

# **Value Additions on Iron-Oxide Nanoparticles in Laterite Soils Available in South-West Sri Lanka: Development of Effective Filtering Techniques**

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## **Abstract**

Environmental contamination by phosphate is on the rise with extensive and diffuse pollution. Answering these badly behaved with serious technologies is very costly. Soil has been commonly used in several wastewater treatment systems and showed to be an in effect substrate for phosphate removal and retention. Using natural sorbent such as laterite could be a way out. The removal of phosphate from aqueous solutions was investigated by using raw laterite in this study. In the adsorption process, the effect of pH, contact time, concentration, adsorbent dosage and salt concentrations were taken into consideration and experiments were carried out in the batch experiment system. Laterite proved to be an effective adsorbent and the removal efficiency remained around 90% of all cases. The optimal dosage was identified as 1g and the removal efficiency was more than 90%. Study of the adsorption as a function of contact time showed that 3 hours was sufficient time for maximum removal of phosphate. Acidic environments of pH values less than 5 facilitated the adsorption of phosphate and the removal efficiency decreased with increasing pH value of the solution. Based on the obtained results from this study, raw laterite is effective in removing phosphate from aqueous solutions and is a cost effective alternative for commercially available adsorbents that are currently used to remove contaminants from drinking water.

*Keywords: Adsorption, phosphate, laterite, removal, efficiency*

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## **1. Introduction**

Phosphorus is an indispensable element for all life on Earth that exists naturally as phosphate ion. Phosphate is added to the environment natural due to decomposition of rocks and minerals, storm water runoff, agricultural runoff (Johnson et al., 2007; Donald et al., 2011), erosion and sedimentation, atmospheric deposition, direct input by animals. Phosphate is also added due to anthropogenic activities such as discharge from wastewater treatment plants and permitted industries (Mueller, 1995).

Out of the main sources of phosphate, municipal wastewater has a significant portion of phosphate than others (Binder et al., 2009; Egle et al., 2014). The natural background levels of total phosphorus are generally less than 0.03 mg/L. The natural levels of phosphate usually range from 0.005 to 0.05 mg/L in water and soil solutions generally contain 0.20 to 0.30 mg/L (Daniel et al. 1998). Many bodies of freshwater are currently experiencing increases of phosphorus from outside sources. The range from 0.025 - 0.1 mg/L stimulates the algal growth and levels greater than 0.1 mg/L accelerates the growth of algae that (Brian, 2005). Small additions of phosphorus can result in large changes in aquatic systems and deteriorate the water quality by changing physical, chemical and biological parameters of the water bodies (Carneiro et al., 2014). Too much phosphorus triggers vigorous growth of algae and larger aquatic plants, which can result in decreased levels of dissolved

oxygen, a process called eutrophication. High levels of phosphorus can also lead to algae blooms that produce algal toxins which can be harmful to human & animal health and suffocates fish populations

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(Chislock et.al., 2013). The most visible effect of cultural eutrophication is the creation of dense blooms of toxic, and foul-smelling phytoplankton that reduces the clearness of water and harms water quality. Algal blooms limit light penetration, reducing growth and causing die-offs of plants in littoral zones and complicates the process of water purification (Tilman et al., 2001).

In order to combat eutrophication like water issues of water reservoirs phosphate should be removed and maintain at its safe level. Various technologies are used to remove phosphorus from wastewater, including: chemical precipitation, membrane separation, biological treatment, and adsorption (Yeoman et al., 1988). But chemical and biological phosphorous treatments are the two main techniques that are used in removing phosphorus from domestic and industrial wastewater. Furthermore, many variations and combinations have been used for the treatment processes. However, these methods of phosphate removal from drain liquids also have certain disadvantages, such as capital costs, operation and maintenance costs, coagulants generate as secondary pollution and complex usage.

Clay minerals have widely been used for removal of phosphate from wastewater. Hamdi and Srasra (2012) dealt with the removal of phosphate ions from aqueous solutions using kaolinitic and smectic clay minerals and synthetic zeolite as an adsorbent. As well as Al-modified Betonite samples were prepared and used for phosphorus removal from the various phosphate contains solutions (El-Sergany and Shanableh, 2012). Laterite is a common secondary clay mineral present in tropical countries under wet climate, that is rich in iron and aluminium predominantly with rusty-red color due to higher levels of iron oxides. Laterite consist specific iron and aluminium bearing primary and secondary minerals such as kaolinite, goethite, hematite, gibbsite and quartz. It usually forms in hot and wet tropical and subtropical regions of Sri Lanka, where the climate is humid. Laterite soil has a higher percentage of clay, higher cation exchange capacity with higher water retaining capacity. In general, laterite is soft and wet in nature (Engelhardt, 2010). It has a strong affinity for sorption of inorganic and organic contaminants. Hence, laterite has been revealed as an effective filter media for contaminated water. Therefore, the scope of this study is to use laterite as a filter medium to remove phosphate in an aqueous solution. The main objectives are to explore an effective phosphate filtering system by laterite soil and to investigate the optimum characteristics of the material for associated sorption processes.

## **2. Methodology**

### *2.1 Sample preparation*

Laterite soil samples were collected from the Western province of Sri Lanka by auger drilling method. Collected soils were air-dried about 48 hours to remove the excess moisture content. Air-dried soil was crushed to prepare powdered material (106  $\mu\text{m}$ ).

### *2.2 Chemical experimentation*

The adsorption of phosphate into the laterite soil was carried out under different conditions. Under room temperature and natural pH, the optimum adsorbent soil dosage, optimum phosphate concentration and optimum contact time were studied at the beginning. The Ascorbic acid method (Eaton et al., 2005) was followed for all the experiments and finally, the removal efficiency of the samples was measured using the UV spectrophotometer at 890 nm wavelength. Readings were taken in triplicates to increase the accuracy.

The absorbed amount of phosphate at the equilibrium was calculated by the equation 1. Where  $C_0$  is the initial concentration of the phosphate (ppm), and  $C_e$  is the equilibrium concentration of the phosphate (ppm). The removal efficiency was calculated according to equation 1.

$$\text{Percentage Removal efficiency} = \frac{(C_0 - C_e) \times 100}{C_0} \quad (1)$$

where:

$C_0$ =Initial concentration of the phosphate (ppm)

$C_e$ =Equilibrium concentration of the phosphate (ppm)

### 2.3 Determination of the optimum soil dosage

A phosphate stock solution of 1000 ppm was prepared by dissolving  $\text{KH}_2\text{PO}_4$  in deionized water and needed solutions were prepared dilutions. Initial phosphate concentration was prepared to 50 ppm and then 50.0 ml of the solution was added to 0.2, 0.5, 1.0, 1.5, 2.0, 2.5 and 3 g of laterite soil in the process to identify the optimum soil dosage. The solutions were shaken at 120 rpm for 1 hour, and the remaining phosphate concentrations were measured.

### 2.4 Determination of the optimum phosphate concentration

To determine the optimum phosphate concentration, series of phosphate solutions were prepared as 0.2, 0.4, 0.6, 0.8, 1, 5, 10, 15, 20, 30, 40 and 50 ppm. 1 g of laterite soil was mixed with 50 ml of solutions and were shaken at 120 rpm for 24 hours. The remaining phosphate concentrations were measured.

### 2.5 Determination of the optimum contact time

50.0 ml of 0.8 ppm phosphate solutions were mixed with 1 g of soil and optimum contact time was observed at different time intervals to find out the optimum contact time for the low concentration. The same procedure was followed for the high concentration phosphate (50 ppm). The solutions were shaken at 120 rpm and the remaining phosphate concentrations were measured.

### 2.6 Determination of the optimum pH

Then optimum pH of phosphate removal by laterite soil was studied by adding 1 g of laterite soil in 50 ml of phosphate solution and it was shaken at 120 rpm for 3 hrs. The pH was adjusted by adding  $\text{H}_2\text{SO}_4$  acid and NaOH drops. 1 g of soil sample was mixed with 50 ml of 0.8 ppm phosphate solution for the testing. The solutions were shaken at 120 rpm and the remaining phosphate concentrations were measured.

### 2.7 Determination of the effect of salts

For the determination of effect of salts of the phosphate absorption, salt solutions with 2 ppm concentrations were prepared. Then 25 ml of the phosphate solution and 25 ml of salt solution ( $\text{NaCl}$ ,  $\text{NaF}$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{NaCl}$ ,  $\text{NaNO}_3$ ,  $\text{CaCO}_3$ ,  $\text{CaCl}_2$ ,  $\text{CaHCO}_3$ ) were mixed and added into the 1 g of laterite sample and kept 3 hrs.

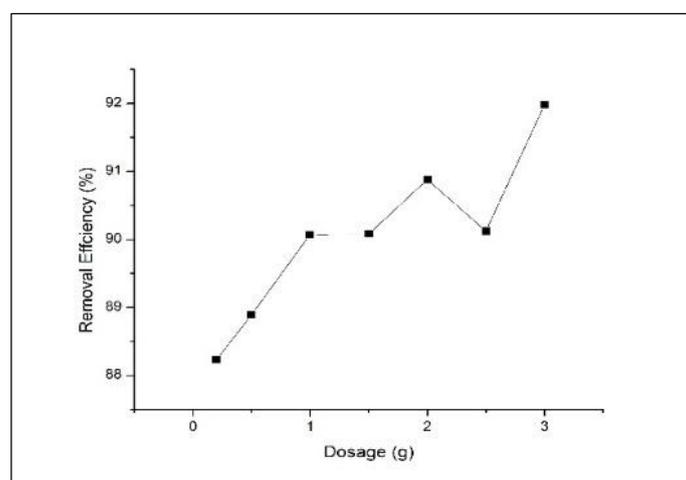


Figure 1. Determination of optimum soil dose

## 3. Results and Discussion

Varying initial soil amount, research was conducted to find out the optimum soil dosage required to attain the highest removal efficiency during laterite phosphate contact. Variation in the

removal phosphate efficiency at different initial soil dosages is shown in Figure 1. This figure shows the removal efficiency of phosphate increases rapidly with an increase in the amount of laterite due to greater availability of the surface area for the adsorption of phosphate. A significant increase in uptake was observed when the dose was increased from 0.3 to 3 g. The figure shows more than 88% higher rate of removal of phosphate and 3 g showed highest removal efficiency (91.98%). 1 g showed more than 90% removal efficiency at first. Therefore 1 g of laterite soil was identified to continue the research.

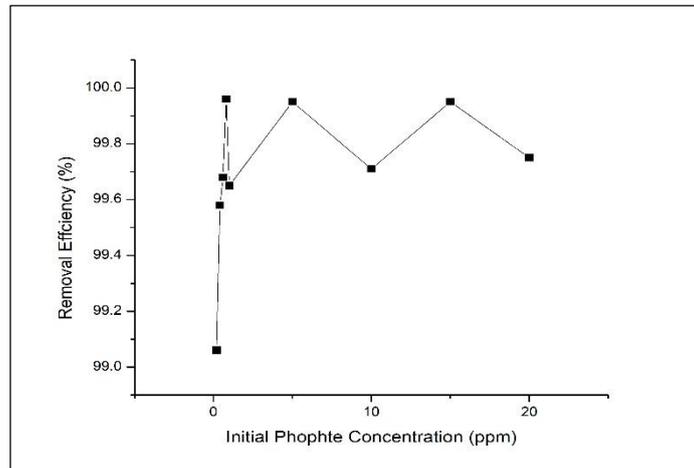


Figure 2. Determination of optimum phosphate concentration

Phosphate removal efficiency at different initial concentrations is shown in Figure 2 and it is evident that more than 99% phosphate removal was seen in all cases. 0.8 ppm was showed highest removal efficiency (99.96%) from all phosphate concentrations. 5 ppm and 15 ppm solution also showed high values (99.95%) that were close to the results of the 0.8 ppm solution. Removal efficiency, increased with the increment in the concentration, because the ratio of surface active sites to total phosphate increases and therefore the interaction of adsorbate with the active sites on the adsorbent surface were sufficient for efficient phosphate removal.

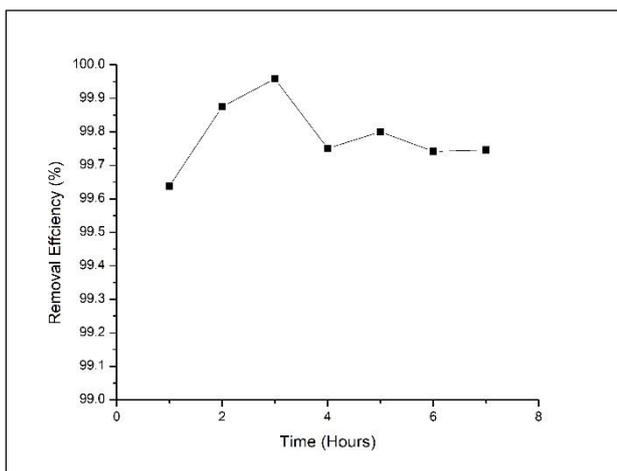


Figure 3. Determination of optimum time for low phosphate concentration

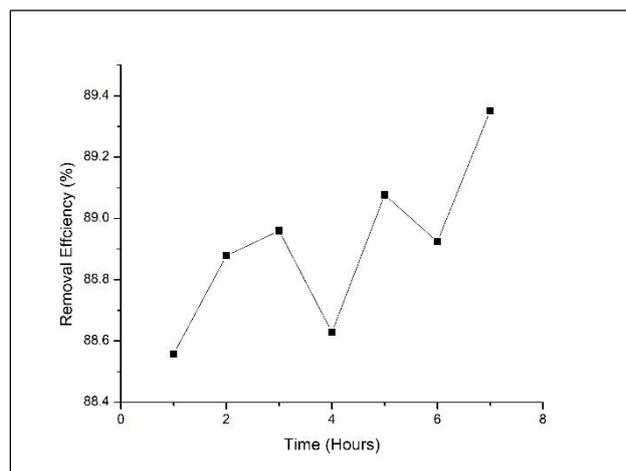


Figure 4. Determination of optimum time for high phosphate concentration

Figure 3 and 4 show the optimum contact time for low and high phosphate concentration respectively. The Figure 3 shows more than 99% removal efficiency followed by a higher subsequent removal rate that gradually approached a constant. At 3 hours it showed the highest removal efficiency of 99.9%. The solution adsorption initiates the rapid removal at the start, while the rate tends to be slightly slowed down with the saturation of the adsorption sites at the late hours. The removal

efficiency of phosphate at low concentrations showed that the removal efficiency, increased up to 3 hours and decreased after 3 hours, where the curve seems to be flattened. Figure 4 showed that more than 88% removal efficiency for high phosphate concentration and 7 hours showed the highest removal efficiency (89.35%).

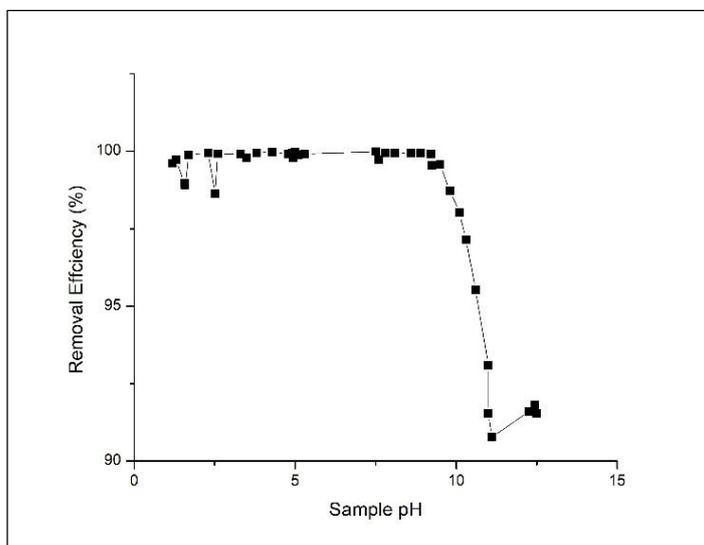


Figure 5. Determination of optimum pH

The results obtained indicate that adsorption of phosphate on laterite increases with decreasing pH, with the highest removal efficiency (99.99%) obtained at pH 7.5 and 99.8% removal efficiency obtained at pH 4.3 and pH 5. The amount of phosphate ions removed from solution by at low pH values was higher than that removed by laterite at high pH values. Lowest removal efficiency (90.76%) was obtained at pH 11.1. This observation can be attributed to the effects of solution pH on phosphate speciation and charge development on the laterite surface.

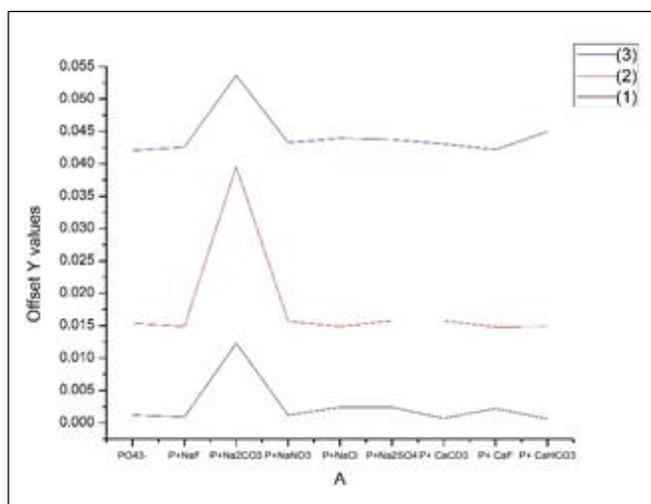


Figure 6. Determination of effect of salts

Phosphate adsorption on laterite soil was investigated in the presence of following competing salts: NaF, Na<sub>2</sub>CO<sub>3</sub>, NaNO<sub>3</sub>, NaCl, Na<sub>2</sub>SO<sub>4</sub>, CaCO<sub>3</sub>, CaCl<sub>2</sub> and Ca(HCO<sub>3</sub>)<sub>2</sub>. The eight competing salts had a different adverse effect on phosphate removal. The phosphate adsorption of laterite soil follows the order: NaF>CaCl<sub>2</sub>>CaCO<sub>3</sub>>NaNO<sub>3</sub>>Ca(HCO<sub>3</sub>)<sub>2</sub>>NaCl>Na<sub>2</sub>SO<sub>4</sub>>Na<sub>2</sub>CO<sub>3</sub> respectively. Among the eight salts, all the salts had not largest impact on the adsorption of phosphate.

Usage of laterite soil as an adsorbent to remove contaminants from water has several advantages. The main benefit is, the lower cost. Other adsorbents require intense purification, preparation, and activation before use in practical applications. However laterite, in its raw form showed significantly high phosphate removal efficiency without any of these steps. This is one reason for the lower cost. It has also been seen that laterite soil is readily available in countries such as Sri Lanka (Dissanayake, 1980), India (Kumar, 2008), and China (Zhang et al., 2014), which is beneficial and cost effective. Other than that, as laterite soil is a natural adsorbent, no environmental damage is anticipated during the preparation stages. Similar studies have been conducted by using Laterite soil available in China (Zhang et al., 2014), however, the phosphate removal efficiency of Sri Lankan raw laterite is very much higher.

#### 4. Conclusion

Adsorption studies were conducted to investigate the adsorption capacity of laterite under different conditions. Based on the result of studies, laterite can be used as an adsorbent for the removal of phosphate from the solutions. Laterite is easily available in large quantities and the treatment method of adsorption seemed to be economical. There is no need for the pre-treatment of the laterite with any chemicals such as acid or base. This minimizes the cost of adsorbent preparation. Laterite proved to be an effective adsorbent and removal capacity was found around 90%. The phosphorus removal increases with adsorbent dosage. Study of the adsorption as a function of contact time showed that 3 hours was sufficient time for maximum removal of phosphate. pH value is an important factor affecting the adsorption of phosphate on laterite. Acidic environment (pH=1-5) is favorable to the adsorption of phosphate. The amount of adsorption decreases with increasing pH value of the solution. The results concluded that laterite soil has a higher removal efficiency for all the given conditions.

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