

REVIEW

Fe-P pools as phosphorus source for rice in acid sulfate soils

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ABSTRACT

Acid sulfate soils (ASS) are rich in Fe, low in available P, and low in soil pH. In acid soils, low P availability is associated with high Fe concentration, which has a high capability in P retention. The reactivity of Fe oxyhydroxides turns large proportions of soil P into insoluble or unavailable form. This review paper is addressed to provide new insights into the Fe and P relationship as the P source for rice (*Oryza sativa* L.) in ASS. Fertilizer application did not significantly alter P solubility in soil solution due to high reactivity and retention power of Fe mineral in ASS, but rice cultivation practice such as water management and soil amelioration has influenced formation, crystallization and character of Fe mineral. Subsequently determining adsorption and desorption of P. Soil waterlogging and drying alternately during rice cultivation have change soil pH and redox potential (Eh), causing dissolution of P from vivianite and reduction of Fe(III)-P to Fe(II)-P. We summarize that Fe-P pools (amorphous-Fe-P) in the rice fields in ASS are categorized as readily available or labile P fraction.

Key words: Acid sulfate soils, iron, *Oryza sativa*, phosphorus, rice, waterlogging.

INTRODUCTION

Acid sulfate soils (ASS) are commonly found in an area where conditions are rich in Fe, sulfate, decomposable organic matter, sulfate reducing bacteria and conducive for sulfate reduction such as waterlogged or swamp environments (Rabenhorst et al., 2013). These soils are widespread in tidal swamps where they are naturally exposed to periods of flooding and drying. Acid sulfate soils typically contain pyrite chemically stable under reducing conditions (Fanning et al., 2017). Alternately of waterlogging and drying of rice cultivation activity as upon land drainage can lead to pyrite oxidation resulting pH < 4 in soil (Fahmi et al., 2018).

Iron occurs predominantly in two oxidation states, ferric (Fe(III)) and ferrous (Fe(II)). Iron is typically in exchangeable, soluble, reductable and residual forms in wetland soils (Reddy and DeLaune, 2008). The abundance of Fe in ASS is related with low soil fertility, especially in high soil acidity, low P availability and Fe toxicity. Its concentration ranges from 10 to 4500 mg kg⁻¹ (Fahmi et al., 2018). The Fe(III) may acts as an oxidizing agent for pyrite mineral at pH < 3 (Bonniessel-Gissingner et al., 1998) and act as source of soil acidity in a hydrolysis reaction (Groeningen et al., 2020).

Phosphorus has many roles in plant's developmental processes at both cellular and whole plant level, seed germination, seedling establishment, root, shoot, flower and seed development, photosynthesis, respiration and N fixation (Malhotra et al., 2018). Phosphorus is a limiting factor for plant growth in wetlands, especially in ASS. Phosphorus-fertilizer application could supply P for plant and help reduce the level of exchangeable Al and Fe, thereby improving the rice growth in ASS (Nguyen et al., 2017). On the other hand, Purnomo et al. (2005) reported local rice varieties yielded even without P fertilizer application, this means P requirement of rice plants can be provided from the native source of P in the soil. Each plant species has developed specific strategies to cope with sparingly P availability. Phosphorus uptake requires

three steps, i.e., P mobilization, P transport to the root surface (predominantly by diffusion), and uptake by root cells. All three processes may be optimized to improve P uptake, but the P mobilization strategy depends on the P-pool and plant species (Schubert et al., 2020).

In acid soil, the majority of soil P is in the form of poorly available for plants. Phosphorus predominantly adsorbed by Al/Fe-oxides and hydroxides (Gypser et al., 2021). These oxides become the main P adsorbents in soils. They have large specific surface areas, which provide large number of adsorption sites. At first, P is adsorbed on the surface of Fe/Al-oxides by forming various complexes. Furthermore, P may be occluded in nanopores of Fe/Al-oxides, and thereby become unavailable to plants (Schubert et al., 2020).

Aluminum and Fe are abundant metal elements in soils. The Fe/Al-hydroxides showed different capacities to retain P, depending on the crystallinity of the hydroxides (Gypser et al., 2021). Based on their reactivity in wetland soils, Fe is more predominant compared to Al, especially in redoximorphic soil. Zin et al. (2015) concluded that high Fe content causing the distribution of the P fraction in ASS is dominated by the Fe-P fraction, the distribution of inorganic P forms was in the order of Fe-P > Al-P > reductant soluble-P > loosely bound P (solid-P) > Ca-P. Especially in ASS, P is mainly adsorbed by Fe (Zin et al., 2015; Fahmi et al., 2018) (Figure 1). This order is related with redox process in wetland soils. Soil waterlogging in rice cultivation on ASS increase Fe(II) concentration from reduction of Fe(III). These processes causing decrease in retention power of Fe for P. According to Li et al. (2012) in wetland soil, P biogeochemistry strongly relate with Fe, particularly in hydromorphic soils where Fe redox reactions control the equilibrium between P retention and release. These facts show that the relationship between P and Fe is complicated and poorly known. The presence of Fe is still considered as the cause of the unavailability of P in ASS. Better understanding is needed to describe the role of Fe on P availability in ASS. Using the literature studies method, this review paper addressed to providing new insights into the Fe and P relationship as source of P for rice in ASS.

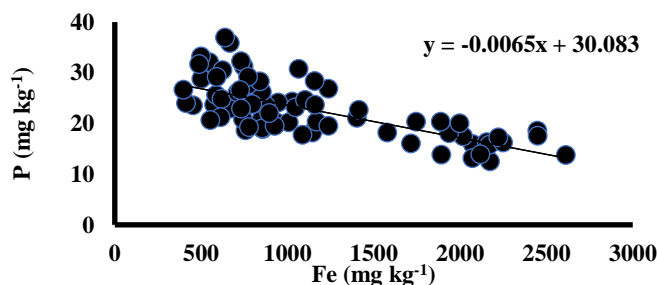


Figure 1. Relationship between P (extracted with Bray I) and Fe (extracted with NH_4OAc) in acid sulfate soils (Fahmi et al., 2018).

PHOSPHORUS IN SOIL

Phosphorus in the soils is in various “pools” of organic and inorganic that vary in availability level. Akinrinde (2005) classified available P into: (i) Readily available (P in soil solution and also in rapidly decomposing organic matter), (ii) moderately available (P that is surface adsorbed of Al and Fe), (iii) difficulty available P are occluded or subsurface P in apatite and organic matter. According to Barker and Pilbeam (2007), P availability is classified in general terms (i) as solution P, (ii) as readily available or labile P (labile fraction might include easily mineralizable organic P, low energy sorbed P, and soluble mineral P), (iii) as non-labile P (the non-labile fraction might include resistant organic P, high energy sorbed P, and relatively insoluble P minerals). According to Mayakaduwage et al. (2021) labile P, non-labile P and residual P represented 70%, 15% and 15% in submerged incubation and 40%, 40% and 30% in moist incubation of

ASS, respectively. Only small amounts of P are readily available, since plants take up P from the soil solution. Replenishing P from non-labile fraction is slower than labile fraction.

Most P in soil is precipitated, fixed, or adsorbed on soil minerals. Only a small fraction of the total soil P in the soil solution is accessible by plants, and more than 90% of total P in soil is present as insoluble and adsorbed forms including primary P-minerals, P-humic, secondary P-minerals including Ca (Ca-P), Fe (Fe-P), and Al (Al-P) and P adsorbed by hydrous oxides and silicate minerals (Mengel and Kirkby, 2001). Phosphorus availability in acid soils is determined by P interaction with Fe-oxides which are influenced by soil water content (Mayakaduwege et al., 2021). Phosphorus is uptaken by plants from soil solution as H_2PO_4^- , HPO_4^{2-} or PO_4^{3-} , depending upon soil pH. Phosphorus uptake by plant from the soil solution alters the equilibrium in soil solution, thus P moves from the less available pools towards plant-available pools. Phosphorus availability depends on the concentration of P in the soil solution and the ability of the soil to replenish the soil solution P from the less labile pools, adsorbed P on the surface of soil particles, P that is precipitated as primary P minerals and secondary P minerals such as Al/Fe-P.

Phosphorus form in soil solution is dynamically undergo an equilibrium, labile and non-labile forms in response to changes in the relative concentration of P in the various pools driven by plant uptake and P applications. The labile forms of P include easily mineralizable organic P, the relatively soluble forms of precipitated P and the adsorbed P that is readily exchangeable. Whereas the non-labile P form include the strongly adsorbed form and less soluble form.

PHOSPHORUS SOLUBILITY IN ACID SULFATE SOILS

Phosphorus solubility in ASS is very low to moderate, while the total P levels is very high (Purnomo et al., 2005), this condition is actually based on soil test methods used to determine P concentration in soil. Phosphorus availability in ASS is usually limited due to strongly absorbed by soil constituents. Fahmi et al. (2005) reported more than 80% of applied P in ASS was adsorbed by soil constituents (Figure 2). Available P concentration in ASS range 12.6 to 19.3 mg kg^{-1} . Mayakaduwege et al. (2021) stated form of P in ASS representing 70% as readily available or labile P, 15% as non-labile P and 15% as residual P in submerged incubation and 40% as readily available or labile P, 40% as non-labile and 30% as residual P in moist incubation.

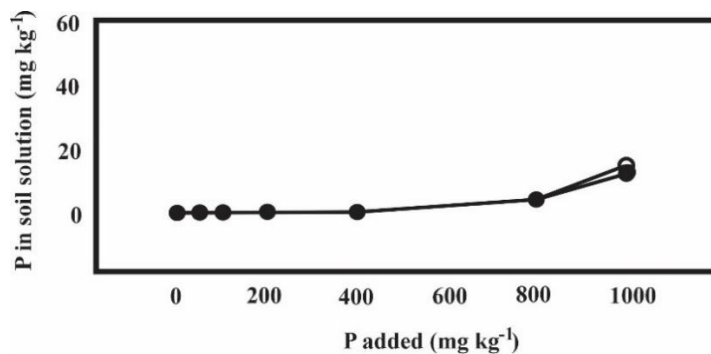


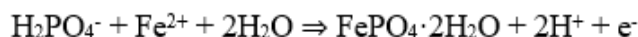
Figure 2. Phosphorus adsorption in acid sulfate soils (Fahmi et al., 2005).

Phosphorus adsorption

Sharma et al. (2013) stated that 75%-90% of applied P is precipitated, it is adsorbed to anion exchange sites of Fe and Al, particularly amorphous-Fe in an anaerobic environment (Li et al., 2012). Phosphorus adsorption by soil constituents occurs only when added P in amendment has higher concentration than in soil solution. Phosphorus adsorption is a term used to describe all the processes resulting in the removal of P from soil solution, mainly by surface adsorption and precipitation. While occluded P is a term used to

describe P physically encapsulated by soil minerals which have no P in their structure (Fenton, 2022). According Blombäck et al. (2021) P availability as well as the soil solution concentration of P will depend on the degree of P saturation, rather than on the total P content. Phosphorus saturation is the proportion of adsorption sites occupied by P, or as the ratio between adsorbed P and the P adsorption capacity of the soil. Phosphorus is strongly adsorbed by number of adsorption site of soil constituents, which greatly varies among soils. The process of P adsorption by Fe-oxides are known as specific adsorption. Specific adsorption can occur on uncharged adsorbents and even on a surface with the same charge as the adsorbent. Phosphorus can be adsorbed on variable-charge minerals such as Fe-oxides even at alkaline pH (adsorbents are negatively charged). According to Asomaning (2020) specific adsorption is characterized by formation of inner-sphere complexes, where no water molecules are interposed between adsorbate and adsorbent.

Forms of Fe-P mineral are commonly found in wetland soil such as ASS, i.e., strengite (FePO₄) and vivianite (Fe₃(PO₄)₂). Phosphorus adsorption on Fe-hydroxides occurs through inner-sphere complex formation or by surface precipitation (Krumina et al., 2016; Gypser et al., 2021). Reddy and DeLaune (2008) showed a reaction between Fe(II) and H₂PO₄ causing the deposition of PO₄³⁻ and Fe(III) ions in the form of strengite. Researchers revealed complex formation of P with goethite is outer or inner-sphere complexes (Boukemara et al., 2016) or bidentate complexes (Ahmed et al., 2019). In addition, P formed bidentate-binuclear complexes with ferrihydrite (Wang et al., 2017).



Phosphorus desorption from Fe-P pools

Phosphorous cycle is often closely coupled with Fe (Li et al., 2012) (Figure 3). In ASS, Al and Fe are major soil constituents which regulate P solubility. Phosphorus mainly adsorbed by Al/Fe-oxides and hydroxides (insoluble P minerals) to form Fe-P, they are sensitive to alteration of soil pH. These minerals are stable under acidic soil, but their solubility decrease with increasing of soil pH (Fahmi et al., 2018). In addition, Fe-P minerals are also greatly sensitive soil redox potential (Eh). According to Reddy and DeLaune (2008) Fe minerals such as Fe(III)-hydroxide, Fe(III)-oxyhydroxide, Fe(III)-hydroxide, and amorphous oxides are known determine P solubility in wetland soils dominated by these minerals. As a result of waterlogged conditions, reduction of hydrated Fe(III)-oxide to more soluble Fe(II)-hydroxide results in the desorption of occluded P.

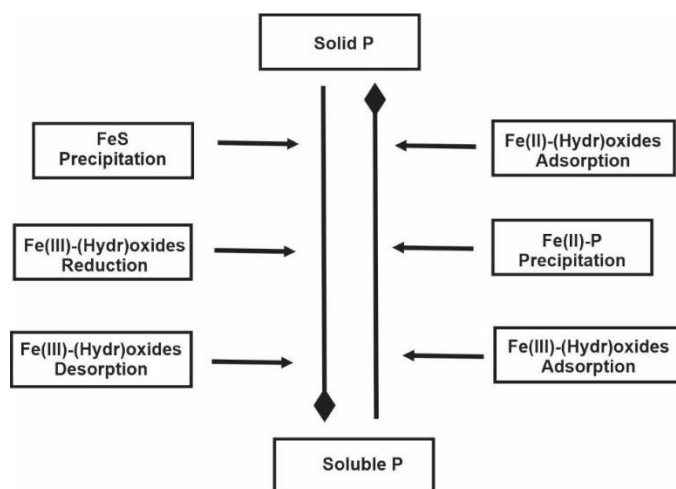


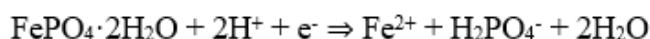
Figure 3. Relationships between the Fe(III)- Fe(II) redox wheel and P cycling in anaerobic soil (modified from Li et al., 2012).

One of the common Fe-P minerals found in ASS is vivianite. Vivianite is formed in particular under Fe-rich condition, reductive conditions (anaerobic), abundant in organic material and most notably in soil where

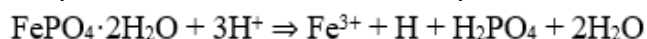
P availability was increased anthropogenically (Rothe et al., 2016). Under anoxic conditions, reductive dissolution of Fe(III)-oxides results in the concurrent release of P and Fe which may initiate precipitation of Fe(II)-P phases, such as vivianite (Walpersdorf et al., 2013). Vivianite solubility is strongly influenced by soil pH, Eh and Fe concentration. According to Rouzies and Millet (1993) vivianite is stable when Fe(III) concentration reaches 50% of the total Fe. Li et al. (2012) reported that precipitation of Fe(III)-P is crystallized to vivianite if ratio of Fe(II):P >1.5. Ratio of Fe:P > 15 caused a decrease in amount of P released.

Under oxic conditions Fe(III)-oxides have a large capacity to co-precipitate and/or adsorb P. In addition, organic matter application on soils in rice cultivation system enhanced vivianite formation due to high Fe(III) reduction rates to Fe(II) and released P as reported previously by Heiberg et al. (2012). Vivianite may constitute a significant sink for P (O'Connell et al., 2015). However, not all of P adsorbed by Fe is in insoluble or classified as unavailable form. Since the ASS is waterlogged, Eh decreased, thus solubilize Fe-oxides and release adsorbed P (Wang et al., 2013; Wisawapipat et al., 2017).

In ASS, groundwater level fluctuation influence on P solubility. In waterlogged condition, P concentration increase and *vice versa* (Fahmi et al., 2018). Increasing P solubility in wetland soil is correlated with reduction process of Fe(III)-P. The occluded Fe(III)-P is unavailable for plants until it has been reduced to more soluble Fe(II)-P (Reddy and DeLaune, 2008). Reduction of Fe(III) to Fe(II) provides a mechanism for availability P, and anaerobic soils potentially exhibiting a higher capacity to bind P than aerobic soils (Li et al., 2012). Although reduction of Fe(III) to Fe(II) creates a larger surface sites for P, those sites have a lower bonding energy than do the smaller number of available sites in the oxidized soil. Thus, a reduced soil will adsorb a large amount of P with a low bonding energy (desorption potential is high), whereas an oxidized soil will adsorb less P with high bonding energy (desorption potential is low) (Reddy and DeLaune, 2008).



Phosphorous availability strongly depends on interactions between P and Fe-oxyhydroxides as well as water content of soil, soil organic matter, soil pH and Fe-oxyhydroxides concentration (Wisawapipat et al., 2017; Gu et al., 2019). Liang et al. (2010) reported that higher P released occurred in very acidic of soil condition compared to basic and slightly acidic condition. Hairani and Susilawati (2013) reported higher P availability on actual ASS compared potential ASS, regarding more higher Fe concentration in actual ASS than potential ASS, despite soil pH of actual ASS was lower than potential ASS.



The solubility of P in ASS is also determined by the crystallinity of Fe mineral. Based on crystallinity phase of Fe-oxyhydroxides, amorphous-Fe forms have a significantly greater P retention capacity compared to crystalline Fe (Yan et al., 2014). Reduction of the insoluble Fe(III)-oxyhydroxide compounds to Fe(II)-oxyhydroxide compounds (more soluble) provides surfaces for P retention (Williams and Patrick Jr., 1973). Acid sulfate soils dominated by amorphous-Fe minerals contain more higher soluble P than ASS dominated by Fe-crystalline minerals. According to Krumina et al. (2016) different types of Fe-oxyhydroxide are the main factors that determine the availability of P. Wang et al. (2013) reported that amorphous-Fe such as ferrihydrite has an important role in the dissolution of P in wetlands, especially in soils with low P content.

AVAILABLE PHOSPHORUS IN RICE FIELD ACID SULFATE SOILS

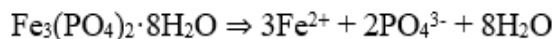
A quick and practical way to improve low P availability in ASS is fertilizer application. Applied fertilizers can be either organic or inorganic. The dosage of P fertilizer for rice in ASS ranges 15-23 kg ha⁻¹ (Fahmi et al., 2018). However, not all fertilizers applied to the soil are available for plants. The application of P fertilizer alters the equilibrium in the soil solution (Chen et al., 2022). Most of the applied P to ASS was adsorbed by soil constituents (Figure 2). In addition, the increasing in soil organic material is also affects the formation process of minerals such as Fe. Organic materials will interfere the crystallization process of

Fe (Wilson et al., 2013) and determine rate of reduction processes Fe(III) (Luo et al., 2018). Furthermore, it will affect the absorption and desorption of P by Fe.

The major limitation of ASS is significant amounts of P fertilizer become unavailable to rice plant due to high P adsorption capacity (Fahmi et al., 2005). However, there was good performance of rice growth even without fertilizer on ASS in tidal swampland as reported by Purnomo et al. (2005). Phosphorus sufficiency for rice plant on soil with high Fe content is associated with rice cultivation system in ASS which has implemented a good water management system and soil amelioration. In this system, soil undergo periodically waterlogging and drying. Labile and non-labile P pools were up to twofold higher in waterlogged condition than in moist condition (Mayakaduwege et al., 2020b). Khan et al. (2022) demonstrated that subjecting soil to drying and flooding cycles increased P availability, thus increased P uptake and DM yields of plant. The increase in P availability due to soil waterlogging is related with many process such as the reduction process of Fe(III)-P to Fe(II)-P and vivianite desorption (Heiberg et al., 2012; Gypser and Freese, 2020).

Sufficiency P for rice plants on soil with high Fe content such as ASS is associated with crystallinity phase of Fe, majority of Fe in these soils are dominated by amorphous form. In redoxomorphic soils, Fe may act as sinks for P (Gasparatos et al., 2019). Adsorbed P by amorphous-Fe is soluble due to amorphous-Fe minerals are very sensitive to redox fluctuations as a result of the changes in soil moisture or water management in rice cultivation. This fact shows that although most of the applied P is directly adsorbed by the soil constituents such as Fe, but P has the potential to be released into the soil solution via desorption process, meaning “the Fe-P pools may become a source of P for rice plants”.

Phosphorus adsorption is a fast and reversible reaction (Barrow, 2006), this condition relates with stability of this mineral that is influenced by soil pH, Eh and soil moisture. Nanzyo et al. (2013) stated that formation and desorption of vivianite crystals could be repeated each year, depending on the water management system employed in the plow layer soil. Desorption of P from vivianite may be related with soil waterlogging during rice cultivation. These results confirm that vivianite may act as native source of P in rice cultivation as shown in following equation:



Part of ASS are located in the zones that frequently flood and dry alternately. In this zone, ASS contain more higher amorphous-Fe than crystalline, whereas ASS located in drier land are dominated by Fe-crystalline (Fahmi et al., 2018). This can be attributed to increase in amorphous-Fe content on the soil surface after waterlogged soil or soil that frequently waterlogged and drained alternately. It has been reported that drying of soils increases crystallization of amorphous-Fe-oxides (Johnston et al., 2009), thus decreasing their solubility and P sorption affinity (Reddy and DeLaune, 2008). Winkler et al. (2018) found that long-term redox fluctuations associated with paddy cultivation can alter Fe crystallinity phase and reactivity. This fact was confirmed by Hairani and Susilawati (2013) and Fahmi et al. (2018), who reported that the availability of P in ASS from zones that frequently waterlogged and dry alternately was lower than in drier ASS. Mayakaduwege et al. (2020a) also reported that labile P fraction in the soil decreased due to adsorption by amorphous-Fe as a result of the changes in soil moisture.

CONCLUSIONS

The P chemistry of wetland soil is different from upland soil. Waterlogging and drying cycles imposed during rice cultivation on soil with high Fe content such as acid sulfate soils (ASS) increase ratio of amorphous-Fe:crystalline-Fe. Soil waterlogging in rice cultivation on ASS increases Fe(II) concentration. Although Fe(II) is able to adsorb large amount of P, but it adsorbed with low bonding energy, thus P will be easily re-released into the soil solution and available for rice plants. In addition, reduction of Fe(III)-P to Fe(II)-P results in the release of adsorbed P. Most of Fe-P pools (especially amorphous-Fe-P) in ASS used for rice cultivation can be a source of P, while providing available P stocks for plants or becoming a P sink when P concentration is high in the soil solution. The Fe-P pools may be categorized as readily available or labile P or low energy sorbed P fraction for rice cultivation in ASS.

Author contribution

Conceptualization: A.F. Methodology: M.A. Data curation: A.H. Writing-original draft: A.F., A.H., M.A., S.N. Writing-review & editing: A.F. Visualization: S.N. All co-authors reviewed the final version and approved the manuscript before submission.

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