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ORIGINAL PAPER

Phosphorus Release and Transformations in Contrasting Tropical Paddy Soils Under Fertiliser Application

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Abstract

Purpose Inconsistent yield responses to inorganic phosphorus (P) fertilisers in tropical rice paddy soils remain a challenge. This study investigated the contributions of applied P fertilisers to soluble soil P and P transformation mechanisms in P-added paddy soils.

Methods An incubation study was conducted on three rice-growing soils (Ultisol, Alfisol, and Entisol) in Sri Lanka with and without single superphosphate (SSP), triple superphosphate (TSP), and urea. Dissolved reactive phosphorus (DRP) was measured over 112 days of submergence. Thermodynamic modelling and chemical P fractionation were employed to assess soil P transformations.

Results Phosphorus-fertilised soils had significantly higher DRP concentrations (1.1–8.0 mg L^{-1}) compared to controls at 7 days after submergence but DRP declined beyond 21 days (0.024–0.300 mg L^{-1}). Single superphosphate increased DRP more than TSP, short-term. Urea did not affect DRP concentration. Ultisols exhibited the lowest DRP, while Alfisols maintained higher DRP than Ultisol which was near or above the critical concentration for rice (0.1 mg L^{-1}) after 28 days. In Entisol, only SSP maintained DRP above 0.1 mg L^{-1}. Modelling suggested Ca phosphates and Fe oxy(hydr)oxides dissolved during submergence. Released P may be resorbed by Fe/Al oxy(hydr)oxides and Ca minerals, with evidence of downward movement of dissolved P and its resorption onto Fe/Al and Ca minerals possibly due to saturation of P sorption sites in the topsoil layer.

Conclusion Low dissolved P in porewater may be linked to inherent soil characteristics, including low organic matter and high amorphous Fe and Al oxides.

Highlights

- Ultisols exhibited the lowest dissolved reactive P (DRP) concentrations in porewater.
- Alfisols showed a slight increase in DRP over time, remaining at or above the critical concentration (0.1 mg L^{-1}) for rice growth.
- Only the single superphosphate added treatment in Entisol exceeded the 0.1 mg L^{-1} threshold.
- Calcium phosphates and Fe(III) oxy(hydr)oxide dissolved and released P to porewater during the submergence.
- The dissolved P leached and resorbed onto amorphous and crystalline Fe/Al oxy(hydr)oxides and Ca minerals.

Keywords Calcium phosphates · Dissolved reactive phosphorus · Fe/Al oxy(hydr)oxides · P fractionation · Tropical paddy soils

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1 Introduction

Global natural phosphorus (P) reserves, specially rock phosphate deposits used for producing inorganic P fertilisers, are limited in both quantity and distribution (Cordell and White 2013; Roberts and Johnston 2015). Approximately 85% of the global rock phosphate reserves are concentrated in only five countries: Morocco (70%), China (5%), Egypt (4%) Algeria (3%), and Syria (3%) (Brownlie et al. 2023). To address the deficiency of plant available P concentration in many agricultural soils, resulting from the continuous removal of P at harvest and by the interactions with soil surfaces, P fertiliser is applied to soils in either inorganic or organic forms (Dougherty et al. 2004). In general, the efficiency of inorganic P fertilisers is low, with estimations suggesting that around 70% of the P fertiliser applied to soils annually are not utilised by crops (Brownlie et al. 2021). Therefore, understanding P transformations in soil is important for efficient management of limited P resources.

Phosphorus release due to submergence is limited in highly weathered tropical soils because of their high P sorption potential (Andriamananjara et al. 2016; Nishigaki et al. 2019; Pierzynski and Hettiarachchi 2018; Rakotoson et al. 2015). This limitation is primarily due to inherent characteristics of soils, such as high concentrations of Fe and Al oxy(hydr)oxides and highly weathered kaolinitic (1:1) clay minerals, which have a high affinity to P (Gérard 2016; Pierzynski and Hettiarachchi 2018). These inherent soil characteristics could be contributing to the poor/unclear yield response to applied P fertilisers in tropical soils, including those found in rice-growing soils in Sri Lanka (Dobermann et al. 2003; Kendaragama et al. 2003; Kodagoda et al. 2022; Kulasinghe et al. 2020; Nishigaki et al. 2021; Palihakkara et al. 2024; Sirisena and Suriyagoda 2018). However, there is a lack of studies offering empirical evidence regarding P release and transformations in fertiliser applied tropical weathered rice paddy soils under submergence.

Phosphate transformation under submerged conditions differs from that observed in aerobic soil conditions. In submerged conditions, the anaerobic microorganisms become more dominant than aerobic microbes (Fageria et al. 2011), which alters the P transformation dynamics. The anaerobic microbes use electron acceptors for their respiration in sequential order of NO_3^-, Mn^{4+}, Fe^{3+}, SO_4^{2-} and CO_2 (Marschner 2021; Sahrawat 2012). Most commonly, microbes do not use less energetically favourable electron acceptors, such as Fe and Mn, in the presence of NO_3^-. Therefore, the presence of NO_3^- in soil solution can buffer or delay the microbially mediated P release under submergence through reductive dissolution of Fe/Mn oxy(hydr)oxides (Smith et al. 2021). Application of urea as a source of nitrogen (N) fertiliser in rice paddy cultivation increases NO_3^- ions in soil solution via hydrolysis of urea followed by nitrification in the oxidised soil–water interface. However, previous studies have not considered the factor of NO_3^- as an important variable that could intervene P transformation in submerged paddy soils.

The cultivation of rice under submerged conditions can influence the dynamics of P in soils (Huang et al. 2014; Jia et al. 2018) by altering soil properties (Ponnamperuma 1972). For example, under submergence, soil pH typically shifts towards the neutral range (Fageria et al. 2011). In acidic soils, pH increases primarily due to the reductive dissolution of Fe(III) and Mn(IV)/Mn(III) minerals, leading to the release of P (Lu et al. 2022). Conversely, in alkaline soils, pH decreases due to decomposition of organic matter (Fageria et al. 2011), which causes Ca and Mg minerals to dissolve, releasing P into the porewater (Jayarathne et al. 2016). In addition to pH, changes in other soil properties under submergence include redox potential, P sorption capacity, organic matter availability, and the crystallinity of minerals which play a significant role in P dynamics (Maranguit et al. 2017; Marschner 2021; Nishigaki et al. 2021). Due to the complex interactions between hydrological and biochemical processes, understanding the dynamics of soil P under submergence has proven challenging. Previous attempts to pinpoint the underlying reasons for inconsistencies in yield responses for different P treatments by examining individual soil properties have been unsuccessful (Kendaragama et al. 2003; Kulasinghe et al. 2020; Sirisena et al. 2013). Therefore, a mechanistic understanding of P dynamics of paddy soils under submergence is necessary to address this unexplained phenomenon.

Triple superphosphate (TSP) is the most used P fertliser in Sri Lanka (De Silva et al. 2019) due to its high P content and rapid solubility. However, this study also considered the use of single superphosphate (SSP) for several reasons. Single superphosphate contains a lower concentration of P compared to TSP, making it a more suitable option for farmers, particularly in regions where P deficiencies are less pronounced but over-application of P fertiliser prevails, such as in Sri Lanka. In fact, a study comparing TSP and locally produced SSP found similar yield responses when applied to paddy soils (Udawatte et al. 2020), suggesting that SSP can be an effective alternative to TSP under certain conditions. Additionally, the use of SSP, which has higher calcium content (21%) compared to TSP (15%), can influence P speciation in the soil differently than TSP due to its calcium content. Thus, the inclusion of both SSP and TSP in this study allows for a comprehensive comparison of their effects on P dynamics, availability, and soil nutrient management in paddy soils in the Sri Lankan context.

The objectives of this study were to investigate release and transformations of P in three contrasting rice paddy soils fertilised with either TSP or SSP, and to assess how

the application of urea influences P release and transformations in the P fertilised soils under submerged conditions. We hypothesised that the reductive dissolution of Fe/Mn oxy(hydr)oxides during submergence would release sorbed P, consequently elevating the P concentration in the soil solution. In addition, we hypothesised that urea application would delay the reductive dissolution of Fe/Mn oxy(hydr) oxides by increasing the NO_3^- concentration in soils, thereby impairing P release through reductive dissolution.

2 Materials and Methods

2.1 Soils Used in the Incubation Study

The present incubation study used three rice-growing soils with contrasting properties collected from three locations in Sri Lanka, namely Hanguranketha (N 7° 08′ 23.57″, E 80° 47′ 34.16″), Mahailluppallama (N 8° 05′ 59.2″, E 80° 26′ 33.2″), and Aranaganwila (N 7° 46′ 30.1″, E 81° 09′ 55.0″). Bulk soil samples from 0 to 30 cm depth of the three locations were classified according to the US Soil Taxonomy as Typic Rhodudults (Ultisol), Typic Haplustalfs (Alfisol), and Typic Udipsamments (Entisol), respectively. The soil samples were air-dried, passed through a 2 mm sieve, homogenised, and stored at room temperature until further analysis. A sub-sample of each soil was analysed for basic soil properties. Soil pH and electrical conductivity were determined in 1:2.5 soil to distilled water suspension using a pH meter (Hanna HI2020 - 02) and a conductivity meter (Eutech COND 6 +), respectively. Organic carbon was determined by Walkey and Black method (Nelson and Sommers 1996). Available P was extracted by Olsen (Olsen and Sommers 1982) and Mehlich- 3 (Mehlich 1984) methods. Water extractable P was extracted by the method introduced by Self-Davis et al. (2000). Phosphorus in the extracts were determined by the molybdate blue colour method by Murphy and Riley (1962). Phosphorus sorption capacity (PSC) was determined by the single point P sorption method by Bache and Williams (1971). Cation exchange capacity (CEC) was determined by the unbuffered salt extraction method (Sumner and Miller 1996). Bulk density was determined by the core method (Grossman and Reinsch 2002). Oxalate extractable Al and Fe were determined using the acid ammonium oxalate extractant in the dark (Jackson and Lim 1986). Mehlich- 3 extractable K and Na concentrations were measured using flame emission spectrophotometer (Jenway PFP 7) and Mehlich- 3 extractable Ca and Mg concentrations were measured using atomic absorption spectrophotometer (Shimadzu AA- 6200). Soil texture was determined by the pipette method (Gee and Bauder 1986). The X-ray diffraction (XRD) analysis was performed for the initial soil samples of the three soils to identify mineralogical compositions of the clay fractions. The XRD intensities were recorded as a function of 2θ operated at a potential of 40 kV and 34 mA producing Cu Kα radiation (λ = 1.5406 Å).

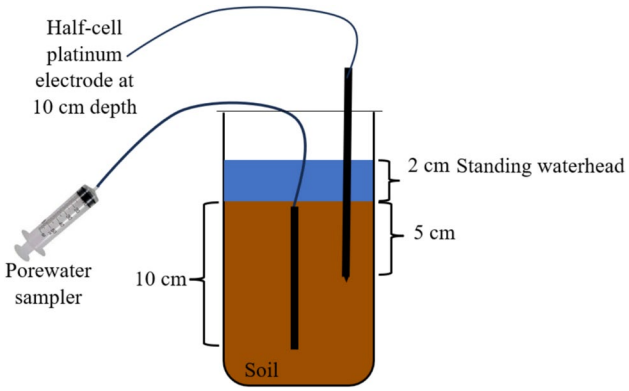

Fig. 1 A schematic diagram of a control treatment used in the incubation study. The diagram is not to scale

2.2 Experimental Design and Treatments

Plastic incubation vessels (1.5 L capacity) were acid cleaned (2% HNO_3) and filled with homogenised soil up to a height of 11 cm (approximately 1.2 kg for Ultisol, 1.3 kg for Alfisol, and 1.4 kg for Entisol). The vessels were covered with a dark paper to control photochemical reactions. Soils were saturated with reverse osmosis (RO) water and a 2 cm waterhead was maintained throughout the experimental period. Porewater samplers (Rhizon-flex – 10 cm in length) were inserted vertically in the middle of each bottle (Fig. 1). A platinum (Pt) electrode was permanently installed at the 5 cm depth from the surface of the soils in control soils (no P and no N fertilisers) for redox potential (Eh) measurements. The vessels were placed under room temperature at 30 ± 5 ^{o}C.

Triple superphosphate, SSP and control (no P fertiliser) with and without urea were used in this experiment to formulate six treatments. The P fertilisers were applied at a rate of 3.2 g P m^{-2} (equivalent to 32 kg P ha^{-1}), 6 days after submergence. Two split applications of urea were applied at the rates of 2.8 and 2.3 g N m^{-2} (equivalent to 28 and 23 kg N ha^{-1}, respectively) at 20 and 48 days after submergence (DASub), respectively. The three fertilisers were applied to the soil surface in granular form, mimicking the farmer practices, and were of fertiliser grade. The time of fertiliser application was decided based on the fertiliser recommendation for rice by the Rice Research and Development Institute, Sri Lanka (RRDI 2013). The P rate was three times higher, and the N rates were 50% lower than the recommended rates of the Rice Research and Development Institute, Sri Lanka (RRDI 2013). A higher P rate was employed

to define treatment effects and elucidate P transformations, while a lower N concentration was used to offset plant N uptake in the field. Although the Rice Research and Development Institute recommends applying urea in four split applications, this experiment adopted two split applications to align with typical farmer practices. The experimental setup was maintained for a duration of 112 days. Four replicates were maintained for each treatment, with one specifically designated for destructive harvesting at 35 DASub for P fractionation analysis.

2.3 Soil and Water Sampling and Analysis

Soil porewater was collected from three replicates, weekly, for the first 70 days and then biweekly for the rest of the 42 days using Rhizon-flex samplers, by applying a vacuum using a 50 ml syringe. The porewater samples were filtered through 0.45 µm nylon membrane filters and analysed for DRP by molybdate blue colour method (Murphy and Riley 1962) using UV visible spectrophotometer (Shimadzu UV- 1900i) within 12 h after sampling. The pH of porewater samples were measured using a glass pH electrode. The soil Eh was measured at each porewater sampling event using a calomel reference electrode (Hanna HI5412). The voltmeter reading was corrected to the standard hydrogen electrode potential by adding + 250 mV. Cation concentrations (Ca, Mg, K, Na, Fe, Al, and Mn) of porewater were measured at 7, 21, 35, and 70 DASub using inductively coupled plasma optical emission spectroscopy (iCAP 7000 series ICP-OES). Porewater anion concentrations (Cl^-, F^-, NO_3^-, and SO_4^{2-}) were measured at 7, 35, and 70 DASub using ion chromatography (Metrohm 930 Compact IC Flex). Dissolved organic carbon concentrations were measured at 7, 35, and 70 DASub using a carbon nitrogen analyser (Skalar Primacs SNC 100-IC). Alkalinity of the porewater samples was measured by titrating against 0.1 N H_2SO_4 in the presence of methyl orange indicator.

2.4 Soil Sampling and Analysis for Phosphorus Fractionation

Phosphorus fractionation was performed using a modified Hedley procedure (Hedley et al. 1982) for the soils collected at 0–2, 2–4 and 4–6 cm depths at 35 and 112 DASub. The soil samples were purged with argon gas, sealed, and stored at − 28 ^{0}C until P fractionation analyses were performed. An aliquot of soil (approximately 0.5 g) was subjected to sequential extraction using 30 mL of each of the following solutions in the specified order: distilled water, 0.5 M $NaHCO_3$, 0.1 M NaOH, and 1.0 M HCl. The suspension was shaken for 16 h, centrifuged for 10 min at 7500 rpm, and the supernatant was passed through a 0.45 µm nylon membrane filter for each extraction step. The first two extraction steps (using distilled water and 0.5 M $NaHCO_3$) were performed while purging argon gas to have minimum contact with oxygen. A portion of the $NaHCO_3$ and NaOH extracts was acidified using concentrated H_2SO_4 to precipitate extracted organic matter and the supernatant was analysed for inorganic P (Pi), while a portion of H_2O extract was directly analysed for Pi without acidification. Another 10 mL of H_2O, $NaHCO_3$, NaOH, and HCl extracts was digested in a digestion block at 150 ^{0}C for 40–60 min until the volume of the digestion tube reduced to 2 mL. Acidified potassium persulfate oxidation was used for the digestion and the residue was analysed for total P (TP). The difference between TP and Pi was estimated to be organic P (Po) (Tiessen and Moir 1993). The P in the soil residue left after sequential extraction was determined after digestion with aqua regia (Chen and Ma 2001).

Phosphorus fractions analysed in this sequential analysis were divided into three broad categories as labile, moderately labile and stable fractions. Organic and inorganic P fractions extracted by water and 0.5 M $NaHCO_3$ are labile fractions which are readily plant available. Organic and inorganic P extracted by 0.1 M NaOH are moderately labile P which are potentially bound to Fe/Al oxy(hydr) oxides. Inorganic P extracted by 1.0 M HCl are also moderately labile P which are likely bound to Ca (Niederberger et al. 2019). Residual P is the stable P fraction which is bound to primary and secondary P minerals.

2.5 Thermodynamic Modelling

To predict the solid P species in porewater samples at 7, 35 and 70 DASub, the thermodynamic modelling software Visual MINTEQ 3.1 (Gustafsson 2013) was used. The input parameters entered in the software were soil Eh, pH, and concentrations of cations (Ca, Mg, Mn, and Fe), anions (Cl^-, F^-, NO_3^-, and SO_4^{2-}), DOC, alkalinity and DRP in porewater. The Stockholm Humic Model (SHM) was used as the state-of-the-art model to simulate the complexation to natural organic matter, and it was assumed that fulvic and humic acids were present in similar quantities in porewater.

2.6 Statistical Analysis

Statistical analysis was performed using SAS 9.4 software. Porewater DRP, pH and cations of the three soils were analysed separately using Proc Mixed repeated

measures analysis to compare treatments (N and P fertiliser) over DASub. Compound symmetry was used as the covariate structure of the subject effect. Statistical analysis of porewater cation concentrations (Ca, Fe, Mn, and Mg) was performed using ANOVA with Type 3 Tests of Fixed Effects to evaluate the main effects and interactions of soil type (Ultisol, Alfisol, Entisol), N treatment (with or without urea), P treatment (SSP, TSP and without P) and submerged duration (DASub). Pearson correlation analysis was performed for the three soils to explore relationships between DRP concentrations in porewater and pH, Eh, porewater Fe, Mn, Ca, and Mg concentrations. Principal component analysis was performed using DRP, Eh, pH, and cations (Fe, Mn, Ca and Mg) to identify the factors governing the porewater DRP.

3 Results

3.1 Basic Soil Properties

Ultisols and Entisols had slightly acidic pH (6.81 and 6.15 respectively), whereas Alfisols had a neutral pH (Table 1). Entisol had 94% of sand, whereas Ultisol and Alfisol had sandy loam and sandy clay loam texture, respectively. Entisols had the lowest extractable P (Olsen P: 3.2 mg kg^{-1}) and PSC (431 mg P kg^{-1}) followed by Ultisols (Olsen P: 26 mg kg^{-1}, PSC: 493 mg P kg^{-1}) and Alfisols (Olsen P: 65 mg kg^{-1}, PSC: 506 mg P kg^{-1}). Similarly, Entisol had the lowest extractable Ca, Mg and K concentrations. All soils had low organic carbon contents, which ranged from 3.6 to 9.8 g kg^{-1}. The mineralogical composition of clay

Table 1 Initial selected physiochemical properties of soils

	Ultisol	Alfisol	Entisol
pH	6.81	7.09	6.15
EC (µS cm^{-1})	80.7	29.6	20.8
Sand %	67.7	68.3	93.7
Silt %	17.9	11.3	3.3
Clay %	14.4	20.4	3.0
Textural class	Sandy loam	Sandy clay loam	Sand
OC (g kg^{-1})	7.7	9.8	3.6
CEC ($cmol_{(+)}$ kg^{-1})	8.8	8.0	3.0
Olsen P (mg kg^{-1})	26.0	65.0	3.2
Mehlich- 3 extractable P (mg kg^{-1})	35.6	125.8	5.1
Water extractable P (mg kg^{-1})	3.6	7.2	2.1
$Al_{oxalate}$ (g kg^{-1})	22.6	22.1	9.7
$Fe_{oxalate}$ (g kg^{-1})	340	250	86
Bulk density (g cm^{-3})	1.54	1.55	1.38
P sorption capacity (mg P kg^{-1})	493	506	431
Extractable Ca (mg kg^{-1})	1016	1525	405
Extractable K (mg kg^{-1})	296.1	206.0	16.4
Extractable Na (mg kg^{-1})	0.9	1.0	0.9
Extractable Mg (mg kg^{-1})	19.0	31.9	3.3
Clay mineralogy	Kaolinite (48.8%) Montmorillonite/Illite (32%) Chlorite/Vermiculite (11%) Gibbsite (3.9%) Quartz (traces) As mineral (4.4%)	Kaolinite (9.8%) Montmorillonite/Illite (47.4) Chlorite/Vermiculite (25.5%) Mn silicate (17.2%) Goethite	Kaolinite (39%) Montmorillonite/Illite (34.6%) Gibbsite (6.2%) Quartz (20.2%)

OC: Organic carbon, CEC: Cation exchange capacity, $Al_{oxalate}$: Ammonium oxalate extractable Al, $Fe_{oxalate}$: Ammonium oxalate extractable Fe. pH and EC were measured in 1:2.5 Soil: Distilled water suspension, Ca, K, Na and Mg were extracted by Mehlich- 3 solution. The abundance of the clay minerals in the tested sample is indicated in parenthesis as a percentage

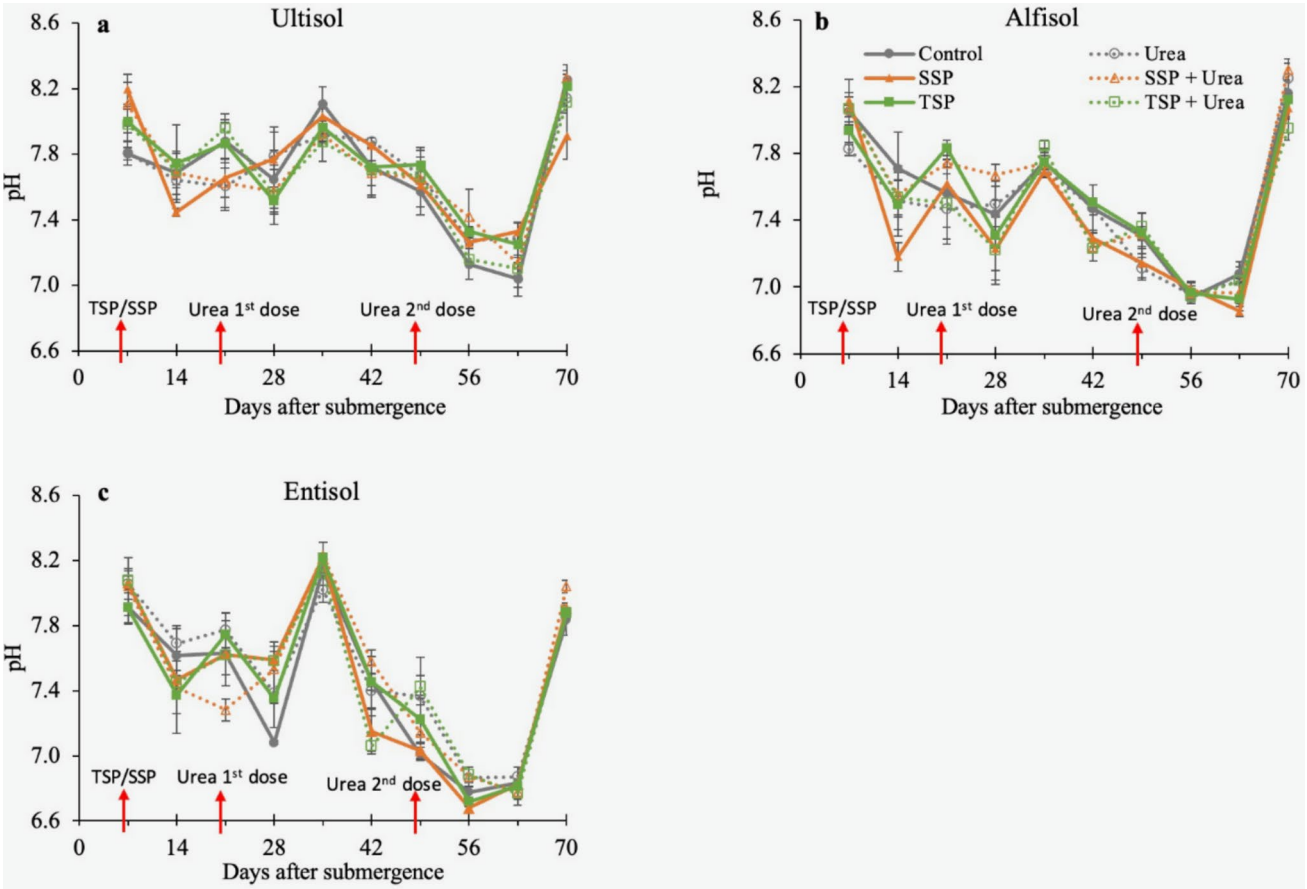

Fig. 2 Variation of porewater pH of Ultisols, Alfisols and Entisols during submergence. The error bars represent the standard errors of the means. The fertiliser application dates (triple superphosphate: TSP, single superphosphate: SSP, and urea) are marked with red arrows on the x-axis. Significant differences are not marked on the graph. A significant decrease in pH ($p < 0.05$) compared to the control was observed in the urea applied Ultisol at 35 days after submergence. Tripple superphosphate and urea treatments in Alfisol and Entisol showed a significant ($p < 0.05$) decrease in pH at 42 days after submergence compared to TSP only treatments. A significant increase in pH ($p < 0.05$) was observed in the urea applied Ultisol compared to the control at 63 days after submergence

fraction of the three soils revealed the presence of kaolinite (1:1 type clay), montmorillonite and illite (2:1 type clay) in all the three soils and presence of chlorite (2:1:1 type clay) vermiculite (2:1 type clay) in Ultisols and Alfisols. Gibbsite was present in Ultisols and Entisols and goethite was present in Alfisols. Further, a manganese silicate (bementite) was present in Alfisols and an arsenate mineral (burgessite) was present in Ultisols.

3.2 Variation in Porewater pH and Soil Eh During Submergence

Overall, in all the soils and treatments, porewater pH decreased during the submergence from ~ 8.0 to ~ 6.5, except at the last sample collection, in which the pH was increased. Urea application inconsistently affected porewater pH for a short time interval (< 1 week): The urea applied Entisol had higher porewater pH compared to the control one day after the second urea application which was at 49 DASub. A significant ($p < 0.05$) lower porewater pH compared to control was observed in urea applied Ultisol at 35 DASub, nearly two weeks after the first split application (Fig. 2a). Further, TSP and urea applied treatments of Alfisol and Entisol showed a significant ($p < 0.05$) decrease in pH at 42 DASub compared to TSP only applied treatments (Fig. 2b and 2c). However, later with time, a significant ($p < 0.05$) increase of pH was observed again at 63 DASub in the urea applied Ultisol compared to the control treatment (Fig. 2a).

The Eh of the three soils decreased over the submergence period. The Eh were 9 (Ultisol), 164 (Alfisol) and 7 mV (Entisol), at 24 h after submergence (Fig. 3a). At the end of 70 days, the Eh values ranged from − 195 to − 247 mV in all three soils indicating a highly reduced condition. Both Ultisol and Entisol soils were anoxic from the beginning of submergence until the end of the experiment. In contrast, Alfisol had relatively higher pe + pH until 28 DASub compared to

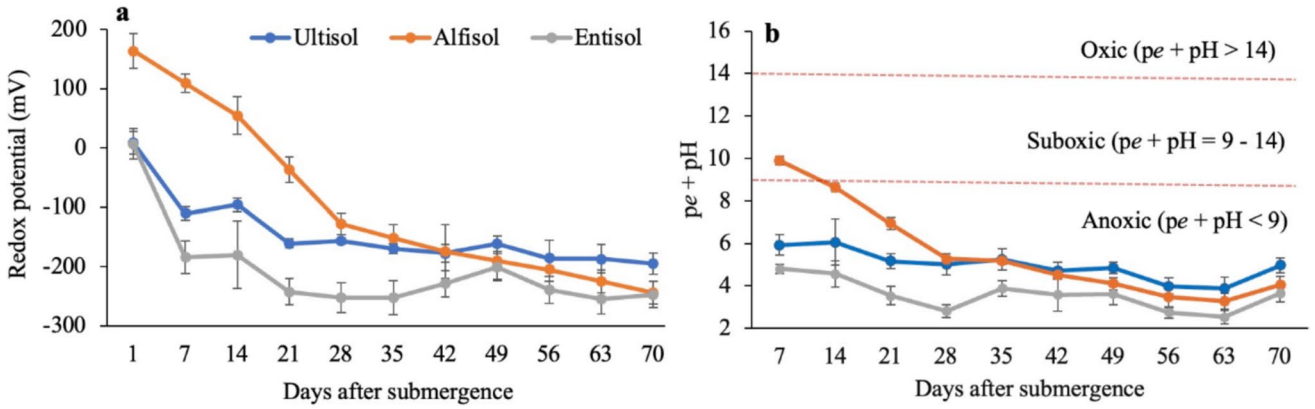

Fig. 3 Variation in redox potentials of the three soils during submergence (**a**) and pe + pH variation of the three soils during submergence. Redox zones are indicated by red colour dash lines; oxic (pe + pH > 14), suboxic (pe + pH = 9–14), anoxic (pe + pH < 9). The error bars represent the standard errors of the means

the other two soils and thereafter it was in the anoxic range. According to XRD results, 17% of the clay fraction of Alfisols was comprised of a Mn mineral. The remaining two soils did not exhibit any indication of the presence of such a mineral. This discrepancy may account for the extended duration Alfisols required to reduce Eh upon submergence compared to Ultisols and Entisols (Fig. 3a).

3.3 Phosphorus Release During Submergence

The DRP release to porewater during submergence was not significantly different among the three soils. The urea treatment did not change DRP concentrations in porewater in all three soils (Supplementary Table S2). Therefore, all six replicates (with and without urea) were combined in each P fertiliser treatment in subsequent analysis (Fig. 4). Results in Fig. 4 shows that application of both P fertiliser treatments significantly increased ($p < 0.05$) the porewater DRP concentration relative to the control only at 7 DASub. In Ultisol, application of the SSP and TSP treatments significantly increased the DRP by 48-fold and 30-fold compared to the control, respectively. Similarly, in Alfisol, the SSP and TSP increased the DRP values by 54-fold and 11-fold relative to the control, respectively. In Entisol, the SSP treatment increased the DRP by threefold compared to the control. However, the TSP treatment did not significantly increase the DRP at 7 DASub in the Entisol. After day- 7, application of SSP and TSP in all three soils did not induce significant changes in DRP concentration compared to the control throughout the experiment.

The porewater DRP concentrations in treatments with P fertilisers decreased significantly ($p < 0.05$) from 1–8 mg L^{-1} at 7 DASub to < 0.10 mg L^{-1} at 14 DASub across all three soils (Fig. 4). Later, around 28 DASub, DRP concentrations slightly increased up to ~ 0.1 mg L^{-1} in all treatments in Alfisols, and P fertiliser applied Entisols (Fig. 4). Ultisols had the lowest DRP concentration throughout the submergence period and it was always below the detection limit (0.024 mg L^{-1}), except on a few occasions in P fertiliser applied treatments, in which the DRP concentrations were slightly (~ 0.04 mg L^{-1}) above the detection limit (Fig. 4).

3.4 Variations of Porewater Fe, Mn, Ca and Mg Concentrations and their Relationship with Redox Potential and pH

The concentrations of porewater Fe, Mn, Ca and Mg during the submergence were significantly ($p < 0.0001$) different among the three soils (Fig. 5). Additionally, the duration of submergence (DASub) significantly influenced porewater cation concentrations, with Fe, Mn, Ca and Mg showing strong time-dependent changes ($p < 0.0001$). Significant interactions between soil type, N fertiliser treatment, and submergence duration were observed for all cations, with the strongest effects on Ca and Mg concentrations ($p < 0.0001$). Marginal effects of P fertilisation were noted for Ca ($p = 0.1172$) and Mg ($p = 0.0429$), while other interactions showed varying degrees of significance across the cations (data is not shown). Correlation analysis conducted to evaluate the relationship among porewater DRP, pH, Eh, and cation concentrations revealed significant correlations between porewater DRP and Ca concentration ($r = 0.45$, $p < 0.01$), and Eh ($r = 0.35$, $p < 0.01$) (Supplementary Table S3).

The principal component (PC) analysis of all three soils reduced the dimensionality of the data set into three

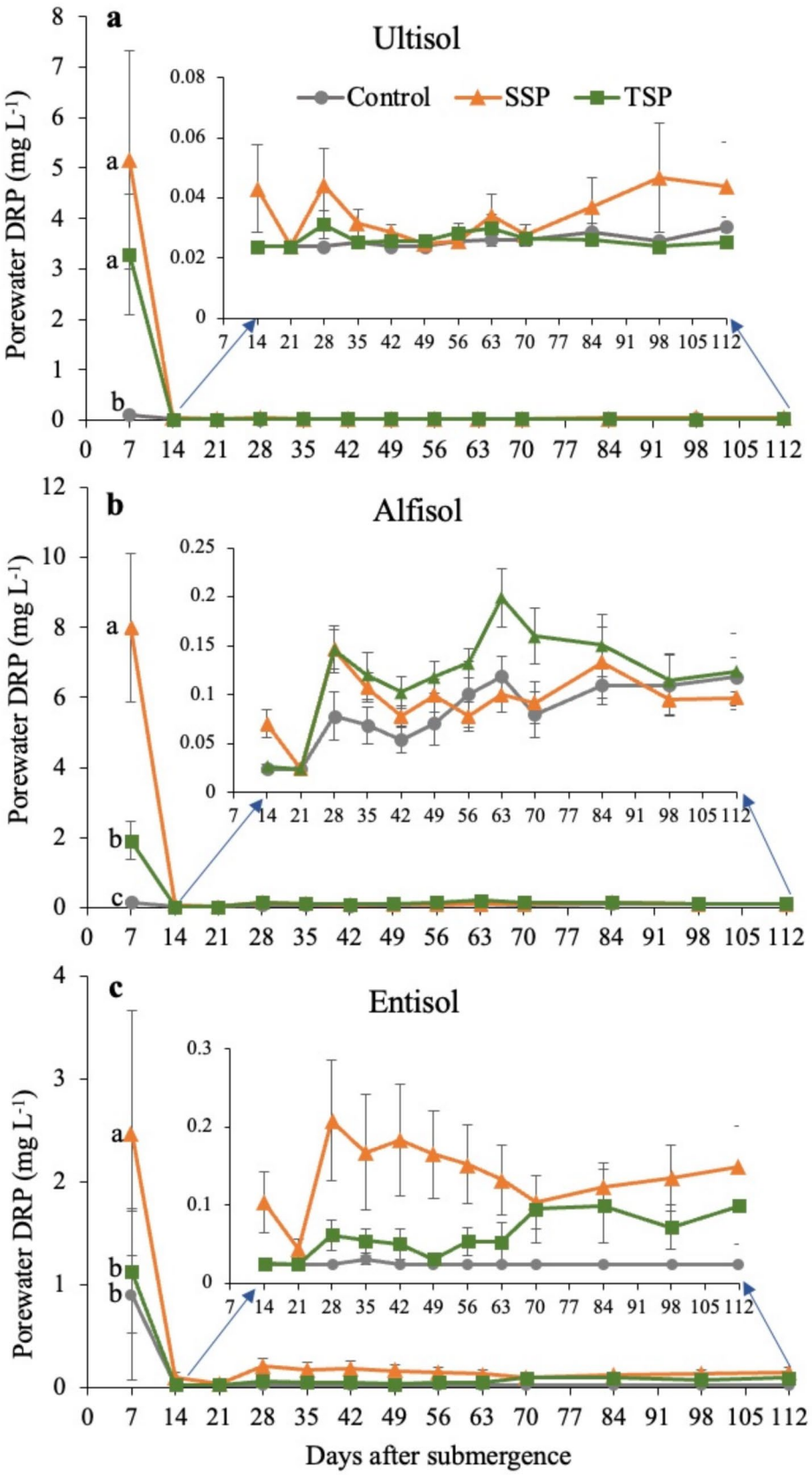

Fig. 4 Variation in dissolved reactive P in porewater of Ultisol (**a**), Alfisol (**b**) and Entisol (**c**) soils during submergence. SSP: Single superphosphate, TSP: Triple superphosphate. The error bars represent the standard errors of the means. Significant differences are indicated only at 7 days after submergence, where there were significant ($p < 0.05$) differences among treatments. The scale after 7 days of submergence is enlarged to better show the differences among treatments. DRP values below the detection limit are marked at the minimum detection limit (0.024 mg L^{-1}) to illustrate the DRP variation pattern

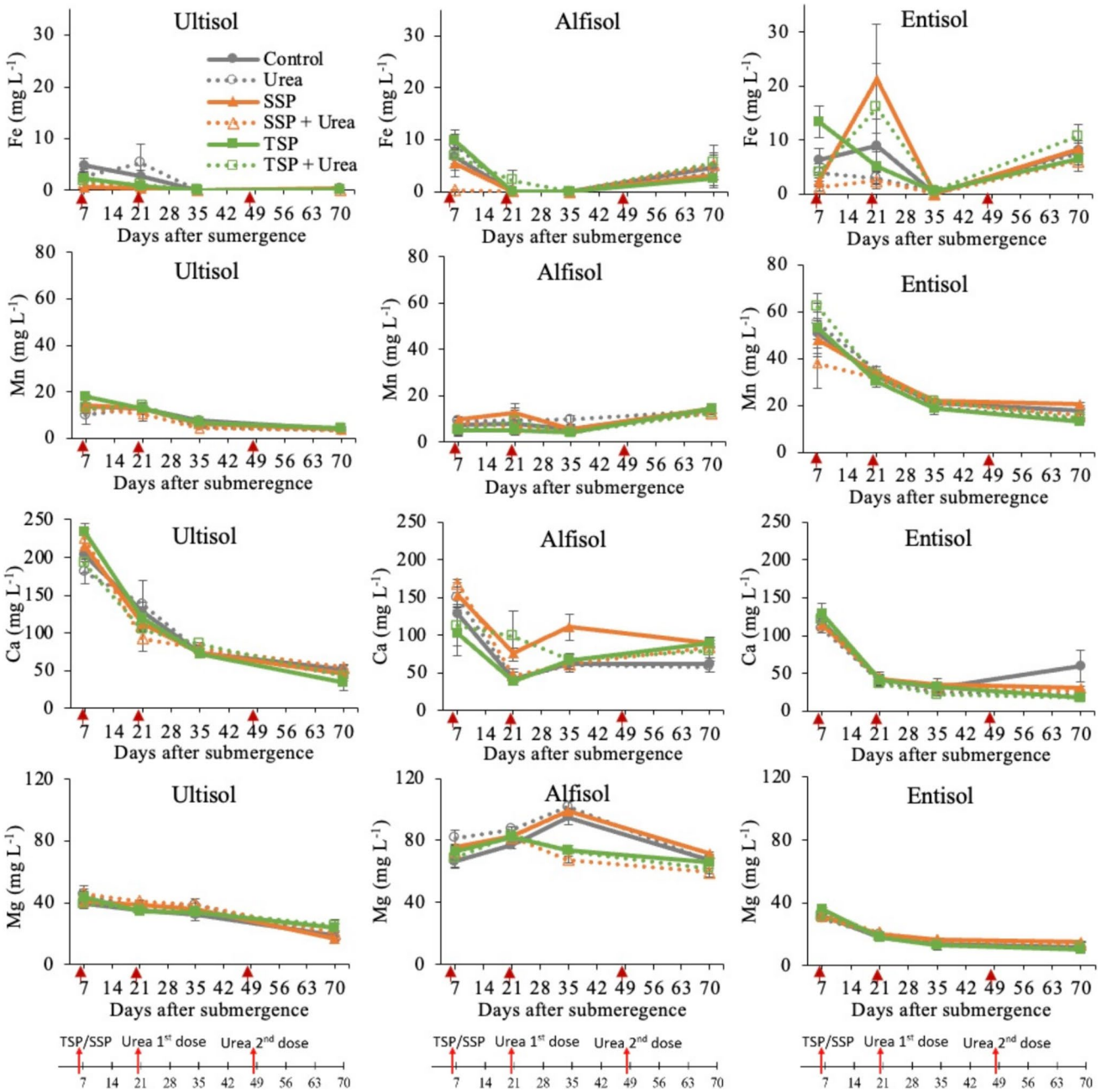

Fig. 5 Variation in porewater Fe, Mn, Ca and Mg concentrations of Ultisol, Alfisol and Entisol soils at 7, 21, 35, and 70 days after submergence. The error bars represent the standard errors of the means. The fertiliser application dates (triple superphosphate: TSP, single superphosphate: SSP, and urea) are marked with red arrows on the x-axis of each graph, with the legend displayed below the x-axis of the bottom set of graphs

principal components (eigenvalue > 1). The variability attributed to PC1, which accounted for 33%, highlighting the relationships among DRP, porewater Ca, Mg, Mn concentrations and Eh (Supplementary Fig. S3). The PC2 attributed to 21% variability of the dataset, elucidated the relationships among DRP, porewater Ca, Fe and Mn.

3.5 Sequential Fractionation of Soil Phosphorus

The changes in P fractions along the three depth categories (0–2, 2–4 and 4–6 cm) of the soil profile at 35 and 112 DASub are illustrated in Fig. 6. The detailed P fractionation results are shown in the supplementary document

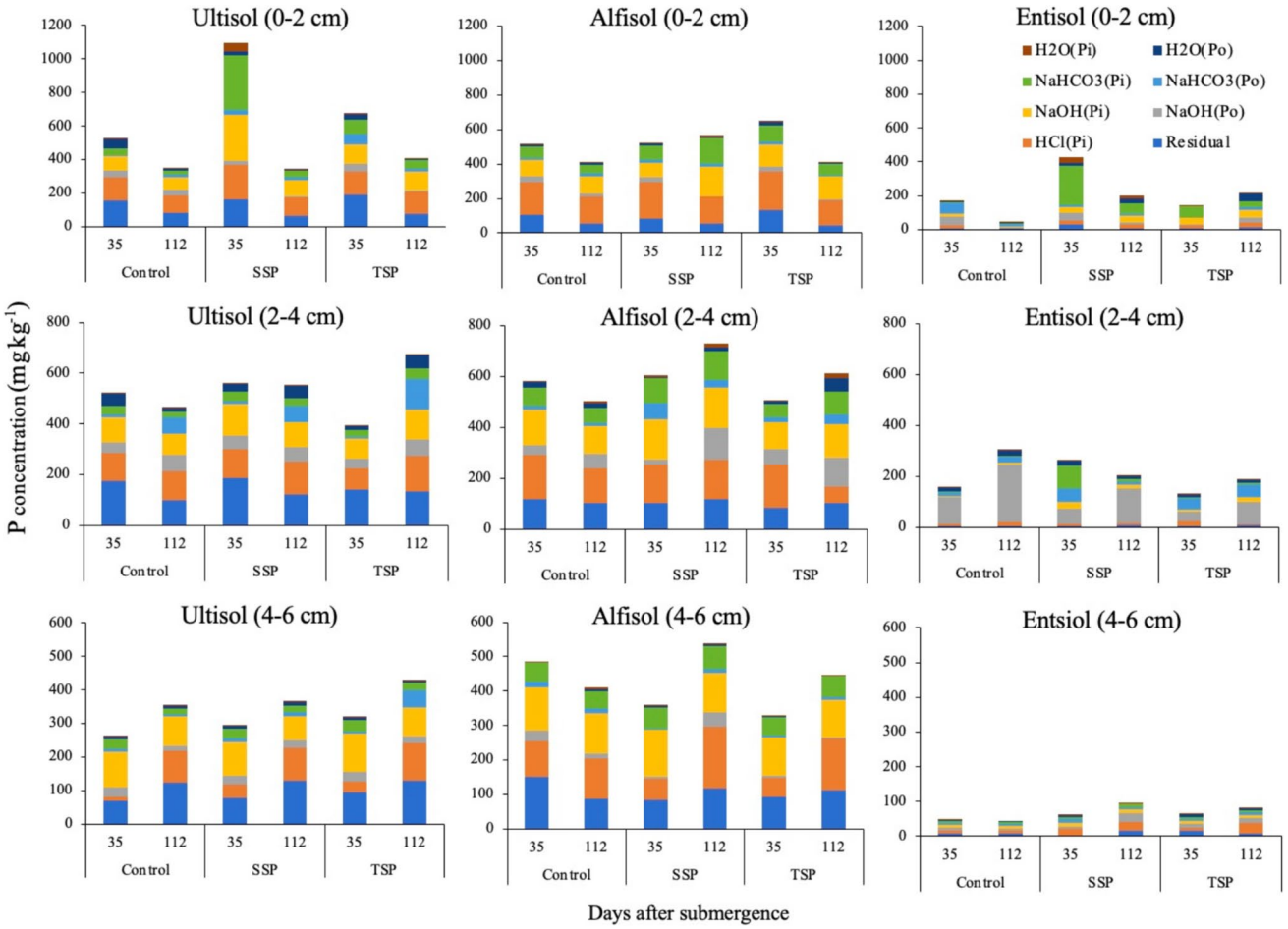

Fig. 6 Changes in different P fractions at depths of 0–2, 2–4 and 4–6 cm at 35 and 112 days after submergence of Ultisol, Alfisol and Entisol soils Pi; inorganic phosphorus, Po; organic phosphorus, H_2O; distilled water extractable P, $NaHCO_3$; 0.5 M sodium bicarbonate extractable P, NaOH; 0.1 M sodium hydroxide extractable P, HCl; 1.0 M hydrochloric acid extractable P, residual; residual P

(Figs. S4-S6). The P fractions varied differently with DASub in the three soils. In the Ultisol, at the 0–2 cm depth, the total P (summation of all the eight fractions) of both SSP and TSP applied treatments decreased from 35 to 115 DASub by 224% and 69%, respectively. In the Alfisol, at the 0–2 cm depth, the total P of SSP applied soil increased by 7% with time while it decreased by 59% in TSP applied soil. Comparatively, in the Entisol (0–2 cm depth), the total P of SSP applied treatment decreased with time by 112% and it increased by 34% in TSP applied treatment. In the 0–2 cm depth, the labile P fraction increased by 2–453% compared to the control with P fertiliser addition in all the soils at both 35 and 112 DASub, except for the TSP applied Entisols at 35 DASub. In the same soil layer, the labile fraction deceased with time by 44–86% in the P applied treatments except for SSP applied Alfisols and TSP applied Entisols, where they increased with time by 52% and 39%, respectively. The Fe and Al associated P fraction deceased with time by 9–83% in all the three treatments except for SSP applied Alfisol and TSP applied Entisol, where they increased with time by 58% and 64%, respectively in the same depth. The Ca bound P fraction also decreased with time by 4–77% in all the three treatments except for the TSP applied Entisol where it increased by 113% at the 0–2 cm depth. In the 2–4 cm depth, the total P increased by 17–30% with time in the TSP applied treatments in all three soils. In the 4–6 cm depth, the total P increased by 19–36% with time, in the P fertiliser applied treatments.

3.6 Changes of Phosphorus Species During Submergence

Tables 2–4 show the saturation indices (SI) of P minerals, Fe/Mn carbonates and selected Fe and Al oxy(hydr) oxides of the three soils. Positive SI indicates the supersaturation of solution, thermodynamically favouring its formation, while negative SI indicates undersaturation of the

Table 2 Saturation indices of P minerals from MINTEQ for Ultisol soils during submergence

	Control			SSP			TSP		
DASub	7	35	70	7	35	70	7	35	70
Ca phosphates									
$Ca_3(PO_4)_2$ (am1)	X	X	X	0.70	X	X	0.70	X	X
$Ca_3(PO_4)_2$ (am2)	0.02	X	X	3.45	X	X	3.45	X	X
$Ca_3(PO_4)_2$ (β)	0.69	X	X	4.12	X	X	4.12	X	X
$Ca_4H(PO_4)_3{:}3H_2O$	X	X	X	3.82	X	X	4.01	X	X
CaHPO4	X	X	X	X	X	X	0.13	X	X
FCO_3-Apatite	21.98	19.74	16.64	30.76	20.09	13.80	30.49	18.71	14.86
Hydroxyapatite	8.50	7.34	5.51	14.06	7.41	3.46	13.88	6.58	4.47
Fe/Mn phosphates									
Strengite (Fe^{III})	1.08	X	0.14	0.38	X	0.28	0.74	X	0.14
Vivianite (Fe^{II})	3.98	X	X	3.95	X	X	5.84	X	X
$MnHPO_4$ (Mn^{II})	3.37	2.93	2.02	4.55	3.01	1.97	4.87	2.87	2.10
Fe/Mn carbonates									
Siderite (Fe^{II})	X	X	X	X	X	X	X	X	X
Rhodochrosite (Mn^{II})	1.95	1.95	1.78	2.27	1.82	1.57	2.14	1.81	1.80
$MnCO_3$ (am)	1.45	1.45	1.28	1.77	1.32	1.07	1.64	1.31	1.30
Fe/Al oxy(hydr)oxides									
$Fe(OH)_2.7\ Cl_{0.3}$	3.91	0.84	2.20	3.81	0.52	1.72	3.95	0.71	2.10
Ferrihydrite (Fe^{III})	0.89	X	X	0.94	X	X	1.00	X	X
Ferrihydrite (aged)	1.40	X	X	1.45	X	X	1.51	X	X
Goethite (Fe^{III})	3.60	0.67	2.12	3.65	0.34	1.55	3.71	0.53	2.02
Hematite (Fe^{III})	9.59	3.75	6.65	9.70	3.08	5.49	9.82	3.45	6.44
Hercynite (Fe^{II})	8.10	4.10	3.70	7.31	2.16	3.73	6.83	3.00	3.67
Lepidocrocite (Fe^{III})	2.72	X	1.24	2.77	X	0.67	2.83	X	1.14
Maghemite (Fe^{III})	1.79	X	X	1.90	X	X	2.02	X	X
Magnetite (Fe^{III}, Fe^{II})	15.97	7.87	12.51	15.73	6.95	11.12	16.11	7.58	12.25
$Al(OH)_3$	1.61	0.74	0.62	1.38	X	X	1.02	0.18	X
Al_2O_3	0.15	X	X	X	X	X	X	X	X
Boehmite	1.32	0.45	X	1.10	X	X	0.73	X	X
Diaspore	3.03	2.15	1.08	2.80	1.31	1.21	2.44	1.60	1.10
Gibbsite	2.16	1.29	0.22	1.93	0.44	0.35	1.57	0.73	0.23
Fe sulphides									
Pyrite	X	X	X	X	X	X	X	X	X

DASub: Days after submergence, X: indicates negative saturation indices

solution, thermodynamically favouring its dissolution (or absence). However, SI values closer to zero, or equilibrium (e.g. 0.02), are less likely to result in precipitation, whereas higher SI values (e.g. 30) increase the probability of precipitation. Notably, a positive SI does not necessarily mean that the mineral will precipitate from the solution, as the slow precipitation kinetics may inhibit the precipitation, or favour precipitation of another mineral (Voigt et al. 2018).

All the three soils were supersaturated with apatite and Mn(II) phosphates throughout the submergence period (Tables 2–4). At 7 DASub, the P fertiliser applied soils were supersaturated with more soluble Ca-P forms (e.g. amorphous $Ca_3(PO_4)_2$, β-$Ca_3(PO_4)_2$, $Ca_4H(PO_4)_3{:}3H_2O$) compared to the control soils, except for Entisols, for which supersaturation with those Ca-P was not observed in P fertiliser applied soils.

All the treatments of Ultisol were supersaturated with Fe(II) phosphates at 7 DASub and later dissolved with time and all the treatments were supersaturated with Fe(III) phosphates at 7 and 70 DASub in Ultisols (Table 2). Comparatively, all the treatments of Alfisol were supersaturated with Fe(II) phosphates at 7 and 70

Table 3 Saturation indices of P minerals from MINTEQ for Alfisol soils during submergence

	Control			SSP			TSP		
DASub	7	35	70	7	35	70	7	35	70
Ca phosphates									
$Ca_3(PO_4)_2$ (am1)	X	X	X	3.67	X	X	1.34	X	X
$Ca_3(PO_4)_2$ (β)	0.32	X	X	4.34	X	X	2.01	X	X
$Ca_4H(PO_4)_3{:}3H_2O$	X	X	X	4.31	X	X	1.08	X	X
$CaHPO_4$	X	X	X	0.22	X	X	X	X	X
FCO_3-Apatite	21.98	16.86	12.03	31.87	16.45	19.57	25.81	17.59	20.81
Hydroxyapatite	8.10	4.75	1.83	14.22	4.32	6.45	10.47	5.09	7.28
Fe/Mn phosphates									
Strengite (Fe^{III})	X	X	X	1.05	X	X	0.60	X	X
Vivianite (Fe^{II})	2.20	X	1.10	3.51	X	3.15	3.07	X	3.36
$MnHPO_4$ (Mn^{II})	2.87	2.58	1.36	4.78	2.14	2.86	3.83	2.51	3.06
Fe/Mn carbonates									
Siderite (Fe^{II})	0.78	X	1.67	X	X	1.21	0.34	X	1.15
Rhodochrosite (Mn^{II})	1.95	1.74	2.35	2.06	1.70	2.19	1.73	1.63	2.21
$MnCO_3$ (am)	1.45	1.24	1.85	1.56	1.20	1.69	1.23	1.13	1.71
Fe/Al oxy(hydr)oxides									
$Fe(OH)_2.7\ Cl_{0.3}$	7.53	X	2.52	6.86	X	2.08	6.85	X	2.10
$Fe_3(OH)_8$	6.60	X	X	4.55	X	X	4.51	X	X
Ferrihydrite (Fe^{III})	4.70	X	X	4.03	X	X	3.96	X	X
Ferrihydrite (aged)	5.21	X	0.28	4.54	X	X	4.47	X	X
Goethite (Fe^{III})	7.40	X	2.48	6.74	X	2.02	6.67	X	2.06
Hematite (Fe^{III})	17.21	1.63	7.36	15.89	1.39	6.45	15.74	0.15	6.52
Hercynite (Fe^{II})	8.01	0.73	5.97	6.44	1.32	5.62	6.43	X	5.45
Lepidocrocite (Fe^{III})	6.52	X	1.60	5.86	X	1.14	5.79	X	1.18
Maghemite (Fe^{III})	9.40	X	X	8.08	X	X	7.94	X	X
Magnetite (Fe^{III}, Fe^{II})	23.42	4.76	14.50	21.37	4.47	13.23	21.33	2.54	13.28
$Al(OH)_3$	1.64	X	0.16	1.23	X	0.16	1.17	X	0.09
Al_2O_3	0.22	X	X	0.96	0.60	0.61	0.68	0.47	0.65
Boehmite	1.36	X	X	0.92	X	X	X	X	X
Diaspore	3.06	0.96	1.58	2.64	1.29	1.58	2.58	0.40	1.50
Gibbsite	2.19	0.09	0.71	1.78	0.42	0.71	1.72	X	0.64
Fe sulphides									
Pyrite	X	X	X	X	X	0.18	X	X	X

DASub: Days after submergence, X: indicates negative saturation indices

DASub (Table 3). Further, some applied P precipitated as Fe(III) phosphates in both SSP and TSP applied treatments at 7 DASub in Alfisols. In Entisols, all the treatments were supersaturated with Fe(II) phosphates at 7 and 70 DASub (Table 4).

Among the selected Fe/Al oxy(hydr)oxides, Ultisols exhibited the highest number of supersaturated Fe/Al oxy(hydr)oxides throughout the submergence period, while Entisol had fewer, and Alfisol had the lowest (Tables 2, 3 and 4). Fe(III) oxy(hydr)oxides were supersaturated at 7 DASub in all the treatments of all the three soils and later undersaturated with time. Additionally, in Entisols, the same pattern was observed for Al oxy(hydr) oxides.

4 Discussion

The three paddy soils exhibited no significant change in porewater DRP concentrations in response to applied P fertilisers during submergence, except immediately following fertiliser application. The critical level of soil solution P concentration for rice plant growth is 0.1 mg L^{-1} (Hossner et al. 1973). In the Alfisol with fertiliser application, DRP concentration remained above the critical level. In Entisols, only the SSP treatment resulted in DRP concentrations exceeding this threshold. In contrast, DRP concentrations in Ultisols remained below the critical level, even in soils where P fertilisers were applied. However, the low DRP concentration (0.03–0.10 mg L^{-1}) maintained throughout the experimental

Table 4 Saturation indices of P minerals from MINTEQ for Entisol soils during submergence

	Control			SSP			TSP		
DASub	7	35	70	7	35	70	7	35	70
Ca phosphates									
$Ca_3(PO_4)_2$ (am2)	0.30	X	X	X	X	X	X	X	X
$Ca_3(PO_4)_2$ (β)	0.97	X	X	0.55	X	X	X	X	X
FCO_3-Apatite	22.48	8.87	13.08	21.66	16.58	14.21	19.66	16.32	15.84
Hydroxyapatite	8.92	0.87	3.41	8.43	5.43	3.79	7.20	5.26	5.24
Fe/Mn phosphates									
Strengite (Fe^{III})	X	X	X	X	X	X	X	X	X
Vivianite (Fe^{II})	5.24	X	3.13	3.66	X	4.30	5.07	X	5.68
$MnHPO_4$ (Mn^{II})	4.29	1.79	2.59	3.88	3.14	3.09	3.69	2.42	3.85
Fe/Mn carbonates									
Siderite (Fe^{II})	1.34	X	1.20	1.09	X	1.57	1.59	1.30	1.15
Rhodochrosite (Mn^{II})	2.65	2.41	1.93	2.73	2.47	2.28	2.57	0.59	1.85
$MnCO_3$ (am)	2.15	1.91	1.43	2.23	1.97	1.78	2.07	3.03	1.35
Fe/Al oxy(hydr)oxides									
$Fe(OH)_2.7\ Cl_{0.3}$	2.97	0.35	1.88	3.00	0.61	2.09	3.42	1.63	1.90
Ferrihydrite (Fe^{III})	0.06	X	X	0.12	X	X	0.45	X	X
Ferrihydrite (aged)	0.57	X	X	0.63	X	X	0.96	X	X
Goethite (Fe^{III})	2.77	0.24	1.75	2.83	0.52	1.98	3.16	1.55	1.78
Hematite (Fe^{III})	7.94	2.88	5.90	8.06	3.45	6.36	8.71	5.50	5.97
Hercynite (Fe^{II})	7.05	2.26	7.19	7.38	2.64	7.06	7.17	4.05	7.11
Lepidocrocite (Fe^{III})	1.89	X	0.87	1.95	X	1.10	2.28	0.67	0.90
Maghemite (Fe^{III})	0.14	X	X	0.26	X	X	0.91	0.17	X
Magnetite (Fe^{III}, Fe^{II})	14.62	7.95	12.70	14.65	8.72	13.31	15.77	1.92	12.74
$Al(OH)_3$	0.94	X	0.94	1.14	X	0.80	0.80	X	0.91
Boehmite	0.65	X	0.65	0.86	X	0.51	0.51	X	0.62
Diaspore	2.35	0.76	2.36	2.56	0.84	2.22	2.22	1.04	2.33
Gibbsite	1.49	X	1.49	1.69	X	1.35	1.35	0.17	1.46
Fe sulphides									
Pyrite (Fe^{II})	X	X	4.99	X	X	4.86	X	X	5.41

DASub: Days after submergence, X: indicates negative saturation indices

period (14–112 DASub) did not support demonstrating significant differences between the two P fertilisers and among the three soils. This suggested that, despite the differences in P and Ca contents between SSP and TSP fertilisers, these factors did not significantly affect DRP release under submerged conditions.

At the beginning of submergence, P fertiliser applied soils exhibited supersaturation with more soluble forms of Ca phosphates and Fe oxy(hydr)oxides in both the P fertilised and control treatments. Later, the soils became undersaturated with these minerals, indicating their dissolution during the submergence period. This could potentially increase the DRP concentration during the submergence (Attanayake et al. 2022; Jayarathne et al. 2016; Kumaragamage et al. 2022; Weerasekara et al. 2021). However, "net" DRP concentration in all three soils was not increased, probably due to resorption of P onto Fe and Ca minerals (Amarawansha et al. 2015; Jayarathne et al. 2016; Nishigaki et al. 2021) as described below in detail. The two P fertilisers used in this experiment was $Ca(H_2PO_4)_2.2H_2O$. This could have resulted in supersaturation of more soluble Ca-P minerals in fertiliser applied Ultisols and Alfisols at the initial stage of submergence.

Previous studies reported increased dissolved P concentrations in porewater upon submergence (Amarawansha et al. 2015; Attanayake et al. 2022; Jayarathne et al. 2016; Kumaragamage et al. 2022; Weerasekara et al. 2021). However, despite anoxic conditions, exceptions have been reported where soils did not release P to porewater (or floodwater) (Shober and Sims 2009; Young and Ross 2001). Those authors attributed that observation to the process of resorbing P into Fe(II) oxy(hydr)oxides and $Al(OH)_2$ phases. The increase of Fe in porewater during the submergence has not been observed in this study as hypothesised. This indirectly explained that the reductive dissolution of Fe oxy(hydr)oxides was not a prominent mechanism of

releasing P in these soils. One reason for this could be the low organic carbon content (~ 4–10 g kg^{-1}) of the three soils. The speed of sequential reduction (O_2, NO_3^-, MnO_2, Fe^{3+}, SO_4^{2-} and CO_2) under submergence is affected by the availability of organic matter as it is the main electron donor for the reduction reactions (Marschner 2021). Therefore, low organic matter content could hinder the reductive dissolution of Fe oxides and consequent P release in these soils. The positive significant ($p < 0.01$) correlation between DRP and porewater Ca concentration in the three soils provides evidence that dissolution of Ca minerals contributed to P release at least at the beginning of the submerged period. This correlation is consistent with the addition of Ca phosphate-based fertilisers, which serve as a source of Ca and facilitate P release through the dissolution of Ca-P minerals. The reason for not observing a significant increase of DRP in our study could be due to resorption of P released by the fertilisers onto other solid phases as described below.

Highly weathered tropical soils are usually characterised by having high potential to sorb P due to the presence of high amounts of Fe and Al oxy(hydr)oxides (Rakotoson et al. 2016). Both Fe and Al oxides are effective adsorbents of phosphate, but Al oxides are more effective than Fe oxides. For example, oxalate extractable Al oxides adsorb nearly twice as much phosphate as oxalate extractable Fe oxides (Darke and Walbridge 2000). Therefore, presence of Al oxy(hydr)oxides even in minor quantities in these soils can sorb considerable sums of P released to porewater under submergence.

The decreasing porewater Al concentration of the three soils with time (from an average of 0.2 mg L^{-1} to below method detection limit) pointed to the possibility of P sorption with Al minerals. The XRD analysis of soil samples revealed gibbsite [$Al(OH)_3$] as a major metal oxy(hydr)oxide in Ultisols and Entisols. Gibbsite was found either as major or trace clay mineral in several soils of Sri Lanka including Ultisols, Alfisols, and Entisols (Indraratne 2020). Phosphorus sorbed on to gibbsite do not undergo reductive dissolution as P sorbed on to Fe oxy(hydr)oxides. Being a very slow exchanging component (Kyle et al. 1975), the P sorbed to gibbsite do not contribute to P release in submerged soils.

The XRD analysis revealed the presence of Fe(III) mineral, goethite as a trace mineral in Alfisols, consistent with previous XRD analyses of these soils (Indraratne 2020). The addition of P fertiliser treatments to Ultisols reported high concentrations of P in the Fe associated fraction compared to the other two soils. The released P to porewater by the dissolution of Ca-P and Fe(III) oxy(hydr)oxides may be resorbed by the abundantly available Fe oxy(hydr)oxides in all the three soils. Conversion of applied P fertilisers to Fe associated P under submergence was previously reported in rice paddy soils in the tropics (Abolfazli et al. 2012). However, this effect may be higher in Ultisols than the other soils, as explained by the comparatively high Fe associated P fraction in Ultisols.

The crystallinity of metal oxy(hydr)oxides plays a key role in P sorption capacity. Poorly crystalline and amorphous oxy(hydr)oxides contribute to a more stable fixation of P compared to well crystalline forms. Amorphous Fe minerals which are not detected by XRD (Ali et al. 2022), may be present in paddy soils that undergo frequent alternative flooding and drainage during the rice cropping cycle, preventing their crystallisation to detectable forms.

The initial soil analyses of the three soils reported high concentrations of oxalate extractable Fe and Al, suggesting the presence of poorly crystalline Fe and Al which have high P sorption capacity. Previous research provided evidence of a trend toward the formation of lower crystallinity Fe(III) oxides after reoxidation of Fe(II) minerals in redox fluctuating environments (Chen et al. 2018) or through partial transformation of Fe oxides into lower crystalline phases with submergence (Zhang et al. 2003).

Further, high concentrations of Fe(II) in soil solution are likely to induce precipitation of Fe(II)/Fe(III) carbonates and hydroxides that limit P release due to sorption or co-precipitation (Amery and Smolders 2012). Supersaturation of Fe(II) carbonates (siderite) was evident at 7 DASub and/or at 70 DASub in Alfisols and Entisols (Table 3 and 4). For TSP added Entisols, supersaturation of siderite persisted throughout the submergence, likely contributing to lower P release compared to the SSP applied treatment.

Changes in the crystallinity of Fe oxy(hydr)oxides also influence P dynamics by affecting the kinetics of microbial Fe(III) reduction. For example, microbes can rapidly reduce ferrihydrite like minerals (typically within hours), but reduce well-ordered hematite, goethite, and lepidocrocite at slower rates (e.g. several months) (Bonneville et al. 2004; Ginn et al. 2017). The variability in crystallinity changes of Fe oxy(hydr)oxides may explain the observed differences in supersaturation and undersaturation of different Fe oxy(hydr)oxides in the soils studied.

Phosphorus fractionation analysis showed that the total P concentration of the P fertiliser applied treatments of the three soils increased with time below the 2 cm depth (2–4 and 4–6 cm depths) with a few exceptions. This indicated that the dissolved P fertilisers leached down through the soil profile. One reason for this P leaching may be due to some saturation of the P sorption sites of the moderately P buffered soils in the study. Previous research reported decreasing P buffering capacity with long-term P fertiliser application in submerged paddy soils and therefore, subsequent increasing risk of P leaching in such soils (Long et al. 2023). The increased P concentration in Ca associated P fraction in the P fertiliser applied Ultisols at both 2–4 and 4–6 cm depths and the 4–6 cm depth of Alfisols and Entisols indicated the possible transformation of leached P into Ca-P

minerals. Further, the increased P concentrations in Fe and Al associated P fractions at these depths in the P fertiliser applied Alfisols and Entisols, hinted that the leached P may have adsorbed onto Fe and Al oxy(hydr)oxides. Since both Ca associated P and Fe and Al associated P are moderately labile, these will not be detected in DRP concentrations but could be available to rice plants under different conditions. For example, under submergence, rice roots can resolubilise some of the immobilise P (e.g. Fe(II) carbonates and hydroxides) by organic anion excretion and acidification caused by oxidation of Fe(II) by oxygen released from roots and the imbalance between the intake of anions and cations (Huguenin-Elie et al. 2003). Increasing residual P fractions at the soil depths below 2 cm of the P fertiliser applied three soils, indicated that a fraction of applied P has been transformed to non-available P pools for plants.

Soil pH plays a crucial role in determining P release in submerged soils (Palihakkara et al. 2024). It significantly affects P release in soils (Barrow and Hartemink 2023; Penn and Camberato 2019; Wang et al. 2023) and application of N fertilisers could indirectly influence P release in soils by altering pH. The effect of urea application on soil pH was not consistent across the three soils and DASub. Previous research has shown significant decrease (Fageria et al. 2010), increase (Michael 2020), no significant changes or mixed results (Khatiwada et al. 2012) of soil pH when urea is applied under different circumstances.

Applied urea granules are dissolved within minutes to hours, depending on soil water content and temperature (Kissel et al. 2008). During hydrolysis, urea is rapidly converted by the urease enzyme into NH_4^+ and HCO_3^- ions leading to an increase in soil pH near to urea granules (Lasisi and Akinremi 2021), depending on the soil's pH buffering capacity (Curtin et al. 2020). High pH favours formation of ammonia (NH_3) from NH_4^+, which may volatilise, while the non-volatised NH_4^+ can be oxidised into NO_3^- via nitrification. This causes soil acidification (Subbarao et al. 2006) and because of acidification, the rate of nitrification will also be reduced (Hanan et al. 2016; Zebarth et al. 2015).

Single superphosphate and TSP generally do not affect soil pH, but they sometimes cause soil acidification specially in alkaline soils (Pahalvi et al. 2021), due to reaction products being very acidic. The mixed effects on soil pH of observed in this study may result from the combine effect of urea hydrolysis, nitrification of NH_4^+ ions, effect of acidification from TSP or SSP, and pH buffering capacity of soils. Additionally, the localised changes in pH in close proximity to the urea granules could have contributed to the observed mixed effects, as pH changes were likely more pronounced in the immediate vicinity of the granules and less consistent at broader soil depths. Notably, the immediate and significant ($p < 0.05$) increase of pH with urea application in Entisols may be due to low pH buffering capacity the soil with the sandy texture.

We hypothesised that increase in NO_3^- through urea application could delay P release by NO_3^- acting as a preferable electron acceptor over Fe and Mn in the sequential reduction process (O_2, NO_3^-, MnO_2, Fe^{3+}, SO_4^{2-} and CO_2). However, urea addition did not significantly ($p > 0.05$) affect the P release of the soils. One reason for this observation may be the time of urea application. Urea was applied as two split applications at 20 DASub and 48 DASub. A study conducted in submerged silt loam paddy soils reported that urea was hydrolised within 5–6 days after application (Siddique et al. 2020). Meanwhile, the formed NH_3 gas could be volatilised, and the rate of volatilisation will be increased with pH when it is greater than 7. Since the pH of porewater was higher than 7 in all the three soils specially after the first urea application (Fig. 2a, b and c), there would be a considerable loss of applied N as NH_3 gas. This may be another reason for not observing a significant effect of urea addition on P release. The remaining NH_3 which did not volatilise, could be slowly converted to NO_3^- ions via nitrification. Therefore, the NO_3^- formation due to urea application may have occurred after 21 DASub. However, the Eh of the three soils were below 0 mV by this time indicating that they passed the NO_3^- and Mn^{4+}/Mn^{3+} reduction in the redox ladder. Furthermore, NO_3^- ions formed via nitrification will also be subjected to microbially mediated denitrification processes resulting in the loss of N from the system in its gaseous forms such as nitrous oxide (N_2O), nitric oxide (NO) and nitrogen gas (N_2) (Coyne 2008).

5 Conclusions

This study offers insights into the potential changes in phosphorus species following submergence in three different paddy soils of Sri Lanka. The results suggested that phosphorus fertilisation did not significantly alter dissolved reactive phosphorus concentrations in porewater, except shortly after fertiliser application, and that phosphorus is primarily released through the dissolution of calcium phosphate minerals and iron(III) oxy(hydr)oxides. However, the released phosphorus appeared to be leached and subsequently resorbed onto the abundantly available iron and aluminium oxy(hydr)oxides surfaces and/or clay minerals, and also precipitated as calcium minerals. Further, urea addition did not affect the phosphorus release of any of the soils. Although the critical phosphorus concentration for rice growth was not consistently reached in, particularly in Ultisols, previous studies have indicated no yield response (no significant increase or decrease) to applied phosphorus fertilisers across paddy-growing soils in Sri Lanka. The unique

micro-environment of the rice plant rhizosphere may play a role in enhancing phosphorus release into the porewater under submergence, particularly by tapping the phosphorus in moderately labile pools. Hence, future research should prioritise investigating phosphorus transformations involving rice plants and rhizosphere soils. Such studies would contribute to a comprehensive understanding of why rice yield could remain unaffected despite the addition or omission of phosphorus fertilisers. Further, this research could be expanded to develop more effective phosphorus management strategies, including methods to access sorbed phosphorus or legacy phosphorus in Sri Lankan paddy soils. Additionally, direct phosphorus speciation methods like X-ray absorption near edge structure spectroscopy could help to validate the results obtained through assumptions-based thermodynamic modelling.

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Data Availability Data will be made available on request.

Declarations

Competing Interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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