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Phosphorus adsorption on oven dried alum sludge

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Abstract

Water bodies contain many contaminates that negatively affect aquatic life (animals, plants) as well as the subsequent uses of contaminated water. Phosphorous is considered one of the most important contaminates due to its common role in the eutrophication process. The treatment of nutrients (phosphorous, nitrogen) in wastewater is one of the costly processes, so in this research, the efficiency of oven dried alum sludge (ODAS) in removing phosphorus from wastewater will be studied. ODAS is the residue from sedimentation and coagulation processes in water treatment plants that is dried in a drying oven at 105 °C for 24 hours, cooled to room temperature, and crushed by mill. Several doses of ODAS were taken and the adsorption process was performed on them for samples containing different concentrations of phosphorous. The effect of pH on the adsorption process was also studied. After completing the results, it was found that the dose at which the highest removal percentage was achieved is 10 g/l of ODAS, where the efficiency was (98, 99, 97 and 97) % for samples with an initial concentration of phosphorous (5, 10, 15, 20) mg/l. The highest removal efficiency was by using the dose of 10 g/l at pH equal to 6, where the efficiencies were (95, 97, 94, 93) %.

Keywords: phosphorus removal, adsorption, alum sludge, eutrophication

1. Introduction

One of the most common issues with lakes and inland seas is eutrophication. Eutrophication is the term used to describe the excessive growth of algae. To avoid such situations, industrial wastewater treatment should strive to remove either phosphorus or nitrogen, depending on the receiving water body (to ensure that the nutrient limiting condition is maintained. Phosphorus must be controlled if the discharge is into a freshwater body. (N. S. VARANDANI 2017)

Eutrophication is most common in lakes, ponds, estuaries, and slow-moving rivers. If sufficient nutrients are introduced into a lake system as a result of human activity, the eutrophication process can be accelerated by up to a decade. Because phosphorus is the nutrient that typically limits algal growth in lakes, adding phosphorus in particular can hasten eutrophication. (Fredette et al. 2012)

Controlling phosphorus inputs, on the other hand, can aid in slowing eutrophication (Week 2012). Phosphorus can be found in organic waste discharged from wastewater treatment plants, overflowing septic tank systems, storm sewers, and drainage from lawns, pastures, and fertilized fields (Impellitteri 2004). When phosphorus is discharged into receiving water, it can encourage the growth of unwanted aquatic life. When large amounts of them are discharged on land, they can pollute groundwater (Bennett 2003). As a result, the challenge is to find a material that is both feasible and cost-effective for use in physical-chemical processes. We have not yet found a complete solution to this problem.

Until now, phosphorous removal from wastewater has been limited to chemical methods (coagulation/sedimentation processes) or biological technologies. However, due to low carbon concentrations, biological technologies may not be suitable for small-scale applications, lengthening and increasing the cost of biological methods(Zilles et al. 2002) . Because physical-chemical methods have a high initial cost, we can obtain an alternative process for small industries (S. A. M. Mohammed and Shanshool 2009)

Aluminum has been linked to a negative impact on barley growth in soils with pH levels lower than 5.5, as well as high aluminum levels reducing phosphorus availability (Week 2012).

In wastewater, phosphorus can be found in three forms: orthophosphate, polyphosphate, and organically bound phosphate.

Other mineral or biological colloids have a lower affinity for phosphorus than Al^{+3}. As a result, when Al^{+3} is added to a phosphate and microorganism-containing suspension, it will first react with the phosphate and then with the colloids after the phosphate has precipitated(Stevenson 1997).

The most logical residual management program in sludge disposal attempts to use the following approach:

1. Decrease residual generation.

2. Recovering chemically from treatment.

3. Reducing volume with residual treatment.

4. Disposal in an environmentally friendly manner at the end(Mackenzie L. Davis 2010).Due to the high initial cost of physical-chemical methods, we can obtain an alternative process for small industries (S. A. M. Mohammed and Shanshool 2009). There are additional advantages for physical-chemical methods, one of which is ease of use; there is no need for extensive maintenance experience, and extra costs associated with sludge handling will be eliminated due to sludge reuse.

If a suitable, easily available, and cost-effective material for phosphorus removal is discovered, the main issue in small-scale applications will be resolved (W. T. Mohammed and Rashid 2012). Waste management is critical because human activities generate a large amount of waste and bio solids (Dietze, Gnirß, and Wiesmann 2002). Alum sludge, which is a residue from the coagulation process in a water treatment plant, is one such species, and in recent years, many researchers have focused on the reuse of alum sludge in a variety of water treatment applications (Fredette et al. 2012). When alum activities persist, disposal of alum sludge may improve phosphorous removal in a wastewater treatment plant (Week 2012) . When it comes to colloidal competition with ionic groups, Al_3 has a stronger attraction for phosphorous. As a result, the Al_3-phosphate reaction occurs first in a suspension containing phosphate and microorganisms, followed by a reaction with colloids after phosphate precipitation. When Al_3 is used in tertiary treatment, the phosphate requirement must be met first, followed by other requests (20). That is a compelling argument for using aluminum-based residues as a phosphorous removal substance. Alum sludge contains a mixture of various forms of aluminum hydroxide (Aguilar et al. 2002).

Because aluminum hydroxide is an effective phosphorous medium, alum sludge can be used as an adsorbent for phosphorous (Galarneau and Gehr 1997).

There are several methods for removing phosphorus from wastewater. There are three types of processes: biological, physical, and chemical. Phosphorus is removed with the help of a new type of sorbent known as a polymeric ligand exchanger (PLE), as well as zirconium oxides and activated alumina (D. Zhao and Sengupta 1998). Two deep-bed filters were used, one with ferrous sulphate as a precipitation agent (Jonsson, Plaza, and Hultman 1997) and the other with natural zeolite modified with lanthanide (90 nm) as an adsorption media (NING et al. 2008). To achieve a high phosphorous removal efficiency, coagulation–filtration was used, and practical applicability was emphasized (Xie et al. 2005). Coagulation and membrane microfiltration are also used to remove dissolved phosphorous (Yu et al. 2000), alum as (Al_2. $(SO_4)_3$.$18H_2O$) is used to remove phosphorous in low alkalinity secondary effluent (Banu, Do, and Yeom 2008) and phosphorous adsorption is accomplished by flocculation following alum Al_2 $(SO_4)_3$ hydrolysis, and phosphorous adsorption is accomplished by flocculation following alum Al_2 $(SO4)_3$ hydrolysis (Boisvert et al. 1997). Two types of dissolved air flocculation DAF were used in nutrient removal: standard air dissolving tank method and modified process with air injection into the suction side of the recirculation pump (Jokela et al. 2001).

The goal of this study is to see how well oven dried alum sludge removes total phosphorous from wastewater at different concentrations and conditions.

Historically, alum sludge was thought to be an inert waste product with little reuse potential. (A. E. Albrecht 1972). Aluminum (Al) is well-known for its high phosphorus affinity (Norris and Titshall 2012). As a result, alum sludge is the most widely generated and locally available water treatment waste on a global scale, but it is typically discarded (201) (Yan Yang et al. 2018)

The first applications of alum sludge were in the field of reuse, and some research was done to determine its engineering properties (Y.Q Zhao 2001).

Previously, alum sludge was used in agricultural applications (Y. Q. Zhao, Zhao, and Babatunde 2009) (Dassanayake et al. 2015). Later, some researchers used sludge to remove phosphorus, and the phosphorus removal capacity of alum sludge was studied in batch sorption tests (Kim et al. 2002). Furthermore, a small-scale continuous flow system with effluent recycling has been set up to investigate the phosphate adsorption ability of air-dried alum sludge from wastewater effluent (Huang and Chiswell 2000). The purpose of the batch experiments was to determine the properties of dewatered alum sludge for phosphorus adsorption (Yongzhe Yang et al. 2006). The researchers investigated the ability of alum sludge removing to absorb phosphorous in aqueous solutions by extending their research into the possibility of phosphorous uptake by sludge under different conditions (Kim et al. 2002) (Babatunde and Zhao 2010).

In addition, the beneficial reuse of alum-containing drinking water treatment sludge extends to the development of a new system for wetlands (CWs) that uses alum sludge as a basic substrate (Y. Q. Zhao et al. 2011) (Kumar, J.L.G.; Wang, Z.Y.; Zhao, Y.Q.; Babatunde and Zhao, X.H.; Jørgensen 2011) (Babatunde, Zhao, and Zhao 2010).

Adsorption is a mass transfer process in which an effluent gas stream is passed through the surface of prepared porous solids (adsorbents) (Spellman and Whiting 2005). Adsorption is well-known as one of the most effective methods for removing various pollutants from bodies of water (Park et al. 2020).

Adsorption can be classified as either physical or chemical. Physical adsorption is based on the attraction force of van der Waals on surfaces, whereas chemical adsorption is associated with a chemical reaction or the transfer of electrons and ions between the adsorbent surface and molecules (Wang, Hung, and Nazih K. Shammas 2013).

The target molecules are attracted to the surface of pore walls within a high sorbent by van der Waals forces and have a low heat of adsorption that is only slightly greater than the adsorbate's heat of sublimation. Chemisorption involves a covalent chemical reaction in which the target gas binds to specific sites on the sorbent with a much higher heat of adsorption, roughly equal to the heat of reaction (Berger and Bhown 2011).

Both kinetic and equilibrium modeling are used in biosorption mathematical modeling. The history of absorption is described by kinetic models, whereas the capacity of sorption as a function of chemistry is described by equilibrium models. Empirical models such as Freundlich and Langmuir isotherms, as well as theoretical models, are used in equilibrium modeling.

The Langmuir isotherm model is represented by the equation below (Park et al. 2020).

$$q_e = \frac{\mathrm{q_{max}\ b\ Ce}}{1 + \mathrm{b\ Ce}}$$

Where q_e represent contaminant amount which adsorbed at equilibrium (mg/g or mmol/g),

C_e represent contaminant concentration in solution at equilibrium (mg/L or mmol/L),

q_{max} represent the maximum capacity of adsorption (mg/g or mmol/g),

b represent Langmuir constant associated with adsorption heat (L/mg or L/mmol).

The following equation describe Freundlich isotherm model:

$$q_{e=} K_f C_e^{1/n}$$

2. Material and Experimental

2.1. Oven Dried Alum Sludge

Prepared by heating an alum sludge (accumulated as a result of the coagulation and sedimentation processes in water treatment plants) in an oven at (104 C) for (24 hr.), then cooling to room temperature and crushing with a mill. Table (1) illustrates the composition of ODAS.

Table 1 : ODAS Composition

Components	Percentage
Al_2O_3	40.1 %
Fe_2O_3	21.7 %
SiO_2	10.8 %
CaO	6.4%
K_2O	5.3%
Na_2O	4.6%
MgO	3.5%
SO_3	1.3%
P_2O_5	0.8%
Others	6.3%

2.2. Wastewater

To study the efficiency of the adsorbent material at different phosphorus concentrations, sodium phosphate diluted in deionized water was used as a sample to make several phosphorous solutions. The mass of sodium phosphate used to prepare various phosphorous concentrations is shown in Table 2, and the physical and chemical properties of Na3PO4 are shown in Table 3.

Table (2): Solutions of phosphorous

P concentration (mg/l)	Na_3PO_4 which added (g)
5	0.02646
10	0.05292
15	0.07939
20	0.10585

Table (3): Properties of Na_3PO_4

Component	
Molecular Weight	163.941 g/mole
Complexity	36.8
Melting point	1583° C
Solubility	freely soluble in water
Density	2.54 g/cm^3
pH	11.5-12.5

3. Experimentation

Batch work experiments were used to obtain isothermal equilibrium and, later, data equilibrium. The following variables were investigated:

1) The effect of ODAS on P adsorption,

2) The effect of pH on P adsorption, and

3) Equilibrium isotherm experiments

All results were checked in a shaker with a water bath at (25° C ±1), shaking speed of (185 rpm), and contact time of (180 min).

Flasks of 100 ml were used in the experiments and phosphorous concentrations of (5, 10, 15, and 20) mg/l were investigated for all conditions.

The ODAS dosage used as adsorbent material was (20, 10, 5, 2.5, and 1.25) g/l and the final phosphorous concentration as adsorbate was measured using a UV-VIS Spectrophotometer.

4. Result and Discussion

4.1. The effect of ODAS dosage

The following figures illustrate the effect of the change for dose on the efficiency of phosphorous adsorption.

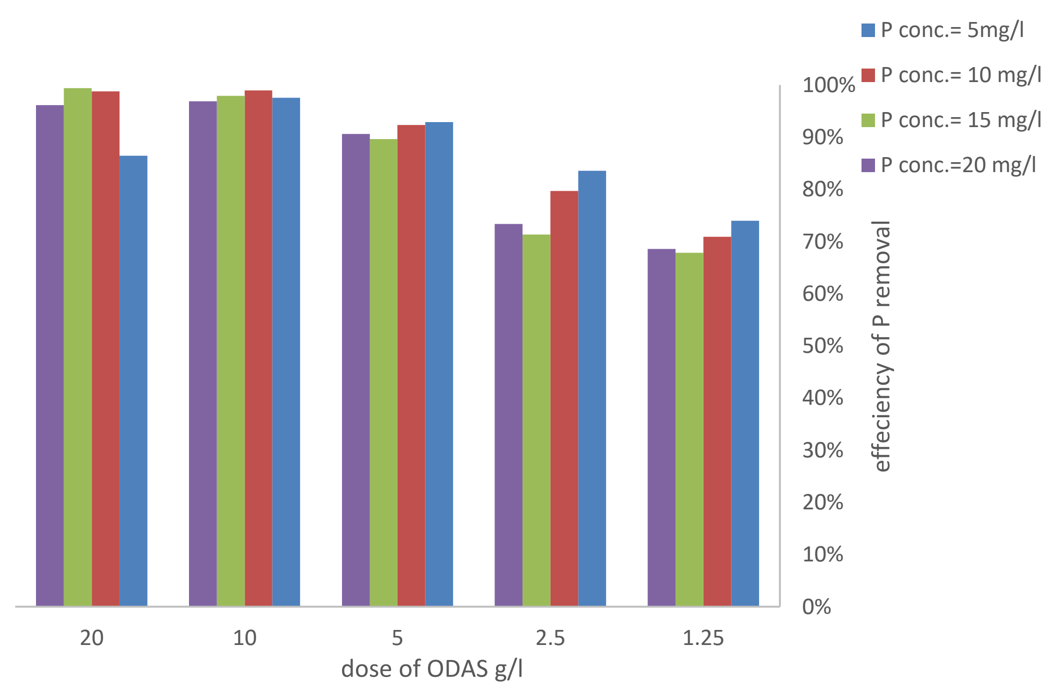

Figure 1: Effect of ODAS dose on P removal efficiency

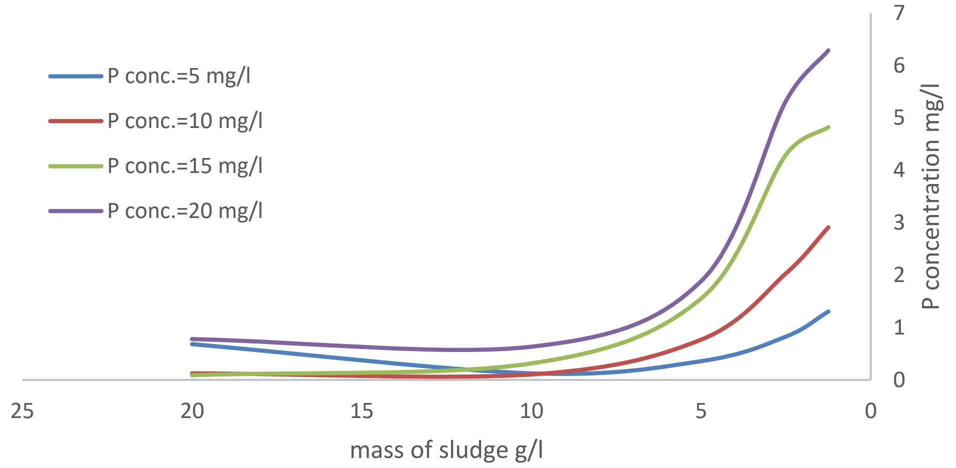

Figure 2: Influence of ODAS dose on residual phosphorus

A dose 1.25 g/l achieved the highest efficiency at concentration 5 mg/l and when the dose was doubled to 2.5 g/l, it was noticed that the adsorption efficiency started to increase as the highest efficiency rate. The phosphorous removal was at a concentration 5 mg/l as well, even at a dose 5 g/l.

At the dose 10 g/l, the removal ratio was significantly and highly increased for all concentrations, and the phosphorous removal efficiency (98, 99, 97 and 97)% was obtained for 5, 10, 15 and 20 mg/l concentrations respectively. At a dose of 20 g/l, it was observed that at a concentration 5 mg/l, the removal efficiency drops to 86%, but it remained the same for the rest of the concentrations (99, 99 and 94)%. The figures show that the final concentration of phosphorous after the adsorption process decreases with increasing ODAS weight. The lowest concentration obtained at a dose of (10, 20) g / l. Advanced wastewater treatment to remove approximately 61 to 68 percent of phosphorous was found to increase capital costs by 42 to 99 percent (Fredette et al. 2012) and thus for more economic purposes 10 mg/L was obtained as the optimal dose for all experiments.

4.2. pH Variation

The effect of pH change on the adsorption process shown in the figure below

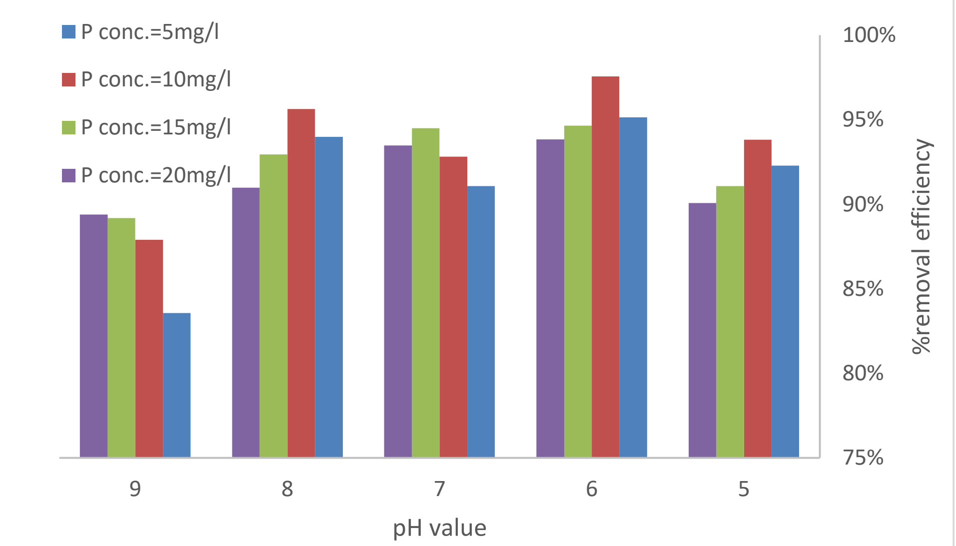

Figure 3: Effect of pH on P removal efficiency

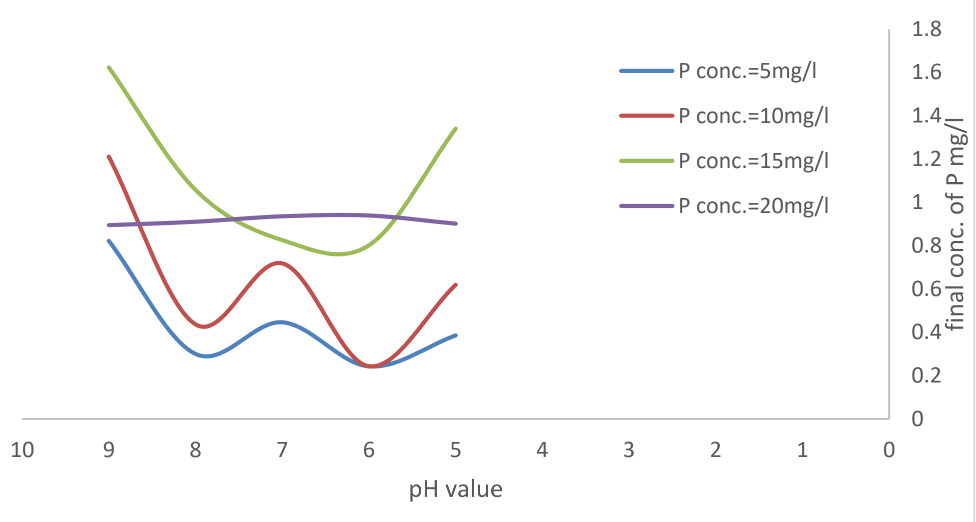

Figure 4: influece of pH on residual P

From the above figures, it was shown at a concentration of 5 mg/L, the efficiency of phosphorous removal was as follows: (92, 95, 91, 94, and 83) % at pH values (4, 5, 6, 7 8 and 9) respectively. The highest removal rate was at pH = 6.

When the initial concentration of phosphorous increase to 10 mg/l, the efficiency of removal become as follow (93, 97, 92, 95, 87) % and the highest percentage was at pH = 6.

At P concentration of 15 mg/l, the highest removal efficiency was at pH values 6 and 7. The efficiency removal at phosphorus concentration at 15 mg/l was (91, 94, 94, 92 and 89) %.

At a concentration of 20 mg/L, the efficiency ratio decreased for most of the pH values except at pH of 9, as the readings recorded the following efficiencies (90, 93, 93, 90 and 89) % .

From all results, it was found that the highest percentage of phosphorous removal was achieved at pH 6 at an initial concentration of phosphorous 10 mg/l, and for all concentration, the pH value of 6 achieved the highest efficiency.

4.3. Equilibrium Isotherm Experiments

Two models of isotherm estimated on the results Langmuir and Freundlich. Langmuir isotherm curves get by plotting the solute concentration at equilibrium (C_e) against C_e /(x/m) and Freundlich isotherm curves for P adsorption on ODAS were get by plotting the solute concentration at equilibrium (C_e) against the solute weight that adsorbed per adsorbent weight (q_e) where

$C_{e:}$ solute concentration at equilibrium

X: mass of adsorbate adsorbed (mg)

m: mass of adsorbent (g)

$q_{e:}$ solute weight that adsorbed per adsorbent weight.

The figures below show the isotherm curves of phosphorus adsorption and the tables illustrate the parameters of Langmuir and Freundlich isotherm models:

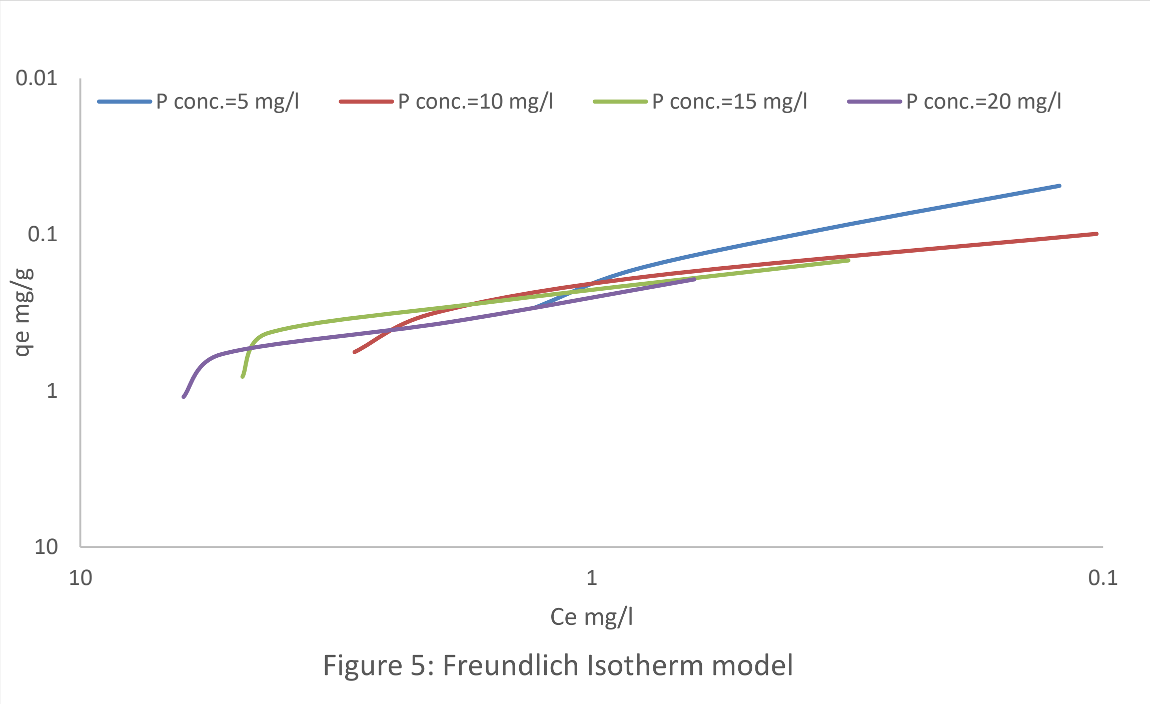

Figure (4-13): Freundlich Isotherm model

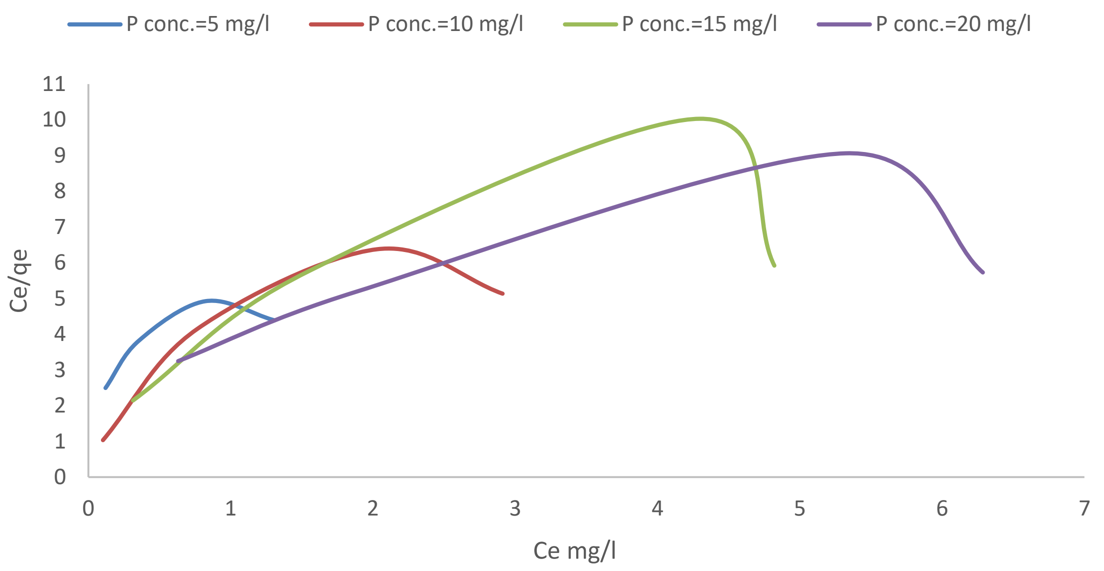

Figure 6: Langmuir Isotherm model

Figure (4-14): Langmuir Isotherm model

Table (3) : Isotherm parameter for P adsorption on ODAS

P conc. (mg/l)	model	Parameters	value
5	**Langmuir**	q_m	**2.582992**
		b	**0.133411**
		R^2	**0.598104**
		R_L	**0.599857**
	Freundlich	**k**	**1.04182034**
		n	**4.87039**
		R^2	**0.98438**
10	**Langmuir**	q_m	**2.2529**
		b	**0.21541**
		R^2	**0.64602**
		R_L	**0.31704**
	Freundlich	**k**	**1.1558**
		n	**6.3338**
		R^2	**0.94715**
15	**Langmuir**	q_m	**0.8929**
		b	**0.38729**
		R^2	**0.6882**
		R_L	**0.14685**
	Freundlich	**k**	**1.2218**
		n	**8.21019**
		R^2	**0.84568**
20	**Langmuir**	q_m	**1.56176**
		b	**0.1804**
		R^2	**0.51367**
		R_L	**0.21701**
	Freundlich	**k**	**1.2322**
		n	**7.52095**
		R^2	**0.83910**

5. Conclusion

Through this study, the following results were obtained:

- ODAS an effective adsorbent for phosphorous in different concentrations
- The efficiency of phosphorous removal does not show a high increase by doubling the dose of ODAS from 10 g/l to 20 g/l.
- Maximum removal of phosphorus obtained in the acidic medium (at low pH).
- The results showed that the Freundlich model, which has the highest correlation coefficient, is well suited for adsorption capacity.

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