



## Research article

## Phosphorus release under short-term submergence of pasture soils in critical source areas

Janani Palihakkara<sup>a,b,\*</sup> , Lucy Burkitt<sup>a</sup>, Paramsothy Jeyakumar<sup>a</sup>, Chammi P. Attanayake<sup>c</sup> <sup>a</sup> Environmental Sciences Group, School of Agriculture and Environment, Massey University, Palmerston North, 4442, New Zealand<sup>b</sup> Department of Soil Science, Faculty of Agriculture, University of Peradeniya, Peradeniya, 20400, Sri Lanka<sup>c</sup> Institute for Sustainability, Energy and Environment, University of Illinois Urbana-Champaign, Urbana, IL, 61801, USA

## ARTICLE INFO

## Keywords:

Allophanic  
Dissolved reactive phosphorus  
Pallic  
Recent  
Short-term frequent submergence

## ABSTRACT

Critical source areas (CSAs) can act as a source of phosphorus (P) in surface waters by releasing soil P to porewater during frequent rainfall events. The extent of P release under short-term, frequent submergence has not been systematically studied in CSAs in New Zealand. A study was conducted to explore the potential of three contrasting dairy and sheep/beef farm soils (Recent, Pallic and Allophanic soils) to release P to porewater and pondwater under short-term and frequent submergence. Five undisturbed soil blocks (20 × 20 × 15 cm) were sampled from each soil. Porewater samplers and half-cell platinum electrodes for in-situ redox potential measurements were installed at 2 and 10 cm depths from the soil surface. Six submerged events were created by maintaining a 5 cm waterhead. Porewater and pondwater samples were collected immediately and three days after each submergence event. After three days of submergence, the soil blocks were drained and maintained at 70% of water holding capacity for 10 days before the next submergence event. Dissolved reactive phosphorus (DRP), pH, dissolved organic carbon, cations, anions, and alkalinity of the water samples were measured. Soil chemical P fractions were assessed in initial soils and soils in the middle and end of the experiment. Thermodynamic modelling was used to infer dissolution and formation of P and P-associated minerals. The Recent soil released P to porewater at both depths and to pondwater. The Pallic soil released P to porewater at both depths but did not change pondwater DRP. Allophanic soil sorbed P and did not increase DRP either in porewater or pondwater. The average pondwater DRP of the three soils during submergence were 17 to 65-fold higher than the New Zealand lowland river target DRP concentration (0.01 mg/L). The mechanisms of P release from the Recent and Pallic soils were desorption and reductive dissolution of Mn(II) minerals. Reductive dissolution of Fe(II) minerals was not supported by fractionation or modelling results. Decreases in labile, moderately labile and stable P fractions contributed to P release in the Recent soil, while increases in the labile and moderately labile P fractions contributed to P retention in the Pallic and Allophanic soils. This study highlighted that the Recent soil is riskier than the other two soils in releasing P upon short-term submergence and the potential use of Allophanic soil as a P sorbing material in CSAs to mitigate P loss.

## 1. Introduction

Non-point source phosphorus (P) contamination originating from agricultural soils is a significant contributor to the degradation of freshwater quality (Yang et al., 2008; Schoumans et al., 2014; Dharmakeerthi et al., 2019). Agricultural soils with elevated P concentrations, exceeding the optimal requirement for plant growth, are prone to P loss via surface runoff and leaching, which is an extensively investigated phenomenon (McDowell and Sharpley, 2001; Djodjic et al., 2004; Tóth et al., 2014). Surface runoff during or following a rainfall event can

lose P in two forms: dissolved P and particulate bound P. The loss of dissolved P is critical for eutrophication, as it is bioavailable for the growth of harmful microorganisms and aquatic plant in freshwater bodies (Li and Brett, 2013). Therefore, there is significant interest in transportation of dissolved P from agricultural soils to waterways.

Critical source areas (CSAs) are nutrient hotspots in agricultural landscapes with active hydrological connectivity to surface waters (Sharpley et al., 2011; Thomas et al., 2016; Smith et al., 2021). Critical source areas can contribute a disproportionately large amount of P loss despite representing a small extent in the landscape (Pionke et al., 2000;

\* Corresponding author. Environmental Sciences Group, School of Agriculture and Environment, Massey University, Palmerston North, 4442, New Zealand.  
E-mail address: [j.palihakkara@massey.ac.nz](mailto:j.palihakkara@massey.ac.nz) (J. Palihakkara).

<https://doi.org/10.1016/j.jenvman.2025.124317>

Received 30 June 2024; Received in revised form 22 December 2024; Accepted 22 January 2025

Available online 29 January 2025

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Sharpley et al., 2011) because CSAs frequently get saturated or form surface ephemeral flow pathways depending on their topography. This creates a unique environment within CSAs where soil properties, including redox potential (Eh) and pH, can fluctuate due to alternating anaerobic and aerobic conditions. These alternative conditions are likely to influence the P transformations and P release to soil porewater. Further, the released P could be diffused into the pondwater overlying the soil and can then be transported to surface waters via flow pathways (Amarawansa et al., 2015; McDowell et al., 2016; Smith et al., 2021), posing a threat of eutrophication of nearby waterways.

Phosphorus is released to porewater under submergence of soil as a combined result of complex hydrological and biogeochemical processes (Jayarathne et al., 2016; Dharmakeerthi et al., 2019). Soil pH changes towards neutrality upon submergence (Ponnamperuma, 1972). The Eh of soil reduces due to the anaerobic condition created in soil by submergence (Maranguit et al., 2017). The Eh controls the stability of several chemical compounds in soil. For example, reductive dissolution of Mn(IV)/Mn(III) and Fe(III) can release P associated with them (Amarawansa et al., 2015).

Continuous alternation of redox conditions such as repeated reductive dissolution and oxidative reprecipitation increase amorphous Fe oxyhydroxides in soil and thereby increase P sorption (Baldwin and Mitchell, 2000). Further, the frequency and duration of soil submergence are known to influence the release of dissolved P into porewater (Gu et al., 2019; Warrinnier et al., 2020). A significant amount of research has been conducted worldwide to explore the P release under prolong submergence (eg: more than two weeks) for a range of land uses such as paddy (Hua et al., 2017; Liu et al., 2019), wetlands (Maranguit et al., 2017; Parsons et al., 2017; Rapin et al., 2019) and river valleys/basins (Weerasekara et al., 2021; Kumaragamage et al., 2022). However, the studies on the effect of short-term (eg: less than four days) submergence with alternative wetting and drying of soil, which are experienced during winter and spring seasons, on P release, are limited. Smith et al. (2021) reported an increase in total dissolved P (up to 0.12 mg/L) in the subsoil (below 20 cm) of a Melanic Orthic Gley soil in the South Island of New Zealand during wet periods, but this study did not measure P in overlying water and so has limited applicability to surface runoff associated with CSAs. In addition, the effect of soil type on the release of P upon short-term but frequent submergence during storm events in agricultural features such as CSAs, has not been studied previously in New Zealand. Furthermore, the extent and mechanisms of P release by different New Zealand grassland soils to porewater and overlying water has not previously been studied. A comprehensive understanding of these aspects is important for implementing improved P management strategies in CSAs to minimise P loss to surface waters.

The intact soil blocks were subjected to short-term (three days) and frequent (every ten days) submergence (1) to explore the potential of three contrasting soils to release P to porewater and pondwater, (2) to explore the relationships between soil characteristics, and P release and (3) to elucidate the mechanisms of P release in these soils using P fractionation and thermodynamic modelling. It was hypothesised that 1) the reductive dissolution of Fe/Mn oxy(hydr)oxides during submergence would release sorbed P, consequently elevating the P concentration in the soil solution and pondwater and 2) the degree of P release due to submergence would depend on soil characteristics.

## 2. Materials and methods

### 2.1. Soils used in the glasshouse study

Three soils with contrasting properties were selected from or adjacent to CSAs on three farms belonging to Massey University in the Manawātū-Whanganui region of New Zealand, namely Dairy 1 (S 40° 22' 27", E 175° 37' 05"), Dairy 4 (S 40° 23' 39", E 175° 36' 55"), and Tuapaka farm (S 40° 21' 00", E 175° 44' 43"). Five undisturbed soil blocks (20 × 20 × 15 cm) with long-term ryegrass/white clover pasture (Dairy 1 and

Dairy 4) and ryegrass, white clover and browntop (Tuapaka) were sampled from each site and placed in black-coloured plastic containers. Two extra soil blocks were collected from each site and treated the same as other replicates to reform the damaged soil blocks due to sampling at the middle of the experiment for the P fractionation study (explained in section 2.5). Soils from the 0–10 cm depth were sampled from each soil type, immediately adjacent to the location where the blocks were collected, for initial soil characterisation. Soils from Dairy 1, Dairy 4 and Tuapaka farms were classified according to the *New Zealand Soil Classification* as Recent soil (Entisol), Pallic soil (Alfisol) and Allophanic soil (Andisol), respectively.

### 2.2. Initial soil characterisation

The soil samples (0–10 cm depth) were air-dried, passed through a 2 mm sieve, homogenised, and analysed for basic soil properties. Soil pH was determined in 1:2 (v/v) soil:water slurry followed by potentiometric determination of pH. Olsen P was determined by Olsen extraction (Olsen and Sommers, 1982) followed by molybdenum blue colorimetry (Murphy and Riley, 1962). Anion storage capacity (ASC) was determined by equilibration of 5 g of soil with 1000 mg/L P solution for 16 h followed by colorimetric analysis (Saunders, 1965). Mehlich-3 P was determined by Mehlich-3 extraction followed by inductively coupled plasma optical emission spectroscopy (ICP-OES) (Mehlich, 1984). Mehlich-3 extractable cations (Ca, Mg, Fe, Mn, Al and Zn) were also determined using ICP-OES. Cation exchange capacity was determined by 1M neutral ammonium acetate extraction of cations (K, Ca, Mg, Na) followed by ICP-OES (Blackmore et al., 1987). Total organic carbon was determined by thermal conductivity detector (Elementar Analyser) after acid pretreatment to remove carbonates present followed by catalytic combustion (900 °C, O<sub>2</sub>) and separation (Nelson and Sommers, 1996). Soil texture was determined by sedimentation procedure by hydrometer after organic matter removal (Standards Association of New Zealand, 1986). The oxalate extractable Fe, Al and Mn concentrations were determined by acid ammonium oxalate method (McKeague and Day, 1966) followed by Microwave plasma atomic emission spectroscopy (MP-AES, Agilent 4200). The acid ammonium oxalate method was repeated for the soil samples collected at the end of the experiment.

### 2.3. Experimental design

The experiment was set up in a glasshouse at the Massey University Plant Growth Unit. Porewater samplers were prepared using 0.5 cm diameter polytetrafluoroethylene (PTFE) tubes connected to 6 µm pore diameter filter cloth at one end and the other end of the tubes were connected to 50 ml syringes. Two porewater samplers were installed at 2 cm and 10 cm depths (Fig. 1) before commencing the experiment by

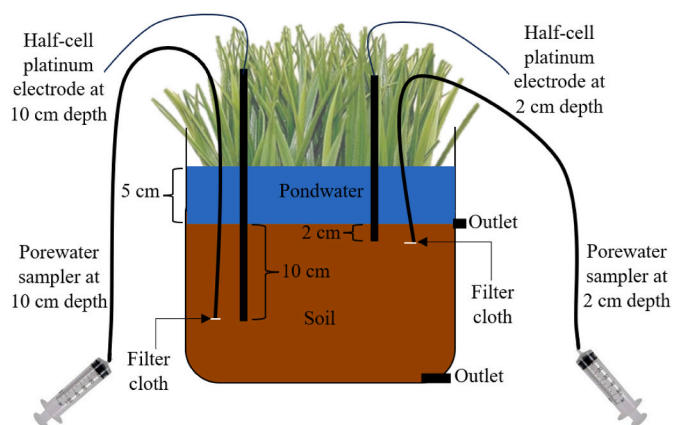


Fig. 1. A diagram of the ponded soil block used in the glasshouse experiment. The diagram is not to scale.

removing the soil up to 2 and 10 cm depths using a tube with a diameter similar to the samplers. The 2 cm depth was selected since it is the soil layer which retains 80% of the P from P fertiliser additions via sorption and also primarily contributes to P runoff via desorption (Vadas et al., 2007a; 2007b). The 10 cm layer was selected as more than half of the root mass of permanent pasture is generally located within the surface 10 cm depth (Crush et al., 2006). Changes in P chemistry and potential release of P within the root zone would influence plant access to P, therefore this is of great interest agronomically. Platinum half-cell electrodes were installed at the same two depths for in-situ redox potential measurements. The glasshouse experimental set up was arranged in a Complete Randomised Design. The average room day/night temperature ranged between 25 and 17 °C during this period. Each ponding event was created by filling the containers using tap water up to a waterhead of 5 cm above the soil surface. Tap water, with its known background P concentration was used to represent the typical agricultural P additions under field conditions and as a replenishment to P removal when water was drained after each submergence event. The soil blocks were maintained under submergence for three days and thereafter the water was drained off using two outlets: one just above the surface soil level and the other at the bottom of the container (Fig. 1). Soil blocks were maintained at a field capacity of 70% during the experimental period. The submergence events were repeated six times after a 10-day interval between each event. The experimental period lasted about 3 months mimicking the short-term submergence of soils during a wet season (eg: winter). Six submergence events were selected to allow adequate time for observations and identification of trends.

#### 2.4. Porewater and pondwater sampling and analysis

Porewater and pondwater samples were collected within an hour and three days after submergence, across the six repeated submerged events (referred to as E1, E2, E3, E4, E5 and E6, respectively). Soil porewater was collected by applying a vacuum using the 50 ml syringe attached to the sampler. Several clogging issues were encountered with the samplers. In such cases, the samplers were removed following such submerged event, the membranes were replaced, and the samplers were reinstalled. Pondwater was collected near to the soil surface using a 50 ml syringe. The porewater and pondwater samples were filtered through 0.45 µm cellulose membrane filters and analysed for DRP by the molybdate blue colour method (Murphy and Riley, 1962) using UV visible spectrophotometer (JENWAY 7315) within 12 h of sampling. The pH of porewater and pondwater samples were measured using a glass pH electrode (ORION STAR A214). The soil Eh was measured in-situ at each porewater sampling event using an Ag/AgCl reference electrode (ORION). The voltmeter reading was corrected to the standard hydrogen electrode potential by adding +200 mV. Cation concentrations (Ca, Mg, K, Na, Fe, Al, and Mn) of porewater were measured using microwave plasma atomic emission spectroscopy (Agilent 4200). Porewater anion concentrations (Cl<sup>-</sup>, F<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) were measured using ion chromatography (Dionex™ Aquion™). Dissolved organic carbon (DOC) concentrations of porewater were measured using a carbon nitrogen analyser (Shimadzu TOC-LCSH). Alkalinity of the porewater samples was measured by titrating against 0.1 N H<sub>2</sub>SO<sub>4</sub> in the presence of methyl orange indicator.

#### 2.5. Phosphorus fractionation study

Phosphorus fractionation was undertaken using a modified Hedley procedure (Hedley et al., 1982) for the soils collected for initial soil characterisation (0–10 cm) and then soil sampled at the middle of the third submergence event which was after 24–36 h after submergence, and at the end of the sixth event, just before draining off the pondwater. Three replicates from each of the 0–2, 2–8 and 8–10 cm depths were collected from each soil block and were bulked to make a composite sample. The three soil depths (0–2, 2–8 and 8–10 cm) were selected to

represent and cover the whole depth of the initial two soil depths (2 and 10 cm depths). Three replicates of soil blocks were used for the fractionation study for each soil type. The gaps created by soil sampling were filled with soil cored at the same depths from the additional two soil blocks maintained under similar experimental conditions as the main ponded blocks. The soil samples taken under submerged conditions were purged with nitrogen gas, sealed, and stored at –28 °C in a freezer until P fractionation analyses were performed.

An aliquot of fresh soil (approximately 0.5 g) was extracted sequentially using 30 mL distilled water, 0.5 M NaHCO<sub>3</sub>, 0.1 M NaOH, and 1.0 M HCl by shaking the suspension for 16 h, centrifuging for 10 min at 5000 rpm, and passing the supernatant through a 0.45 µm cellulose membrane filter for each extraction step. The first two extraction (distilled water and NaHCO<sub>3</sub> extraction) steps were conducted while purging with nitrogen gas. Acidified potassium persulfate oxidation was used for the digestion of distilled water, NaHCO<sub>3</sub> and NaOH extracts, and the residue was analysed for total P. The difference between total P and inorganic P was estimated to be organic P (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 were grouped into three broad categories as labile, moderately labile and stable fractions. The labile P fraction is a summation of both organic and inorganic P fractions extracted by water (H<sub>2</sub>O-P) and NaHCO<sub>3</sub> (NaHCO<sub>3</sub>-P). Organic and inorganic P extracted by NaOH are moderately labile P which are bound to Fe and Al oxy(hydr)oxides (Fe/Al-P). Inorganic P extracted by HCl are also grouped as moderately labile P which are bound to Ca (Ca-P). Residual P is the stable P fraction bound to primary and secondary P minerals (Niederberger et al., 2019).

#### 2.6. Phosphorus speciation study by thermodynamic modelling

The thermodynamic modelling software Visual MINTEQ 3.1 (Gustafsson, 2013) was used to predict the changes of P species in soils based on the composition of porewater samples across the six submergence events collected at the 2 cm depth. The input parameters entered in the software were soil Eh, pH, DOC, alkalinity, concentrations of cations (Ca, Mg, Na, K, Mn, Al and Fe), anions (Cl<sup>-</sup>, F<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>), 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 (50% each) in porewater.

#### 2.7. Overall redox status

The overall redox status of the soil solution during each ponding event was calculated by summing up *pe* and pore water pH values (*pe* + pH). *pe* is a measure of electron activity calculated using the Nernst equation (Lindsay, 1979);

$$pe = \frac{Eh \times F}{2.303 \times R \times T}$$

where, the Eh is the redox potential measured at 2 or 10 cm depth (V), F is the Faraday constant (96,485 Coulombs), R is the universal gas constant (8.314 J mol<sup>-1</sup> K<sup>-1</sup>) and the T is the absolute temperature in Kelvin (298.15 K, as the average daytime temperature was 25 °C).

#### 2.8. Statistical analysis

Statistical analysis was performed using SAS 9.4 software. Porewater DRP, pH, Eh and cations were analysed for the six repeated submergence events separately using three-way ANOVA to 1) compare soils, 2) two depths (2 and 10 cm), and 3) point of submergence (the beginning and the end of submergence). A two-way ANOVA was performed to compare the pondwater DRP of the soils at the beginning and at the end of

submergence separately for each submerged event. To identify the factors governing the porewater DRP, principal component analysis and correlation analysis were performed. Principal component analysis was performed separately for the three soils using DRP, DOC, Eh, pH,  $pe + pH$ , and cations (Fe, Al, Mn, Ca and Mg) data at the end of submergence. Correlation analyses were performed separately for each soil between DRP concentrations in porewater and pH, Eh,  $pe + pH$ , DOC, porewater Fe, Mn, Ca, Mg and Al concentrations at the end of submergence.

### 3. Results

#### 3.1. Basic soil properties

The three soils were acidic and varied in their P fertility and texture (Table 1). Anion storage capacity was highest in the Allophanic soil followed by the Pallic and Recent soil, respectively. The sum of oxalate extractable Fe and Al was three times and two times higher in the Allophanic soil compared to the Recent and Pallic soils, respectively. The Allophanic soils had the highest total organic carbon (TOC) content at 8.2 g/100g, surpassing the Recent soil by more than 4-fold (2 g/100g) and the Pallic soil by nearly 2-fold (4.7 g/100g).

pH was measured in 1:2 soil: water slurry. Anion exchange capacity was determined by soil equilibration with 1000 mg/L P solution. CEC: Cation exchange capacity, Al<sub>oxalate</sub>: Ammonium oxalate extractable Al, Fe<sub>oxalate</sub>: Ammonium oxalate extractable Fe, Mn<sub>oxalate</sub>: Ammonium oxalate extractable Mn. Ca, Mg, Fe, Mn, Al and Zn were extracted by Mehlich-3 solution.

#### 3.2. Variation of porewater and pondwater pH and soil Eh

Overall, the average porewater and pondwater pH of the six submergence events of the three soils (Supplementary Fig. S1) varied within the neutral pH range (6.7–7.7). The porewater and pondwater pH were similar in the three soils. Immediately after submergence of the first event, soil pH of the three soils increased up to ~7.5. During the submergence events the porewater pH change was minute and the porewater pH change ranged from <0.1 to 0.8.

Soil Eh decreased significantly ( $p < 0.0001$ ) during the submergence events (Supplementary Fig. S1). The Eh of the two soil depths were not significantly ( $p > 0.05$ ) different in any soil at a given submergence event. The average of Eh of the six submergence events at the beginning of submergence were 315 (Recent soil), 398 (Pallic soil) and 443 mV (Allophanic soil) (Supplementary Fig. S1). At the end of submergence, the average Eh were 30 (Recent soil), -25 (Pallic soil) and -4 mV (Allophanic soil).

The redox zones are defined as oxic ( $pe + pH > 14$ ), suboxic ( $pe +$

pH = 9–14), and anoxic ( $pe + pH < 9$ ). After 1 h of submergence, the overall redox status of the Recent soil was suboxic except in the E1 where it was oxic while the other two soils were in the oxic range with few exceptions (Fig. 2). At the end of three days of submergence, all three soils were in the anoxic range except the 2 cm depth of the Recent soil at the E2 (Fig. 2). The  $pe + pH$  values varied from ~11 to ~15 across the six submergence events at the two soil depths of the Recent soil at the beginning and it varied from ~6 to ~10 at the end of submergence. Comparatively, the  $pe + pH$  values of both depths of the Pallic and Allophanic soils changed from ~13 to ~16, respectively at the beginning and ~5 to ~9, respectively at the end of submergence (Fig. 2).

#### 3.3. Variation of porewater and pondwater DRP during submergence

The interaction effect between the two factors “soil” and “point of submergence” was significant ( $p < 0.05$ ) for porewater DRP for all the submergence events, indicating that the change in porewater DRP concentrations due to submergence varied among soils. In the Recent soil, porewater DRP concentration at the end of submergence was 2.4–5.1-fold higher than that at the beginning of submergence for all six events, indicating the release of soil P during submergence. In the Pallic soil, the magnitude of P release was ~2.3-fold less than that in the Recent soil. In the Allophanic soil, submergence did not result in a release of soil P as indicated by the statistically similar DRP concentrations at the beginning and end of submergence for all of the events. The three way interaction (soil  $\times$  point of submergence  $\times$  depth) or two-way interaction with depths (point of submergence  $\times$  depth) were not significant for any submergence events. Porewater DRP concentrations were significantly ( $p < 0.05$ ) different in the 2 and 10 cm depths only for three of the submergence events (E1, E2, and E5.) Porewater DRP concentrations were 1.5, 1.6 and 1.4-fold higher at 2 cm than at 10 cm for the E1, E2 and E5, respectively.

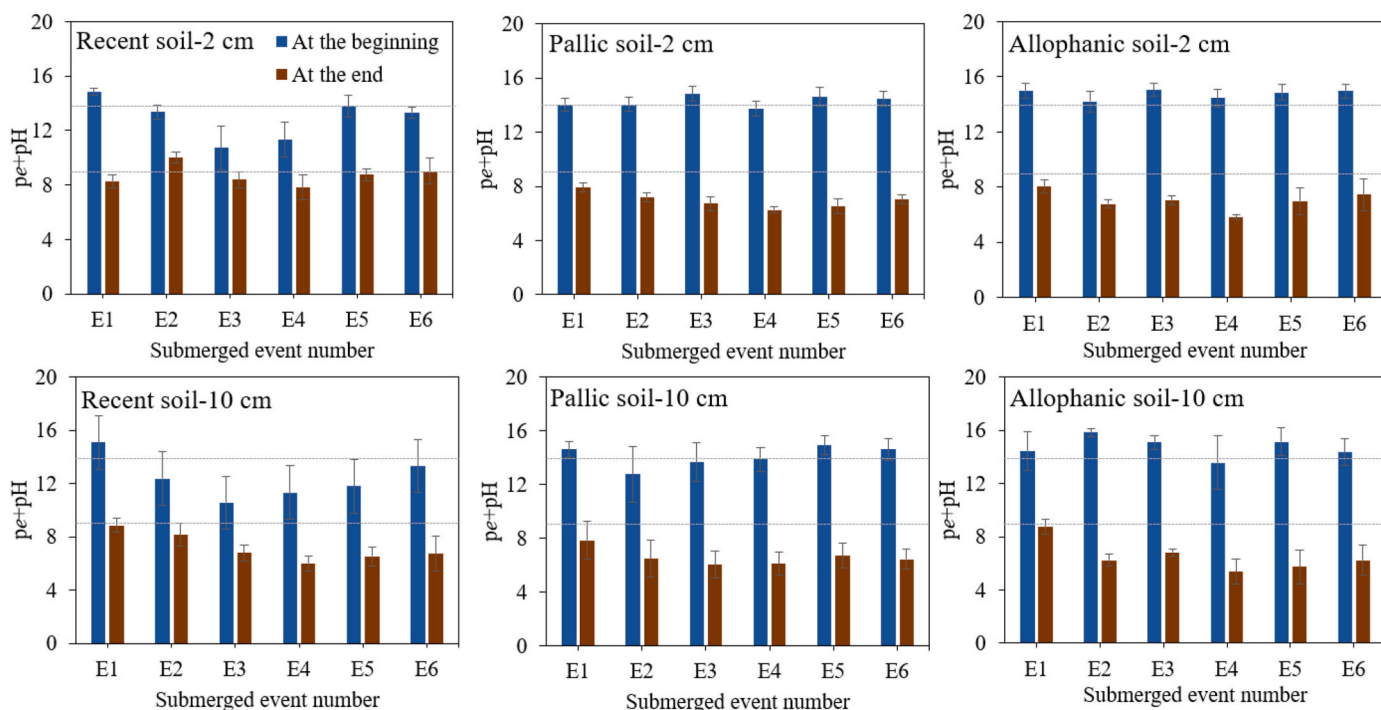
The interaction effect between the two factors “soil” and “the point of submergence” was not significant for pondwater DRP for all the submergence events, except E4. The pondwater DRP among the three soils were significantly ( $p < 0.05$ ) different from each other in the six submergence events. The tap water P concentration at the beginning of the six submergence events were 0.17, 0.22, 0.08, 0.07, 0.03, 0.16 mg/L, respectively. Since the same source of tap water was used for all replicates across the three soil types, the effect of the tap water was consistent among the soils types and replicates. The average pondwater DRP at the beginning and at the end of submergence across the six submergence events were 0.72, 0.52 and 0.19 mg/L for the Recent, Pallic, and Allophanic soils, respectively. The DRP concentration in porewater at both the 2 and 10 cm depths showed significant positive correlations ( $p < 0.01$ ) with pondwater DRP both at the beginning and at the end of submergence, except for the Allophanic soils where only the DRP at 2 cm and pondwater at the end of submergence was not significantly correlated (Supplementary Table S1).

#### 3.4. Variations in porewater ion concentrations and the relationship of porewater ion concentrations with porewater DRP and soil redox status

The average porewater  $NO_3^-$  concentration measured from the two depths from the three soils ranged from 0.06 to 0.50 mg/L at the beginning of the submergence in the first ponding event (Supplementary Fig. S3). Thereafter, the porewater  $NO_3^-$  concentrations decreased throughout the events in all depths of the three soils, with few exceptions. In the Recent soil, porewater Mn concentration at the 2 and 10 cm depths increased with submergence, but a significant ( $p < 0.05$ ) increase was recorded only in E1, E2 and E4 (Figs. 4 and 5). The porewater Mn concentration increased significantly ( $p < 0.05$ ) with submergence in both depths of the Pallic soil, with exceptions in E4 and E6 where the increase was not significant (Figs. 4 and 5). In the Allophanic soil, Mn concentration was significantly ( $p < 0.05$ ) increased only at 2 cm depth

**Table 1**  
Initial physico-chemical properties of the Recent, Pallic and Allophanic soils.

	Recent soil	Pallic soil	Allophanic soil
pH	6.5	5.8	5.9
Sand %	39	14	8
Silt %	43	54	57
Clay %	18	32	35
Olsen P (mg/L)	69	66	32
Anion storage capacity %	19	29	56
CEC (cmol <sub>(+)</sub> /kg)	12	22	23
Total organic C (g/100g)	2.0	4.7	8.2
Fe <sub>oxalate</sub> (mg/kg)	3200	4910	6010
Al <sub>oxalate</sub> (mg/kg)	725	1210	6455
Mn <sub>oxalate</sub> (mg/kg)	80	315	485
Mehlich-3 extractable cations (mg/L)			
Ca	1233	1735	1555
Mg	170	254	216
Fe	681	545	180
Mn	46	75	43
Al	568	800	1278
Zn	2.9	10.7	2.9



**Fig. 2.** Variation of  $pe + pH$  measured from the 2 and 10 cm depths at the beginning and end of six submergence events (E1-E6). Redox zones are indicated by dashed lines: oxic ( $pe + pH > 14$ ), suboxic ( $pe + pH = 9-14$ ), anoxic ( $pe + pH < 9$ ). The error bars represent the standard errors of the means ( $n = 5$ ). At the beginning: within an hour after submergence, at the end: three days after submergence.

in E2 and Mn concentrations during the other submergence events were not changed. Porewater Fe concentration at the two depths of the three soils increased with submergence (Figs. 4 and 5). A significant ( $p < 0.05$ ) increase of Fe concentration was observed at the last two submergence events of the three soils compared to the Fe concentration at the beginning of submergence (Figs. 4 and 5). The increase of porewater Fe concentration in the Recent soil was significant ( $p < 0.05$ ) in both the 2 and 10 cm depths in E1, E2, E5 and E6. The porewater Ca and Mg concentrations measured from both depths of the three soils increased significantly ( $p < 0.05$ ) with submergence, with few exceptions in the Recent soil (Figs. 4 and 5). The Allophanic soil showed the highest average porewater Al concentration among the three soils after submergence which were 0.06 mg/L (2 cm) and 0.03 mg/L (10 cm). The average DOC concentration of the 2 cm depth across the six submergence events of the three soils varied from 37 (Pallid soil) to 48 mg/L (in both Recent and Allophanic soils) (Figs. 4 and 5).

The porewater DRP concentrations showed significant correlations with several soil parameters. The porewater DRP of the Recent soil and Pallid soil showed significant negative correlations with Eh and  $pe + pH$  and significant positive correlations with porewater Ca and Mg (Table 2). Additionally, in the Recent soil, porewater DRP was positively

correlated ( $r = 0.497, p < 0.01$ ) with DOC and in the Pallid soil, porewater DRP was positively correlated ( $r = 0.235, p < 0.05$ ) with porewater Mn. In contrast, the porewater DRP of the Allophanic soil showed significant positive correlations with pH, Eh and  $pe + pH$  and significant negative correlations with porewater Ca and Fe (Table 2).

The principal component (PC) analysis of the Recent, Pallid and Allophanic soils including both depths reduced the dimensionality of the data sets into 2, 3 and 3 principal components (eigenvalue > 1), respectively (Supplementary Table S3). In the Recent soil, the variability attributed to PC1, which accounted for 45%, highlighted the relationships among DRP, DOC, porewater Ca, Mg, and Mn concentrations, Eh and  $pe + pH$ . While the PC2 attributed to 17% variability of the dataset, elucidating the relationship among  $pe + pH$ , pH, Eh, porewater Fe, and Al. In the Pallid soil, PC1 attributed to 54% of the variance highlighting the relationships among pH, Eh,  $pe + pH$ , DOC, porewater Ca, Mn and Mg. The PC2 and PC3 of the Pallid soil attributed to 15 and 11%, respectively. In the Allophanic soil, the variability attributed to PC1 was 42% which highlighted the relationships among Eh,  $pe + pH$ , DOC, porewater Ca, Mn, and Mg. The PC2 and PC3 attributed to 18 and 13% of the variability of the data set, respectively.

### 3.5. Transformation of soil P fractions during submergence

In the Recent soil, P in the  $\text{NaHCO}_3\text{-P}$ , Ca-P, and stable-P fractions significantly ( $p < 0.05$ ) changed, while in the Pallid soil, the changes in P occurred only in  $\text{H}_2\text{O-P}$  and stable P fractions (Fig. 6). In the Allophanic soil, significant ( $p < 0.05$ ) changes in P occurred in the  $\text{H}_2\text{O-P}$ ,  $\text{NaHCO}_3\text{-P}$ , Fe/Al-P and stable P fractions (Fig. 6). In the Recent soil, the  $\text{NaHCO}_3\text{-P}$  fraction reduced by 42% in the 2–8 cm depth, Ca-P fraction reduced by 12% at the 0–2 cm depth and the stable P fraction reduced by 66 and 69% at the 0–2 and 8–10 cm depths, respectively. In the Pallid soil, the  $\text{H}_2\text{O-P}$  fraction increased by 203% at the 0–2 cm depth and again decreased by 42% towards the end of the experimental period. In addition, in the Pallid soil, the stable P fraction decreased by 38% at the 2–8 cm depth. In the Allophanic soil, at the 0–2 cm depth,  $\text{H}_2\text{O-P}$ ,

**Table 2**  
Correlation coefficients between porewater DRP and pH, Eh,  $pe + pH$ , DOC and selected cations at the end of submergence.

Soil	Recent soil	Pallid soil	Allophanic soil
pH	0.049	0.009	0.474**
Eh	-0.528**	-0.340**	0.498**
$pe + pH$	-0.496**	-0.319**	0.528**
DOC	0.479**	0.185	-0.229
Ca	0.528**	0.327**	-0.331**
Fe	0.153	0.130	-0.262*
Mn	0.235*	0.180	-0.201
Mg	0.450**	0.380**	-0.125
Al	0.171	0.194	-0.196

\*\* $p < 0.01$ ; \* $p < 0.05$ .

NaHCO<sub>3</sub>-P and Fe/Al-P fractions increased by 172, 66 and 47–78%, respectively. Comparatively, in the Allophanic soil, the NaHCO<sub>3</sub>-P fraction at the 2–8 and 8–10 cm depths decreased by 48 and 64%, respectively. In addition, in the Allophanic soil, the stable P fraction at the 8–10 cm depth decreased by 32% (Fig. 6).

The total organic P fraction (summation of water, NaHCO<sub>3</sub> and NaOH extractable organic P fractions) of the 0–2 cm depth of the Recent soil decreased by 32% at the middle of the third submergence event and it increased by 26 and 83% in the Pallic and Allophanic soils, respectively (Supplementary Fig. S7). The increase in total organic P in the Allophanic soil was significant ( $p < 0.05$ ).

Supplementary Tables S4–S6 show the saturation indices (SI) of P minerals, Fe/Mn carbonates and selected Fe and Al oxy(hydr)oxides of the three soils. The SI is an index which shows if porewater will tend to dissolve or precipitate a particular mineral. Positive SI indicates the supersaturation of solution, thermodynamically favouring its formation, while negative SI indicates undersaturation of the solution, thermodynamically favouring its dissolution or absence. A positive SI may not necessarily mean that the mineral is present in soil, as the slow precipitation kinetics may intervene the precipitation (Voigt et al., 2018).

Porewater was supersaturated with FCO<sub>3</sub>-Apatite and MnHPO<sub>4</sub> in all three soils throughout the submergence period (Supplementary Tables S4–S6). In addition, porewater was supersaturated with hydroxyapatite in both Recent (Supplementary Table S4) and Pallic soils (Supplementary Table S5) throughout the submergence period but became undersaturated with time in the Allophanic soil (Supplementary Table S6). Formation of vivianite and siderite (Fe(II) minerals) was observed in the Recent and Pallic soils and siderite in the Allophanic soil towards the last submergence events (Supplementary Tables S4–S6). Supersaturation with some other common Fe/Al oxy(hydr)oxides, namely, hematite, and magnetite occurred in all the three soils throughout the submergence period. Undersaturation with  $\beta$ -Ca<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub> with submergence event was observed in the Recent soil.

#### 4. Discussion

The magnitude of P release across the submergence events of the three contrasting soils under short-term frequent submergence was different. The Recent soil released P to porewater at both depths and to overlying pondwater during the 3-day submergence. The Pallic soil released P to porewater at both depths but did not change pondwater DRP during submergence. In contrast to the Recent and Pallic soils, the Allophanic soil sorbed P in pondwater as well as in porewater with submergence, suggesting this soil was effective at mitigating P loss to water.

##### 4.1. Effect of pH and Eh on DRP release

The pH and Eh are two factors governing P release in submerged soils. The pH of most soils tends to change toward the neutral range (6.5–7.5) upon submergence (Patrick and Reddy, 1978). Comparably, in this study, the average pH of porewater and pondwater of the three soils varied within a neutral pH range (Supplementary Fig. S1). Marschner (2021) stated that in neutral soils (pH = 7), in the presence of equal concentrations of oxidant and reductant, Mn(IV) ions reduce to Mn(II) ions within the redox range of 200 to 100 mV, and Fe(III) ions reduce to Fe(II) ions within the redox range of 0 to –100 mV. At the end of three days of submergence, the three soils had surpassed the Mn(IV) reduction redox stage (Supplementary Fig. S1). In further support of this, the three soils showed significant ( $p < 0.05$ ) increases in dissolved Mn in porewater with submergence (Figs. 4 and 5). Further, the Recent soil showed a significant positive correlation ( $r = 0.235$ ,  $p < 0.01$ ) between Mn and porewater DRP, suggesting the possible contribution of Mn reduction and subsequent release of P associated with Mn minerals to porewater, resulting in an increase in the DRP.

Although the Eh of the three soils reduced with submergence, no

significant amount of Fe bound P had released due to the short-term submergence period as evidenced by P fractionation (Fig. 6) and modelling results (Supplementary Tables S4–S6). The soils were back to oxidised status after the 3-day submergence (Supplementary Fig. S1), which might have prevented significant reductive dissolution of Fe minerals. The Eh reduction rate of a submerged soil depends on several factors. For example, soils with low contents of reducing Fe and Mn and high organic-matter content can lead to a rapid decline in Eh (Fageria et al., 2011). On the contrary, the presence of NO<sub>3</sub><sup>-</sup> or Mn(IV)/(III) which are preferable by microbes can prevent the redox potential declining further (Smith et al., 2021). Further, abundance of Fe reducing bacteria affects Fe reduction (Li et al., 2021), which affects the rate of Eh reduction under submergence. The decreasing porewater Mn concentration and increasing Fe concentration towards the last two submergence events (E5 and E6) suggested the initiation of Fe(III) reduction, possibly due to exhausting the reducible Mn ion pool in soils (Figs. 4 and 5). This was further evidenced by a tendency towards the formation of vivianite and siderite at the last two submergence events for the three soils (Supplementary Tables S4–S6).

In most redox-related mineral transformations in soil (eg: Fe and Mn mineral transformations), the redox is poised at characteristic/pe + pH values (Lindsay and Sadiq, 1983). Therefore, those values are used to identify mineral transformations in soil. The first redox poise of the Fe minerals starts at pe + pH of 14.04 and the transformation of Mn minerals happens even at pe + pH value of 16.62 (Lindsay and Sadiq, 1983). Since the pe + pH values of the three soils across the submergence events were below 10 at the end of the submergence, it can be stated that there was a favourable condition in soils for both Mn and Fe mineral transformations.

##### 4.2. Phosphorus release under short-term and frequent submergence

The magnitude of DRP release to porewater and subsequently to pondwater increased in the order of Allophanic < Pallic < Recent soils. Previous studies suggested that algal growth requires total P concentrations >0.02 mg/L, and DRP concentrations beyond 0.01 mg/L, when other growth requirements are met (Owens and Shipitalo, 2006). The average pondwater DRP of the three soils across the six submergence events during submergence were 17 to 65-fold higher than the New Zealand lowland river target for DRP concentration; 0.01 mg/L (ANZG, 2018) which is a significant elevation. In this study, the released P from soil accumulated in the overlying water (~2L of volume), but this does not necessarily represent the field condition where a continuous surface flow would be present. However, under field conditions, soil will not be the only source of dissolved P as there will be other P sources such as inorganic P from recent fertiliser application and dissolved organic P from manure (Reid et al., 2018). Therefore, these significant releases of dissolved P from both the Recent and Pallic soils have to be considered in CSA management.

There was no significant difference between the porewater DRP change during submergence and soil depths of any of the soils. Phosphorus translocation through the soil profile (0–10 cm) was also not evident by the P fractionation analysis (Fig. 6). Therefore, the results suggested that the 10 cm layer is releasing P upon submergence in both the Recent and Pallic soils at similar concentrations as the 2 cm layer. These results suggested that although the 0–2 cm soil depth is generally considered as the primarily contributing layer to P runoff via desorption (Vadas et al., 2007a; 2007b), the 10 cm layer is equally important in releasing P upon submergence at least for the Recent and Pallic soils. Similarly, Smith et al. (2021) previously recorded a maximum total dissolved P concentration of 0.12 mg/L at a depth of 20 cm in a Melanic Orthic Gley soil from a dairy farm in New Zealand during winter. Although there will not be a direct interaction of porewater DRP at 10 or 20 cm depths with the surface runoff, the porewater DRP at those depths contribute to the subsurface lateral flow and ultimately transporting DRP to lowland freshwater bodies (McDowell et al., 2004).

Partial drying of previously submerged soils and alternative oxic-anoxic conditions exhibited due to water management processes in this experiment may have influenced P release in these soils as reported in previous studies (Schönbrunner et al., 2012; Kraal et al., 2015). For example, the dissolved Fe during the reduction process under submergence is precipitated as amorphous Fe minerals during re-oxidation. Before those minerals undergo crystallinity, the following reduction event dissolves them. Therefore, alternative oxidation and reduction increase the amorphous Fe oxyhydroxides in soil which have a high affinity to bind P and thus can affect P release (Baldwin and Mitchell, 2000). In support of this, it was evident that the oxalate extractable (amorphous) Fe, Al, and Mn concentrations increased with submergence (Table 1 and Supplementary Table S7) in this experiment. This may be one reason among other reasons such as removal of P with draining water, for the significant ( $p < 0.05$ ) decrease in DRP concentration recorded in the Recent soil in the later submergence events (Fig. 3).

Over the long-term, the dissolution of P species will be the rate limiting step in P release in soil while in the short-term, desorption processes are likely to dominate (McDowell and Sharpley, 2003). However, differentiation of P release by the two processes would be difficult. Adsorption and desorption of P from minerals is pH and redox sensitive (Schönbrunner et al., 2012; Oxmann and Schwendenmann, 2015). Phosphorus sorption/desorption process in soil can be influenced by several factors. Phosphorus can be associated with Fe and Al in the mineral form or can sorbed onto Fe and Al oxides by exchange with hydroxyl groups or associated with Ca or adsorbed onto calcium carbonate minerals (McDowell and Condon, 2001). Phosphorus can also

be associated with organo-mineral complexes (Gerke, 2010) or sorbed onto clay mineral surfaces such as illite and montmorillonite (Gimsing and Borggaard, 2002). In this study, although porewater Ca and Mg concentrations of three soils significantly ( $p < 0.05$ ) increases with submergence and they showed significant positive correlations (Table 2) with porewater DRP (especially in the Recent and Pallic soils), neither the P fractionation or modelling data supported the dissolution of Ca and/or Mg-P and subsequent release of P. Since there was no clear evidence of reductive dissolution of Fe and subsequent release of P as explained in 4.1 section, the most probable mechanism of P release of the three soils under 3-day submergence may be sorption/desorption other than the reductive dissolution of Mn.

### 4.3. Risk of P release to runoff

The soil water extractable P fraction is commonly used as a measure of labile P susceptible to risk of P loss via runoff (Castillo and Wright, 2008; Pietrzak et al., 2020). Given the 0–2 cm layer of soil is a critical depth in terms of contributing to P runoff via desorption (Vadas et al., 2007a), it is important to measure the changes of P fractions in that layer upon short-term submergence. However, the P fractionation method used in this experiment may not have been sensitive enough to detect the changes in P fractions (Attanayake et al., 2022) as most of the changes in P fractions were not significant during the short-term submergence (Fig. 6). The decrease in the H<sub>2</sub>O-P and other fractions with submergence in the Recent soil suggests that P in all the fractions (labile, moderately labile, and stable P) is released from this soil upon

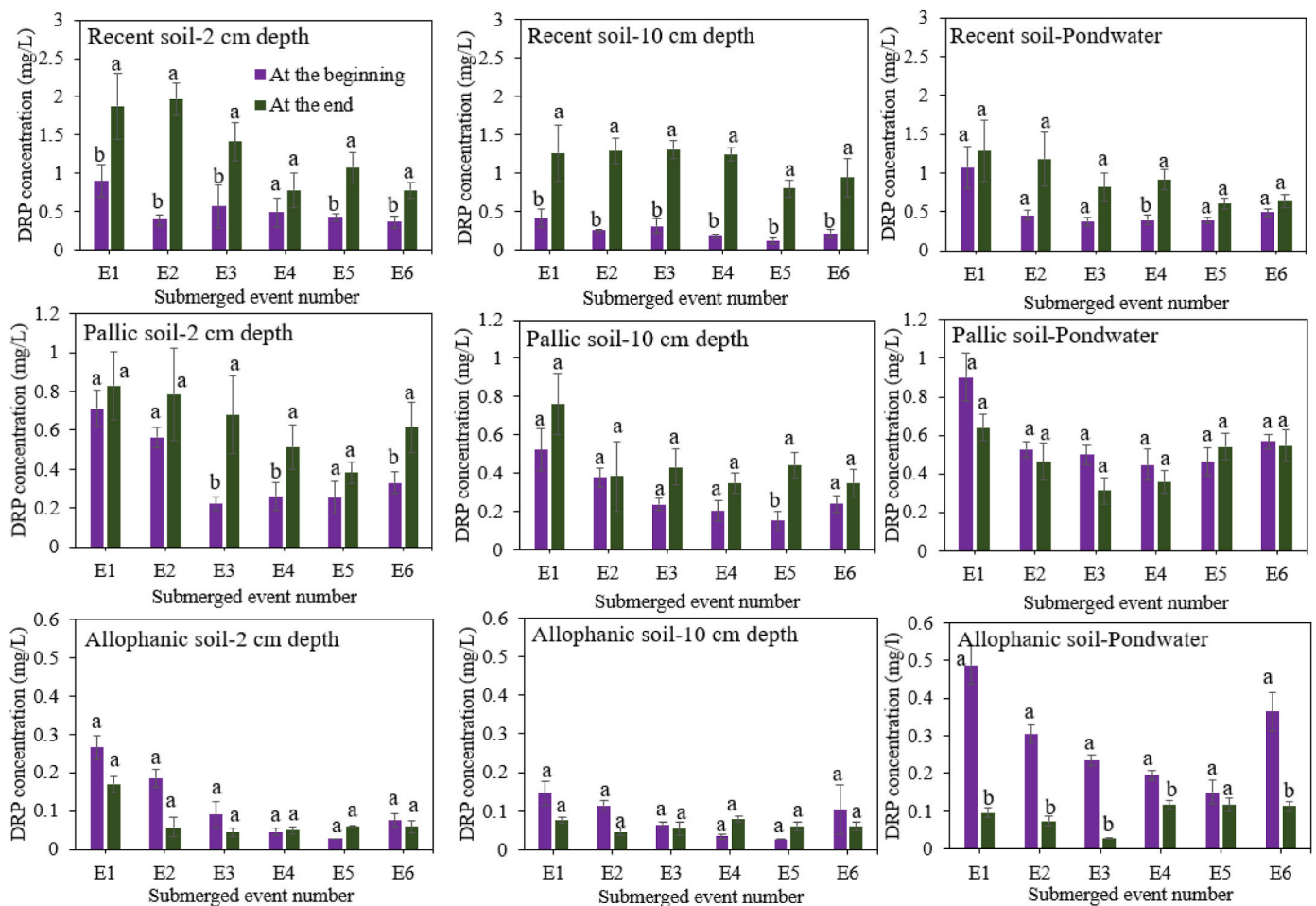
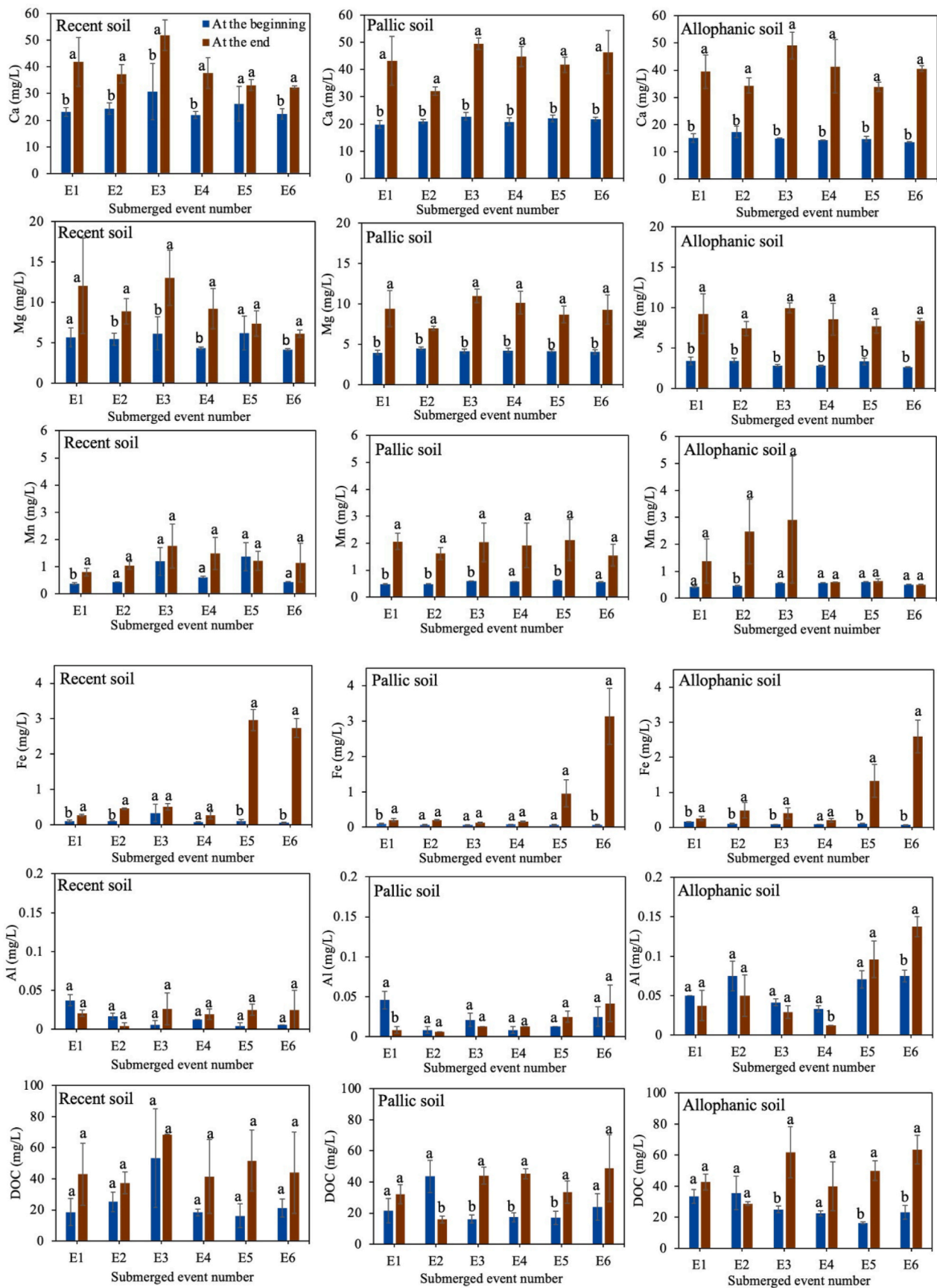


Fig. 3. Variations of dissolved reactive phosphorus in porewater measured from the 2 cm and 10 cm soil depths and pondwater measured from the Recent, Pallic and Allophanic soils at the beginning and at the end of six submergence events. The error bars represent the standard errors of the means ( $n = 5$ ). Different scales are used for the DRP concentration (Y axis) of the three soils. At the beginning: within an hour after submergence, at the end: three days after submergence.



**Fig. 4.** Porewater concentrations of selected cations (Mn, Fe, Ca, Mg and Al) and dissolved organic carbon (DOC) measured from the 2 cm depth following six submergence events in the Recent, Pallid and Allophanic soils. The error bars represent the standard errors of the means. At the beginning: within an hour after submergence, at the end: three days after submergence.

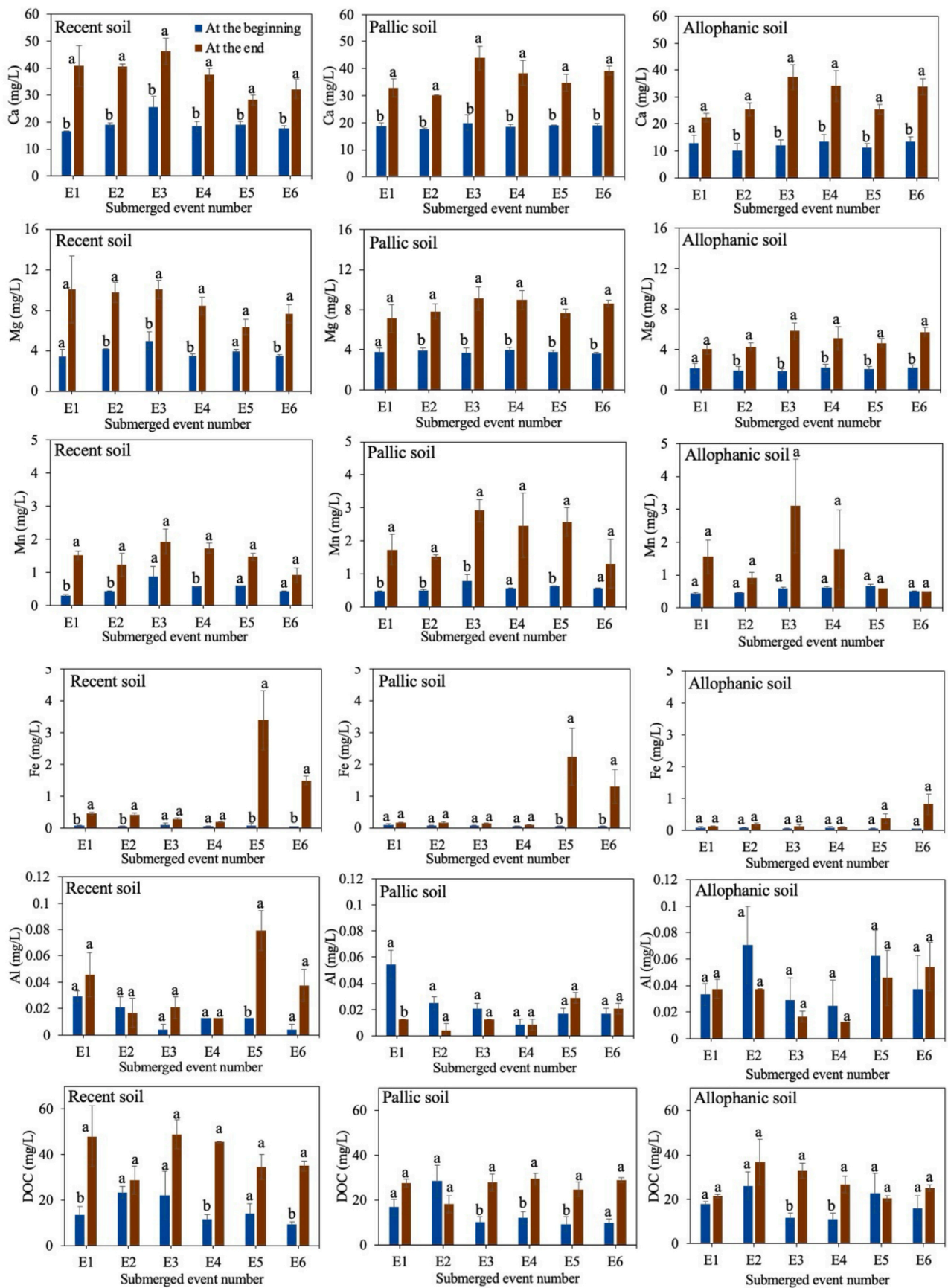
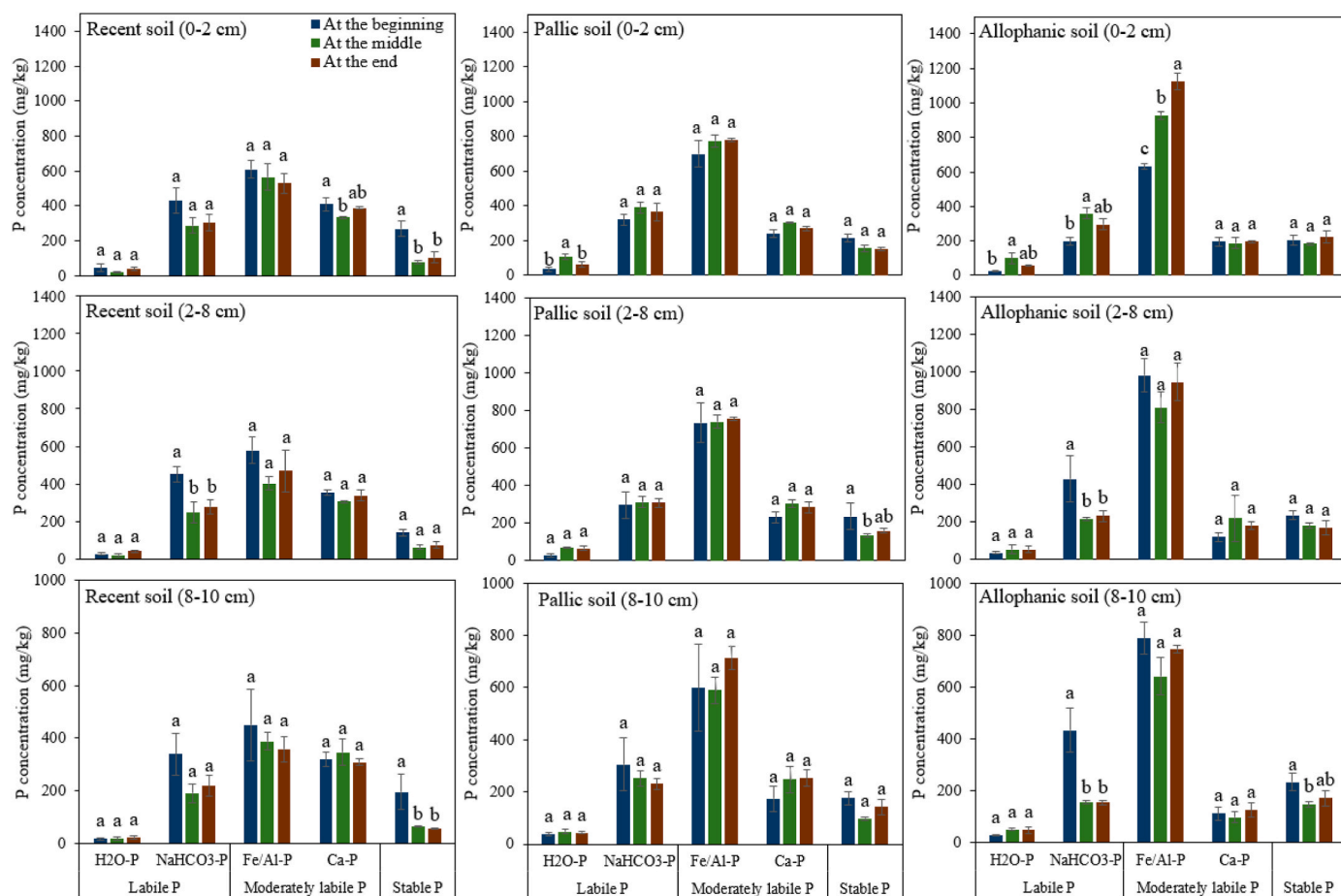


Fig. 5. Porewater concentrations of selected cations (Mn, Fe, Ca, Mg and Al) and dissolved organic carbon (DOC) measured from the 10 cm depth following six submergence events in the Recent, Pallid and Allophanic soils. The error bars represent the standard errors of the means. At the beginning: within an hour after submergence, at the end: three days after submergence.



**Fig. 6.** Labile (water extractable (organic and inorganic) P: H<sub>2</sub>O-P, sodium bicarbonate extractable (organic and inorganic) P: NaHCO<sub>3</sub>-P, moderately labile P (Fe/Al (organic and inorganic)-P, Ca-P) and stable P fractions of the 0–2, 2–8 and 8–10 cm depths in the Recent, Pallid and Allophanic soils at the beginning, at the middle and at the end of the experimental period. The error bars represent the standard errors of the means ( $n = 3$ ). At the beginning: before commencing the experiment, at the middle: after 24–36 h of the third submerged event, at the end: at the end of the sixth submerged event.

submergence and this coincides with increased P release to porewater and pondwater in the Recent soil (Supplementary Table S2). Similarly, the reduction of the water extractable P fraction was evident in other two depths (2–8 and 8–10 cm) of the Recent soil indicating the potential risk of subsurface P loss. Therefore, in combining the fractionation results with the porewater and pondwater DRP concentrations, it was clear that having Recent soil in CSAs is riskier under short-term frequent submergence.

In contrast, the Pallid and Allophanic soils, significantly ( $p < 0.05$ ) increased the water extractable P suggesting the retention of P in the most labile P fraction rather than releasing them to porewater. According to literature, the clay mineralogy of the Pallid soil is dominated by illite (Hewitt et al., 2021). Therefore, clay minerals and the presence of high amounts of Fe and Al minerals might have controlled P sorption/desorption in the Pallid soil. The increase in the water-soluble P fraction at the 0–2 cm depth of the Allophanic soil may be attributed to the sorption of P from the added tap water. Further, the increases of the same fraction in the deeper soil depths (2–8 and 8–10 cm) of the Allophanic soil may be attributed to the transformation of P from the NaHCO<sub>3</sub>-P fraction, which reduced (Fig. 6). However, in the Allophanic soil, the significant ( $p < 0.05$ ) increase of Fe/Al-P at the 0–2 cm depth with time suggests that the possible transformation of the labile P into moderately P fraction decreased the risk of P release to runoff (Fig. 6). However, it was observed that both labile P and Fe/Al-P fractions decreased with submergence in soil layers below 2 cm in the Allophanic soil (Fig. 6). This may be an indication that P present below 2 cm may have migrated to the top layer due to a concentration gradient (high

concentration in the deep soil and low concentration in the shallow soil) and accumulated (resorbed) in the top layer.

Allophanic soils are dominated by allophane and also imogolite or ferrihydrite and aluminium or iron humus complexes (Hewitt et al., 2021). Supersaturation of Al oxides such as Al(OH)<sub>3</sub>, boehmite, diaspore and gibbsite, may be contributed to low dissolved P concentration in the Allophanic soil by sorbing or precipitating phosphates in porewater (Supplementary Table S6). The high amounts of amorphous Fe and Al might have played a significant role in sorbing P in the Allophanic soils as evidenced in previous studies (Hashimoto et al., 2012; Hosokawa et al., 2022). The significantly ( $p < 0.05$ ) increased total organic P fraction recorded in the Allophanic soils (Supplementary Fig. S6) upon submergence in the present study, might be due to the high TOC (8.2%) content of the soil and the presence of aluminosilicates which allows retention of large quantities of soil organic P (Redel et al., 2016).

In contrast to the Recent soil, volcanic soils (eg: Allophanic soils) usually have a high ASC and have the ability to sorb high amounts of P, minimising losses (Hewitt, 2010). The sum of oxalate extractable Fe and Al is known to be a good predictor of P sorption in soils (Blombäck et al., 2021). Here, the P sorption ability (sum of oxalate extractable Fe and Al) of the Allophanic soil was 2–4-fold higher than that of the Pallid and Recent soils, respectively (Table 1 and Supplementary Table S7). Further, sorption of P added with the tap water by the Allophanic soil was evident in each submergence event. Therefore, this study suggests that Allophanic soils (especially the topsoil layer; 0–2 cm) could be used as P sorbing materials in CSAs to mitigate P losses from farms. However, the success of using Allophanic soils for the above purpose will depend

on its P concentration and P sorption capacity in order to estimate how much of the soil P sorption capacity has already been used (McDowell and Condron, 2004).

## 5. Limitations and implications

This submergence study differed in some ways to a field CSA as water was not flowing over the soil, submergence events occurred in frequent fixed intervals and overlying tap water was not being replenished with sources of P like it would in a field surface runoff situation (eg: dissolved P fertilisers, organic P from manures and dissolved P from runoff water). However, the current design allowed us to control field soil heterogeneity somewhat by maintaining five replicate field soil blocks with actively growing pasture.

This study revealed that three soils located from or adjacent to CSAs with contrasting soil characteristics showed three different P releasing patterns and magnitudes upon short-term frequent submergence. The risk of P loss increased in the order of Allophanic < Pallic < Recent soil. The finding of this study can be applied to edge of farm mitigation practices such as wetlands and detainment bund constructions, especially in selecting suitable sites/soils. Use of Allophanic soils as a P sorbing material was proved to be advantageous in this study, considering the P sorbing capacity of such soils. The main mechanisms of P release in the Recent and Pallic soils under 3-day submergence may be desorption of P sorbed to minerals (eg: Ca, Mg, Fe, Al and Mn minerals) and reductive dissolution of Mn-P minerals. According to literature, the reduction of Fe and consequent phosphate release starts after the reduction of the entire nitrate pool which was completed three days after the submergence of soils (Reddy and DeLaune, 2008). There is a possibility of an occurrence of iron reduction and concomitant P release under field condition in CSAs during rainfall induced submergence events. Therefore, it is important that further research focusses on the collection of field data and verification of the findings of this study.

## CRedit authorship contribution statement

**Janani Palihakkara:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Lucy Burkitt:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Paramsothy Jeyakumar:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Conceptualization. **Chammi P. Attanayake:** Writing – review & editing, Visualization, Supervision, Resources, Methodology, Conceptualization.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Janani Palihakkara reports financial support was provided by AHEAD Scholarship for PhD study, Sri Lanka. Lucy Burkitt, Jeyakumar Paramsothy, Janani Palihakkara reports financial support was provided by Kathleen Spragg Research Award, New Zealand. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

The authors are thankful for the financial support from the Kathleen Spragg Research Award of New Zealand for the research and the World Bank funded AHEAD Scholarship (AHEAD/PhD/R3/Agri/336) of Sri Lanka for the PhD study of Janani Palihakkara.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2025.124317>.

## Data availability

Data will be made available on request.

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