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Title Saltwater intrusion affects nitrogen, phosphorus and iron transformations under oxic and anoxic conditions: An incubation experiment

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Conflicts of interest/Competing interests

The authors declare no conflict of interest

Availability of data and material

Data are available upon request to authors.

Code availability

All R code used to analyze the data in this manuscript is available upon request to authors.

Abstract

Low-lying coastal ecosystems are rapidly salinizing due to sea level rise and associated saltwater intrusion (SWI). In agricultural soils, SWI can alter biogeochemical cycling of key nutrients such as nitrogen (N), phosphorus (P), and iron (Fe). The main objective of this study was to quantify the amount of nitrate-N ($\text{NO}_3\text{-N}$), ammonium-N ($\text{NH}_4\text{-N}$), soluble reactive P

(SRP), and total dissolved iron (TDFe) released from agricultural soils undergoing SWI to determine their potential loss to downstream waterways. Agricultural soils were incubated for 0, 15, and 30 days (under oxic and anoxic conditions) with various salt solution combinations of sodium chloride, sodium sulfate (Na_2SO_4), calcium sulfate (CaSO_4), and Instant Ocean® to mimic (1) different ionic constituents of saltwater at different ionic strengths and (2) the presence or absence of gypsum, a soil amendment, through the addition of CaSO_4 . We also included a set of incubations treated with deionized water as a no ionic strength control ($0.00M$). To increase statistical power, we grouped individual salt treatments based on our initial hypotheses at the end of the incubation period (day 30) to determine the effects of high ($0.26\text{--}0.28M$) and low ($0.03\text{--}0.04M$) ionic strength on inorganic N release and combinations of Ca and SO_4^{2-} additions on SRP release to microcosm soil solution. Calcium additions decreased SRP release relative to saltwater that contained only sodium chloride (NaCl) additions under oxic and anoxic conditions. Additionally, high ionic strength treatments, which were about 7 times the ionic strength of low ionic strength treatments, released two times as much $\text{NH}_4\text{-N}$ to the soil solution, which suggests a non-linear relationship between ionic strength and $\text{NH}_4\text{-N}$ release. At day 30, anoxic microcosm soils treated with Instant Ocean® to simulate 15 ppt seawater (ionic strength $0.26M$) released significantly more $\text{NH}_4\text{-N}$ (by 782 times), SRP (by 6 times), and TDFe (by 197 times) to the soil solution than oxic microcosm soils ($P < 0.05$). This treatment was designed to reflect a field inundated by brackish seawater for almost a month, which is representative of conditions on farm fields undergoing SWI. Under these anoxic conditions, as much as 22% of bioavailable soil P (as SRP) and 45% of total inorganic N (as $\text{NH}_4\text{-N}$) could be released to overlying water when inundated with saltwater. Our work indicates that the influx of salts and inundation of SWI-

69 affected farm fields could lead to a large export of N and P from agricultural soils and potentially
70 affect downstream water quality.

71

72 ***Abbreviations***

73 Ammonium (NH_4^+); ammonium nitrogen ($\text{NH}_4\text{-N}$); calcium (Ca); iron (Fe); nitrate (NO_3); nitrate
74 nitrogen ($\text{NO}_3\text{-N}$); nitrogen (N); phosphate (PO_4^{3-}); phosphorus (P); saltwater intrusion (SWI);
75 sodium (Na); soluble reactive phosphorus (SRP); sulfate (SO_4^{2-}); total dissolved iron (TDFe)

Introduction

Saltwater intrusion (SWI) into freshwater systems is a growing problem in coastal regions worldwide (White and Kaplan 2017; Tully et al. 2019a). Driven by sea level rise, the frequency and intensity of storms and droughts, water extraction for human use, and the connectivity of the landscape to sources of saltwater (e.g. canals, ditches, etc.), SWI can push sea salts inland far beyond the high tide line (Schoepfer et al. 2014; Bhattachan et al. 2018; Tully et al. 2019b). By 2050, global sea levels are expected to rise by 20 to 30 cm (IPCC 2013). The effects of SWI include, but are not limited to, marsh migration, forest die-off, the spread of invasive species, crop yield declines, and nutrient pollution through changes in biogeochemical cycles in soils (Tully et al. 2019a). In this study, we focus on biogeochemical effects of SWI that alter nitrogen (N) and phosphorus (P) cycling in agricultural soils and can lead to pollution in coastal waterways downstream of SWI-affected farm fields (Ardón et al. 2013; Williams et al. 2014).

Many coastal farms are located on former wetlands that were drained for agricultural use (Moomaw et al. 2018). In areas very close to saline water bodies, such as tidal creeks and salt marshes, ditches can regularly flood with saltwater, even during baseflow conditions (Bhattachan et al. 2018). In coastal regions with low elevations, extensive ditch networks were historically carved into the landscape to allow excess water to flow off farm fields (Reddy and DeLaune 2008). However, as SWI moves inland, ditches serve the reverse purpose and frequently act as conduits that allow saltwater to concentrate on fields during high tides, storms, and droughts (Tully et al. 2019a). Saltwater can remain on these fields for days or even weeks and spur major changes in soil chemistry and nutrient release (Weissman and Tully 2020).

Though the theoretical understanding of the biogeochemical processes that lead to nutrient release in soils exposed to inundation and saltwater has been well established (Ardón et al. 2013; Hartzell et al. 2017), the rate and extent of their occurrence vary greatly from site to site depending on prevailing environmental conditions such as soil texture, organic matter content, pH, and temperature (Roman and Burdick 2012). Further, most studies on SWI have been conducted on freshwater wetlands that are becoming salinized (Neubauer et al. 2013; Helton et al. 2014; White and Kaplan 2017). Agricultural soils properties are distinct from freshwater wetland soils, which causes nutrient cycles to behave differently in each of these environments. For example, since wetlands are frequently inundated, nitrate (NO_3^-) is leached quickly from or denitrified in wetland soils, while sulfate (SO_4^{2-}) is consumed by sulfate reduction (Megonigal and Neubauer 2009). In contrast, drier conditions in well-drained agricultural soils allow for accumulation of NO_3^- and SO_4^{2-} (McLay et al. 1991). Because wetland soils usually contain a thicker horizon of carbon (C)-rich organic material than agricultural soils, biological activity is often more C-limited in agricultural soils (Roman and Burdick 2012). Finally, wetland soils tend to be more oxygen depleted and release soluble reactive P (SRP) into solution more readily than well-drained agricultural soils (Nair et al. 2015). Thus, in order to better understand the potential consequences of large-scale SWI in coastal farmlands, we conducted a detailed incubation study with agricultural soil to mimic different aspects of SWI (e.g. oxygen levels, ionic strength, SO_4^{2-} addition, and a soil amendment containing calcium; Ca) and assess their impacts on N and P release to overlying waters.

In SWI-affected areas, N loss from soils and subsequent movement to downstream waterways increases with inundation and ionic strength (Tully et al. 2019a). Because of its negative charge, NO_3^- is repelled by negatively charged soil particles and hence tends to leach

quickly from saturated soils (Goyne et al. 2008). On the other hand, ammonium (NH_4^+) ions tend to readily sorb to exchange sites on agricultural soil but are easily displaced by cations found in saltwater, such as Na^+ . Both of these processes can lead to a large export of N into local waterways (Ardón et al. 2013).

Interactions between SO_4^{2-} and iron (Fe) can enhance P release from SWI-affected soils (Williams et al. 2014). In well-aerated soils, Fe is present in its oxidized form (Fe(III)) within poorly-crystalline oxide minerals that strongly bind phosphate (PO_4^{3-}), the bioavailable form of P (McDowell and Sharpley 2001). However, Fe(III) is biotically and abiotically reduced to Fe(II) under low oxygen conditions, such as in a field that has been inundated. The Fe reduction process releases PO_4^{3-} into the soil porewater (Nair et al. 2013). Unlike freshwater, saltwater is rich in SO_4^{2-} , which provides another terminal electron acceptor in saturated soils. Microbial populations reduce SO_4^{2-} to sulfide (a compound containing one or more S^{2-} ions), which can bind to Fe(II), forming Fe sulfides (FeS_x) in reducing environments (Schoepfer et al. 2014). This process immobilizes Fe and makes it unavailable to immediately rebind P (Caraco et al. 1990; Jordan et al. 2008). Since soils in SWI-affected areas tend to undergo wetting and drying cycles (Tully et al. 2019b), FeS_x could be reoxidized upon soil drying and release hydrogen protons (acid), SO_4^{2-} , and Fe(III), which can then reprecipitate as Fe oxides and rebind PO_4^{3-} (Luther et al. 1992; Roden and Edmonds 1997). However in a SWI-affected system, soil drying and consequently Fe reoxidation reactions would occur relatively slowly compared to the rate at which mobilized PO_4^{3-} tends to diffuse or move with tidal pulses out of porewater of inundated soil, into overlying water, then into downstream water bodies (Tully et al. 2019b).

Other transformations of N and P are also greatly affected by changing soil redox conditions that co-occur with SWI. Aerobic microbial respiration pathways (where oxygen is

reduced) are the most thermodynamically favorable, and thus are the dominant form of respiration in well-aerated (oxic) soil. However, soils are depleted in oxygen as they become saturated, and other respiration pathways begin to dominate. The next most widespread and energetically favorable pathways are NO_3^- reduction, followed by Fe reduction, and SO_4^{2-} reduction (Arndt et al. 2013). In saturated soils, much of the NO_3^- present is rapidly reduced to N_2 gas through the process of denitrification. However, many studies have shown that a large portion of NO_3^- can also be reduced to NH_4^+ through dissimilatory nitrate reduction to ammonium (DNRA), particularly in saline soils (Tobias et al. 2001, Burgin and Hamilton 2007, Giblin et al. 2013).

Finally, soil amendments can also affect soluble reactive P (SRP; a proxy for phosphate) concentrations in soil porewater (Grubb et al. 2012). Calcium is often added to fields as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) to remediate sodic soils (Fowler et al. 2014) and as agricultural lime ($\text{Ca}(\text{OH})_2$) to raise soil pH (Brady and Weil 2016). Because Ca also binds and sequesters SRP in soils, it has been studied for its potential to reduce P pollution in agricultural ditches (Moore and Miller 1994; Grubb et al. 2012). Thus, the addition of CaSO_4 to soil as management response to SWI can also affect soil P bioavailability.

There is a growing body of literature using laboratory incubation (Weston et al. 2011; Williams et al. 2014; Steinmuller and Chambers 2018; Neubauer et al. 2019) studies to assess the effects of SWI on wetland soils. Additionally, in field studies, we have previously demonstrated how the synergistic effects of salinity additions and frequent inundation result in redox-mediated chemicals reactions that drive N and P loss from soils of coastal farmlands (Tully et al. 2019b; Weissman and Tully 2020). However, no prior laboratory incubation studies have examined how incoming saltwater affects nutrient release from agricultural soils undergoing SWI. Therefore,

our incubation study represents a novel effort to quantify N and P release from coastal agricultural soils into downstream waterways under controlled conditions.

The main objective of this study was to quantify the effect of different ionic constituents of saltwater and CaSO_4 on the release of dissolved inorganic N (DIN), SRP, and total dissolved Fe (TDFe) release from an agricultural soil under oxic and anoxic conditions. We also measured total Fe oxides to determine if the amount of Fe dissolved into soil solution caused a significant change in overall pools of soil Fe after 30 days. Because of DNRA and cation exchange on soil particle surfaces, we hypothesized that anoxic high ionic strength treatments would enhance $\text{NH}_4\text{-N}$ release to the soil solution while driving $\text{NO}_3\text{-N}$ concentrations to near-zero levels. Additionally, we expected to observe increased SRP release under anoxic conditions due to Fe and SO_4^{2-} reduction. Finally, because Ca^{2+} readily binds to SRP, we expected that calcium sulfate (+ CaSO_4) treatments would suppress SRP release from both oxic and anoxic soils. To achieve our study objective, we simulated four moderators of nutrient release: (1) change in ionic strength (Ardón et al. 2013); (2) enhanced SO_4^{2-} reduction (Williams et al. 2014); (3) decreased oxygen levels (Nair et al. 2013); and (4) soil amendment with Ca (Grubb et al. 2012). Saltwater is comprised of a variety of ions that can affect biogeochemical cycling in soils such as base cations like sodium (Na^+), calcium (Ca^{2+}), magnesium, and potassium, and the anions SO_4^{2-} and chloride (Cl^-). Combined with lowered oxygen levels in soils that result from inundation, ions have synergistic effects on nutrient release. Our study results can be used to develop improved nutrient reduction targets for ecologically-sensitive coastal areas.

Materials and methods

Study site

In Maryland, the Chesapeake Bay surrounds the western shores of Dorchester, Wicomico, and Somerset counties (Figure 1a). We collected soils on a farm field near Princess Anne, in Somerset County, the southernmost county in Maryland (38.2° N, 75.7° W; Figure 1b). The farm field has been planted in a corn/soy rotation for the past ten years. In 2018, the year prior to soil sample collection, the field was planted in corn. Urea ammonium nitrate fertilizer was applied to the field at a rate of 90 kg N/ha. The field had been limed three years prior with calcium carbonate (CaCO₃). The soil was comprised of a mesic Aquic Hapludult loam mainly from the Manokin soil series (Soil Survey Staff 2018). Additional physical and chemical characteristics of the soil are described in Table 1. We collected soils from the top 10 cm of the agricultural field in April 2019 and stored the field-moist soils at 4°C. The sampling point was 500 meters from the closest salt marsh (Figure 1b), and while neighboring fields had been affected, the focal field did not yet show signs of SWI. The nearby marsh soils were comprised of a combination of Transquaking and Mispillion soil series and had an average salinity of 22 parts per thousand (ppt) in the top 50 cm of the soil porewater (Soil Survey Staff 2018). By 2050, most of the county is expected to undergo regular tidal flooding (Maryland Department of Natural Resources and NOAA 2008). Based on estimates, sea levels are projected to rise 0.34-0.91m near Cambridge, a nearby town in Dorchester County by 2050 (NOAA 2020). The area receives an average of 1040 mm of precipitation annually. The mean annual maximum temperature is 21°C and the mean annual minimum temperature is 6°C (NOAA-NCEI 2018).

209 *Soil solution preparation and analysis*

210 In order to examine the effect of SWI on soil N, P, and Fe dynamics, we created a suite of
211 salt solution treatments to determine the relative effect of different ionic constituents and ionic
212 strength on N, P, and Fe release from soils (Table 2). Throughout this paper, we refer to
213 “release” as the sum of biotic and abiotic processes that spur the movement of ions out of the soil
214 and into the soil solution. Treatments containing calcium sulfate (+CaSO₄ treatments) were
215 designed to simulate gypsum additions to SWI-affected fields. Treatments containing sodium
216 sulfate (+Na₂SO₄ treatments) were intended to simulate SO₄²⁻ addition from saltwater without
217 including the suppressive effects of Ca on SRP release. Treatments containing sodium chloride
218 (+NaCl treatments; 5 treatments total) were designed to determine the effects of high ionic
219 strength (0.26-0.28M) on nutrient release to soil porewater, and the Na₂SO₄ and CaSO₄
220 treatments (2 treatments total) mimicked low ionic strength (0.03-0.04M; Table 2). We also
221 included deionized water (DIW) as a freshwater control and Instant Ocean® Sea Salt (Spectrum
222 Brands, VA, USA) as a treatment to mimic natural seawater.

223 In total, we created eight salt treatment solutions with the ionic strength noted in
224 parentheses: (1) DIW (0.00M); (2) Na₂SO₄ (0.04M); (3) CaSO₄ (0.03M); (4) NaCl + CaSO₄
225 (0.26M); (5) NaCl + Na₂SO₄ (0.27M); (6) NaCl (0.26M); (7) NaCl + Na₂SO₄ + CaSO₄ (0.28M);
226 and (8) Instant Ocean® Sea Salt (0.26M; Table 2). Instant Ocean® Sea Salt is comprised of the
227 major cations Na⁺, K⁺, Mg²⁺, Ca²⁺, Sr⁺, the major anions Cl⁻, SO₄²⁻, and trace metals, all found in
228 a similar ratio to that of natural seawater (Atkinson et al. 1997). The Na₂SO₄, CaSO₄, NaCl +
229 Na₂SO₄, and NaCl + CaSO₄ treatments contained SO₄²⁻ at the same concentration found in 15 ppt
230 (brackish) saltwater (1.14 g SO₄²⁻/l) and the addition of NaCl brought those treatments up to 15
231 ppt total. The 15 ppt salt treatments ranged in ionic strength from 0.26-0.28M. Exact stock

solution mixtures are detailed in Table 2. We selected 15 ppt salinity (and hence high ionic strength) salt treatments for this experiment based on the results of a previous field study we conducted on actively farmed fields undergoing SWI in Somerset County where soil porewater salinity in the top 25 cm of SWI-affected parts of these fields ranged from approximately 5-25 ppt throughout the data collection period (Tully et al. 2019b).

Each microcosm consisted of a slurry of 120 ml of treatment water added to 20 g well-mixed and homogenized field-moist soil and incubated at 25 °C. Microcosm slurries were made in duplicate and one was maintained aerated and the other anoxic to simulate high and low redox potential saturated environments. Though aerated microcosm slurries were also saturated with solution, we herein will refer to them as “oxic” for consistency. Oxic microcosm slurries were created in 250 ml beakers, sealed with gas-permeable membranes and stirred daily, to maintain aeration and prevent SRP from being released due to Fe reduction from FePO_4 complexes under lower redox conditions, and also to prevent the reductive dissolution of Fe oxide minerals, a process that can release sorbed SRP (Musolff et al. 2017). Anoxic microcosm slurries were placed into 125 ml Erlenmeyer flasks and sealed with airtight rubber septa. These slurries were purged for ten minutes with purified N_2 gas. Each oxygen level by salt solution combination was replicated four times for each microcosm soil solution sampling date (day 0, 15, and 30; $n_{\text{total}} = 192$). Microcosms were destructively sampled to avoid changing the total soil and soil solution volume throughout the experiment. To limit the introduction of air to the water sample, anoxic microcosms were sampled via a syringe through the rubber septa. Oxic microcosms were sampled by syringes placed directly into the soil solution. Day 0 microcosms were sampled immediately after they were set up (< 30 minutes after salt treatment was applied).

All microcosm soil solution samples were immediately filtered through 1 μ m glass fiber filters, and frozen until analysis. A subsample of each sample to be analyzed for dissolved inorganic phosphorus (measured as SRP) was acidified to pH \sim < 2 with hydrochloric acid to prevent PO_4^{3-} coprecipitation with Fe upon sample exposure to oxygen. Samples were analyzed colorimetrically on a LACHAT QuikChem (LACHAT Instruments Loveland, CO) using the sulfanilamide method for $\text{NO}_3\text{-N}$, the salicylate-nitroprusside method for $\text{NH}_4\text{-N}$, and the molybdate-blue method for SRP.

All microcosm soil solutions were analyzed for total dissolved Fe (TDFe) using a modified method to reduce all dissolved Fe(III) to Fe(II) (Viollier et al. 2000). A reducing agent, 1.4M hydroxylamine hydrochloride ($\text{H}_2\text{NOH.HCl}$), was prepared in 2M hydrochloric acid. A 10M ammonium acetate ($\text{C}_2\text{H}_7\text{NO}_2$) buffer was prepared and adjusted to pH 9.5 with a solution of ammonium hydroxide (NH_4OH). Then, 2 ml of the reducing agent and 1.67 ml of the buffer was added to 10 ml of the water sample. All microcosm soil solutions were analyzed for Fe, which represents the total dissolved Fe(II) and Fe(III) oxyhydroxides present in the water samples, on an atomic adsorption spectrometer (PinAAcle 900; Perkin Elmer; CT, USA) using an air-acetylene flame.

Soil analyses

Soil samples from the agricultural field were analyzed at Brookside Laboratories in New Bremen, OH, USA to determine baseline soil nutrient levels. Where noted, the Mehlich III method was used to extract bioavailable (inorganic) nutrients (Mehlich 1984; Table 1). A potassium chloride method was used to extract $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ from the soils and extracts were run on a flow injection analyzer (FIAlyzer; FIALab; WA, USA). Inductively coupled plasma optical emission spectroscopy (ICAP 7600; Thermo Fisher Scientific; MA, USA) was

used to quantify concentrations of Ca, Mg, K, Na, B, Fe, Mn, Cu, Zn, and Al in the samples. Concentrations of Cl^- and SO_4^{2-} were water extracted from the soil and measured on an ion chromatograph (Mehtrrom AG, Herisau, Switzerland). Soil pH and EC were measured in a 1:1 water extraction (wt:vol) on a SevenExcellence probe (Mettler Toledo; OH; USA) and soil organic matter percentage was estimated using the loss-on-ignition method (Heiri et al. 2001). Total soil C and N were analyzed at AgroLab in Harrington, DE, USA on a LECO CN-2000 dry combustion analyzer (MI, USA).

At each sampling point in the experiment (day 0, 15, and 30), we determined total Fe oxides in all of the microcosm soils in order to determine whether measurable iron transformations occurred in the overall pool of soil Fe over time. Soil samples from each microcosm were air-dried, ground, and passed through a 2-mm sieve. They were then extracted for total (crystalline + poorly-crystalline) Fe oxides via the dithionite citrate bicarbonate (DCB) extraction method, which uses a powerful reductant, dithionite, to reduce Fe(III) oxides to Fe(II). The supernatants from the extractions were decanted, filtered through Whatman 42 filter paper (2.5 μm) and analyzed for Fe on an atomic absorption spectrometer (PinAAcle 900; Perkin Elmer; CT, USA) using an air-acetylene flame.

Scaling nutrient release to the field level

In the final portion of this study, we calculated potential nutrient release from the soils used for this experiment under oxic and anoxic conditions (top 10 cm of topsoil from the field) in kg/ha as this is a unit that is useful to land managers when making decisions about fertilizer application (measured in kg/ha or lbs/acre). To area-scale concentrations, we used a soil bulk density of 1.55 g/cm³ from Web Soil Survey (Soil Survey Staff 2018) and results from analyte

concentrations for different salt treatment and oxygen treatment combinations at day 30 of the experiment. The calculation used is as follows (Equation 1):

$$\frac{c_x v_x}{m_x} * \frac{1.55g}{cm^3} * d_x * \frac{10^8 cm^2}{ha} * \frac{1kg}{10^6 mg} \quad \text{Equation 1}$$

where x is the particular salt and oxygen treatment, c is the analyte concentration (in mg/l), v is the volume of microcosm soil solution (in l), m is the mass of the dried microcosm soil sample (in g), d_x is the depth to which the soil sample was taken in the field (10 cm) and the last two terms are conversion units for square cm to hectares and mg to kg.

To compare results to initial microcosm soil solution analyte concentrations, values in Table 1 were scaled from mg/kg to kg/ha using a similar equation (Equation 2):

$$a_i * \frac{1.55g}{cm^3} * d_i * \frac{10^8 cm^2}{ha} * \frac{1kg}{10^6 mg} * \frac{1kg}{10^3 g} \quad \text{Equation 2}$$

where a_i is the initial analyte soil solution concentration (in mg/kg), d_i is the depth to which the original soil sample was taken in the field (10 cm) and the last three terms are conversion units.

Statistical approach

To examine differences in NO₃-N, NH₄-N, SRP, and TDFe in microcosm soil solutions and total Fe oxides in microcosm soil samples, we used a linear fixed-effects (LM) model *lme4* package for R (Bates et al. 2018). Salt treatment, oxygen level, and day (0, 15, and 30) were included as fixed effects. We examined the main effect of salt treatment, oxygen level, and day and interactive effects for each variable. For each day sampled, we used *Tukey post-hoc* tests to examine pairwise differences in water chemistry among treatment combinations (*multcomp* package; Hothorn et al. 2017). A Welch's two sample t-test (Welch 1947) was used to determine whether microcosm soil pH in anoxic versus oxic treatments was significantly different at day 30. We used the Box-Cox method (Box and Cox 1964) for log transformations prior to analysis

to meet the assumptions of the statistical model when needed. All statistics were computed in R Studio (R Studio Team 2019).

Oxygen level and individual salt treatments were the initial variables in this study. However, when individual salt treatments were not significantly different, individual treatments were grouped to test the effects of ionic strength in the case of the different N species and combinations of Ca and SO_4^{2-} additions in the case of SRP at the end of the incubation period (day 30). Because we hypothesized that higher ionic strength treatments would result in greater $\text{NH}_4\text{-N}$ release to microcosm soil solution, we grouped salt treatments into two categories: high ionic strength (0.26-0.28M: NaCl, NaCl + Na_2SO_4 , NaCl + CaSO_4 , NaCl + Na_2SO_4 + CaSO_4 , and Instant Ocean®) and low ionic strength (0.03-0.04M: Na_2SO_4 and CaSO_4) to better interpret our findings on inorganic N release. The DIW treatment (no ionic strength) was not included because it was the only treatment in its group and did not allow for adequate statistical power to detect significance between this treatment and the other treatment groups. Since PO_4^{3-} sequestration in soils is enhanced by the presence of Ca (Penn et al. 2011) and SO_4^{2-} often provides a control on PO_4^{3-} release, to better interpret our findings on SRP microcosm soil solution concentrations, we grouped data from the salt treatments at day 30 based on whether the treatments contained added Na_2SO_4 (Na_2SO_4 and NaCl + Na_2SO_4 treatments), CaSO_4 (CaSO_4 and NaCl + CaSO_4 treatments), neither constituent (NaCl and DIW treatments), or a combination of both (NaCl + Na_2SO_4 + CaSO_4 treatments). These treatments are referred to as: (1) + Na_2SO_4 treatments; (2) + CaSO_4 treatments; (3) - SO_4^{2-} treatments; (4) or + Na_2SO_4 and CaSO_4 treatments, respectively. We conducted a Pearson correlation to determine the linear relationship between TDFe and SRP concentrations but omitted data from day 0 as this was our baseline before there was adequate time for chemical transformations to take place in the microcosm soils.

Results

Overall, we found that oxygen levels and salt treatments had major effects on N, P, and TDFe release into the microcosm soil solution, with anoxic soils releasing more $\text{NH}_4\text{-N}$, SRP, and TDFe to the overlying water than oxic soils after 15 and 30 days ($P < 0.05$). Values expressed in parentheses are a mean of all salt treatments with standard error of the mean, unless otherwise indicated. Concentrations of $\text{NH}_4\text{-N}$, SRP, and TDFe were lowest in anoxic microcosm soil solutions at day 0 (0.23 ± 0.06 mg N/l, 0.30 ± 0.02 mg P/l, and 0.99 ± 0.23 mg Fe/l, respectively), higher at day 15 (3.93 ± 0.36 mg N/l, 5.67 ± 0.65 mg P/l, and 7.48 ± 1.14 mg Fe/l, respectively), and highest at day 30 (5.08 ± 0.41 mg N/l, 9.31 ± 0.67 mg P/l, and 16.64 ± 1.51 mg Fe/l, respectively), while $\text{NO}_3\text{-N}$ concentrations decreased from 7.65 ± 0.46 mg N/l at day 0 to near zero levels by day 15. Nitrate-N release from oxic soils was lowest at day 0 (7.68 ± 0.5 mg N/l), intermediate at day 15 (8.87 ± 0.88 mg N/l), and highest at day 30 (10.04 ± 1.19 mg N/l) while $\text{NH}_4\text{-N}$ and TDFe concentrations remained near zero throughout the incubation period. Though there was no discernable significant difference in DIN concentrations between anoxic and oxic treatments, DIN concentrations in oxic treatments were consistently higher than those of the anoxic treatments for each salt treatment at day 15 and day 30. In oxic microcosm soils, SRP release was lowest at day 0, but similar between day 15 and day 30 (0.29 ± 0.02 mg P/l, 2.19 ± 0.19 mg P/l, and 2.57 ± 0.20 mg P/l, respectively; Figure 2). After exploring trends through time, we shifted our focus to the effects of the grouped salt treatments as described in the methods and oxygen level at day 30, as this was the end of the incubation period.

By day 30, significantly more $\text{NO}_3\text{-N}$ was released to the overlying microcosm soil solution under oxic conditions in both low and high ionic strength treatments (7.85 ± 2.47 and 12.14 ± 1.39 mg $\text{NO}_3\text{-N/l}$, respectively) as compared to anoxic conditions (0.06 ± 0.05 to $0.08 \pm$

0.05 mg NO₃-N/l, respectively; $P < 0.05$). However, there was no effect of salt treatment or ionic strength on NO₃-N release regardless of oxygen level (Table 3 and Figure 3a). Twice as much NH₄-N was released to overlying waters in high ionic strength solution than in the low ionic strength solution, under both oxic and anoxic conditions ($P < 0.05$; Figure 3b). Additionally, in both low and high ionic strength treatments, NH₄-N concentrations were much higher under anoxic conditions (3.25 ± 0.49 and 6.09 ± 0.50 mg NH₄-N/l; respectively) than under oxic conditions (0.04 ± 0.01 and 0.06 ± 0.04 mg NH₄-N/l, respectively; $P < 0.0001$; Table 3 and Figure 3b).

At the end of the incubation period, concentrations of SRP were significantly lower in CaSO₄-only (simulating gypsum soil amendment) treatments than in Na₂SO₄-only treatments with other treatments releasing intermediate amounts of SRP to microcosm soil solution (Figure 4; $P < 0.05$). Additionally, SRP release was significantly lower under oxic conditions (1.46 ± 0.20 - 3.54 ± 0.31 mg SRP/l) than under anoxic conditions (6.22 ± 0.93 - 11.67 ± 1.78 mg SRP/l; $P < 0.05$; Figure 4). There was a moderate and significant positive correlation between TDFe and SRP concentrations in microcosm soil solution ($R^2 = 0.49$, $P < 0.0001$; Figure 5). Concentrations of TDFe were significantly higher under anoxic conditions (16.64 ± 1.51 mg Fe/l) than under oxic conditions (0.43 ± 0.12 mg Fe/l; $P < 0.05$; Figure 2). We observed no effect of salt treatment on the release of total dissolved Fe to solution (Table S1 and Figure S1). Total Fe oxides (6.48 ± 0.06 mg Fe/g dry soil) did not vary significantly by day, oxygen treatment, or salt treatment (Table 3). Additionally, at day 30, soil pH in anoxic treatments was significantly higher than that in the oxic treatments (6.2 ± 0.0 versus 5.7 ± 0.1 , respectively; $P = 0.00617$).

When scaled to kg/ha, potential N release was similar between the Instant Ocean® and NaCl + Na₂SO₄ + CaSO₄ (representing a field with added gypsum amendments) salt treatments.

Under anoxic conditions, soils released a negligible amount (0.1 ± 0.0 kg of $\text{NO}_3\text{-N/ha}$) to overlying solution in both Instant Ocean® and $\text{NaCl} + \text{Na}_2\text{SO}_4 + \text{CaSO}_4$ treatments, whereas they released an average of 74 kg $\text{NH}_4\text{-N/ha}$ to solution in the two salt treatments. On the other hand, SRP and TDFe release was much lower in the soils treated with $\text{NaCl} + \text{Na}_2\text{SO}_4 + \text{CaSO}_4$ than when exposed to Instant Ocean (90.4 ± 7.1 and $119.7 \text{ kg} \pm 6.4$ P/ha, respectively), illustrating the suppressive effects of Ca on nutrient release. Under oxic conditions, $\text{NO}_3\text{-N}$, $\text{NH}_4\text{-N}$, SRP, and TDFe release was similar between the Instant Ocean® and $\text{NaCl} + \text{Na}_2\text{SO}_4 + \text{CaSO}_4$ salt treatments. In the Instant Ocean® treatment, $\text{NO}_3\text{-N}$ release was almost 1450 times greater under oxic than anoxic conditions (143.6 ± 30.3 kg N/ha and 0.1 ± 0.0 kg N/ha, respectively) while $\text{NH}_4\text{-N}$ release was over 700 times greater under anoxic versus oxic conditions (78.2 ± 6.0 kg N/ha and 0.1 ± 0.0 kg N/ha, respectively). Finally, SRP was over 6 times greater under anoxic versus oxic conditions (119.8 ± 6.0 kg P/ha and 19.9 ± 0.9 kg P/ha, respectively). TDFe was almost 200 times greater under anoxic versus oxic conditions (295.3 ± 27.1 kg Fe/ha and $1.5 \text{ kg Fe/ha} \pm 0.4$, respectively; Table 4). For the complete dataset of nutrient release scaled to kg/ha for each salt treatment, see Table S2.

Discussion

Overall, our results have major implications for how N, P, and Fe may be transformed and released from agricultural soils undergoing SWI. The soil used for this study was from a farm field that is at risk for SWI in the coming years as it is located only 500 m from a tidal salt marsh and hydrologically connected to the marsh through agricultural ditches. This farm soil shows a high potential for N and P release when exposed to saltwater. For example, when the soil becomes anoxic due to inundation, concentrations of $\text{NH}_4\text{-N}$, SRP, and TDFe may increase in overlying waters. Although release of $\text{NH}_4\text{-N}$, SRP, and TDFe was lower under oxic soil

conditions, NO₃-N release was greatly increased. Ionic strength was a strong driver of NH₄-N release from anoxic soils while the addition of Ca suppressed SRP release from both oxic and anoxic soils.

Nitrogen

The main form of DIN release differed between oxic and anoxic microcosm soil solutions due to divergent redox pathways for N. By day 15, NO₃-N concentrations were near zero under anoxic conditions (Figure 3a), which indicates that NO₃-N was either denitrified or converted to NH₄-N via DNRA, as has been observed in previous studies of anoxic soils (Tobias et al. 2001; Koop-jakobsen and Giblin 2010; Giblin et al. 2013). Research has shown that DNRA rates tend to be higher in saline soils because bisulfide (HS⁻) produced from anaerobic SO₄²⁻ reduction is utilized as a reactant and oxidized back to SO₄²⁻ through one of the major DNRA pathways. This reaction occurs as:



High ionic strength treatments resulted in a greater NH₄-N release to overlying water than low ionic strength treatments (Figure 3b) due to cations in saltwater replacing NH₄⁺ ions on soil exchange sites (Ardón et al. 2013). This effect was more pronounced under anoxic conditions where concentrations of NH₄-N in solution were higher than under oxic conditions. Of note, high ionic strength treatments contained 10 times the salts and were about 7 times the ionic strength of low ionic strength treatments, but NH₄-N release was only two times as much in the high ionic strength treatments, which suggests a non-linear relationship between ionic strength and NH₄-N release (Figure 3b). Similarly, other studies have demonstrated a non-linear increase in NH₄-N released in soils exposed to incremental increases in ionic strength. As ionic strength increases, Na⁺ cations compete with NH₄⁺ cations for exchange sites on soil colloidal surfaces (Seitzinger

et al. 1991; Weston et al. 2010). The lyotropic series describes the order of cations according to their adsorptive power to soil colloidal particles, where ions with lower hydrated radii have higher adsorptive power, which is mechanistically defined by Coulomb's Law (Huang 1980). Further, the non-linear nature of the replacement rate of NH_4^+ cations with Na^+ cations has been modeled through the Langmuir adsorption isotherm (Abukhadra et al. 2020).

As expected, $\text{NO}_3\text{-N}$ was the dominant form of DIN in oxic microcosm soil solutions and $\text{NH}_4\text{-N}$ concentrations were close to zero. Additionally, salt treatments had no effect on $\text{NO}_3\text{-N}$ release. Nitrate-N concentrations remained relatively constant under oxic conditions throughout the incubation period as most of the $\text{NO}_3\text{-N}$ was immediately released into solution (Figure 3a). Because $\text{NO}_3\text{-N}$ is prone to rapid leaching from soils (Jessen et al. 2017), SWI may spur a large amount of N loss to downstream waterways through inundation, before soils even become anoxic. Since there are no studies that have attempted to quantify $\text{NO}_3\text{-N}$ losses from an agricultural field initially undergoing SWI, this hypothesis should be tested through future research.

Consistently greater though not statistically significant DIN released to microcosm soil solution in the oxic versus anoxic microcosms suggests that other chemical pathways such as N mineralization and nitrification may also have played an important role in final N soil solution concentrations at the end of the incubation period. The significantly lower pH we observed in oxic soils versus anoxic soils could be attributed to greater rates of nitrification from aerobic microbial metabolic pathways. This difference in nitrification rates has been observed in other studies because N mineralization rates are greater in the presence of oxygen (Robertson and Vitousek 1981; Fellman and D'Amore 2007). Additionally, N mineralization and nitrification are tightly coupled. When $\text{NH}_4\text{-N}$ is produced from the decomposition of organic compounds under

oxic conditions, it is quickly converted to $\text{NO}_3\text{-N}$ through aerobic microbial respiration (Groffman and Rosi-Marshall 2013).

Phosphorus

Overall, the soil used in this study had a very high level of bioavailable P ($\text{PO}_4\text{-P}$; 352.5 ± 7.2 mg P/kg dry soil), which was over three times higher than bioavailable N ($\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$) concentrations (116.0 ± 6.7 mg N/kg dry soil; Table 1). Due to the historic over-application of poultry manure, a fertilizer with a low N to P ratio (Waldrip et al. 2015), these P concentrations are more than four times what is needed for optimal crop growth in the study region (Sharpley et al. 2003). Remarkably, no poultry manure or P fertilizer was applied to the field the year that the soils were collected. Thus, the high bioavailable P levels reflect a legacy of nutrient buildup in the soil from past years of fertilizer application. Therefore, with exposure to SWI, agricultural soils with a history of P applications may become a source of nutrient pollution to downstream water bodies.

Calcium played an important role in P dynamics under both oxic and anoxic conditions. By day 30, we observed a significant suppression of SRP release in $+\text{CaSO}_4$ treatments versus $+\text{Na}_2\text{SO}_4$ treatments under both oxygen levels (Figure 4) because Ca ions tend to bind to SRP and form insoluble Ca-phosphate precipitate minerals ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH},\text{F},\text{Cl})_2$; Zak et al. 2009), thus removing this form of P from the soil solution (Grubb et al. 2011). Additionally, by day 30, $+\text{CaSO}_4$ oxic treatments released significantly less SRP to solution than the oxic $-\text{SO}_4$ treatments (Figure 4). However, this was not the case for the corresponding anoxic treatments, potentially due to differences in redox pathways under oxic and anoxic conditions. Consequently, as an agricultural field is initially undergoing SWI, Ca amendments such as gypsum may provide a moderate ability to prevent SRP loss downstream. However, the fact that we observed greater

SRP release from anoxic treatments than oxic treatments suggest that as the field becomes anoxic, this ability is reduced.

Many incubation studies in wetland soils have demonstrated increases in SRP release with SO_4^{2-} addition under anoxic conditions because of interacting relationships between Fe, sulfur (S), and P cycling (Weston et al. 2006; Williams et al. 2014; Steinmuller and Chambers 2018). Field observations show that sulfide (reduced S) tends to bind reduced Fe in solution in anoxic sediments and form insoluble FeS_x complexes that prevent Fe from sequestering SRP (Jordan et al. 2008). In contrast, we did not clearly observe this effect from the agricultural soil used for our study, possibly because the background levels of SO_4^{2-} in this soil were several orders of magnitude higher than those of freshwater wetland soils (compare to Alphin et al. 2007; Williams et al. 2014) due to the application of SO_4^{2-} -rich poultry manure on the farm field from which the soil was collected. In our study, the soil used for our incubations already contained a high background level of $\text{SO}_4\text{-S}$ (23.3 ± 1.3 mg $\text{SO}_4\text{-S}$ / kg dry soil; Table 1; Flynn 2015) and salt treatments with SO_4^{2-} increased SO_4^{2-} in the incubations by only ~11% based on scaling salt treatment solution concentrations of $\text{SO}_4\text{-S}$ to mg/g dry soil (Calculation 1). Therefore, any effects of the added SO_4^{2-} from these treatments may have been muted by high background levels of SO_4^{2-} in the incubation soils.

Iron

Over the course of our incubations, we observed significantly greater TDFe concentrations in solution in the anoxic microcosms than in the oxic microcosms (Figure 6). Therefore, it is clear that anoxic conditions spurred the reductive dissolution of Fe (Hartzell et al. 2017). Under oxic conditions, Fe oxyhydroxides tend to coprecipitate with SRP, thus removing it from solution (Weston et al. 2006), which is why we observed near zero concentrations of TDFe

in the oxic microcosms (Figure 2). In contrast to TDFe concentrations, total Fe oxide pools in microcosm soils remained constant throughout the entire incubation period (Figure S1). Crystalline and poorly-crystalline Fe(III) oxides have low solubility in water with circumneutral pH, and it can take years for a significant proportion of the Fe oxides to undergo dissolution to dissolved forms as a soil undergoes SWI (Weston et al. 2006). The 30 day length of our incubations appears to have been an insufficient amount of time for dissolution to significantly shift the overall pool of total Fe oxides even though it significantly increased TDFe concentrations. In the anoxic treatments, TDFe concentrations were less than 3% of total Fe oxide concentrations (Figures 2 and S1), which illustrates that TDFe constitutes a small but chemically important pool of Fe in SWI-affected systems. The fact that we observed a moderate and significant positive correlation between TDFe and SRP concentrations (Figure 5) suggests that TDFe plays an important role in sequestering and releasing SRP from inundated soils, as is supported by studies conducted in wetland ecosystems (Chambers and Odum 1990; Hartzell and Jordan 2012).

Additionally, S plays an important role in preventing Fe from re-binding SRP under changing oxygen conditions. As previously mentioned, SO_4^{2-} in soil and soil amendments is reduced to sulfide under anoxic conditions and tends to bind Fe in FeS_x complexes (Schoepfer et al. 2014). The fact that we still observed high levels of TDFe in solution under anoxic conditions suggests that the rate of Fe dissolution is faster than its precipitation with S. Therefore, Fe reduction is likely the driver of SRP release from inundated soils while FeS_x formation prevents Fe from sequestering SRP if soil conditions become oxic again. Fields affected by SWI are subject to these interacting processes because they undergo wetting and drying cycles, which cause them to fluctuate between oxic and anoxic redox states (Tully et al. 2019b).

Potential soil nutrient release

Because we generated constant oxygen levels and salinity exposure in our incubations, it is important to note that our estimates reflect a potential for nutrient release from soils over time. Thus, this portion of our study represents a thought experiment representing the effects of an extreme case of SWI on nutrient release on the agricultural field. Unlike in a controlled laboratory setting, soil conditions in agricultural fields are dynamic. For example, the level of salinity exposure in an SWI-affected can change spatially and temporally, depending on factors such as tidal inundation, and precipitation and evapotranspiration rates (Weissman and Tully 2020). Consequently, the quantity and form of nutrients released from SWI-affected soils is highly dependent on the timing and magnitude of environmental variables. For example, it is possible that much of the $\text{NO}_3\text{-N}$ in the soil would rapidly be lost to leaching with saltwater inundation, before the soils could become oxygen-depleted. In turn, less $\text{NO}_3\text{-N}$ would be present in the soil to undergo DNRA, thus releasing less $\text{NH}_4\text{-N}$ to porewater when soils finally became anoxic. Therefore, the total proportion of inorganic N released as $\text{NO}_3\text{-N}$ versus $\text{NH}_4\text{-N}$ over time would vary depending on field conditions.

The pattern of enhanced $\text{NH}_4\text{-N}$, SRP, and TDFe release and reduction of $\text{NO}_3\text{-N}$ concentrations in saline inundated soils has also been observed in other incubation studies. For example, in one study, a marsh soil that had been formerly diked and drained was subjected to full strength (~32 ppt) seawater over a period of 20 months. The soil showed a roughly 7-fold increase in $\text{PO}_4\text{-P}$ and a 28-fold increase in $\text{NH}_4\text{-N}$ (Portnoy and Giblin 1997). However, in contrast to the increase in soil solution TDFe after 30 days of anoxic conditions observed in our study, after the 20 month incubation in this study, TDFe concentrations decreased by 42% which was attributed to Fe sequestration in FeS_x compounds (Portnoy and Giblin 1997). Another study

focused solely on $\text{NH}_4\text{-N}$ exchange from wetland soils showed that at 0 ppt salinity, about 69% of $\text{NH}_4\text{-N}$ was bound to soil particles, 14% as salinity increased to 3 ppt, 6% at 10 ppt salinity, and less than 1% at 34 ppt (Weston et al. 2010). This decrease in soil bound $\text{NH}_4\text{-N}$ is comparable to the 7-fold increase in soil solution $\text{NH}_4\text{-N}$ concentrations we observed in our study between 1.5 and 15 ppt. In another study, where salinity was experimentally raised to 0.9, 2.25, 4.5, and 9 PSU with seawater in wetland soils over 2.5 years, researchers found lower porewater P concentrations as salinity increased. This was attributed to an increase in Ca concentrations due to seawater addition as well as enhanced mobilization of Ca through mechanisms of cation exchange on soil particles (van Dijk et al. 2019). We also observed a suppressive effect of Ca on P release to soil solution in our study. Finally, a 2.5 year study on wetland soils where salinity was increased from 0.0-0.1 ppt to a range of 1.7 to 3.8 ppt, showed a marked increase in the abundance of the microbial community responsible for DNRA (Neubauer et al. 2019). Taken together with our results, these studies suggest that processes associated with SWI mechanistically cause similar patterns of nutrient release in wetland soils as in agricultural soils. However, because of the legacy of intensive soil inputs, agricultural soils have a capacity to release nutrients at a greater magnitude than wetland soils.

Overall, our results show that large quantities of N and P are poised for loss from agricultural fields undergoing SWI. Soils were collected in April 2019, when precipitation rates in our study region are high, spring tides frequently flood coastal areas, and about a month before farmers tend to plant cash crops (Tully et al. 2019b). Consequently, nutrient export is likely highest during the spring, when there is less plant nutrient uptake because fields are planted in cover crops, conditions are prime for saltwater to move onto fields, and soils tend to remain saturated, sometimes for weeks. After 30 days of inundation with saltwater (e.g. Instant Ocean®

treatment), as much as 45% of the total inorganic N could be released (as $\text{NH}_4\text{-N}$) if soils were under anoxic conditions. As much as 22% of the bioavailable soil P could be released as SRP to overlying water after a month of inundation with saltwater (when initial soil N and P concentrations in Table 1 are scaled to kg/ha and compared to Table 4).

As mentioned previously, bioavailable P levels in the soil were over four times the maximum recommended level to optimize crop yield while minimizing P runoff losses (Sharpley et al. 2003). Thus, SRP release from SWI-affected fields could be a major source of nutrient pollution to nearby waterways. In order to better understand rates of nutrient loss, future research could focus on determining how conditions such as soil porewater salinity and redox potential change with nutrient concentrations in SWI-affected fields over time.

Conclusion

As sea levels rise, SWI will cause extended periods of soil inundation in coastal agroecosystems. Here we simulated this effect on an agricultural soil collected from a farm field vulnerable to SWI. Overall, we found that oxygen depletion in inundated soils contributes to SRP release from soils and shifts N cycling so that the DIN pool is dominated by $\text{NH}_4\text{-N}$. Higher ionic strength, in turn, enhances $\text{NH}_4\text{-N}$ release from both oxic and anoxic soils. Additionally, even without the addition of added salts, high levels of Fe in agricultural soils can cause a large release of SRP from anoxic soils as a result of reductive dissolution. However, Ca addition from soil amendments such as agricultural lime or gypsum can bind to P and suppress its release into water overlying inundated soil. These findings imply that SWI-affected fields may be hotspots of nutrient pollution, particularly during the spring, when they have the potential to release a large proportion of inorganic soil N and P to downstream waterways. Therefore, defining mechanisms

through which ions interact in solution is critical for quantifying N and P export in coastal agricultural soils affected by SWI.

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Table 1: Soil physical and chemical properties. Abbreviations used: cation exchange capacity (CEC), organic matter (OM), carbon (C), EC (electrical conductivity), nitrogen (N), nitrate-nitrogen (NO₃-N), ammonium-nitrogen (NH₄-N), phosphorus (P), calcium (Ca), magnesium (Mg), potassium (K), sodium (Na), boron (B), iron (Fe), manganese (Mn), copper (Cu), zinc (Zn), aluminum (Al), chloride (Cl⁻), and sulfate-sulfur (SO₄-S). Standard error of the mean is indicated in parentheses after each value except for results of particle size analysis.

CEC (meq/100g)	pH	OM (%)	Total C (%)	EC (mS/cm)	% Sand	% Silt	% Clay	mg/kg			
								Total N	NO ₃ -N	NH ₄ -N	P*
10.8 (0.2)	6.0 (0.0)	2.9 (0.1)	1.8 (0.2)	0.5 (0.1)	43.4	42.6	14.0	1614.7 (48.5)	114.7 (6.6)	1.3 (0.1)	352.5 (7.2)
mg/kg											
Ca*	Mg*	K*	Na*	B*	Fe*	Mn*	Cu*	Zn*	Al*	Cl ⁻	SO ₄ -S
1251.0 (18.0)	171 (2.5)	266.5 (7.5)	18.3 (1.5)	0.7 (0.0)	376.0 (4.4)	35.3 (1.0)	12.6 (0.4)	29.5 (1.2)	641.0 (11.7)	7.2 (0.7)	23.3 (1.3)

Note: * = Mehlich III extracted

826 **Table 2:** Microcosm salt treatments.

Treatment	Salt added (g/l)				Total salinity (ppt)	Ionic strength (M)
	NaCl	CaSO ₄	Na ₂ SO ₄	Instant Ocean®		
Deionized water (DIW)	0.00	0.00	0.00	0.00	0.00	0.00
CaSO ₄	0.00	1.47	0.00	0.00	1.47	0.04
Na ₂ SO ₄	0.00	0.00	1.53	0.00	1.53	0.03
NaCl	15.00	0.00	0.00	0.00	15.00	0.26
NaCl + CaSO ₄	13.53	1.47	0.00	0.00	15.00	0.27
NaCl + Na ₂ SO ₄	13.47	0.00	1.53	0.00	15.00	0.26
NaCl + Na ₂ SO ₄ + CaSO ₄	12.00	1.47	1.53	0.00	15.00	0.28
Instant Ocean®	0.00	0.00	0.00	15.00	15.00	0.26

827 Note: Deionized water (DIW) is the no salt control and Instant Ocean® is the 15 parts per thousand (ppt)
828 salinity treatment designed to mimic the full suite of ions in saltwater. Treatments containing sodium chloride
829 (NaCl) are high ionic strength treatments (0.26-0.28M and 15ppt total salts). Treatments without NaCl are low ionic
830 strength treatments (0.03-0.04M and ~1.5 ppt total salts). Treatments containing calcium sulfate (CaSO₄) are
831 designed to simulate soil amendments used to reclaim sodic soils. Treatments containing sodium sulfate (Na₂SO₄)
832 are designed to simulate the sulfate component of saltwater.

Table 3: Results of linear fixed-effects model for the effect of oxygen level (O2.level) and salt treatment (Trt.) on each variable and all interactive effects on day 30 of the experiment.

Variable	Factor	num df	den df	Sum of squares	<i>F</i> value	<i>P</i> value
NO ₃ -N	Trt.	7	7	10.53	0.69	0.6791
NO ₃ -N	O2.level	1	7	758.52	348.37	< 0.0001***
NO ₃ -N	Trt.:O2.level	7	7	14.10	0.93	0.4958
NH ₄ -N	Trt.	7	7	3.33	0.75	0.6316
NH ₄ -N	O2.level	1	7	325.96	512.46	< 0.0001***
NH ₄ -N	Trt.:O2.level	7	7	6.85	1.54	0.1770
DIN	Trt.	7	7	54.41	2.03	0.0707
DIN	O2.level	1	7	30.12	7.86	0.0072**
DIN	Trt.:O2.level	7	7	10.48	0.39	0.9034
SRP	Trt.	7	7	11.87	4.65	0.0301*
SRP	O2.level	1	7	36.75	86.37	< 0.0001***
SRP	Trt.:O2.level	7	7	0.77	0.30	0.9316
TDFe	Trt.	7	7	6.55	0.75	0.6344
TDFe	O2.level	1	7	352.74	281.19	< 0.0001***
TDFe	Trt.:O2.level	7	7	33.12	3.77	0.0505
Total Fe oxide	Trt.	7	7	0.07	0.61	0.7419
Total Fe oxide	O2.level	1	7	0.03	1.84	0.1812
Total Fe oxide	Trt.:O2.level	7	7	0.13	1.22	0.3141

Note: * = $P < 0.05$, ** = $P < 0.01$, *** = $P < 0.001$. Variables are nitrate-nitrogen (NO₃-N), ammonium-nitrogen (NH₄-N), dissolved inorganic N (DIN; NO₃-N + NH₄-N); soluble reactive phosphorus (SRP), total dissolved iron (TDFe), and total Fe oxide.

840 **Table 4:** Potential nitrate-nitrogen (NO₃-N), ammonium-N (NH₄-N), dissolved inorganic N
841 (DIN; NO₃-N + NH₄-N), soluble reactive phosphorus (SRP), and total dissolved iron (TDFe)
842 release (in kg/ha) from soils (0-10 cm of the agricultural field sampled) exposed to the NaCl +
843 Na₂SO₄ + CaSO₄ and Instant Ocean® salt treatment (in kg/ha) based on data from day 30 of the
844 experiment. Standard errors of the means are indicated in parentheses after each value.

845

Analyte	Nutrient release (kg/ha)			
	Oxic		Anoxic	
	NaCl + Na ₂ SO ₄ + CaSO ₄	Inst. Ocean®	NaCl + Na ₂ SO ₄ + CaSO ₄	Inst. Ocean®
NO ₃ -N	149.7 (32.8)	143.6 (30.3)	0.1 (0.0)	0.1 (0.0)
NH ₄ -N	1.2 (1.0)	0.1 (0.0)	70.7 (12.3)	78.2 (6.0)
DIN	150.8 (33.6)	143.7 (30.3)	70.8 (12.3)	78.3 (6.0)
SRP	20.9 (3.6)	19.9 (0.9)	90.5 (7.4)	119.8 (6.0)
TDFe	1.1 (0.4)	1.5 (0.4)	240.8 (27.4)	295.3 (27.1)

Figure legends

Figure 1: (a) Map of the Chesapeake Bay region, United States. Somerset County is outlined by the dotted rectangle. Blue dot is location of soil collection site (b) Satellite imagery of the soil collection site (Google Earth 2019).

Figure 2: Microcosm soil solution nitrate-nitrogen ($\text{NO}_3\text{-N}$) concentrations in $\text{mg NO}_3\text{-N/l}$, ammonium-N ($\text{NH}_4\text{-N}$) concentrations in $\text{mg NH}_4\text{-N/l}$, dissolved inorganic N (DIN; $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$) concentrations in mg N/l , soluble reactive phosphorus (SRP) concentrations in mg P/l , and total dissolved iron (TDFe) concentrations in mg Fe/l at days 0, 15, and 30 of the experiment under oxic and anoxic conditions (oxygen level). Error bars represent standard error of the mean. Statistically significant differences within each day are indicated by different letters at $P < 0.05$.

Figure 3: (a) Microcosm soil solution nitrate-nitrogen ($\text{NO}_3\text{-N}$) concentrations in $\text{mg NO}_3\text{-N/l}$ at day 30 of the experiment grouped by low ionic strength (Na_2SO_4 , CaSO_4 ; $0.03M$ and $0.04M$, respectively) and high ionic strength (NaCl , $\text{NaCl} + \text{Na}_2\text{SO}_4$, $\text{NaCl} + \text{CaSO}_4$, $\text{NaCl} + \text{Na}_2\text{SO}_4 + \text{CaSO}_4$, and Instant Ocean®; $0.26M$, $0.26M$, $0.27M$, $0.28M$, $0.26M$, respectively) treatments. Statistically significant differences in means are indicated by different letters at $P < 0.05$. (b) Microcosm soil solution ammonium nitrogen ($\text{NH}_4\text{-N}$) concentrations in $\text{mg NH}_4\text{-N/l}$ at day 30 of the experiment grouped by low ionic strength (Na_2SO_4 , CaSO_4 ; $0.03M$ and $0.04M$, respectively) and high ionic strength (NaCl , $\text{NaCl} + \text{Na}_2\text{SO}_4$, $\text{NaCl} + \text{CaSO}_4$, $\text{NaCl} + \text{Na}_2\text{SO}_4 + \text{CaSO}_4$, and Instant Ocean®; $0.26M$, $0.26M$, $0.27M$, $0.28M$, $0.26M$, respectively) treatments. Statistically significant differences in means are indicated by different letters at $P < 0.05$. (c) Microcosm soil solution DIN (dissolved inorganic N; $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$) concentrations in mg N/l at day 30 of the experiment grouped by low ionic strength (Na_2SO_4 , CaSO_4 ; $0.03M$ and $0.04M$, respectively) and high ionic strength (NaCl , $\text{NaCl} + \text{Na}_2\text{SO}_4$, $\text{NaCl} + \text{CaSO}_4$, $\text{NaCl} + \text{Na}_2\text{SO}_4 +$

869 CaSO₄, and Instant Ocean®; 0.26M, 0.26M, 0.27M, 0.28M, 0.26M, respectively) treatments.

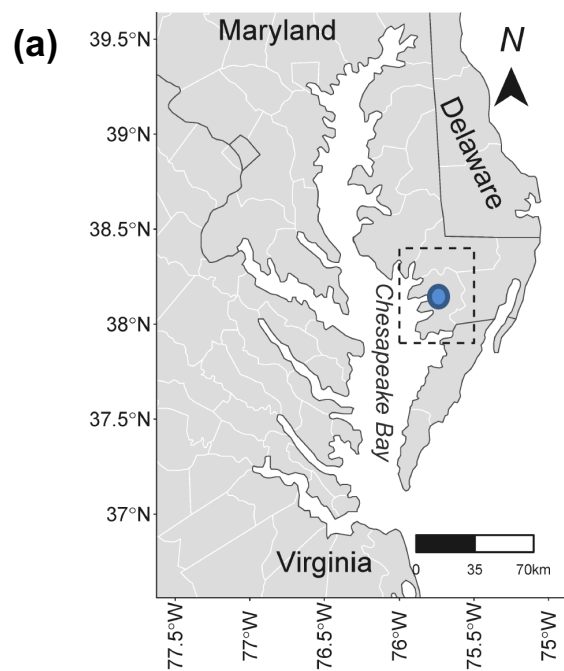
870 Statistically significant differences in means are indicated by different letters at $P < 0.05$.

871 **Figure 4:** Microcosm soil solution soluble reactive phosphorus (SRP) concentrations in mg P/l at
872 day 30. Treatments are grouped by the type of sulfate (SO₄²⁻) salt they contained: sodium sulfate
873 (Na₂SO₄), calcium sulfate (CaSO₄), neither constituent (sodium chloride or deionized water), or
874 a combination of both (Na₂SO₄ and CaSO₄). Treatments containing CaSO₄ are designed to
875 simulate soil amendments used to reclaim sodic soils. Treatments containing Na₂SO₄ are
876 designed to simulate the sulfate component of saltwater. Statistically significant differences in
877 means are indicated by different letters at $P < 0.05$.

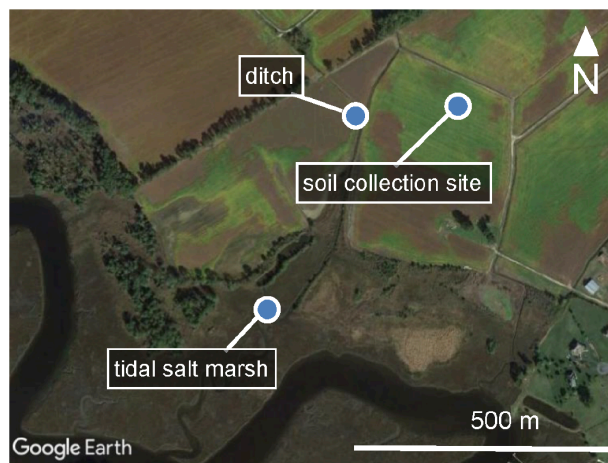
878 **Figure 5:** Total dissolved iron (Fe) concentrations in mg Fe/l versus soluble reactive phosphorus
879 (SRP) concentrations in mg SRP/l for days 15 and 30 of the experiment. Dotted red line is the
880 line of best fit based on Pearson correlation.

881

882 **Figure 1**

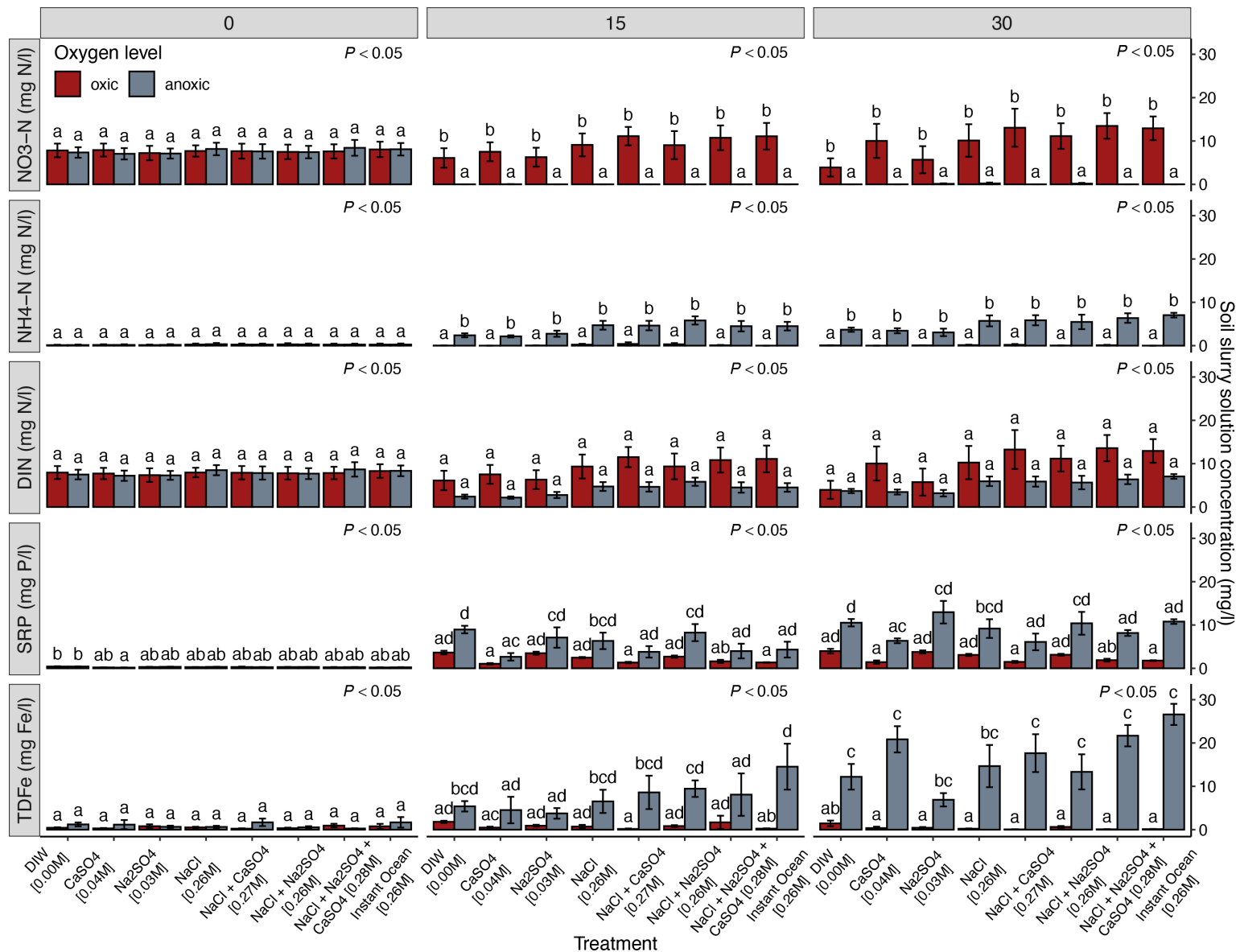


(b)

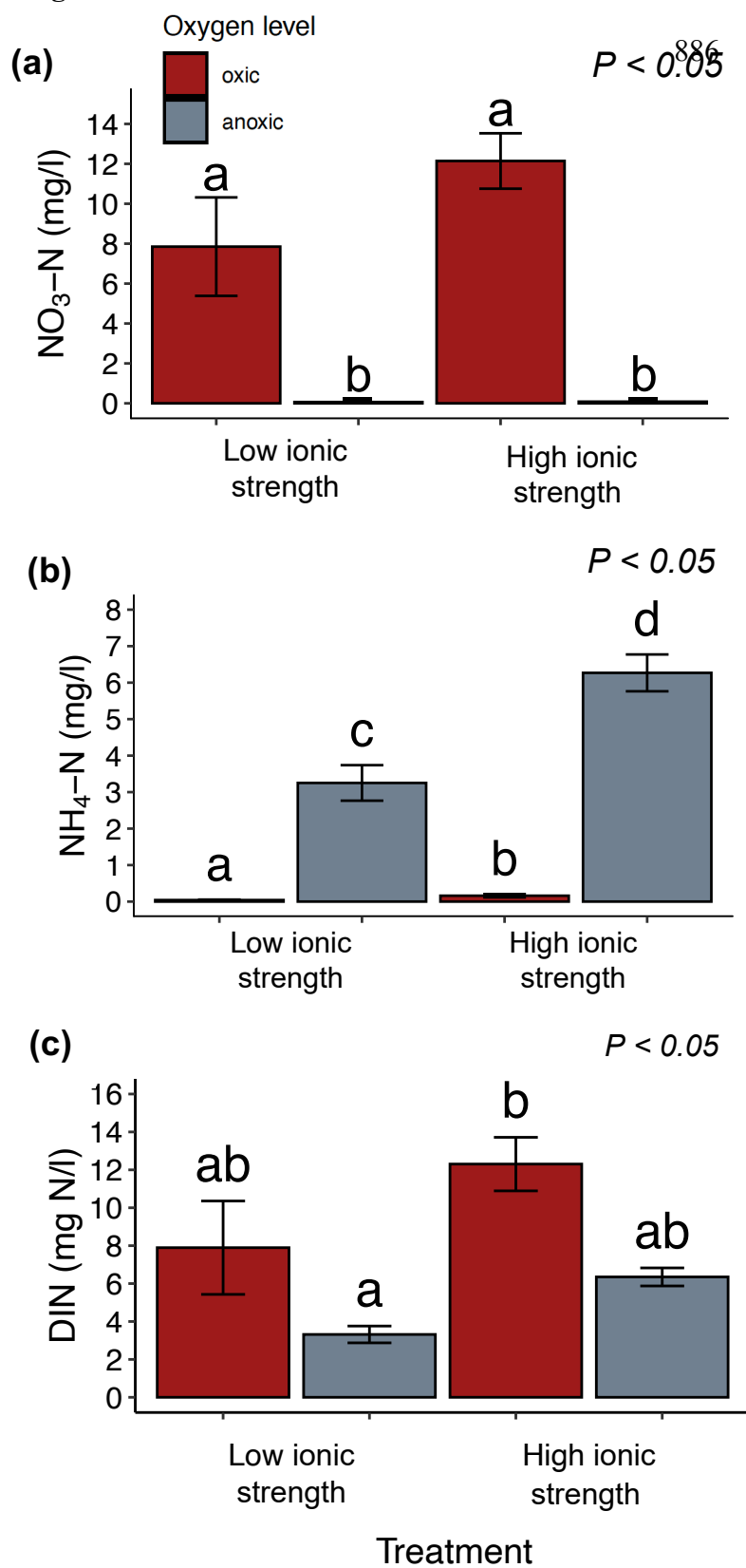


883

884 **Figure 2**

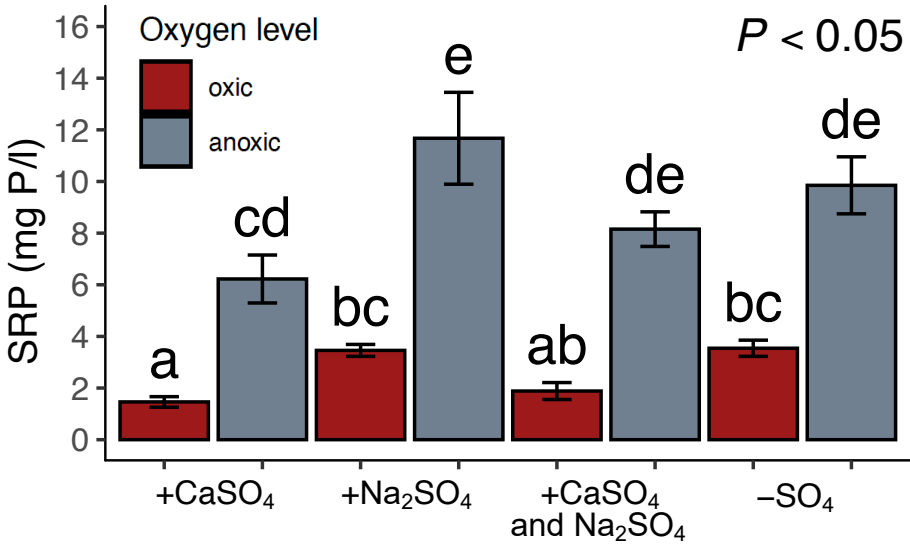


885 **Figure 3**

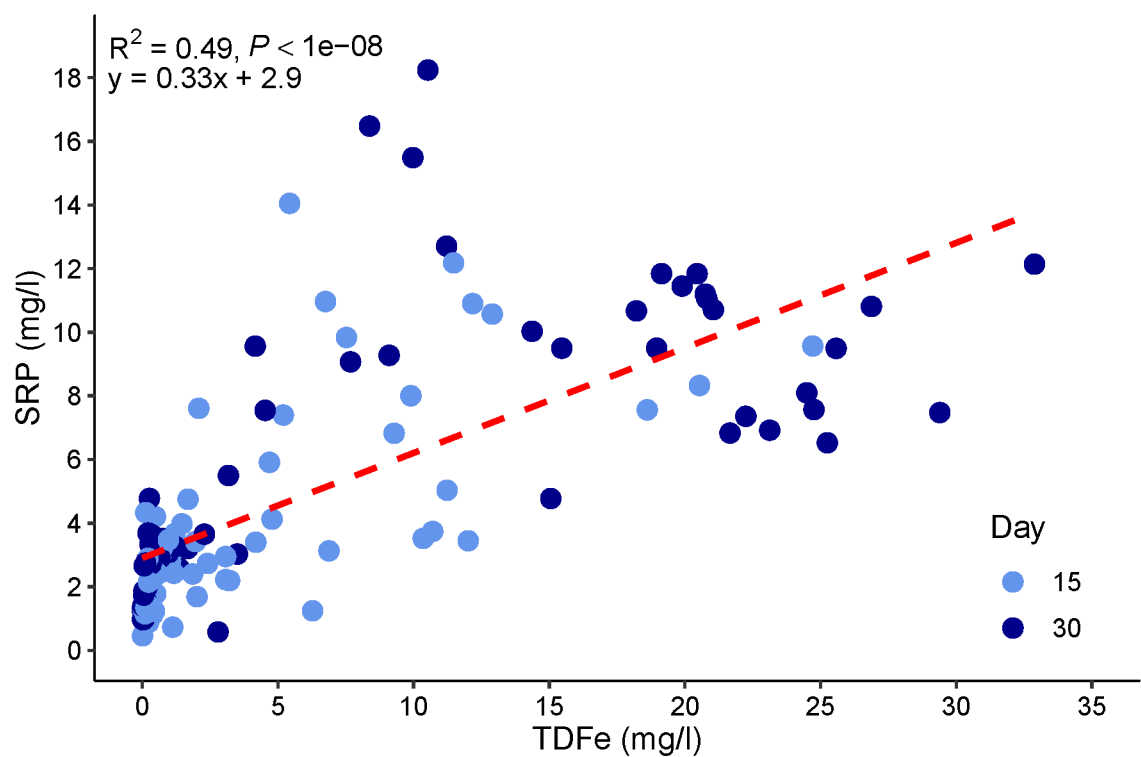


887 **Figure 4**

888



889 **Figure 5**



890

891 **Table S1:** Results of linear fixed-effects model for the effect of oxygen level (O2.level), salt
892 treatment (Trt.), and day (Day) on each variable and all interactive effects.

Variable	Factor	num df	den df	Sum of squares	<i>F</i> value	<i>P</i> value
NO ₃ -N	O2.level	1	14	578.25	271.94	< 0.0001***
NO ₃ -N	Trt.	7	14	9.27	0.62	0.7368
NO ₃ -N	Day	1	14	220.87	103.87	< 0.0001***
NO ₃ -N	O2.level:Trt.	7	14	10.26	0.69	0.6810
NO ₃ -N	O2.level:Day	1	14	212.33	99.86	< 0.0001***
NO ₃ -N	Trt.:Day	7	14	4.94	0.33	0.9384
NO ₃ -N	O2.level:Trt.:Day	7	14	7.22	0.49	0.8444
NH ₄ -N	O2.level	1	14	675.82	230.48	< 0.0001***
NH ₄ -N	Trt.	7	14	29.87	1.46	0.1868
NH ₄ -N	Day	1	14	109.63	37.39	< 0.0001***
NH ₄ -N	O2.level:Trt.	7	14	15.83	0.77	0.6123
NH ₄ -N	O2.level:Day	1	14	232.99	79.46	< 0.0001***
NH ₄ -N	Trt.:Day	7	14	1.35	0.07	0.9995
NH ₄ -N	O2.level:Trt.:Day	7	14	10.46	0.51	0.8264
DIN	O2.level	1	14	84.38	24.40	< 0.0001***
DIN	Trt.	7	14	85.35	3.36	0.0022**
DIN	Day	1	14	5.74	1.66	0.1994
DIN	O2.level:Trt.	7	14	10.43	0.43	0.8819
DIN	O2.level:Day	1	14	26.65	7.70	0.0061**
DIN	Trt.:Day	7	14	30.61	1.26	0.2713
DIN	O2.level:Trt.:Day	7	14	8.71	0.36	0.9241
SRP	O2.level	1	14	32.12	66.17	< 0.0001***
SRP	Trt.	7	14	23.11	6.80	< 0.0001***
SRP	Day	1	14	256.30	527.92	< 0.0001***
SRP	O2.level:Trt.	7	14	0.56	0.16	0.9919
SRP	O2.level:Day	1	14	16.21	33.39	< 0.0001***
SRP	Trt.:Day	7	14	2.40	0.71	0.6660
SRP	O2.level:Trt.:Day	7	14	0.91	0.27	0.9652
TDFe	O2.level	1	14	255.75	184.07	< 0.0001***
TDFe	Trt.	7	14	20.27	2.08	0.1153
TDFe	Day	1	14	75.79	54.55	< 0.0001***
TDFe	O2.level:Trt.	7	14	34.47	3.54	0.0210*
TDFe	O2.level:Day	1	14	128.91	92.78	< 0.0001***

TDFe	Trt.:Day	7	14	5.07	0.52	0.8173
TDFe	O2.level:Trt.:Day	7	14	20.60	2.12	0.1097
Total Fe oxide	O2.level	1	14	0.00	0.23	0.6352
Total Fe oxide	Trt.	7	14	0.06	0.50	0.8345
Total Fe oxide	Day	1	14	0.01	0.65	0.4222
Total Fe oxide	O2.level:Trt.	7	14	0.11	0.85	0.5480
Total Fe oxide	O2.level:Day	1	14	0.01	0.50	0.4795
Total Fe oxide	Trt.:Day	7	14	0.11	0.86	0.5391
Total Fe oxide	O2.level:Trt.:Day	7	14	0.11	0.89	0.5155

Note: * = $P < 0.05$, ** = $P < 0.01$, *** = $P < 0.001$. Variables are nitrate-nitrogen ($\text{NO}_3\text{-N}$), ammonium-nitrogen ($\text{NH}_4\text{-N}$), dissolved inorganic N (DIN; $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$), soluble reactive phosphorus (SRP), total dissolved iron (TDFe), and total Fe oxide

896 **Table S2.** Potential nitrate-nitrogen (NO₃-N), ammonium-N (NH₄-N), dissolved inorganic N (DIN; NO₃-N + NH₄-N), soluble reactive
897 phosphorus (SRP), and total dissolved iron (TDFe) release (in kg/ha) from soils (0-10 cm of the agricultural field sampled) for all salt
898 treatments (in kg/ha) based on data from day 30 of the experiment. Standard errors of the means are indicated in parentheses after each
899 value.

900

analyte	oxic								anoxic							
	DIW	CaSO ₄	Na ₂ SO ₄	NaCl	NaCl + CaSO ₄	NaCl + Na ₂ SO ₄	NaCl + Na ₂ SO ₄ + CaSO ₄	Inst. Ocean®	DIW	CaSO ₄	Na ₂ SO ₄	NaCl	NaCl + CaSO ₄	NaCl + Na ₂ SO ₄	NaCl + Na ₂ SO ₄ + CaSO ₄	Inst. Ocean®
NO ₃ -N	43.2 (23.3)	111.3 (43.7)	63.1 (34.7)	112.3 (41.7)	145.1 (48.7)	123.6 (32.9)	149.7 (32.8)	143.6 (30.3)	0.1 (0.0)	0.1 (0.0)	1.3 (1.2)	2.4 (2.3)	0.1 (0.0)	1.9 (1.8)	0.1 (0.0)	0.1 (0.0)
NH ₄ -N	0.7 (0.4)	0.2 (0.1)	0.7 (0.2)	1.5 (1.1)	2.2 (2.0)	0.4 (0.2)	1.2 (1.0)	0.1 (0.0)	40.9 (5.4)	38.2 (6.5)	34.0 (9.6)	63.5 (14.1)	65.1 (13.0)	60.8 (18.6)	70.7 (12.3)	78.2 (6.0)
DIN	44.0 (23.3)	111.5 (43.8)	63.8 (34.6)	113.8 (42.6)	147.3 (49.8)	124.0 (32.9)	150.8 (33.6)	143.7 (30.3)	41.0 (5.4)	38.3 (6.5)	35.3 (8.4)	65.9 (12.3)	65.2 (13.0)	62.7 (17.2)	70.8 (12.3)	78.3 (6.0)
SRP	44.3 (5.7)	15.9 (4.3)	42.0 (4.0)	34.4 (2.7)	16.6 (2.4)	34.8 (2.4)	20.9 (3.6)	19.9 (0.9)	116.9 (9.5)	70.5 (6.2)	143.9 (28.9)	101.9 (24.0)	67.7 (21.4)	115.4 (29.2)	90.5 (7.4)	119.8 (6.0)
TDFe	16.2 (7.2)	4.1 (3.7)	4.9 (1.9)	2.6 (0.6)	0.8 (0.3)	7.1 (2.3)	1.1 (0.4)	1.5 (0.4)	135.5 (32.9)	231.5 (33.6)	76.6 (17.1)	162.9 (54.1)	196.0 (48.5)	148.2 (44.9)	240.8 (27.4)	295.3 (27.1)

901

902 **Figure S1:** Total iron (Fe) oxide concentrations in microcosm soil in mg Fe/g dry soil at days 0,
 903 15, and 30 of the experiment under oxic and anoxic conditions. Error bars represent standard
 904 error of the mean.

