlich (Freundlich, 1907), and Van Huay (Kassa et al., 2019) isotherms were fitted to the experimental data related to equilibrium phosphorus adsorption onto all four soil samples (Equations 2, 3, 4, and 5). The models fitted well with the phosphorus adsorption data of the soils and the adsorption coefficients of the governing equilibrium equations were determined based on a trial-and-error procedure using the solver add-in with spreadsheet, Microsoft Excel 2010 software.

$$q_e = K_d C_e \quad (2)$$

$$q_e = \frac{q_m K_l C_e}{1 + K_l C_e} \quad (3)$$

$$q_e = K_f C_e^n \quad (4)$$

$$q_e = m + n\sqrt{C_e} \quad (5)$$

Where q_e is the equilibrium adsorption capacity (mg g⁻¹), C_e is the equilibrium liquid phase concentration (mg l⁻¹), K_d is the adsorption constant of the solute to the stationary phase surface (l g⁻¹), q_m is the maximum adsorption capacity (mg g⁻¹), K_l and K_f are the Langmuir and Freundlich constants (l g⁻¹), n is Freundlich exponent, m is Van Haury adsorption coefficient and n is Van Huay constant parameter (l g⁻¹).

2.5. Adsorption kinetic studies

To study the adsorption kinetics of phosphorus, the soils of Mehdi Abad (MA) and Kamal Abad (KA) villages were selected with average adsorption capacity and two different textures (clay and clay loam). The contact time of phosphorus solution with soil was considered 0.17, 0.5, 1, 2, 4, 8, 16, 24, 48, and 72 hours. The preparation of the containers for each contact time was similar to the equilibrium study. The initial phosphorus concentration for all containers was 20 mg l⁻¹ in 0.01 M CaCl₂ (Merck) solution. The suspension samples that were taken from containers related to the desired time were centrifuged at 3000 rpm for 10 minutes and then passed through a filter (Eco-sensa CA033S0451 CA 33 mm 0.45 μm syringe filter). The phosphorus concentrations were measured using a spectrophotometer.

2.6. Kinetic models

To determine the appropriate model for describing the adsorption kinetics of phosphorus in the studied soil samples, the pseudo-first-order, and pseudo-second-order models as well as the equations proposed by Kuo and Lotse (1974), Barrow and Shaw (1975), and Panda et al. (1978) were studied, which are presented in Equations 6, 7, 8, 9, and 10, respectively (Kuo and Lotse, 1974; Barrow and Shaw, 1975; Panda et al., 1978; Esmaeilian et al., 2015; Zhang et al., 2015).

$$q_t = q_e [1 - \exp(-k_1 t)] \quad (6)$$

Where q_t is the amount of adsorbed phosphorus at time t (mg g⁻¹), q_e is the amount of adsorbed phosphorus at equilibrium (mg g⁻¹), k_1 is the rate constant of adsorption (1 min⁻¹), and t is the time (min).

$$q_t = \frac{q_e^2 k_2 t}{1 + q_e k_2 t} \quad (7)$$

Where k_2 is the rate constant of adsorption (g mg⁻¹ min⁻¹).

$$q_t = k C_0 t^{\frac{1}{m}} \quad (8)$$

Where C_0 is the initial phosphorus concentration (mg l⁻¹), k is the reaction rate (l g⁻¹ min⁻¹), and m is the constant coefficient.

$$q_t = k C_t^m t^n \quad (9)$$

Where k is the reaction rate (l g⁻¹ min⁻¹), C_t is the phosphorus concentration measured at time t (mg l⁻¹), m and n are constants related to the adsorption rate.

$$C_t = k t^{-n} \quad (10)$$

Where the constant k denotes phosphorus concentration in the equilibrium solution (mg l⁻¹ min⁻¹) and n is the index related to the phosphate adsorption rate in soil.

By fitting these equations to the data obtained from the adsorption kinetic experiments on the soils of Mehdi Abad (MA) and Kamal Abad (KA) villages using the Microsoft Excel add-in Solver, the constant parameters of kinetic equations were determined for the desired soils.

2.7. Effect of temperature, salinity, and pH on phosphorus adsorption

The effect of temperature at three levels of 12, 25, and 38°C, salinity at five levels of 0, 5, 10, 20, and 30 ppt (g salt per 1000 g water), and pH at six levels of 2.5, 3.5, 5.36, 7.5, 9.5, and 11.5 were studied on phosphorus adsorption in the soil of Mehdi Abad (MA) village. A 0.01 M CaCl₂ solution with the required phosphorus concentration was added to the soil samples in a ratio of 1:25. While we used soils with different initial phosphorus concentrations of 0, 15, 20, 30, 50, 80, and 100 mg l⁻¹ to examine the effect of temperature, the effects of salinity and pH were only examined on samples with a phosphorus concentration of 20 mg l⁻¹. Different levels of salinity and pH were created by adding different amounts of NaCl and also small amounts of HCl and NaOH to the CaCl₂ solution, respectively. As in previous experiments, the samples were placed in the shaker for 24 hours at 250 rpm. The temperature experiment was carried out in a factorial design to investigate two factors of temperature and P concentration, and salinity and pH tests were performed in a completely randomized design with three replications for each treatment.

2.8. Adsorption thermodynamics

The most commonly used adsorption thermodynamic model is the van't Hoff model. The thermodynamic parameters for the adsorption process including Gibbs free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) changes in Mehdi Abad (MA) soil were calculated using Equations 11, 12, and 13 for phosphorus initial concentrations of 15, 20, 30, 50, 80, and 100 mg l⁻¹ at three temperatures of 285 K (12°C), 298 K (25°C), and 311 K (38°C) (Sarathi Guru and Dash, 2012).

$$K_d = \frac{q_e}{C_e} \quad (11)$$

$$\ln(K_d) = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (12)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (13)$$

K_d is the adsorption equilibrium constant (l g⁻¹) obtained from Equation 11. By plotting $\ln K_d$ vs $1/T$, the values of slope and intercept of the corresponding line are extracted, and enthalpy and entropy changes are determined according to van't Hoff equation (equation 12). In this Equation, $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ is the gas constant. Gibbs free energy changes are also calculated for different temperatures using Equation 13.

2.9. Statistical analysis

Statistical tests were performed using SAS software 9.4. One-way analysis of variance (ANOVA) was employed to examine differences among phosphorus adsorption treatments under different levels of salinity, pH, and temperature using Tukey test at $p < 0.05$. Correlation and linear regression analysis were conducted to determine the relationship between soil physico-chemical properties and adsorption coefficients derived from the Langmuir isotherm using the same software. For each soil sample, the most appropriate equilibrium and kinetic models were determined by calculating the determination coefficient (r^2) and the standard error of estimate (s) for each of these equations as follows (Mirzaghaderi et al., 2010).

$$r^2 = \left(\frac{n \sum xy - \sum x \sum y}{\sqrt{(n \sum x^2 - (\sum x)^2)(n \sum y^2 - (\sum y)^2)}} \right)^2 \quad (14)$$

$$s = \sqrt{\frac{\sum (y - x)^2}{n - 2}} \quad (15)$$

Where n is the number of observations and x and y are the observed and predicted values, respectively.

3. Results and discussion

3.1. Equilibrium adsorption and its isotherms

The adsorption parameters obtained by fitting the studied isotherms to experimental data along with r^2 and s for different soils are listed in Table 3. According to the results, Langmuir isotherm was the most accurate isotherm in estimating equilibrium adsorption ($r^2 = 0.867-0.998$). The Freundlich and Van Huay equations also showed a good correlation with the laboratory data ($p < 0.01$). Our results are similar to the experiences by Moazed et al. (2010), who studied the phosphorus adsorption of silty, clayey, and loamy acidic soils with low cation exchange capacity. Bai et al. (2017) found that the Langmuir isotherm model provided a better fit for phosphorus adsorption onto marsh soils with $\text{pH} > 8.3$ and clay $> 1.9\%$ compared to the Freundlich model. However, in another study on different textured soils (loam, clay loam, and silt loam) with pH ranging from 4.4 to 7.6, the Freundlich equation was expressed as the most appropriate adsorption governing equation (Wolde and Haile, 2015).

In general, the difference in the equations fitted to phosphorus adsorption data in various experiments is related to a combination of different physical and chemical properties in the soils and adsorption sites (Hussain et al., 2003). For example, in soils with homogenous adsorption sites, phosphorus adsorption data are described by the Langmuir equation. While phosphorus adsorption properties in soils with heterogeneous adsorption sites fit on the Freundlich isotherm better than other equations (Sparks, 2003).

Table 3

Adsorption parameters of four equilibrium isotherms for the soil samples (Z, K, MA, KA)

Soil sample	Langmuir				Freundlich			
	q_m	K_l	r^2	s	K_f	n	r^2	s
Z	4.91E-01	8.28E-02	0.984**	0.019	8.52E-02	3.88E-01	0.955**	0.032
K	3.07E-01	7.64E-02	0.867**	0.037	5.73E-02	3.61E-01	0.839**	0.040
MA	4.17E-01	9.29E-02	0.998**	0.007	7.96E-02	3.70E-01	0.962**	0.026
KA	3.98E-01	6.15E-02	0.975**	0.020	5.33E-02	4.34E-01	0.916**	0.035
Soil sample	Linear			Van Huay				
	K_d	r^2	s	m	n	r^2	s	
Z	7.89E-03	0.722*	0.132	2.81E-02	5.26E-02	0.929**	0.040	
K	4.59E-03	0.597*	0.096	2.35E-02	3.08E-02	0.805**	0.044	
MA	6.77E-03	0.703*	0.122	3.01E-02	4.50E-02	0.925**	0.036	
KA	5.75E-03	0.709*	0.097	8.17E-03	4.08E-02	0.904**	0.037	

* p<0.05.

** p<0.01.

In Fig. 1, the amount of adsorbed phosphorus versus the equilibrium phosphorus concentration obtained from laboratory measurements for all soil samples is plotted as scatter points. In Fig. 1, the Langmuir isotherms fitted to the data of different soil types are also depicted as lines. As can be seen in Fig. 1, the amount of adsorbed phosphorus increased with increasing equilibrium concentration in all four soil samples. However, in lower equilibrium concentrations, the slope of the changes in the amount of adsorbed phosphorus was more than the ones for higher equilibrium concentrations. For example in Mehdi Abad (MA) soil, an increase in initial phosphorus concentration from 5 to 10 mg l⁻¹ led to a 78% increase in the amount of adsorbed phosphorus, while the concentration increased from 30 to 80 mg l⁻¹ increased the adsorbed phosphorus by 30%. Similar results were obtained by other researchers in a wide variety of soils with different physicochemical properties (Idris and Ahmed, 2012; Wolde and Haile, 2015; Yang et al., 2019). In soils with this shape of adsorption curve, a strong bond is formed between phosphorus and soil particle surfaces at low concentrations of

phosphorus, but with increasing phosphorus concentration in solution and decreasing available adsorption sites, phosphorus adsorption decreases and eventually stabilizes (Sparks, 2003).

The comparison of the graphs in Fig. 1 showed that the soil of Zaferan (Z) village had the highest adsorption values at different initial concentrations and after that, the adsorption values in descending order were related to Mehdi Abad (MA), Kamal Abad (KA), and Koochar (K) soils. According to the data in Table 3, the values of the phosphorus maximum adsorption capacity (q_m coefficient in Langmuir equation) for the soil of Zaferan (Z), Mehdi Abad (MA), Kamal Abad (KA), and Koochar (K) villages were 0.49, 0.42, 0.40, and 0.31 mg g⁻¹, respectively. The second parameter of the Langmuir isotherm (K_l) is related to the binding energy of the soil sample and the affinity of soil for phosphorus (Munhoz et al., 2011; Wang and Liang, 2014; Debicka et al., 2015). The highest values of K_l were observed in Mehdi Abad (MA) and Zaferan (Z) soils, with 0.0929 and 0.0828 l g⁻¹, respectively. These constants (q_m and K_l), which have been investigated in several studies (Moazed et al., 2010; Debicka et al., 2015;

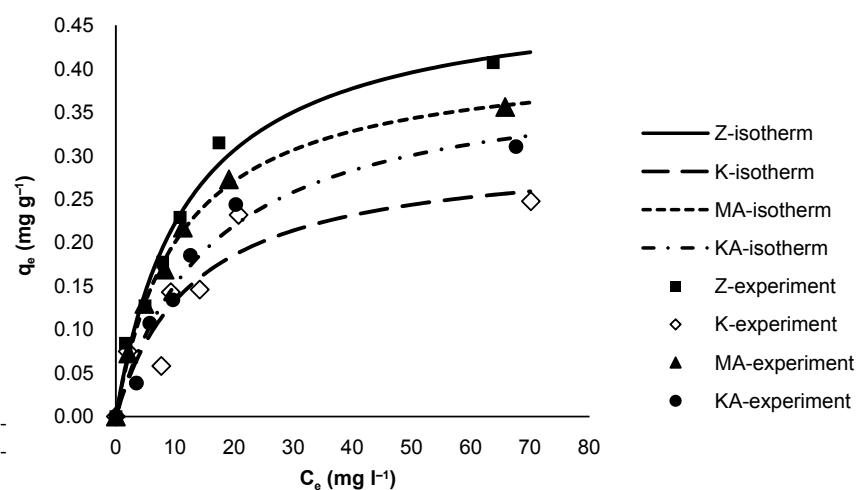


Fig. 1. Relationship between equilibrium phosphorus concentration and adsorbed phosphorus (laboratory and Langmuir isotherm) for the four soil samples (Z, K, MA, KA.)

Wolde and Haile, 2015; Bai et al., 2017), can be used to discriminate the studied soils based on their phosphorus sorption capacities which can lead to the proper design of phosphorus management strategies for improving phosphorus nutrition of crops and limiting phosphorus mobility in agricultural soils.

3.2. Correlation between soil parameters and adsorptions coefficients

The results of the relationship between soil properties and adsorption coefficients of the Langmuir equation which had the highest accuracy in modeling the equilibrium phosphorus adsorption in the studied soils are presented in Table 4. According to these results, the parameters of organic matter percentage, calcium carbonate percentage, sand and clay percentages had the highest correlation with maximum adsorption capacity (q_m) with r values of -0.97 , 0.75 , -0.70 , and 0.61 , respectively. Despite the high values of these correlation coefficients, only soil organic matter percentage had a significant correlation ($p < 0.05$).

The maximum adsorption capacity is used to evaluate the number of adsorption sites per unit weight of soil (Hiradate and Uchida, 2004; Yan et al., 2013). As shown in Table 4, the maximum adsorption capacity of phosphorus increased with decreasing organic matter content and increasing calcium carbonate percentage and clay content in the soil. According to the results of several studies, the presence of calcium in the soil increases the phosphate adsorption capacity which can be related to the deposition of applied phosphorus in contact with soluble and exchangeable calcium and the formation of calcium phosphate precipitated on the soil surface (e.g. Burt et al., 2002; Cheung and Venkitachalam, 2006; Idris and Ahmed, 2012; Hamdi et al., 2014). In a study on soils from cultivated areas with the same degree of weathering, a significant increase in phosphorus adsorption was observed following an increase in pH and CaCO_3 (Antoniadis et al., 2016). However, the results of a study on several acidic soils with high percentages of clay showed that the concentration of exchangeable calcium was negatively and significantly correlated with phosphorus adsorption ($r = -0.6$) (Moazed et al., 2010). Organic matter can

Table 4
Correlation coefficients between two Langmuir adsorption coefficients and selected soil properties

Soil properties	q_m	K_l
EC	0.46	-0.74
pH	-0.53	-0.19
CaCO_3 (%)	0.75	-0.07
Organic matter (%)	-0.97*	-0.13
P	-0.28	0.83
Fe	0.24	0.48
Sand (%)	-0.70	-0.82
Silt (%)	0.49	0.48
Clay (%)	0.61	0.81

* $p < 0.05$

decrease phosphorus adsorption in the soil by competing with phosphate over the positively charged adsorption sites and affecting the required bonding energy. Also, organic matter can cause electrostatic repulsion of phosphate or a decrease in the amount of phosphorus adsorption to the soil surfaces by increasing the negative charge or reducing the positive charge on the mineral surfaces (e.g. Schwertmann et al., 1986; Gillman et al., 1989; Afif et al., 1995; Antelo et al., 2007; Jindo et al., 2023). The results of studies on the relationship between soil organic matter and phosphorus adsorption are contradictory. First an increase and then a decrease in the adsorption capacity values of calcareous soils were observed following the increase in soil organic matter content by Xia and Wang (2009). Based on other test experiences, the phosphorus maximum adsorption capacity was negatively correlated with soil organic carbon (e.g. Gillman et al. 1989; Nobile et al., 2018; Lulu et al., 2022). Fink et al. (2016) stated that the increased amounts of organic carbon present under no-tillage compared to conventional tillage led to lower values of phosphorus adsorption capacity. However, the results of some previous studies on a wide range of soils showed a significant and positive correlation between soil organic carbon and maximum adsorption capacity (Borling et al., 2001; Zhang et al., 2005; Thomas et al., 2015; Campos et al., 2016; Yang et al., 2019). The contradiction of these results may be due to the influence of factors such as soil type, clay content, pH, and Al and Fe chemical forms which can affect the phosphorus adsorption capacity of soils (Hiradate and Uchida, 2004; Wang and Liang, 2014). Clay particles due to their high specific surface areas and reactive surfaces provide more adsorption sites in the soil and therefore play an important role in increasing the phosphorus adsorption onto the soil surface (Sparks, 2003; Guedes et al., 2016). The high adsorption of phosphorus in soils with a high percentage of clay has been emphasized in separate studies which are confirmed by the result of our study (e.g. Idris and Ahmed, 2012; Brenner et al., 2019; Lulu et al., 2022). The correlation coefficients of the Langmuir adsorption constant (K_l) with different parameters in a descending order were related to phosphorus content ($r = 0.83$), sand percentage ($r = -0.82$), clay percentage ($r = 0.81$), soil electrical conductivity ($r = -0.74$). Therefore, considering the soil properties' correlation with maximum phosphorus adsorption capacity and Langmuir constant, it can be concluded that the high percentage of clay and calcium carbonate, as well as the low amount of organic matter, were the main reasons for high phosphorus adsorption of soil sample from Zaferan (Z) village. Based on the results of our research and some other studies, addition of organic amendments to soils is considered as an effective method in reducing phosphorus fixation due to the influence of organic matter on phosphorus sorption ability and the energy of phosphorus sorption (Gillman et al., 1989; Hansen et al., 2002; Wolde and Haile, 2015;).

3.3. Adsorption kinetics and its equations

The adsorption laboratory data for Mehdi Abad (MA) and Kamal Abad (KA) soils were fitted to several kinetic models. The results of the adsorption parameters, r^2 and s are presented in

Table 5. As can be seen in Table 5, all three equations of Kuo and Lotse (1974), Borrow and Show (1975), Panda et al. (1978) were fitted well to the experimental data of both soils with $r^2 > 0.8$ which were statistically significant ($p < 0.01$). However, the highest accuracy was related to Kuo and Lotse (1974), and Barrow and Show (1975) equations in both soil samples ($r^2 = 0.904$ and 0.946). The pseudo-first-order ($r^2 = 0.191$ and 0.124) and the pseudo-second-order ($r^2 = 0.376$ and 0.463) equations had the lowest accuracy in estimating phosphorus adsorption kinetics. The results of this study are in agreement with the results of Santos et al. (2011), where kinetic equations for phosphorus adsorption of several soil samples with pH in the range of 4.4 to 8.4 were investigated. However, in another study, the pseudo-first-order, pseudo-second-order, and Elovich equations were found to describe the adsorption kinetics of phosphorus in several alkaline soil samples well (Hamdi et al., 2014). According to Zhang et al. (2015), the parameters of the nonlinear pseudo-second-order kinetic model correlated most strongly with phosphorus adsorption properties for soils with different iron (2.4–9.8%), aluminum (8.1–10.6%) and calcium (0.7–10.6%) contents. Soil physical and chemical properties including pH, EC, calcium percentage, clay content, and porosity are among the parameters affecting the adsorption capacity and rate. In other words, these parameters determine the trend of changes in phosphorus adsorption over time and the type of governing kinetic equations.

The amount of adsorbed phosphorus versus time along with the fitted model of Kuo and Lotse (1974) are shown in Fig. 2. According to the results, 34, 44, and 51% of phosphorus were adsorbed onto the Mehdi Abad (MA) soil particles after 10 minutes, 8 hours, and 72 hours. Phosphorus adsorption percentages for Kamal Abad (KA) soil for the mentioned contact times were 26, 34, and 46%, respectively. Therefore, according to these observations, most changes in the amount of adsorbed phosphorus occurred within 8 hours of soil contact (86 and 74% of total phosphorus adsorption in Mehdi Abad (MA) and Kamal Abad (KA) soils, respectively) and then the rate of phosphorus adsorption decreased. The amount of adsorbed phosphorus in Mehdi

Abad (MA) soil increased by about 8% over 24 to 72 hours, and in Kamal Abad (KA) soil, changes in phosphorus adsorption over a period of 48 to 72 hours was about 4%. Hence, it seems that the adsorption of phosphorus by the studied soils reached equilibrium within approximately 24 to 48 hours.

Previous studies showed that phosphorus adsorption rates (kinetics) are significantly different for each type of soil. For example, Santos et al., (2011) experienced that while in some soil samples, the time required for adsorption of about 75% of phosphorus was 45 minutes, while for other soil samples this time was about 1020 minutes (Santos et al., 2011). Cheung and Venkitachalam (2006) in a kinetic study of the effect of several soil amendments, found that phosphorus adsorption was relatively rapid for all test materials in the first 4 hours. The rate of reaction decreased during the 4–24 hour period and slowed dramatically after 4 days. According to another study on three alkaline soils with clay and fine sandy loam textures, equilibrium was achieved within approximately 24 hours (Hamdi et al., 2014). The main reason for the differences in the time required to achieve equilibrium is the differences in physical morphology and chemical composition of the various soils and minerals (Cheung and Venkitachalam, 2006).

In general, by observing this Fig. 2, the adsorption process involved two fast and slow steps until equilibrium was reached, which was in line with the results of other studies on the adsorption kinetics of phosphorus onto minerals, soils with different physical and chemical properties, and soil amendments (Karaca et al., 2004; Cheung and Venkitachalam, 2006; Santos et al., 2011). The fast phosphorus adsorption in the first phosphorus-soil contact periods has been attributed to the high clay contents and the precipitation process in the soils with high values of pH and Ca^{2+} (Santos et al., 2011). Adsorption of phosphorus on the surfaces of recently precipitated high reactivity forms of iron oxides is also believed to be responsible for phosphorus fixation (Ryden and Pratt, 1980; Hernandedz and Meurer, 2000). In a kinetic study of phosphorus sorption by various materials such as fly ash, gypsum and red mud over periods up to 360 days, the decrease in

Table 5
Adsorption parameters of five kinetic equations for the two soil samples (MA, KA)

Soil sample	pseudo-first-order					pseudo-second-order				
	k_1	q_e	$q_e(exp)$	r^2	s	k_2	q_e	$q_e(exp)$	r^2	s
MA	1.41E-01	2.17E-01	2.57E-01	0.191	0.033	7.74E-01	2.27E-01	2.57E-01	0.463*	0.027
KA	1.26E-01	1.72E-01	2.31E-01	0.124	0.038	6.12E-01	1.84E-01	2.31E-01	0.376	0.032
Soil sample	Kuo and Lotse (1974)				Panda et al. (1978)					
	k	m	r^2	s	k	n	r^2	s		
MA	6.57E-03	1.22E+01	0.946**	0.009	1.57E+01	5.50E-02	0.902**	0.459		
KA	4.27E-03	8.78E+00	0.904**	0.013	1.76E+01	4.98E-02	0.819**	0.687		
Soil sample	Barrow and Shaw (1975)									
	k	m	n	r^2	s					
MA	1.32E-01	0.00	8.12E-02	0.946**	0.009					
KA	8.54E-02	0.00	1.14E-01	0.904**	0.013					

* $p < 0.05$, ** $p < 0.01$

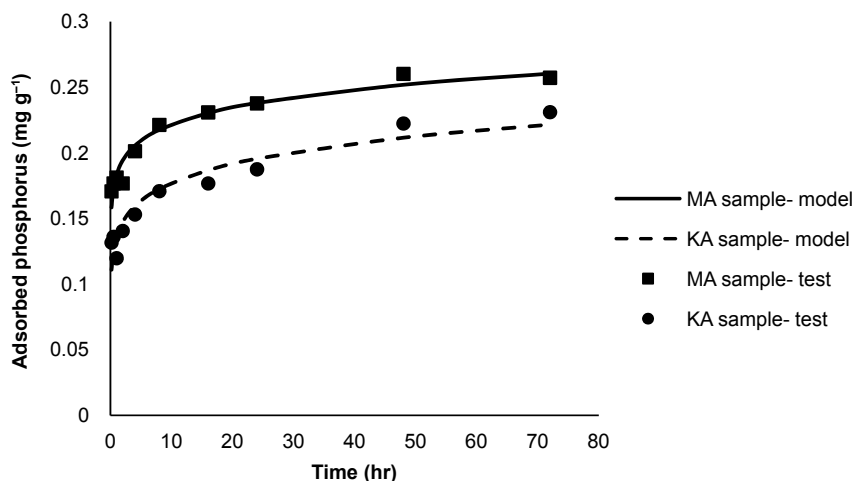


Fig. 2. Comparison of changes in the amount of adsorbed phosphorus over the 72-h time interval in the kinetic experiment and the equation of Kuo and Lotse (1974) for the two soil samples (MA, KA)

phosphorus adsorption amount over time has been attributed to factors including phosphorus penetration into the soil pores and saturation of the adsorbent surfaces by phosphorus (Cheung and Venkitachalam, 2006). The kinetic information of phosphorus sorption in soils plays an important role in understanding the dynamics of phosphorus in soils, which is required to optimize phosphorus fertilizer use efficiency in agricultural production, reduce environmental pollution and develop guidelines for the disposal of P-rich wastes onto the land (Agbenin and Reij, 2001).

3.4. Effect of salinity, pH, and temperature on phosphorus adsorption

The results of the study showed that salinity ($p < 0.05$) and pH ($p < 0.01$) had a significant effect on the rate of phosphorus adsorption in the tested soil (MA). The results of the analysis of variance of phosphorus adsorption under the influence of temperature and initial concentration factors showed significant effects of temperature, concentration as well as the interaction of two factors on phosphorus adsorption amount ($p < 0.01$). The results of ANOVA test indicated that the effect of temperature on phosphorus adsorption in soil was significant at all initial concentrations except 50 and 80 mg l⁻¹ ($p < 0.05$). The changes in the amount of phosphorus adsorption against the studied levels of salinity, pH, temperature, and initial concentration are plotted in Fig. 3., and the results of non-parametric tests are also shown in Latin letters in Fig. 3.

As shown in Fig. 3. (A), salinity values of 5, 10, 20, 30 ppt resulted in a 12, 16, 16, and 18% decrease, respectively, in the amount of adsorbed phosphorus compared to the control experiment (zero salinity). According to the results of our investigation, the highest phosphorus adsorption was observed at zero salinity with 0.227 mg g⁻¹ and the lowest adsorption was experienced at 30 ppt salinity with 0.186 mg g⁻¹. Moreover, the results of the comparison of means showed, that most of the observed changes in the amount of adsorbed phosphorus occurred up to the salinity of 10 ppt. Over 10 ppt salinity, there was no significant change in phosphorus adsorption. The main reason for the decrease in phosphorus adsorption following salinity increase is the competition of negatively charged ions in salt-containing

solution with phosphate ions over the adsorbent sites on soil particles (Reddy and DeLaune, 2008). In a research conducted by Zhang and Huang (2011), it was reported that the amount of adsorbed phosphate was approximately 1.5 times higher by halving salinity and in general, an increase in phosphorus adsorption was observed following salinity reduction in all tested sediments. Bruland and DeMent (2009) in a study on the effect of surface water salinity on phosphorus adsorption in coastal wetlands found that the phosphorus sorption index decreased with increasing surface water salinity. However, salinity was not definitive in explaining the variance in phosphorus sorption index for these wetlands.

As it is clear in Fig. 3 (B), the lowest amount of phosphorus adsorption with an average of 0.213 mg g⁻¹ was at pH 2.5 to 5.36 and with increasing pH, the amount of phosphorus adsorption in the soil sample increased, so that the highest amount of adsorption with an average of 0.417 mg g⁻¹ was at pH 7.5 to 11.5. However, in another experiment done by Liu et al. (2002), the lowest phosphorus adsorption on tidal flat surface sediments was reported in the pH range of 7 to 8, and phosphorus adsorption gradually increased by increasing acidity (pH < 7) and alkalinity (pH > 8). The results of another study showed that phosphorus adsorption decreased by up to 21% and 34% when pH increased from 4.7 to 5.9 and 7, respectively (Sato and Comerford, 2005). The fluctuation in pH results in changes in precipitation and adsorption characteristics by altering the properties associated with the surface charge (Illés and Tombác, 2006). It was reported that at high pH values, phosphorus adsorption occurs through ligand exchange between phosphorus in solution and OH⁻ on the surfaces of the soil particles (Du et al., 2016). Additionally, as shown in Fig. 3 (B), the highest rate of change in adsorption (82% increase) occurred at pH 5.36 to 7.5 while rates of change in phosphorus adsorption were relatively low at pH 2.5 to 5.36 (13%) and pH 7.5 to 11.5 (only 8%). The observed increase in adsorption capacity following the increase in pH indicates that the increase in pH leads to the formation of PO₄³⁻ species from hydrolysis products of phosphate at high pH values (Karaca et al., 2004). In a study done by Karaca et al. (2004), the increase in probability of adsorption of phosphate onto dolomite in the forms of Ca₃(PO₄)₂ and Mg₃(PO₄)₂ was reported following an increase in

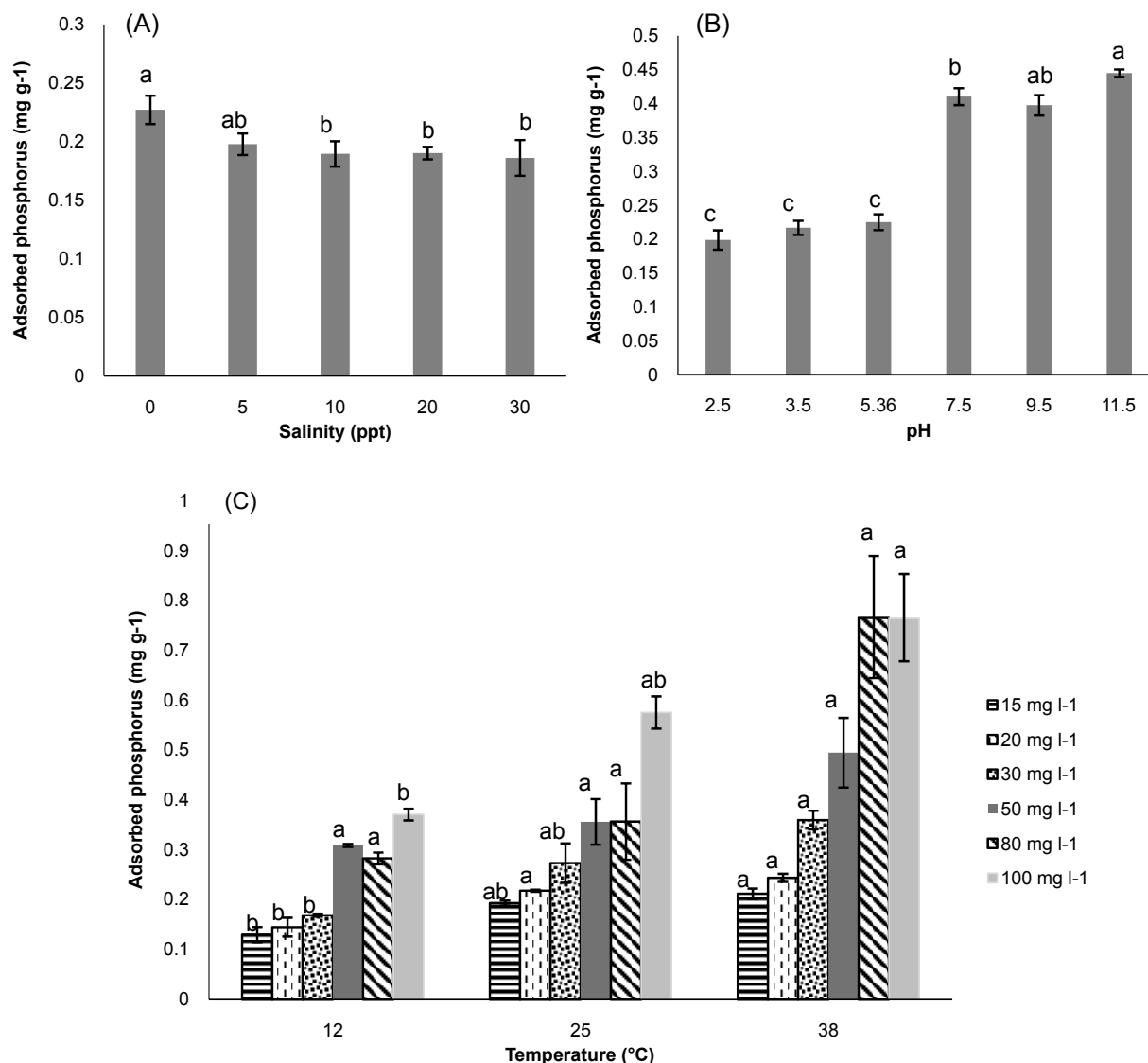


Fig. 3. The effect of different levels of salinity (A), pH (B), temperature, and initial concentration (C) on phosphorus adsorption in Mehdi Abad (MA) soil. The error bars indicate the standard deviation. Different letters indicate a significant difference (Tukey test)

pH from 1 to 11, which implies that chemisorption is a dominant mechanism in phosphate/dolomite system. In soils with low pH, Al and Fe oxides are the major sorbents of phosphorus; whereas at higher pH values, the precipitation of phosphate with Ca is expected. A pronounced transformation from adsorbed Al/Fe-P (pH<6.6) to mineral Ca-P (pH>6.6) occurs due to decreasing acidity. In general, Ca-P formation may mitigate a desorption-derived phosphorus release from sediments under alkaline conditions, while the dissolution of Ca-P minerals such as apatite and Fe/Al oxides occurs at low pH values (Gustafsson et al., 2012; Oxmann and Schwendenmann, 2014; Wolde and Haile, 2015).

As can be seen in Fig. 3 (C), the highest phosphorus adsorption in soil was observed at 38°C for all concentrations, and the increase in temperature increased the adsorption amount. However, the percentages of increase in phosphorus adsorption for a specific temperature increase (12°C to 25°C or 25°C to 38°C) were not the same at different initial concentrations. In other

words, the effect of temperature increase on the rate of increase in adsorption was affected by the initial concentration of phosphorus. For example, the temperature increasing from 25°C to 38°C at two lower initial concentrations (15 mg l⁻¹ and 20 mg l⁻¹) resulted in an increase of 10% and 12% in phosphorus adsorption, respectively. However, the phosphorus adsorption approximately doubled (115% increase) for this temperature change at an initial concentration of 80 mg l⁻¹. It is also observed that the same temperature increase of 13°C, for all studied initial concentrations, resulted in different changes in the amount of adsorbed phosphorus for two temperature intervals of 12–25°C and 25–38°C.

According to our results, in concentrations of 50 and 80 mg l⁻¹, increase in phosphorus adsorption in the temperature range of 25°C to 38°C was more compared to the temperature range of 12 to 25°C. At other phosphorus concentrations, more changes of phosphorus adsorption were observed in the temperature

Table 6

The constant coefficients of Langmuir and Freundlich equations at the three temperatures studied

Temperature (°C)	Langmuir				Freundlich			
	q_m	K_l	r^2	s	K_f	n	r^2	s
12	0.472	0.033	0.935**	0.035	0.046	0.462	0.93**	0.037
25	0.645	0.042	0.916**	0.058	0.073	0.448	0.932**	0.052
38	1.235	0.026	0.979**	0.046	0.074	0.567	0.974**	0.051

** p<0.01

range of 12 to 25°C. The change in temperature affects the phosphorus adsorption and desorption by influencing the ionic exchange strength at the solution-sediment interface (Liu et al., 2002). Increasing the temperature increases the bonding energy of phosphorus adsorption and precipitation in soil. High temperatures can also reduce the time required to form crystalline compounds (Reddy and DeLaune, 2008). The results of this study are consistent with the results of Zhang and Huang (2011), who reported that every 10°C increase in temperature resulted in an approximately 1.3 times increase in phosphate adsorption on marine sediments. In another experiment on different trophic lake sediments, an increase in phosphate adsorption was reported following an increase in temperature from 10°C to 35°C (Jin et al., 2005). Huang et al. (2016) found that phosphorus adsorption capacity in wetland sediment (pH 6.2 and organic matter 3.53%) increased rapidly between 5°C and 25°C and increased slowly between 25°C and 35°C.

The values of the absorption coefficients at three temperatures of 12°C, 25°C, and 38°C for both Langmuir and Freundlich isotherms were estimated and presented in Table 6. along with r^2 and s. As can be seen in Table 6, the values of r^2 in both isotherms for all three temperatures were high (> 0.9) and all were significant (p < 0.01). Comparing q_m values at different temperatures showed that with increasing temperature from 12 to 25°C and from 25°C to 38°C, the phosphorus maximum adsorption capacity increased 1.4 and 1.9 times, respectively. Therefore, phosphate fertilizer application when the soil temperature is lower should be taken into consideration to increase phosphorus bio-availability in soils and improve fertilizer recommendation and management.

3.5. Thermodynamic parameters of adsorption

The van't Hoff plot ($\ln K_d$ vs $1/T$) related to the adsorption of phosphorus onto Mehdi Abad (MA) soil for different phosphorus concentrations at three temperatures of 285 K (12°C), 298 K (25°C), and 311 K (38°C) is shown in Fig. 4. Thermodynamic parameters of phosphorus adsorption are also presented in Table 7. The negative values of ΔG° at different temperatures and concentrations indicated the spontaneous nature of phosphorus adsorption by the studied soil. As shown in Table 7., as the temperature increased the Gibbs free energy decreased, which means that the soil had more tendency to adsorb. In the same way, comparing the values of this parameter at different concentrations of phosphorus indicated a decrease in the tendency of phosphate adsorption by soil particles at higher concentrations of phosphorus. Moreover, the positive values of the enthalpy suggested that the adsorption process was endothermic and the positive values of the entropy indicated an increase in randomness of the system at the solid-solution interface during the adsorption process. In an experiment done by Doula and Dimirkou (1996) on phosphate adsorption by four types of soils including Alfisols, Entisols, Vertisols, and Inceptisols, the adsorption process was observed endothermic. Besides, the values of ΔG° were negative and more negative for small initially added phosphorus concentration, which indicated that the process can be spontaneous (Doula and Dimirkou, 1996). Despite the experienced endothermic nature of phosphate adsorption onto biochar and composite fiber (Karunanithi et al., 2017; Liu and Hu, 2019), in a study conducted by Yuan et al. (2015), the adsorption of phosphorus onto dolomite was reported to be exothermic.

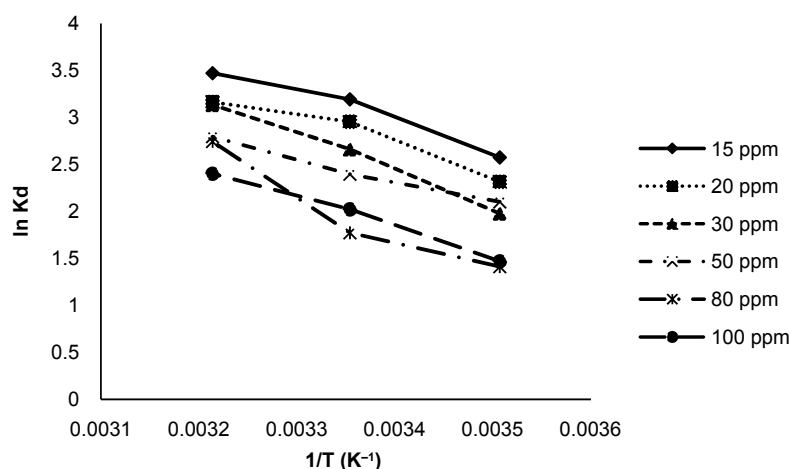


Fig. 4. Van't Hoff plot for phosphorus adsorption by the studied soil (MA)

Table 7

Thermodynamic parameters of phosphorus adsorption by the studied soil (MA)

Phosphorus concentration (mg l ⁻¹)	ΔH° (kJ mol ⁻¹)	ΔS° (J mol ⁻¹ K ⁻¹)	ΔG° (kJ mol ⁻¹)		
			285 K (12°C)	298 K (25°C)	311 K (38°C)
15	25.588	111.524	-6.196	-7.646	-9.096
20	24.252	104.806	-5.618	-6.98	-8.343
30	32.895	132.01	-4.728	-6.444	-8.16
50	19.589	85.983	-4.916	-6.034	-7.152
80	37.464	142.236	-3.073	-4.922	-6.771
100	26.483	105.264	-3.517	-4.886	-6.2541

4. Conclusions

Comparison of different sorption isotherms and kinetic models demonstrated that Langmuir and Kuo and Lotse models obtained the best estimate of the amount of phosphorus adsorbed in all the studied soils. The highest value of phosphorus maximum sorption capacity was observed in the soil sample with the high levels of calcium carbonate percentage and clay content, and the low amount of organic matter. Furthermore, an increase in pH and temperature, as well as a decrease in salinity, resulted in a significant increase in phosphorus adsorption onto the adsorbent sites. The results obtained from this study in relation to how the adsorption process is influenced by different soil parameters can be helpful and practical in providing appropriate strategies for fertilizer and soil management in the studied agricultural soils with high phosphorus adsorption in order to reduce the phosphorus accumulation in the soil profile and increase its availability for cultivated plants.

Based on the results of the present study, it is recommended to add elemental sulfur, aluminium sulfate, ammonium nitrate fertilizers, and organic compounds to these calcareous soils in order to reduce soil pH and maintain the phosphorus concentration in soil solution at an optimal level. Also, due to the high adsorption of phosphorus onto the soil particle surfaces in a short period of time, it is recommended that the time of application of phosphate fertilizers be near the time of plant's need for phosphorus and plant uptake.

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