

Contribution to the Study of Phosphorus Adsorption Kinetics on the Superficial Sediments of a Recently Development Artificial Estuary: Vridi Channel (Côte d'Ivoire)

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Authors' contributions

This work was carried out in collaboration among all authors. Author NDAS designed this study, performed the sampling and managed the analyses. He also wrote the first draft. Author YMK performed the statistical analysis, managed the analyses and contributed for the results discussion. Author TA contributed to the results discussion. All authors read and approved the final manuscript.

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ABSTRACT

The aim of this study is to characterize phosphates adsorption kinetics on the superficial sediments of Vridi channel from its new hydromorphology. So, three scenarios have been carried out taking account the different waters seasons of this estuary by using pH and temperature as relevant parameters. In the first scenario, the experiments carried out at pH = 8 and T = 20°C to simulate phosphates adsorption on these sediments in its great cold season. In the second, the experiments carried out at pH = 8 and T = 25° C to simulate phosphates adsorption on these substrates in its hot season and small cold season. In the third, the experiments carried out at pH = 7 and T = 30°C to simulate phosphates adsorption on these sediments in its flood season. The experiences were carried out according to US EPA/530/SW-87/006-F protocol. KH₂PO₄ solutions were used as phosphates synthetic solutions. The experimental data were modeling by Lagergeen kinetics model (pseudo-order 1), Blanchard kinetics model (pseudo-order 2), Elovich kinetics model, Weber and

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Morris kinetics model and, external diffusion kinetics model. The results have showed phosphates adsorption on these sediments increase in the experiments carried out at pH = 8 and T = 20°C to those carried out at pH = 7 and T = 30°C. All experimental data have been well modeling by Blanchard kinetics model. So, these phosphates adsorption kinetics are pseudo-order 2 and the chemisorption is the dominant mechanism. The results obtained by the experimental data modeling by Weber and Morris kinetics model and external diffusion kinetics model showed that this process is not limiting by the molecular diffusion process. The experimental data have been again well modeling by Elovich kinetics model. That has confirmed the chemisorption character of this adsorption, and showed this chemisorption is activated.

Keywords: Atlantic Ocean; Côte d'Ivoire; ébrié system; eutrophication; kinetics adsorption models; phosphorus; pollution; vridi channel.

1. INTRODUCTION

With regard to the devastating effects of eutrophication on the entire ecosystem, studies on phosphorus have occupied a special place for limnologists, hydrochemists and marine geologists. Indeed, this ecological scourge, destroying surface waters, visibly results in an anarchic proliferation of aquatic plants of all kinds in these aquatic ecosystems. It is mainly due to an excessive presence of nutrients, particularly those of nitrogen and phosphorus [1,2]. If nitrogen and phosphorus are considered as its limiting factors, the fact remains that phosphorus is its control factor because of its sedimentary biogeochemical cycle [3]. So, the knowledge of phosphorus adsorption in these aquatic ecosystems has always aroused much interest [4,5]. One of the approaches carried out in this kind of study has been the simulation of phosphorus adsorption on sediments under specific conditions at laboratory. Thus, models have always been used to characterize experimental kinetics in these studies, in order to obtain important information about this process relative to the experimental conditions used.

The modeling of adsorption kinetics in liquid-solid systems is based on the theory of Langmuir [6]. The kinetics adsorption models are grouped into three categories: the kinetics models based on binding energies; the kinetics models based on the composition of the system and; the kinetic models linked to the molecular diffusion process [7]. The kinetics models based on the composition of the system and those linked to the molecular diffusion process are the most used in the studies focusing on substance adsorption on substrates, such as the case of phosphorus on sediments. The models based on the composition of the system are based on the principle that the instantaneous sorption rate is determined by the difference between the state

of the system at that instant and its equilibrium state. These models are also called "driving force models" or "surface process models". Lagergren kinetics model [6] (the pseudo-order 1 kinetics model) and Blanchard kinetics model [8] (the pseudo-order 2 kinetics model or Lagergren kinetics model of pseudo-order 2)) form part of this category of models. As regards the kinetics models based on the molecular diffusion process, they are based on the fact that the adsorption rate is determined by the molecular diffusion process. They correspond to several models including Weber and Morris kinetics model [9] (the intra-particle diffusion kinetics model), the diffusion in the liquid film kinetics model or the external diffusion kinetics model [10-11] and Elovich kinetics model [12].

Vridi channel is the only route of communication between Atlantic Ocean and Ébrié system. It exhibited the superficial sediments with high relative phosphorus contents in its former hydromorphology [13]. As part of the extension and the growth in traffic from the autonomous harbour of Abidjan, this estuary has undergone development. This development led to a modification of its hydromorphological characteristics. The immediate corollary of which was the change of the geochemical characteristics of its superficial sediments. This has led to a decrease in the adsorbing power of phosphorus by these substrates, as illustrated by the very recent studies of N'Da et al. [13,14]. In addition, the studies carried out by Mahi et al. [15] showed the high influence of pH, salinity and phosphates concentration in this process. However, this study only focused on the loose sedimentary layer of this estuary. Thus, it is necessary to carry out studies to contribute more to the knowledge of phosphorus adsorption kinetics in the superficial sediments of this estuary, especially in its new configuration. It is

within this framework that this study was carried out.

2. MATERIALS AND METHODS

2.1 Presentation of the Study Area

Vridi channel is located in the south of Côte d'Ivoire, precisely at 4°0'50" west longitude at the north latitude of 5°15'23" (DMS) (5.257636 N; 4.011545 W in decimal degrees). It results from the breakthrough of the Jacquville dyke during the creation of the autonomous harbour of Abidjan in 1953. It is therefore an artificial marine estuary as mentioned by Yao et al. [15] and Yao and Trokourey [16,17,18]. As part of the increase in traffic from this harbour, this estuary underwent hydromorphological development from 2016 to 2018. This development focused on its deepening along its entire length and the enlargement of its western jetty. This fact has led to a modification of the biogeochemical characteristics of its surface sediments. This is particularly the case of the nature and particle size distribution of these substrates, which previously was dominated by coarse sands in its former hydromorphology, currently presents a texture predominantly of clays and silts in its new hydromorphology. This is also the case for their phosphorus contents, which experienced a real drop with this development. On the other hand, this development has very little influence their pH [14,19].

Due to its position, Vridi channel has an impressive hydrological network. This network is made up of Ébrié system and Atlantic Ocean on the one hand, and fluvial water supplies, the most important of which are Comoé river, Mé river and Agnéby river, on the other. Its water seasons are characterized by five seasons: a Hot Season (HS) from February to April; a Rainy Season (RS) from May to July; a Great Cold Season (GCS) from August to September; a Flood Season (FS) from October to November and, a Small Cold Season (SCS) from November to December [18].

2.2 Experimental Techniques

2.2.1 Principe of this study

The study of phosphorus adsorption kinetics was carried out using the protocol established by US EPA/530/SW-87/006-F [20]. The batch mode was used as the dynamic mode in this study. The various phosphates synthetic solutions, obtained

from the dissolution of KH_2PO_4 in demineralized water, were saturated with NaCl solution to approximate the character of the marine waters of Vridi channel. The samples of the superficial sediments Vridi channel used in this study were composed on mean of $(10.23 \pm 3.07)\%$ of rudites, $(24.77 \pm 6.86)\%$ of sands and, of $(70.00 \pm 5.84)\%$ of clays and silts. So, the influence of particle size distribution of these samples on phosphates adsorption in this study was neglected in this study, due to their high dominance by silts and clays. This study was carried out taking into account the water seasons of this estuary. pH and temperature, being among essential characteristics of these waters, were considered as relevant parameters in this study. So, three study scenarios were developed:

- 1- The experiments carried out with phosphates synthetic solutions of pH = 8 at $T = 20^\circ\text{C}$, to simulate phosphates adsorption on these superficial sediments in GCS;
- 2- The experiments carried out with phosphates synthetic solutions of pH = 8 at $T = 25^\circ\text{C}$, to simulate phosphates adsorption on these superficial sediments in HS, RS and SCS;
- 3- The experiments carried out with phosphates synthetic solutions of pH = 7 at $T = 30^\circ\text{C}$, to simulate phosphates adsorption on these surface sediments in FS.

All experiments were performed in triplicate and the reagents used were of analytical purity.

2.2.2 Experiments

The experiments were carried out taking into account the prescription of Coulibaly [21], which recommends a solid/liquid ratio of 1/150 for optimal adsorption in such a study. Thus, 1 g of dry sediment sample, previously reduced to diameters $\varnothing \leq 0.2\text{ mm}$, were brought into contact with 150 ml of the phosphates synthetic solutions at the initial concentrations (C_0) of 1.5; 2; 3 and 5 mg/l. These solutions were saturated with NaCl by adding 4 ml of a solution of 0.01M NaCl. The microbial activity was inhibited by adding 2 drops of a pure commercial toluene solution. According to the experiment carried out, pH of the solution was adjusted with a solution of 0.01M NaOH or a solution of 0.01M H_2SO_4 and, brought to the desired temperature in a thermostatted bath. In order to simulate the water strong

hydrodynamics of this estuary, the tests were carried out with horizontal agitation at 400 rpm. At predefined time intervals, 5 ml of the phosphates synthetic solution was first taken, then filtered through Whattman type filter paper at 0.47 µm in diameter, finally assayed according to AFNOR NF T90-023 standard [22]. So, the different assays were performed by a spectrophotometry method using the JENWAY 7315 brand spectrophotometer.

The amount of phosphates instantly adsorbed on these substrates ($q_{ads}(t)$), expressed in mg/g, is given by:

$$q_{ads}(t) = (C_0 - C_t) \frac{V}{m} \quad (I - 1)$$

With: C_0 and C_t , the initial liquid-phase phosphates concentration and the instantly liquid-phase phosphates concentration (mg/l) respectively; V , the volume of the phosphates synthetic solution (ml); m , the mass of the dried sediment sample (g).

2.2.3 Experimental adsorption kinetics modeling

The different models used to characterize the experimental adsorption kinetics are given in Table 1.

In this study, a model is considered adequate for the description of the experimental data if the following criteria are simultaneously satisfied:

- the function plotted to according the model must be a is straight line;
- The determination coefficient (R^2) must be significant ($R^2 \geq 0.900$), i.e the model must Translate more than 90% of the experimental data;
- The Root Mean Square Error (RMSE) must be relatively very low. It is defined by:

$$RMCE = \sqrt{\frac{\sum_1^N (q(t)_{cal} - q(t)_{exp})^2}{N}} \quad (I - 2)$$

- The mean prevision error (e_m) must be very low. It is defined by:

$$e_m = \frac{\sum_1^N |q(t)_{cal} - q(t)_{exp}|}{N} \quad (I - 3)$$

- The absolute deviation error (Δ (%)) must be relatively very low. It is given by:

$$\Delta(\%) = \frac{100}{N} \times \sum_1^N \left| \frac{q(t)_{cal} - q(t)_{exp}}{q(t)_{exp}} \right| \quad (I - 4)$$

With: $q(t)_{cal}$, the calculated value of $q(t)$ obtained from the model; $q(t)_{exp}$, the experimental value of $q(t)$; N , number of observations.

In the particular case of the models for determining the order of these experimental kinetics (Lagergren kinetics model [6] and Blanchard kinetics model [8]), the following condition is appended, namely $q_{m,cal}$ must very close to $q_{m,cal}$.

3. RESULTS AND DISCUSSION

3.1 Results

3.1.1 Hour dynamic of phosphates adsorption on these sediments

The results showed that the hourly dynamic of phosphates adsorption on these superficial sediments presents the same profiles under the three experimental conditions used. In most cases, it took place in three stages. The first stage was characterized by a very rapid phosphates adsorption on these substrates in the first two hours, especially the first. As for the second stage, it was marked by a slowing down of this adsorption followed by the third stage which was identified by a pseudo-equilibrium state of this phenomenon (Fig. 1).

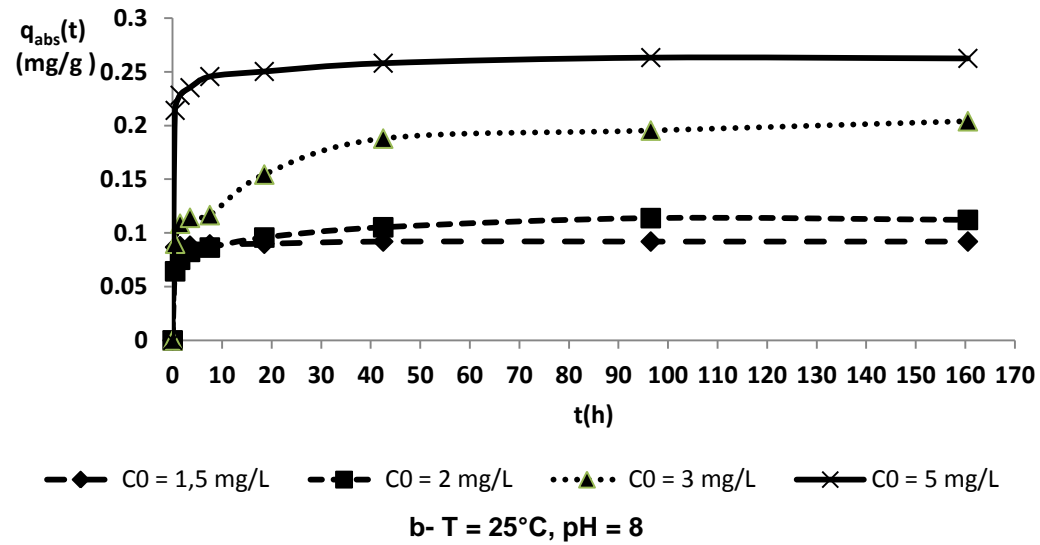
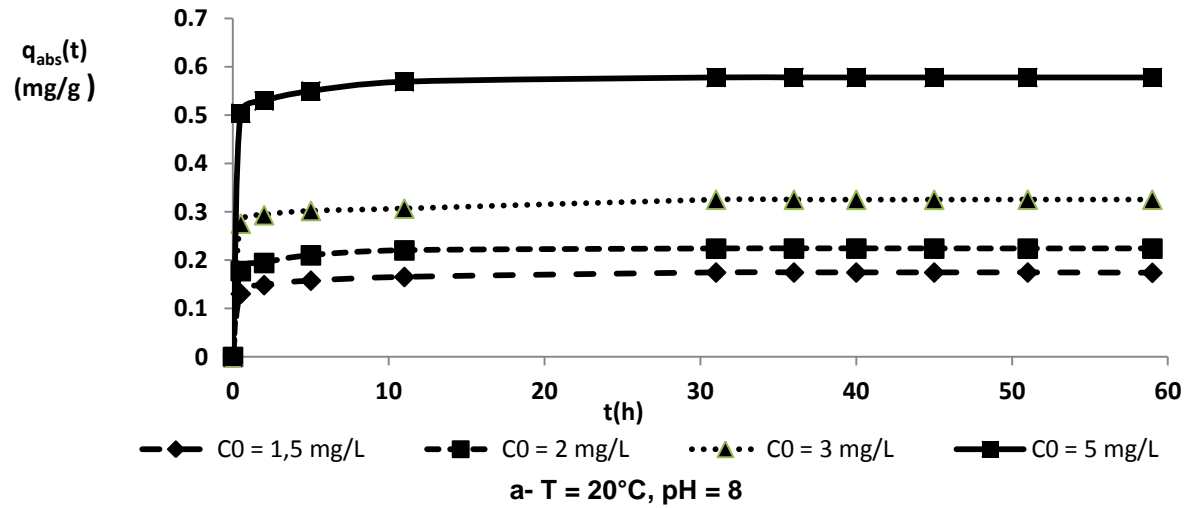
The equilibrium time (t_e) for all the experiments carried out at for $T = 20^\circ\text{C}$ and $\text{pH} = 8$ was observed at 31 h. This was also the case for those carried out at $T = 30^\circ\text{C}$ and $\text{pH} = 7$. As for the experiments carried out at $T = 25^\circ\text{C}$ and $\text{pH} = 8$, the equilibrium time (t_e) was observed at 42.5 h. $q_{abs}(t)$ decreases from the experiments carried out at $T = 20^\circ\text{C}$ and $\text{pH} = 8$ to those carried out at $T = 30^\circ\text{C}$ and $\text{pH} = 7$. These t_e values obtained in this study are very important than those determined by Mahi et al. [15] with the loose sedimentary layer of this estuary in its former hydromorphology in practically the same experimental conditions.

In each of experiments series, $q_{abs}(t)$ increases with the initial concentration of phosphates synthetic solutions (C_0). $q_{max,exp}$ has been of 0.578 mg/g at $T = 20^\circ\text{C}$ and $\text{pH} = 8$; of 0.258 mg/g at $T = 25^\circ\text{C}$ and $\text{pH} = 8$; and of 0.184 mg/g at $T = 30^\circ\text{C}$ and $\text{pH} = 7$. This suggested that phosphates adsorption on these sediments would decrease a priori with increasing temperature and to a certain extent with pH. Also, these $q_{max,exp}$ values obtained in this study are less important than those determined by Mahi et al. [15].

Table 1. Characteristic of the kinetics models used in this study

| | Kinetics model | Kinetics equation | Characteristic parameters | function to plot |
|---|--|--|----------------------------------|---|
| Surface process models | Lagergren kinetics model [6] (Pseudo-order 1) | $\log(q_m - q(t)) = \log(q_m) - \frac{k_1}{2,303}t$ | k_1, q_m | $\text{Log}(q_m - q(t)) = f(t)$ |
| | Blanchard kinetics model [8] (Pseudo-order 2) | $\frac{t}{q(t)} = \frac{1}{k_2 q_m^2} + \frac{1}{q_m}t$ | k_2, q_m | $\frac{t}{q(t)} = f(t)$ |
| Models of the molecular diffusion process | Elovich kinetics model [12] | $q(t) = \frac{1}{\beta} \ln(t) + \frac{1}{\beta} \ln(\alpha\beta)$ | $\alpha; \beta$ | $q(t) = \ln(t)$ |
| | Webber et Morris kinetics model [9] (the diffusion intra particle model) | $q(t) = k_d t^{\frac{1}{2}} + C$ | $K_d; C$ | $q(t) = f(t^{\frac{1}{2}})$ |
| | The external diffusion kinetics model [10-11] | $\ln\left(1 - \frac{q(t)}{q_m}\right) = \ln(1 - F) = -k_{fd} t$ | $K_{fd};$ | $\ln\left(1 - \frac{q(t)}{q_m}\right) = f(t)$ |

With: $q(t)$, the amount of adsorbate instantly adsorbed ; q_m , the maximum adsorbent uptake by adsorbant (mg/g); k_1 , the pseudo-order 1 kinetics constant (t^{-1}); k_2 , the pseudo-order 2 kinetics constant ($g/(mg.t)$); α , the adsorption initial rate (mg.t/g); β , the constant related to the external surface and to the chemisorption activation energy; k_d , the intra-particle diffusion constant ($mg/(gt^{1/2})$); C , the thickness of the boundary layer of diffusion constant (mg/g); k_{fd} , the external diffusion constant (m^{-1})



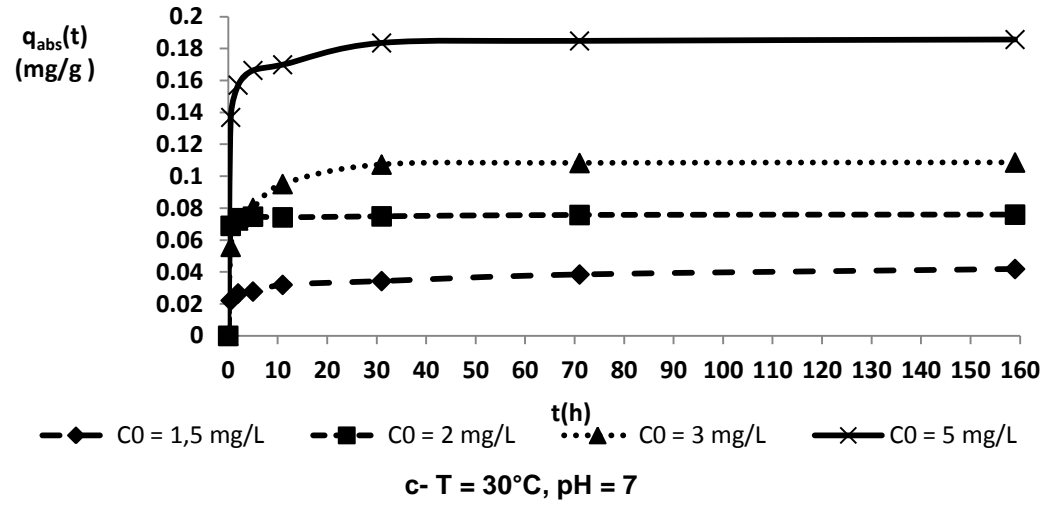


Fig. 1. Hourly dynamic of phosphates adsorption on the superficial sediments of Vridi channel from its recent hydromorphology in experimental conditions used

3.1.2 Adsorption kinetics modeling

3.1.2.1 Driving force models

The analysis of Table 2 shows that Lagergren kinetics model [6] is inadequate for the modeling of the experimental data obtained (Fig. 2). Indeed, the values of determination coefficient obtained by this model with these experimental data are relatively very low in the most case. Also, the different values of $q_{\max,cal}$ are distinct to those of $q_{\max,exp}$ corresponding in each experiment series. Again, the very high values of e_m , Δ and RMCE are very important. So, this model is not validated in this study according to the validation criteria mentioned above.

Regarding Blanchard kinetics model [8], it gives a very good representation of these experimental data obtained (Fig. 3). This was shown by the high values of the determination coefficient obtained by this model with these experimental data. Also, the different values of $q_{\max,exp}$ are identical to those of $q_{\max,cal}$ corresponding in each experiment series. This observation is reinforced by the very low values of e_m , RMCE and Δ . This model is validated according to the validation criteria mentioned above. As a result, phosphates adsorption kinetics on these substrates under the established experimental conditions is of pseudo-order 2.

The results obtained with Blanchard kinetics model [8] also show that the initial adsorption rates of phosphates (h) by these substrates are relatively greater in the experiments carried out at $T = 30^\circ\text{C}$ and $\text{pH} = 7$ compared to those carried out at $T = 25^\circ\text{C}$ and $\text{pH} = 8$, which in turn are more significant than those obtained in the experiments carried out at $T = 20^\circ\text{C}$ and $\text{pH} = 8$. Also, the pseudo-order 2 kinetics constant (k_2) is globally important when h is low.

3.1.2.2 Models of molecular diffusion process

The Table 3 presents the results of the modeling of the experimental data by Weber and Morris kinetics model [9], the external diffusion kinetics model [10-11] and Elovich kinetics model [12].

Although presenting low values of e_m , Δ and RMCE in the mainly cases, all the values of the determination coefficient obtained by Weber and Morris kinetics model [9] with the experimental data are very low (Fig. 4). Therefore, this model is not validated according to the validation criteria used in this study. Thus, the intra-particle

diffusion is not the only mechanism limiting phosphates adsorption kinetics on these sediments in the experimental conditions used. The thickness of the diffusion boundary layer (C) increase and, the intra-particle diffusion constant (K_d) decrease respectively with the increase in the initial concentration of phosphates synthetic solutions (C_0) in the experiments carried out at $T = 20^\circ\text{C}$ and $\text{pH} = 8$, and at $T = 30^\circ\text{C}$ and $\text{pH} = 7$. These two kinetics parameters of Weber and Morris kinetics model [9] also increase with the initial concentration of the phosphates synthetic solutions (C_0) in the experiments carried out at $T = 25^\circ\text{C}$, $\text{pH} = 8$; exception for C obtained for $C_0 = 2 \text{ mg/l}$ and, for K_d obtained for $C_0 = 5 \text{ mg/l}$, which are weak.

All the values of the determination coefficient determined by the inter-particle diffusion kinetics model [10-11] with the experimental data are also very low (Fig. 5). The different values of e_m , Δ and RCME are relatively very important. So, this model is not validated in this study according to the validation criteria used. Consequently, the process of phosphates adsorption on these sediments is not controlled by the diffusion in the liquid film under the experimental conditions used.

Elovich kinetic model [12] makes it possible to obtain very high values of the determination coefficient with the experimental values in all experiment series, with the exception of the experiments carried out at $T = 30^\circ\text{C}$ and $\text{pH} = 7$ for $C_0 = 2 \text{ mg/l}$ (Fig. 6). Also, all the values of e_m , Δ and RMCE are very low for these experiments. This shows the adequacy of Elovich kinetic [12] model to translate the experimental data of this study. So, this model is validated according the validation criteria used in this study.

3.2 Discussion

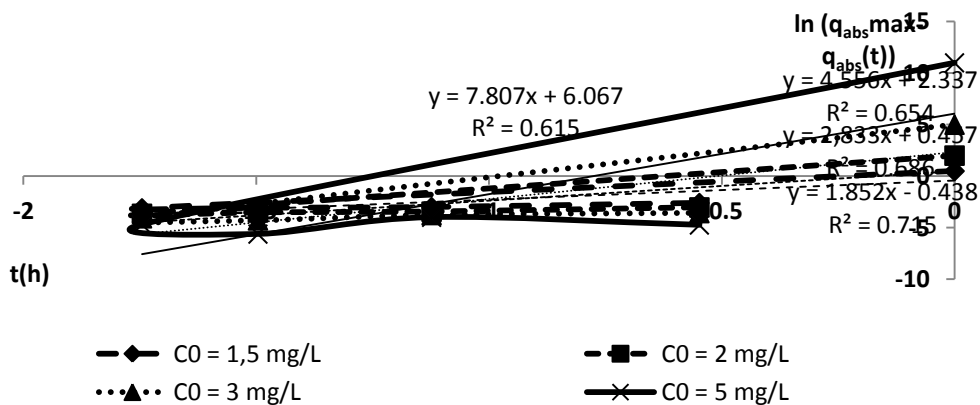
The hourly dynamic profiles of phosphates adsorption on the superficial sediments of Vridi channel used in this study are similar to those obtained in many similar studies in general, and those very recent of Cao et al. [23-24], Huang et al. [25], Omari et al. [26], Topçu et al. [27] and Mahi et al. [15], in particular.

The different values of t_e (equilibrium time) obtained in this study are in accordance with the observations of Bai et al. [28], Hou et al. [29], Wang et al. [30] and Zhang et al. [31]. These authors noted that in studies of phosphorus retention on substrates on batch mode, t_e is

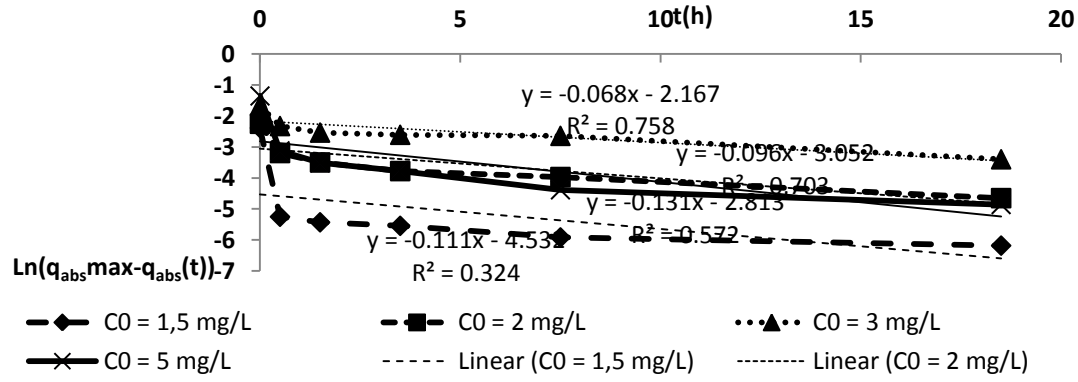
Table 2. Kinetics parameters for Lagergren kinetics model [6] and Blanchard kinetics model [8] obtained with the experimental data in this study

| Lagergren kinetics model [6] | | | | | | | | |
|------------------------------|--------------------------|--------------------------------|--------------------------------|--------------------------------------|----------------|------------------------|------------------------|------------------------|
| T (°C) et pH | C ₀ (mg/l) | q _{max,exp} (mg/g) | q _{max,cal} (mg/g) | K ₁ (h ⁻¹) | R ² | e _m | Δ(%) | RMCE |
| T = 20°C; pH = 8 | 1.5 | 0.175 | 0.645 | -1.852 | 0.715 | 1.67 10 ⁴⁶ | 1.05 10 ⁴⁹ | 5.54 10 ⁴⁶ |
| | 2 | 0.224 | 1.579 | -2.833 | 0.686 | 5.60 10 ⁷¹ | 2.75 10 ⁷⁴ | 1.86 10 ⁷² |
| | 3 | 0.325 | 10.350 | -4.556 | 0.654 | 5.17 10 ¹¹⁶ | 2.54 10 ¹¹⁹ | 1.72 10 ¹¹⁷ |
| | 5 | 0.578 | 41.385 | -7.807 | 0.615 | 4.32 10 ²⁰¹ | 2.12 10 ²⁰⁴ | 6.38 10 ¹³⁷ |
| T = 25°C; pH = 8 | 1.5 | 0.092 | 0.011 | 0.111 | 0.324 | 0.052 | 66.683 | 0.064 |
| | 2 | 0.105 | 0.047 | 0.096 | 0.703 | 0.010 | 17.262 | 0.046 |
| | 3 | 0.188 | 0.115 | 0.068 | 0.758 | 0.048 | 5.253 | 0.058 |
| | 5 | 0.258 | 0.060 | 0.131 | 0.572 | 0.021 | 1.029 | 0.030 |
| T = 30°C; pH = 8 | 1.5 | 0.034 | 1.149 | -1.593 | 0.803 | 1.44 10 ¹⁰⁹ | 3.93 10 ¹¹² | 4.07 10 ¹⁰⁹ |
| | 2 | 0.075 | 3.452 | -2.303 | 0.796 | 4.61 10 ¹⁵⁸ | 6.93 10 ¹⁶¹ | 1.26 10 ⁷¹ |
| | 3 | 0.107 | 34.709 | -3.429 | 0.758 | 2.63 10 ²³⁷ | 2.76 10 ²⁴⁰ | 7.09 10 ¹⁰⁶ |
| | 5 | 0.184 | 49.130 | -5.423 | 0.834 | 1.17 10 ¹⁷⁰ | 6.34 10 ¹⁷² | 2.08 10 ⁷⁶ |
| Blanchard kinetics model [8] | | | | | | | | |
| T (°C) et pH | C ₀ (mg/l) | q _{max,exp} (mg/g) | q _{max,cal} (mg/g) | K ₂ (g/(mg.h)) | R ² | e _m | Δ(%) | RMCE |
| T = 20°C; pH = 8 | 1.5 | 0.175 | 0.176 | 14.570 | 0.999 | 0.002 | 2.002 | 0.010 |
| | 2 | 0.224 | 0.225 | 18.195 | 1.000 | 0.001 | 0.958 | 0.008 |
| | 3 | 0.325 | 0.327 | 9.374 | 0.999 | 0.007 | 2.700 | 0.024 |
| | 5 | 0.578 | 0.578 | 10.180 | 1.000 | 0.007 | 1.458 | 0.022 |
| T = 25°C; pH = 8 | 1.5 | 0.092 | 0.092 | 66.879 | 1.000 | 0.002 | 2.227 | 0.005 |
| | 2 | 0.105 | 0.114 | 5.117 | 0.999 | 0.005 | 7.922 | 0.014 |
| | 3 | 0.188 | 0.206 | 1.368 | 0.998 | 0.007 | 9.034 | 0.026 |
| | 5 | 0.258 | 0.264 | 8.033 | 1.000 | 0.008 | 4.066 | 0.025 |
| T = 30°C; pH = 8 | 1.5 | 0.034 | 0.042 | 7.393 | 0.997 | 0.002 | 1.489 | 0.006 |
| | 2 | 0.075 | 0.076 | 71.812 | 1.000 | 0.025 | 3.397 | 0.029 |
| | 3 | 0.107 | 0.109 | 8.341 | 0.999 | 0.001 | 3.668 | 0.007 |
| | 5 | 0.184 | 0.186 | 10.082 | 1.000 | 0.012 | 8.374 | 0.011 |

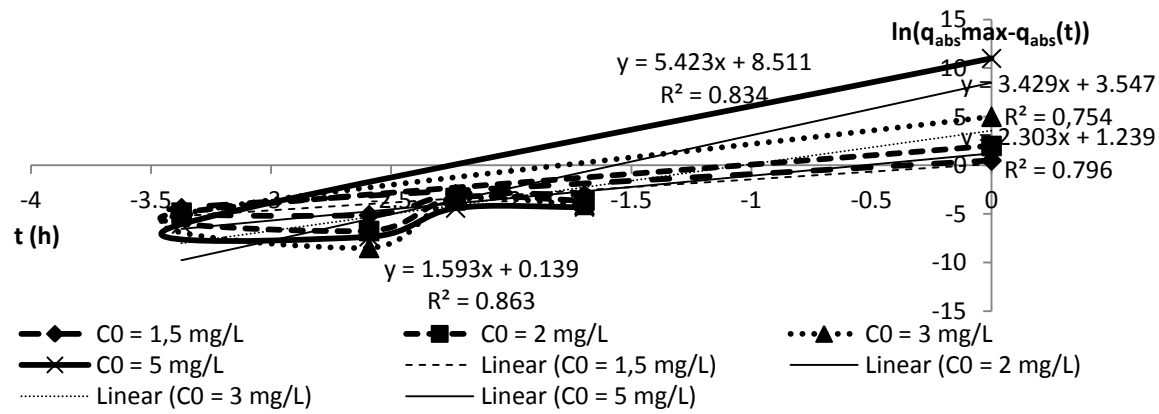
With: q_{m,exp}, the experimental maximum adsorbent uptake by adsorbant (mg/g); q_{m,cal}, the maximum adsorbent uptake by adsorbant (mg/g) obtained from model; C₀, the initial concentration of the phosphates synthetic solutions; k₁, the pseudo-order 1 kinetics constant ; k₂, the pseudo-order 2 kinetics constant; R², the determination coefficient; RMSE, the Root Mean Square Error; e_m, the mean prevision error; Δ, the absolute deviation error



a- T = 20°C, pH = 8

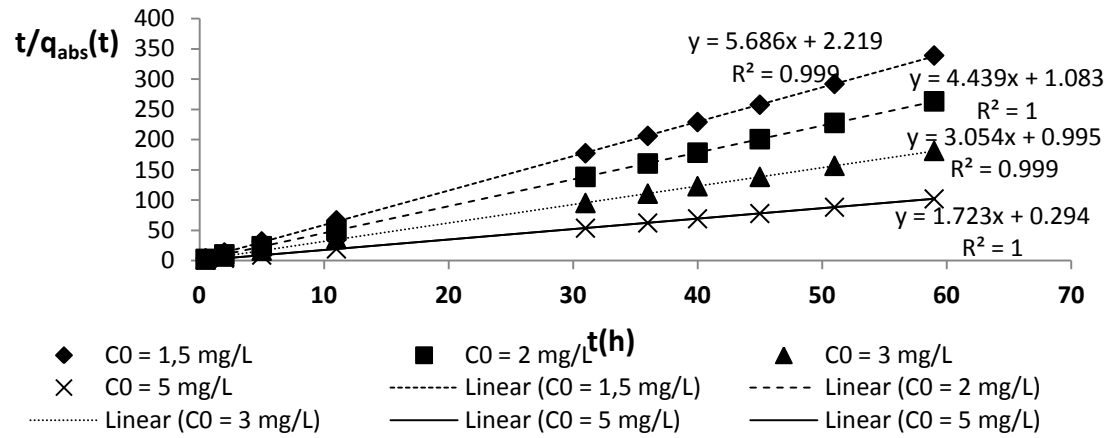


b- $T = 25^\circ\text{C}$, $\text{pH} = 8$

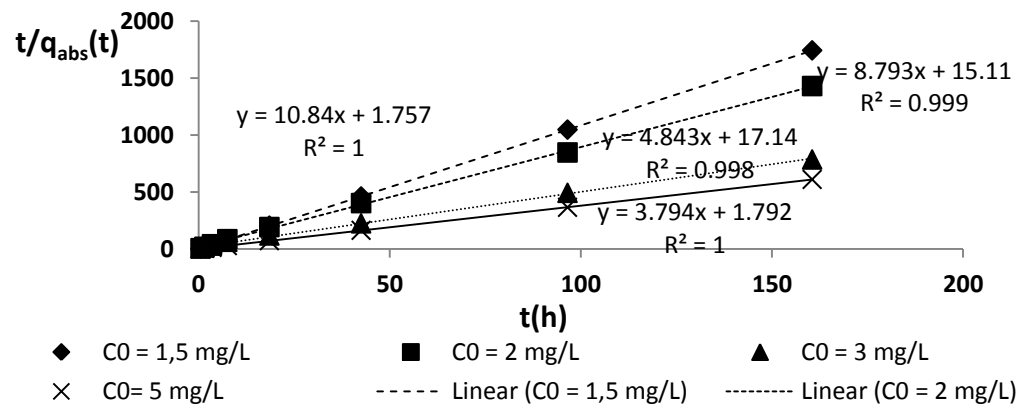


c- $T = 30^\circ\text{C}$, $\text{pH} = 7$

Fig. 2. Modeling of the experimental data by Lagergren kinetics model [9]



a- T = 20°C, pH = 8



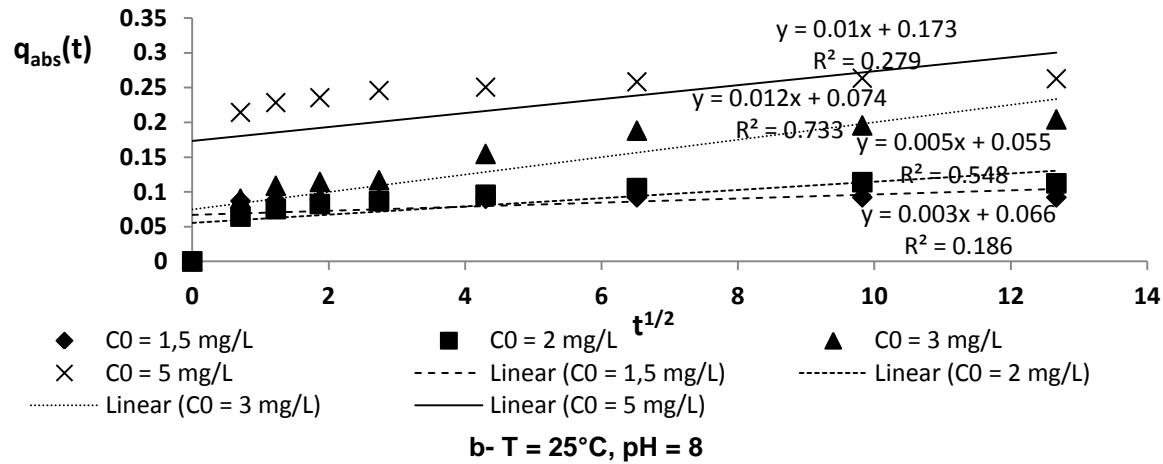
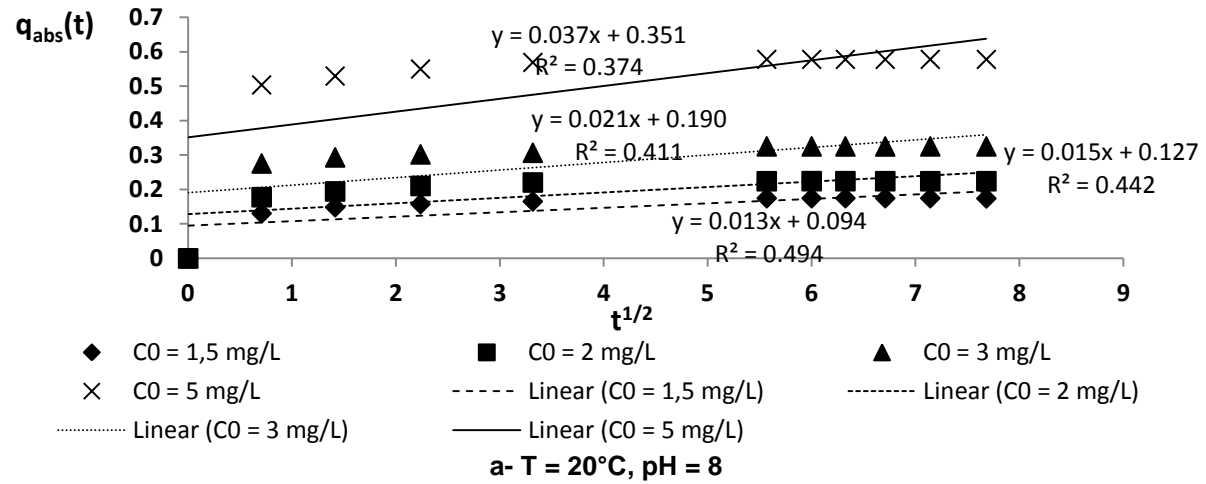
b- T = 25°C, pH = 8

Fig. 3. Modeling of the experimental data by Blanchard kinetics model [8]

Table 3. Kinetics parameters for Weber and Morris kinetics model [9], the external diffusion kinetics model [10-11] and Elovich kinetics model [12] with the experimental data in this study

| Weber and Morris kinetics model [9] | | | | | | | |
|--|---------------------------------|----------------------|---|----------------------|----------------------|------------------|-------------|
| | C₀ (mg/L) | R² | K_d (mg/(g.h^{1/2})) | C | e_m | Δ (%) | RMCE |
| T = 20°C, pH = 8 | 1.5 | 0.374 | 0.013 | 0.094 | 0.0003 | 6.742 | 0.021 |
| | 2 | 0.411 | 0.015 | 0.127 | 0.004 | 8.575 | 0.028 |
| | 3 | 0.442 | 0.021 | 0.190 | 0.004 | 8.240 | 0.041 |
| | 5 | 0.495 | 0.037 | 0.351 | 0.002 | 7.023 | 0.074 |
| T = 25°C, pH = 8 | 1.5 | 0.186 | 0.003 | 0.066 | 0.0005 | 10.074 | 0.013 |
| | 2 | 0.548 | 0.005 | 0.055 | 0.005 | 13.776 | 0.014 |
| | 3 | 0.733 | 0.012 | 0.074 | 0.003 | 9.936 | 0.020 |
| | 5 | 0.279 | 0.010 | 0.173 | 0.0003 | 9.520 | 0.033 |
| T = 30°C, pH = 7 | 1.5 | 0.610 | 0.008 | 0.112 | 0.104 | 75.620 | 0.121 |
| | 2 | 0.201 | 0.006 | 0.051 | 0.006 | 8.013 | 0.028 |
| | 3 | 0.536 | 0.002 | 0.033 | 0.041 | 51.376 | 0.047 |
| | 5 | 0.338 | 0.002 | 0.017 | 0.125 | 84.394 | 0.134 |
| The external diffusion kinetics model [10-11] | | | | | | | |
| | C₀ (mg/L) | R² | K_{fd} (h⁻¹) | e_m | Δ (%) | RMCE | |
| T = 20°C, pH = 8 | 1.5 | 0.697 | 0.205 | 0.012 | 4.956 | 0.055 | |
| | 2 | 0.841 | 0.308 | 0.071 | 38.215 | 0.085 | |
| | 3 | 0.481 | 0.175 | 0.054 | 20.459 | 0.102 | |
| | 5 | 0.726 | 0.289 | 0.075 | 15.898 | 0.157 | |
| T = 25°C, pH = 8 | 1.5 | 0.324 | 0.111 | 0.029 | 37.255 | 0.043 | |
| | 2 | 0.703 | 0.096 | 0.002 | 2.045 | 0.014 | |
| | 3 | 0.758 | 0.068 | 0.048 | 35.228 | 0.057 | |
| | 5 | 0.572 | 0.131 | 0.136 | 62.729 | 0.144 | |
| T = 30°C, pH = 7 | 1.5 | 0.808 | 0.193 | 0.007 | 28.919 | 0.01 | |
| | 2 | 0.432 | 0.332 | 0.014 | 22.455 | 0.025 | |
| | 3 | 0.854 | 0.163 | 0.015 | 25.376 | 0.024 | |
| | 5 | 0.588 | 0.175 | 0.038 | 28.560 | 0.060 | |
| Elovich kinetics model [12] | | | | | | | |
| | C₀ (mg/L) | R² | α (mg/(g.h)) | β (g/mg) | e_m | Δ (%) | RMCE |
| T = 20°C, pH = 8 | 1.5 | 0.971 | 5.128 10 ⁴ | 111.111 | 0.001 | 0.588 | 0.003 |
| | 2 | 0.934 | 1.187 10 ⁷ | 111.111 | 0.002 | 0.777 | 0.004 |
| | 3 | 0.983 | 2.158 10 ¹⁰ | 100.000 | 0.002 | 0.710 | 0.003 |
| | 5 | 0.958 | 1.705 10 ¹³ | 66.667 | 0.002 | 0.391 | 0.006 |
| T = 25°C, pH = 8 | 1.5 | 0.956 | 6.076 10 ³⁴ | 1000 | 0.0004 | 0.415 | 0.0006 |
| | 2 | 0.988 | 50.486 | 125 | 0.003 | 2.637 | 0.004 |
| | 3 | 0.941 | 1.846 | 47.619 | 0.002 | 0.914 | 0.01 |
| | 5 | 0.969 | 1.157 10 ¹⁰ | 125 | 0.001 | 0.559 | 0.003 |
| T = 30°C, pH = 7 | 1.5 | 0.982 | 6.408 | 333.333 | 0.002 | 4.967 | 0.002 |
| | 2 | 0.746 | 6.838 10 ²⁷ | 1000 | 0.0006 | 0.827 | 0.001 |
| | 3 | 0.940 | 12.326 | 111.111 | 0.003 | 2.772 | 2.772 |
| | 5 | 0.933 | 8.660 10 ⁵ | 125 | 0.002 | 1.084 | 1.084 |

With: $q_{m,exp}$, the experimental maximum adsorbent uptake by adsorbant (mg/g); $q_{m,cal}$, the maximum adsorbent uptake by adsorbant (mg/g) obtained from model; C_0 , the initial concentration of phosphates synthetic solutions; α , the adsorption initial rate; β , the constant related to the external surface and to the chemisorption activation energy; k_d , the intra-particle diffusion constant; C , the thickness of the boundary layer of diffusion constant; k_{fd} , the external diffusion constant; RMSE, the Root Mean Square Error; e_m , the mean prevision error; Δ , the absolute deviation error



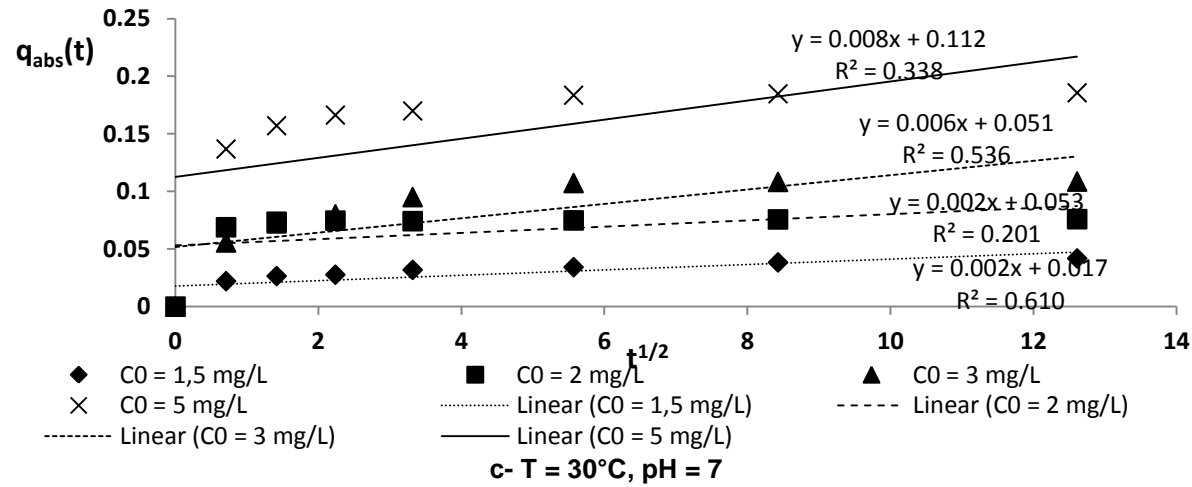
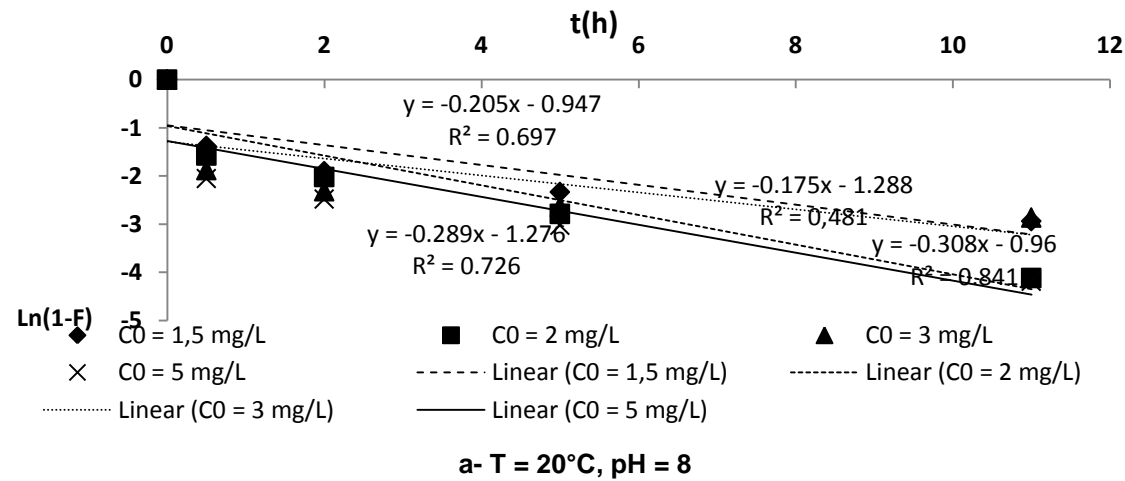


Fig. 4. Modeling of the experimental data by Weber and Moris kinetics model [9]



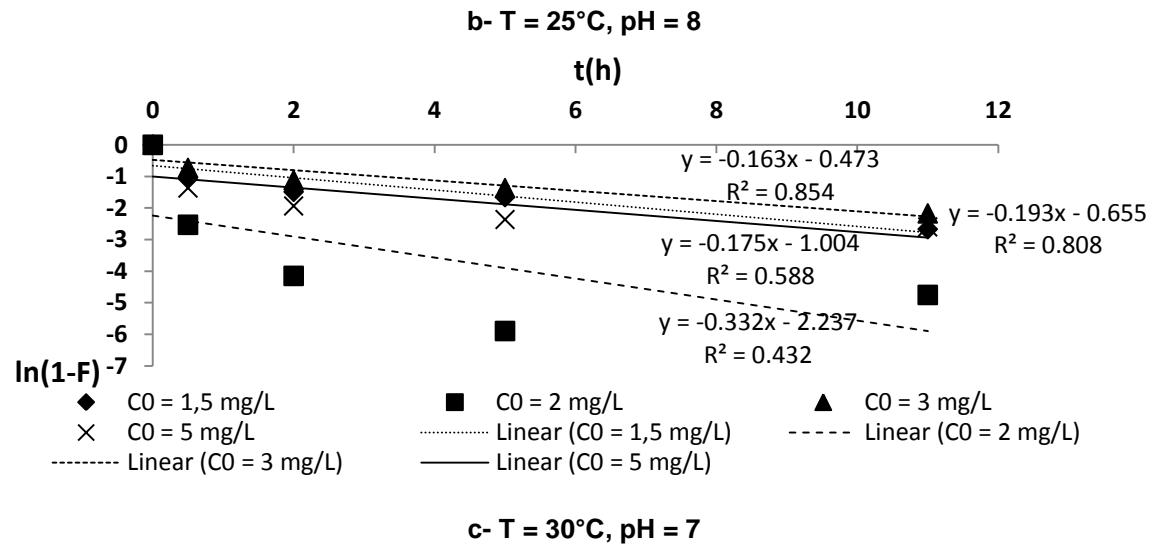
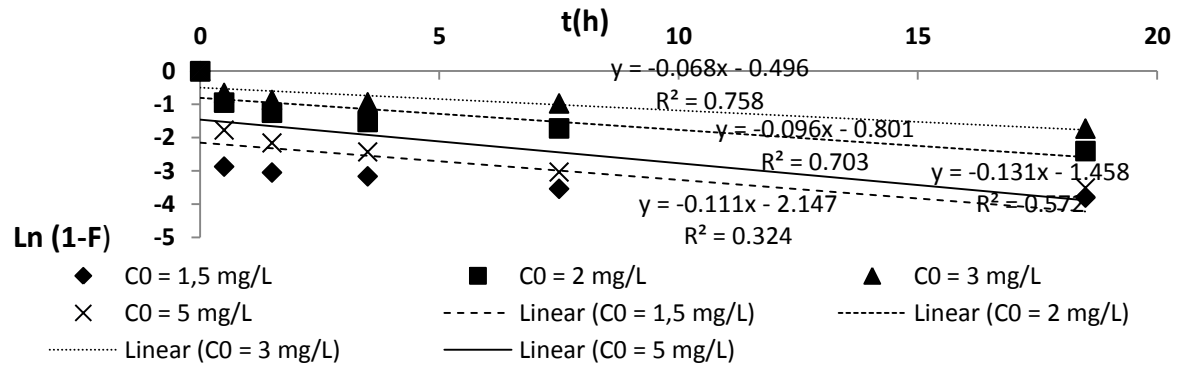
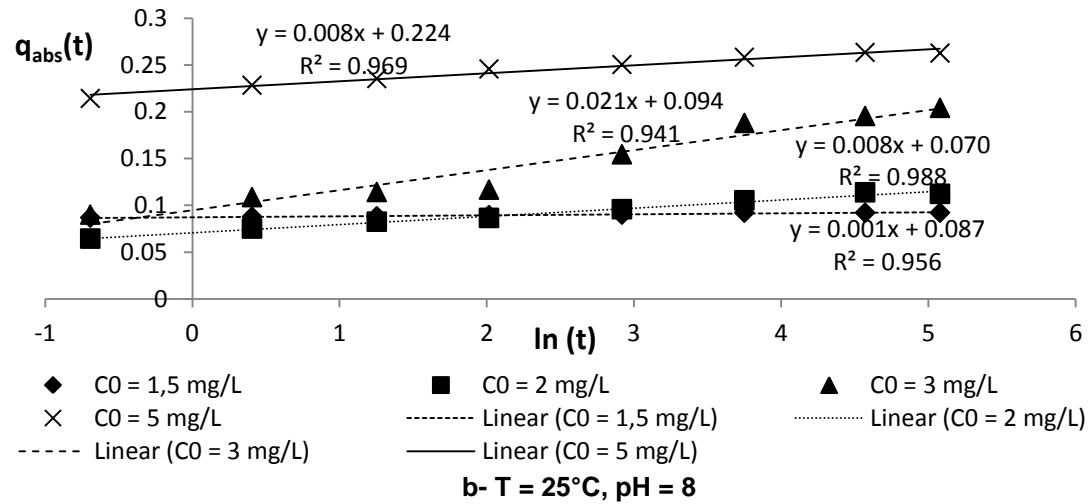
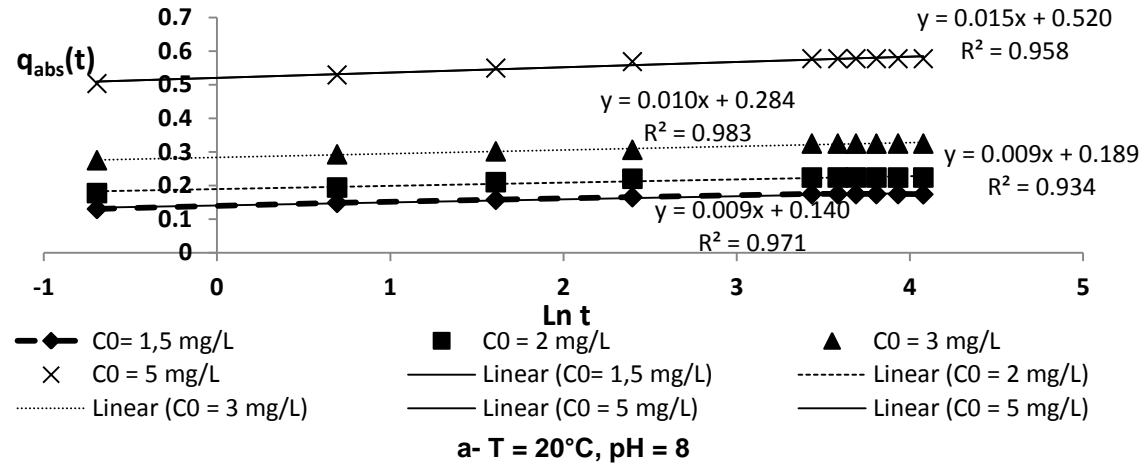


Fig. 5. Modeling of the experimental data by the external diffusion kinetics model [10-11]



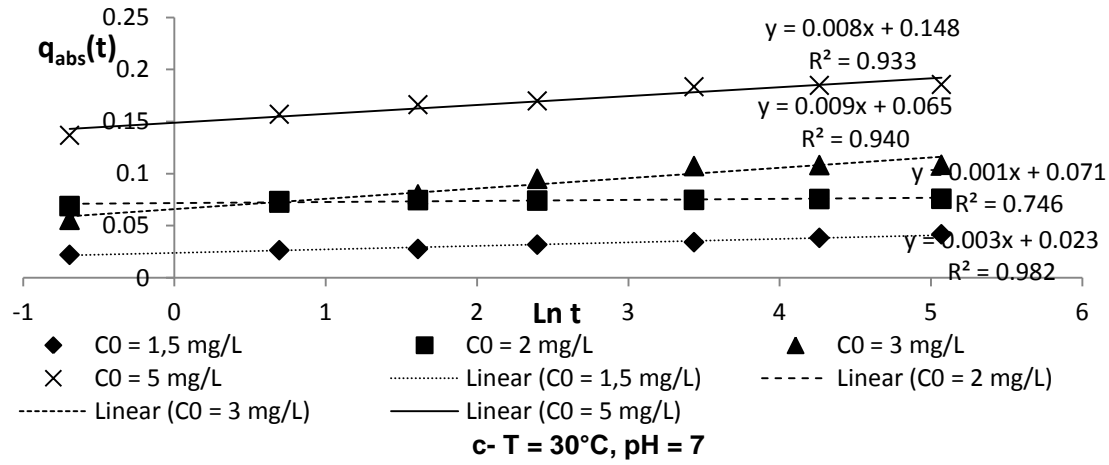


Fig. 6. Modeling of the experimental data by Elovich kinetics model [12]

generally achieved between 4 and 48 hours. These different t_e would show you that for the same basic pH, a drop in temperature would accelerate phosphates adsorption on these superficial sediments. This would be the opposite effect at neutral pH, where the increase in temperature accelerated this phenomenon. In each of these experiments, the growth of $q_{abs}(t)$ with that of the initial concentration of the phosphates synthetic solutions (C_0) would be explained by the favoring of collisions between phosphates and the active sites of these sediments, due to the increase in phosphates concentration in solutions [21]. This would confirm the influence of phosphate concentrations on their retention on the substrates [29,32- 33].

The decrease in $q_{abs}(t)$ with increasing temperature would tend to go up as physisorption would be the predominant mechanism in these experiments [34-35]. However, the geochemical composition of the samples and the specificity of each experiment series in this study don't a priori allow a conclusion to be established from this observation. Moreover, Karaca et al. [36] noted that an increase in temperature would have resulted in a strong collision between phosphates, thereby reducing the interactions between these molecular ions and their driving force in solution towards the surface of sediments, as well as the intensity of the impact with the active sites. This could explain in our case the decrease in phosphates adsorption rate on these sediments with the increase in temperature. Also, the increase in temperature would have had the effect of generally reducing phosphates adsorption on these substrates by inducing a displacement of predominance domains of the species (Fe^{3+} , $FeOH^{2+}$, $Fe(OH)_2^{2+}$, $Fe(OH)_4^-$) involved in their retention in basic environment. This fact would also lead to the appearance of neutral species ($Fe(OH)_2^0$, $FeOH_3$), due to the drop in the water dielectric constant that it would cause [21,37]. Furthermore, the drop in pH from 8 to 7 would have led to a relative decrease in the active adsorption sites on these substrates because of the isoelectric potential of some of their constituents [21]. The competitive effect of phosphates with other anions, particularly chloride ions and hydroxide ions, following the decrease in active sites due to the drop in pH could also lead to a decrease in phosphates adsorption rate on these sediments. The effect of the decrease in phosphates adsorption on substrates due to salinity has been demonstrated

by several authors including Bai et al. [28] and Oxmann and Schwendenmann [38]; and that linked to competitiveness with hydroxide ions by Coulibaly [21] and Omari et al. [26]. The simultaneous action of temperature and pH, as well as the geochemical nature of the samples, would contribute in our case to the drop in $q_{abs}(t)$ from $T = 20^\circ C$ and $pH = 7$ to $T = 30^\circ C$ and $pH = 7$. Also, the crystallochemical characteristics of these different sediments could also contribute to the explanation of this process, as Coulibaly [21] has already suggested in the case of phosphates adsorption on laterite. All these phenomenon could explain the weak phosphates adsorption by these substrates in this study relatively to those of the loose sedimentary layer of this channel in its former hydromorphology, used in practically the same experimental conditions by Mahi et al. [15].

In all the experiments carried out, the rapidity of the first stage of phosphates adsorption kinetics on these sediments would testify to the great reactivity of these substrates with this nutrient. This step would reflect the occupation of the most reactive and/or easily accessible sites such as the external surfaces of the particles and of the macropores. In this case, phosphates adsorption on these substrates would be considered spontaneous. In the second phase, the slow phosphates adsorption on these sediments would be characteristic of their retention after diffusion in mesopores and micropores. This would be a gradual adsorption [15, 21, 25].

Of the three models used for the characterization of surface reaction processes, only Blanchard kinetics model [8] and Elovich kinetics model [12] which have been appropriate for the full description of the experimental data. The inadequacy of Lagergren kinetics model [6] for the experimental data description in this study shows that the speed of this process doesn't depend only on phosphates concentration in solution [21]. The suitability of Blanchard kinetics model [8] for the description of phosphates adsorption kinetics on the superficial sediments of Vridi channel was also noted by Mahi et al. [15]. Moreover, the adequacy of this model in this study would show that chemisorption is the dominant mechanism during phosphates retention on these substrates. The suitability of this model to describe kinetics on complex materials would be explained essentially by the real heterogeneity of the complexation sites taken into account by the model [21,25,39-40].

This is in agreement with the nature of these superficial sediments. Also, this model assumes that the maximum adsorption capacity is proportional to the number of active sites occupied by phosphates. Therefore, the maximum adsorption is the formation of a monolayer on the surface of the sediments [26, 41]. The values of h indicate that the mass transfer rate was greater in the experiments carried out at $T = 20^{\circ}\text{C}$ and $\text{pH} = 7$, followed by those carried out at $T = 25^{\circ}\text{C}$ and $\text{pH} = 8$ then those carried out at $T = 30^{\circ}\text{C}$ and $\text{pH} = 8$. This fact is attributed to the external resistance to the mass transfer (access to the Gouy-Chapmann boundary layer) [15,21,42-43], representing a step in chemisorption [43]. h is the more important as the Gouy-Chapmann boundary layer is weak [44-45]. According to Saha et al. [46], the increase in temperature would favor the reduction in the thickness of the boundary layer due to the tendency of phosphates to come back into solution. This could also explain the drop in phosphates adsorption rate in the experiments carried out from $T = 20^{\circ}\text{C}$ and $\text{pH} = 8$ to those carried out at $T = 30^{\circ}\text{C}$ and $\text{pH} = 7$. The relatively high values of h for all the experiments carried out would show a priori that the mass transfer through the film is not a step limiting of phosphates retention by these sediments in this study. The dependence of k_2 on phosphates concentration in solution would justify its importance with the low values of h and vice versa. The experimental results also agree with Elovich kinetics model [12] in their quasi totality. This confirms that chemisorption is the dominant mechanism as shown by the adequacy of Blanchard kinetics model [8] with these experimental data on the one hand, and this chemisorption would be activated [21, 43], on the other. For the particular case of $C_0 = 2 \text{ mg/l}$ for the experiment carried out at $T = 25^{\circ}\text{C}$ and $\text{pH} = 7$, the inadequacy of the experimental data with Elovich kinetics model [12] would show that physisorption would be the mechanism dominant before the equilibrium time while chemisorption would be dominant after this time. A similar remark was reported by Salah [43] in his study relating to the photocatalytic degradation of organic pollutants in the presence of TiO_2 .

Phosphates adsorption by these sediments is not limited to the diffusion phenomena, having regard to the various results obtained with the

two models of the molecular diffusion process. The inadequacy of the external diffusion kinetics model [10-11] with the experimental data confirms that the mass transfer through the film is not a step limiting of phosphates retention on these sediments, as the results obtained with Blanchard kinetics model [8]. The values of C obtained with Weber and Morris kinetics model [9] are in agreement with those of h obtained with Blanchard kinetics model [8], because evolving in the opposite direction of the experiments carried out at $T = 20^{\circ}\text{C}$ and $\text{pH} = 8$ to those carried out at $T = 30^{\circ}\text{C}$ and $\text{pH} = 7$. As mentioned above, the gradual increase in temperature would have the effect of gradually decreasing C [46]. The increase in temperature and decrease in pH in the experiments performed at $T = 30^{\circ}\text{C}$ and $\text{pH} = 7$ would have had the opposite effect relative to the first two experiments. This is shown by the drops in K_d and C with the growth of the initial concentration of phosphates in solution (C_0).

4. CONCLUSION

Ultimately, the results obtained in these experiments confirm the important roles of temperature and pH in phosphates adsorption on substrates in general, and on sediments in particular. This is the case for the superficial sediments of Vridi channel in its new hydromorphology. In the particular case of this estuary, this study also highlights the very great influence of seasonal water supplies on this phenomenon. The results obtained are in agreement with the recent studies of N'Da et al. [13-14] which showed a decrease in the capacity of phosphorus adsorption by the superficial sediments of this ecosystem with its development. The results obtained could serve as a basis for any study aimed at understanding the function mode of this estuary.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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