

# Saltwater intrusion affects nutrient concentrations in soil porewater and surface waters of coastal habitats

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**Abstract.** Coastal ecosystems are undergoing major biogeochemical shifts due to climate change and sea level rise. At the same time, agricultural fertilizer applications are increasing coastal nutrient inputs. In this study, we examine the potential impact of saltwater intrusion (SWI) on nitrogen (N) and phosphorus (P) concentrations in soil porewater and surface water of different habitats within the Chesapeake Bay estuary. Study sites are located along Maryland's Lower Eastern Shore and were monitored over three summers from 2016 to 2018. These sites encompass various habitats on the land–sea interface, consisting of healthy forests, intruded forests, abandoned fields, intruded fields, agricultural ditches, tidal creeks, and tidal salt marshes. Intruded fields were being actively farmed at the time of the study. Soil porewater and surface water grab samples were collected from the habitats and analyzed for electrical conductivity, pH, dissolved organic P (DOP), dissolved inorganic P in the form of soluble reactive P (SRP), dissolved inorganic N as ammonium-N ( $\text{NH}_4\text{-N}$ ), and total dissolved iron (TDFe). Electrical conductivity was greatest in the marshes (16.58 mS/cm averaged across all years) and did not significantly differ among intruded forests, intruded fields, and agricultural ditches. As a legacy of heavy fertilizer use, DOP concentrations exceeded 0.45 mg P/L in all habitats. Concentrations of inorganic N and P differed significantly by habitat. Concentrations of  $\text{NH}_4\text{-N}$  were significantly higher in salt marsh soil porewater than in any other habitat measured. Overall, SRP concentrations were the highest in the soil porewater of intruded fields and marshes and in the surface water of agricultural ditches (0.30, 0.29, and 0.33 mg P/L averaged across all years, respectively). These concentrations greatly exceeded recommended U.S. Environmental Protection Agency nutrient pollution thresholds for the region. In its oxidized state, dissolved iron can bind to SRP and prevent it from becoming bioavailable. However, TDFe concentrations in the ditches, tidal creeks, and marshes were too low to adequately buffer against SRP loss to downstream areas. As SWI moves salts inland and increases hydrologic connectivity across coastal landscapes, it is important to consider the mechanisms through which nutrients may be released from coastal soils and their potential to impact downstream water quality.

**Key words:** agroecosystems; Chesapeake Bay; legacy nutrients; nitrogen; phosphorus; saltwater intrusion; water quality.

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## INTRODUCTION

Coastal agricultural landscapes are changing rapidly in response to climate change. Low-lying

farmland is particularly vulnerable to saltwater intrusion (SWI) and coastal flooding. In many areas of the world, cultivated landscapes, forests, and other ecosystems are transforming into tidal

salt marshes as sea levels rise (Smith 2013, Moomaw et al. 2018, Kirwan and Gedan 2019). As a result, extensive upland area will be lost rapidly in coming decades (Wasson et al. 2013). This phenomenon is spurring radical shifts in ecosystem dynamics, including biogeochemical cycling in soils and water bodies (Bhattachan et al. 2018).

Though the effects of SWI on coastal forests have been studied (Hussein and Rabenhorst 2010, Marton et al. 2012, White and Kaplan 2017), little research has focused on its impact on farmland. Salinization of agricultural soils has been studied widely (Allison et al. 1954, Hanin et al. 2016). However, soil salinization studies only consider well-drained inland soils that receive salts via irrigation water (e.g., they are saline but not saturated) and have very different biogeochemical processes than SWI-affected soils in coastal areas (e.g., they are saline and frequently saturated).

Globally, accelerating rates of sea level rise are increasing the amount of coastal upland area that is affected each year (White and Kaplan 2017). The combination of agricultural land use legacies, sea level rise, and SWI has large and measurable effects on soil biogeochemical processes (Ardón et al. 2013, Helton et al. 2014, Sharpley et al. 2014). Thus, it is critical to develop an understanding of how SWI affects these processes in coastal regions in order to determine how to best help communities adapt to this phenomenon. In this study, we explore interactions between SWI, nitrogen (N), and phosphorus (P) that can lead to nutrient pollution in waterways.

As sea levels rise, SWI into freshwater ecosystems has increased in frequency, duration, and spatial extent. This phenomenon can enhance nutrient export from watersheds (Ardón et al. 2013, 2017, Williams et al. 2014). Prior studies indicate that salts and sulfate ( $\text{SO}_4^{2-}$ ) in saltwater can lead to N and P release from inundated soils as ammonium ( $\text{NH}_4^+$ ), nitrate ( $\text{NO}_3^-$ ), and phosphate ( $\text{PO}_4^{3-}$ ) due to mechanisms of ion exchange (Ardón et al. 2013, 2017, Williams et al. 2014). These ionic forms are inorganic and readily bioavailable. Phosphorus is often retained in farm soils after decades of excessive fertilizer use (legacy P) by binding to metals such as iron (Fe) and aluminum (Al; Sharpley et al. 2014, Tully et al. 2019b). Pools of P tend to be recalcitrant in

the aerobic upper soil layers of agricultural fields but can become mobile and bioavailable due to Fe transformations associated with saturated and reducing conditions as fields undergo SWI (Tully et al. 2019b). Ammonium can be released from inundated soil through cation exchange with salts (Ardón et al. 2013), and studies have demonstrated that N and P export is enhanced in oxygen-depleted soils saturated with saltwater (Brand-Klibanski et al. 2007, Dierberg et al. 2011, Ardón et al. 2017). Since coastal groundwater tables in the study region are typically shallow (<3 m; Sharpley et al. 2014), SWI and tidal inundation can create anaerobic conditions in upper soil layers. Although wetlands typically serve as nutrient filters and sinks (Tanner and Sukias 2011), they may become net sources of nutrients to water bodies downstream as they receive high concentrations of legacy nutrients from flooded agricultural soils upstream (Weston et al. 2006, Nair et al. 2013). Effective nutrient management requires a better understanding of the role that SWI plays in transporting nutrients off farm fields and into nearby habitats.

This study focuses on differences in nutrient concentrations in the soil porewater and surface water of farmland and surrounding hydrologically connected habitats undergoing SWI in the Chesapeake Bay, which is the largest estuary in the United States (Boesch 2006) and is situated along the Atlantic Coastal Plain region. Our study took place in Somerset County, Maryland, along its border with the Chesapeake Bay, where the primary drivers of SWI are land subsidence and sea level rise (Ezer and Corlett 2012, Tully et al. 2019a).

By 2050, sea level is projected to rise anywhere from 28 to 65 cm along the mid-Atlantic Coastal Plain under most emissions scenarios (Miller et al. 2013). In response, tidal marshes are predicted to migrate into upland forest and farmland (Glick et al. 2008). Agricultural fields in coastal areas of the Atlantic Coastal Plain are bounded by ditches on one or more sides to allow for drainage (e.g., removing water from fields). However, these ditches are often connected to tidal creeks that drain into tidal salt marshes and larger tributaries such as rivers, bays, and subestuaries (Bhattachan et al. 2018, Tully et al. 2019a). Sea level rise can convert them into conduits for saltwater moving onto fields.

Tide gates, which allow water to flow unidirectionally (i.e., off of a farm field into an agricultural ditch; Roman and Burdick 2012), are sometimes installed to allow fields to drain during low tide and prevent saltwater from moving onto fields during high tide (Walsh and Miskewitz 2013). As sea levels rise, maintaining functional tide gates on low-lying farmland has become challenging (Walsh and Miskewitz 2013). Due to high soil salinity, farmers and landowners have stopped planting crops on a significant portion of fields in Somerset County (Maryland Department of Natural Resources and NOAA 2008). These fields have been overtaken by novel plant communities, which include a mixture of agricultural weeds, native and non-native wetland vegetation, and upland species (Gedan and Fernández-Pascual 2019). Nearby forested areas are also undergoing SWI. In this study, our main objective was to determine how soil porewater and surface water electrical conductivity (EC, a proxy for salinity), N, and P concentrations change in distinct yet connected habitats across the land–sea interface. We also measured pH and total dissolved iron (TDFe) to allow us to better interpret our findings as they are important controls on some of the processes that affect N and P cycling in systems undergoing SWI (Chambers and Odum 1990, Tully et al. 2019b).

This goal of this research was to determine how agricultural fertilizer inputs and legacy nutrients affect concentrations of N and P in habitats within coastal agroecosystems undergoing SWI. To accomplish this, we first identified seven distinct habitats: (1) forest; (2) intruded forest; (3) abandoned field; (4) intruded field (actively farmed); (5) ditch (agricultural ditch); (6) tidal creek; and (7) marsh (tidal salt marsh). We measured dissolved inorganic N and P in the form of ammonium nitrogen ( $\text{NH}_4\text{-N}$ ), nitrate nitrogen ( $\text{NO}_3\text{-N}$ ), soluble reactive phosphorus (SRP), and dissolved organic P (DOP) in soil porewater and surface water collected from these habitats. We expected dissolved inorganic N to be primarily in the form of  $\text{NH}_4^+$  as  $\text{NO}_3^-$  is rapidly converted to other forms under low oxygen conditions that form in saturated soils. We hypothesized that SRP concentrations would be the highest on intruded fields and in their surrounding ditches due to active farming and P applications on these fields. We also expected to

observe high concentrations of SRP and DOP that had moved off of farm fields in downstream tidal creeks and salt marshes. Finally, we also considered the potential that a proportion of the different forms of N and P measured in each habitat were estuarine-derived through analysis of concentration data from the Chesapeake Bay Program at water quality monitoring stations located near our study sites (Chesapeake Bay Program 2019). We hypothesized that this contribution would be minor compared to concentrations we observed in our selected habitats. Measurements of nutrient concentrations in different areas of SWI-affected agroecosystems can indicate which habitats are net sources of N and P to downstream waterways and lead to an improved understanding of key biogeochemical controls on N and P transformations in these systems.

## MATERIALS AND METHODS

### Study site

We studied a suite of biogeochemical responses to SWI along the coast of Somerset County, Maryland, located in the mesohaline zone of the Chesapeake Bay. From 1985 to 2006, mean annual surface water salinities along the coastline of Somerset County ranged from 7.6 to 18 parts per thousand (ppt; 11.9–28.1 mS/cm) in the spring and 12.6–18 ppt (19.7–28.1 mS/cm) in the winter (Weinberg 2008).

Our study sites are located near Princess Anne in Somerset County, the southernmost county in Maryland (38.2° N, 75.7° W; Fig. 1A, B). Somerset County is a major producer of poultry, grains, oilseeds, and beans (USDA-NASS 2012). The area receives an average of 1085 mm of precipitation annually. The mean annual high temperature is 19°C, and the mean annual low temperature is 10.2°C (NOAA-NCEI 2018). Over the course of the study, conditions were wet and warm relative to an average year. Total annual precipitation ranged from 1370 to 1610 mm, mean high temperatures ranged from 20.7 to 20.9°C, and mean low temperatures ranged from 8.8 to 9.5°C (2016–2018).

This region has a land use history of clearcutting and reforestation (Trimble 2008). Until the 1930s, most upland areas of Somerset County were agricultural (Markewich et al. 1990). Now,

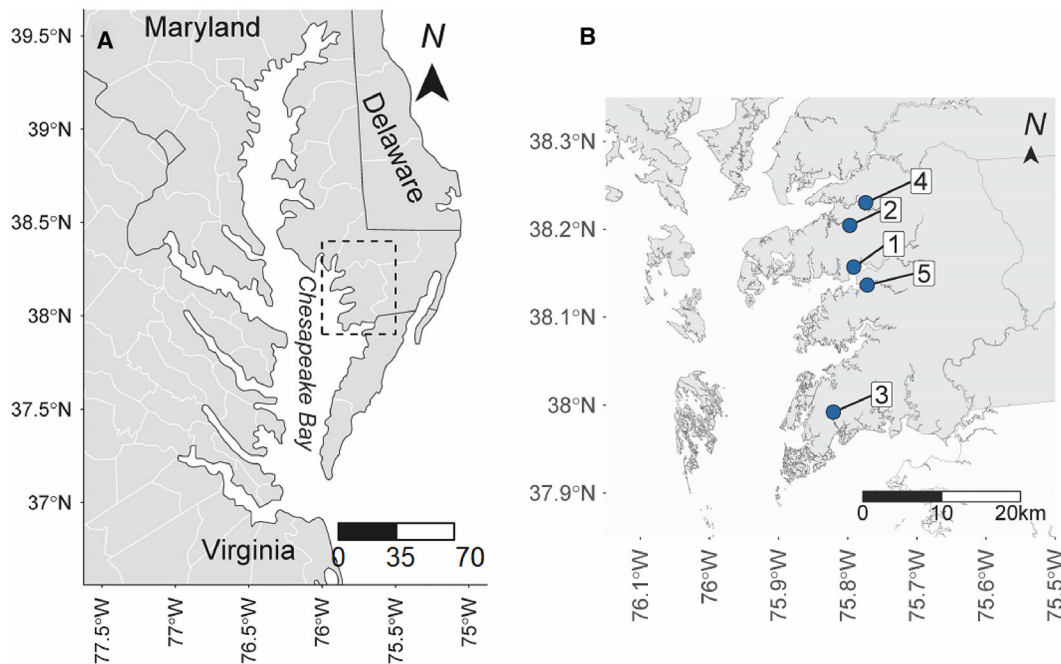


Fig. 1. (A) Map of the Chesapeake Bay region, United States. Study area is outlined by the dotted rectangle; (B) map of the study area and agroecosystem site locations, Somerset County, located within Maryland in the Chesapeake Bay.

roughly 32% of the county's land area is in agriculture (United States Department of Agriculture - National Agricultural Statistics Service 2012) and ~50% is loblolly pine-dominated forest (Kirwan et al. 2007, Lister et al. 2011). Tidal salt marshes cover about 15% of Somerset County and are located along its coastline (Shepard et al. 2013).

In 2017, agricultural runoff contributed to 42% of N and 56% of P loading in the Chesapeake Bay (Chesapeake Bay Program 2018). Excess nutrients can cause hypoxic zones, algal blooms, and fish kills (Boesch et al. 2001, Burgin and Hamilton 2007, Perez 2015). Though many parts of the Bay are showing water quality improvement, as of 2016, 58% of Bay tidal waters did not meet standards for dissolved oxygen, chlorophyll a, and water clarity, key indicators of good water quality (Zhang et al. 2018). In 2017, the U.S. Geological Survey found increasing N and P loads at several monitoring stations along major tributaries that drain into the Bay (Moyer and Blomquist 2018). In most tributaries within our study area, nutrient concentrations exceed water

quality standards set by the U.S. Environmental Protection Agency (EPA; Maryland Department of the Environment 2018). Moreover, P management has been particularly challenging in this area due to the large and intensifying poultry industry and the historic over-application of P-rich poultry litter on farm fields (Kleinman et al. 2011, Sharpley et al. 2014, Waldrip et al. 2015).

#### *Water sample collection and analysis*

Water samples were collected four to five times (approximately once a month) over late spring to early fall each year (2016–2018;  $n_{\text{total}} = 13$ ) at five different agroecosystem sites along the west coast of Somerset County. Each site comprised at least four of the following habitats: (1) forest; (2) intruded forest; (3) abandoned field; (4) intruded field; (5) ditch; (6) tidal creek; and (7) marsh (Table 1). An illustrated example of the layout of a typical site is shown in Fig. 2A. Each habitat was represented at a minimum of three sites (Table 1). Descriptions of the soil type for each habitat are included in Table 2. Broadly, marshes consisted of Histosols, intruded forests consisted

of Alfisols, and all other habitats were silt loam Ultisols (Table 2). Complete chemical properties of these soils can be found on the Web Soil Survey website (Soil Survey Staff 2018).

Samples from ditches and tidal creeks were grab samples taken ~30 cm below the surface of the water. The total water depth ranged from ~1 to 1.5 m in the creeks and 0.5 to 1 m in the ditches depending on streamflow conditions at the time. Soil porewater samples from all other habitats were collected using ceramic cup lysimeters (22 mm diameter; Soil Solution Access Tubes, Irrometer, Riverside, California, USA). Lysimeters were installed in three pairs in each habitat. Each pair consisted of a lysimeter at 25 and 50 cm depth in the soil profile. All water samples (surface and porewater) were kept at 4°C after collection and transported within 24 h to the University of Maryland where they were filtered (Whatman No. 42; 2.5 µm), and frozen until analysis. A subsample of each sample to be analyzed for dissolved inorganic P (DIP measured as SRP) was acidified to pH ~2 with hydrochloric acid (HCl) to prevent PO<sub>4</sub><sup>3-</sup> coprecipitation with Fe upon sample exposure to oxygen. Because we used a larger pore size filter paper, these samples may contain both soluble reactive phosphorus (SRP) and very low concentrations of acid hydrolyzable phosphorus (Chesapeake Bay Program 2019). However, we will refer to this form of DIP as SRP herein. Samples were analyzed colorimetrically on a LACHAT QuikChem (LACHAT Instruments, Loveland,

Colorado, USA) using the molybdate-blue method for SRP (detection limit 0.01 mg PO<sub>4</sub>-P/L), the salicylate–nitroprusside method for NH<sub>4</sub>-N (detection limit 0.02 mg NH<sub>4</sub>-N/L), and the sulfanilamide method for NO<sub>3</sub>-N (detection limit 0.025 mg NO<sub>3</sub>-N/L). Concentrations of NO<sub>3</sub>-N were only measured in 2016 water samples. Over 90% of NO<sub>3</sub>-N concentrations were below the minimum detection limit of the colorimeter and did not differ significantly by habitat, so we did not continue to measure NO<sub>3</sub>-N in 2017 and 2018. We measured EC and pH on unacidified samples (Orion Versa Star Pro; Thermo Fisher Scientific, Hampton, New Hampshire, USA). In 2018, lysimeters were damaged by a tractor on one of the three abandoned fields we were monitoring, so we were unable to analyze that habitat as part of our statistical model for that year.

#### Total dissolved P and Fe

Water samples from 2017 were digested to determine total P using potassium persulfate (K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>)–sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) method, which converted all forms of DOP to measurable SRP. Digested extracts were diluted 2:5 with a 1.0 mol/L sodium bicarbonate solution before colorimetric analysis using the molybdate-blue method for PO<sub>4</sub>-P (representing TDP) on a LACHAT QuikChem (LACHAT Instruments). Dissolved organic P was calculated as the difference between TDP and SRP. A subset of water samples from one sampling date (5 July) in 2017 were analyzed for total dissolved Fe (TDFe)

Table 1. Description and number of habitats (*n*) measured for each year of study.

Habitat	Description	Water sample type	<i>n</i> <sub>Year</sub>		
			<i>n</i> <sub>2016</sub>	<i>n</i> <sub>2017</sub>	<i>n</i> <sub>2018</sub>
Forest	Forested areas that do not show evidence of saltwater intrusion. (i.e., no tree mortality or encroachment of marsh plant species)	Soil porewater	4	4	4
Intruded forest	Forested areas that show evidence of saltwater intrusion (i.e., displaying tree mortality and encroachment of marsh plant species)	Soil porewater	3	3	3
Abandoned field	Former agricultural fields in their second year of abandonment as of summer 2016 and not affected by saltwater intrusion. They were left fallow due to being formerly leased to solar companies	Soil porewater	3	3	2
Intruded field	Actively farmed agricultural fields undergoing saltwater intrusion (i.e., displaying crop death and encroachment of native marsh plant species such as <i>Spartina patens</i> and <i>Distichlis spicata</i> )	Soil porewater	3	3	3
Ditch	Agricultural ditches directly surrounding and draining actively farmed fields	Surface water	5	5	5
Tidal creek	Creeks hydrologically connected to agricultural ditches and larger tributaries	Surface water	0	3	3
Marsh	Tidal <i>Spartina patens</i> salt marshes	Soil porewater	5	5	5

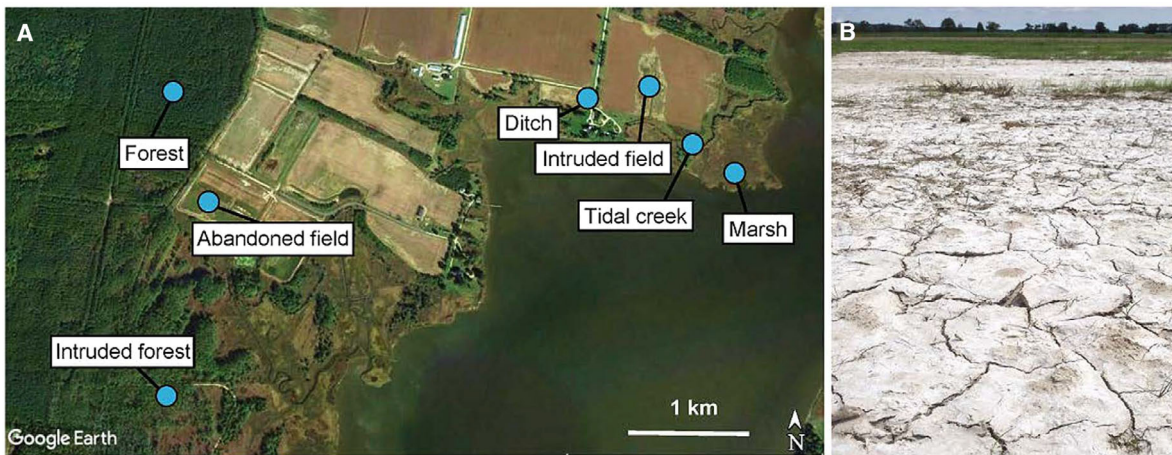


Fig. 2. (A) Satellite imagery of one of the study sites with labeled habitats. (B) Salt crust on an intruded farm field at one of the study sites. Panel A imagery taken from Google Earth (Google Earth 2019).

Table 2. Soil types of each habitat within each agroecosystem site with soil classification in parentheses.

Habitat	Agroecosystem site				
	1	2	3	4	5
Intruded forest	Sunken silt loam (Typic Endoaqualfs)	NA	Sunken silt loam (Typic Endoaqualfs)	NA	Sunken silt loam (Typic Endoaqualfs)
Forest	Quindocqua silt loam (Typic Endoaqualfs)	Othello silt loam (Typic Endoaqualfs)	Othello silt loam (Typic Endoaqualfs)	Quindocqua silt loam (Typic Endoaqualfs)	Quindocqua silt loam (Typic Endoaqualfs)
Abandoned field	Quindocqua silt loam (Typic Endoaqualfs)	NA	NA	Woodstown silt loam (Aquic Hapludults)	NA
Intruded field/ditch	Manokin/Queponco silt loam (Aquic Hapludults, Typic Hapludults)	Queponco silt loam (Typic Hapludults)	Othello, Fallsington silt loam (Typic Endoaqualfs, Typic Endoaqualfs)	Manokin/Queponco silt loam (Aquic Hapludults, Typic Hapludults)	Manokin/Queponco silt loam (Aquic Hapludults, Typic Hapludults)
Tidal creek/marsh	Transquaking/Mispillion organic deposits underlain by loamy mineral sediments (Typic Sulfihemists, Terric Sulfihemists)	Transquaking/Mispillion organic deposits underlain by loamy mineral sediments (Typic Sulfihemists, Terric Sulfihemists)	Honga peat (Terric Sulfihemists)	Transquaking/Mispillion organic deposits underlain by loamy mineral sediments (Typic Sulfihemists, Terric Sulfihemists)	Transquaking/Mispillion organic deposits underlain by loamy mineral sediments (Typic Sulfihemists, Terric Sulfihemists)

Note: Table information is compiled from the Web Soil Survey (Soil Survey Staff 2018).

using the modified ferrozine method to reduce all dissolved Fe(III) to Fe(II) (Viollier et al. 2000). This sampling date reflected water samples with the highest overall SRP concentrations from 2017. Therefore, we assumed that TDFe concentrations would represent each habitat’s potential to form Fe-P complexes during periods of higher SRP concentrations in soil porewater and surface water based on stoichiometric calculations of the molar ratio of TDFe:SRP present in the sample.

Samples were run for Fe, which represents the total dissolved Fe(II) and Fe(III) present in the water samples, on an atomic absorption spectrometer (PinAAcle 900; PerkinElmer, Shelton, Connecticut, USA).

**Chesapeake Bay Program data processing**

To estimate potential estuary-derived contributions of N and P, we analyzed data from the Chesapeake Bay Program from 2016 to 2018 (CBP;

Chesapeake Bay Program 2019). Four monitoring stations were chosen near our study sites in the mainstem portion of the Bay, where tributaries within our study sites drained (stations EE3.1, EE3.2, EE3.3, and EE3.4). We calculated the mean and standard error of the concentrations of  $\text{NH}_4\text{-N}$  and SRP in surface water samples collected at these monitoring stations from late spring through early fall of 2016–2018 (i.e., the same time frame as the study).

### Statistical approach

To examine differences in water EC, pH,  $\text{NH}_4\text{-N}$ , SRP, DOP, and TDFe by habitat, we used a linear mixed-effects (LME) model lme4 package for R (Bates et al. 2018) with a repeated-measures design. Habitat was included as a main effect, and sampling date and lysimeter identification numbers nested within each agroecosystem site were included as random effects. We found no significant differences in soil porewater characteristics between each lysimeter pair (25 cm vs. 50 cm below ground surface). Thus, we took the average value of each pair, which were considered replicates of the habitat-level chemistry with habitat as the true replicate. We used Tukey's post hoc tests to examine pairwise differences in water chemistry among habitats (multcomp package; Hothorn et al. 2017). We used the Box-Cox method (Box and Cox 1964) to transform the data prior to analysis to meet the assumptions of the statistical model when needed. Less than 10% of values were at or below the detection limit for each analysis. These values were assigned a value of half of the detection limit as it was the best approximation that could be tailored to the datasets based on their non-normal distributions. All statistics were computed in RStudio (RStudio Team 2019).

## RESULTS

Results from the LME models show a significant main effect of habitat on EC, pH,  $\text{NH}_4\text{-N}$ , and SRP within each year and across all years and for TDFe for the one date sampled in 2017 (Table 3; in all cases,  $P < 0.001$ ). Subsequent  $P$  values reported in this section are from the results of Tukey's post hoc tests. Across all years, we found significantly higher surface water and soil porewater EC levels in the intruded fields

and marshes (15.45 and 16.59 mS/cm, respectively) than in forests (2.15 mS/cm; Table 4, Fig. 3,  $P < 0.01$ ). These EC levels correspond to salinity values of roughly 9.00–9.73 and 1.10 ppt at 25°C. Electrical conductivity was always significantly higher in intruded forests than in forests and in intruded fields than in abandoned fields (Fig. 3,  $P < 0.01$ ). Across all years, the lowest pH was in the forests and intruded forests (4.95 and 4.95, respectively) and the highest pH was in the marshes and tidal creeks (7.26 and 7.66, respectively; Table 4, Fig. 4). Marsh pH ranged from 7.06 in 2016 to 8.74 in 2018. In the marshes, pH was consistently significantly higher than in the intruded forests for each year and across all years ( $P < 0.01$ ). Marsh pH was also significantly higher than forest pH in 2017 and 2018 (Fig. 4,  $P < 0.01$ ).

Ammonium-N concentrations were significantly higher in the marshes than in any other habitat each year and across all years ( $P < 0.01$ ). They ranged from 1.76 mg  $\text{NH}_4\text{-N/L}$  in 2017 to 2.65 mg  $\text{NH}_4\text{-N/L}$  in 2016. There were no other significant differences in  $\text{NH}_4\text{-N}$  concentrations among habitats across the study period. In all habitats except for marshes,  $\text{NH}_4\text{-N}$  concentrations were over three to seven times higher in 2016 as compared to 2017 or 2018. For example, in 2016 forests were 0.98 mg  $\text{NH}_4\text{-N/L}$ , but in 2018, they were 0.15 mg  $\text{NH}_4\text{-N/L}$  (Fig. 5). Because  $\text{NO}_3\text{-N}$  concentrations were very low across habitats in 2016 (in over 90% of samples lower than the method detection limit), we did not continue to measure them in 2017 and 2018. Across study years, SRP concentrations were significantly higher in the ditches (0.33 mg P/L) than on the intruded fields (0.30 mg P/L), but a significant difference was not detected between the ditches and the marshes (0.29 mg P/L,  $P < 0.01$ ). Concentrations of SRP in the intruded fields were approximately three times lower in 2018 than in 2016 (0.12 mg P/L and 0.38 mg P/L, respectively). Overall, SRP concentrations were an order of magnitude higher in marshes and ditches than in abandoned fields, forests, and intruded forests (Table 4 and Fig. 6).

Dissolved organic P was measured only in 2017 and ranged from 0.46 to 0.87 mg P/L (Table 4). In forests, intruded forests, and abandoned fields, DOP concentrations were an order of magnitude higher than SRP concentrations,

Table 3. Results of type III one-way ANOVA with Satterthwaite's method for unbalanced designs for the effect of habitat on each variable.

Year	Variable	Numerator df	Denominator df	Sum of squares	F-value	P
2016	EC	5	80	96.14	18.79	<0.0001
2017	EC	6	72	104.49	24.18	<0.0001
2018	EC	5	80	135.00	31.40	<0.0001
all	EC	5	416	126.22	25.72	<0.0001
2016	pH	5	80	1411.20	6.33	<0.0001
2017	pH	6	72	600,000	7.62	<0.0001
2018	pH	5	80	1275.00	10.175	<0.0001
all	pH	5	416	2665.20	8.04	0.00001
2016	SRP	5	80	125.73	32.85	<0.0001
2017	SRP	6	72	133.38	20.10	<0.0001
2018	SRP	5	80	63.75	15.44	<0.0001
all	SRP	5	416	186.18	42.40	<0.0001
2016	NH <sub>4</sub> -N	5	80	24.75	21.91	<0.0001
2017	NH <sub>4</sub> -N	6	72	76.26	15.18	<0.0001
2018	NH <sub>4</sub> -N	5	80	77.16	22.30	<0.0001
all	NH <sub>4</sub> -N	5	416	91.54	33.19	<0.0001
2017	DOP	6	72	8.65	3.469	0.03152
2017	TDFe	6	72	154.31	8.921	<0.0001

Notes: Variables are electrical conductivity (EC), pH, ammonium nitrogen (NH<sub>4</sub>-N), and soluble reactive phosphorus (SRP) across all years of the study, dissolved organic phosphorus (DOP) in 2017, and total dissolved iron (TDFe) from the 5 July 2017 sampling date.

Table 4. Porewater and surface water concentrations of electrical conductivity (EC), pH, ammonium nitrogen (NH<sub>4</sub>-N), and soluble reactive phosphorus (SRP) across all years of the study, dissolved organic phosphorus (DOP) in 2017, and total dissolved iron (TDFe) from the 5 July 2017 sampling date with  $\pm$ standard error in parentheses for each habitat.

Habitat	EC (mS/cm)	pH	NH <sub>4</sub> -N (mg N/L)	SRP (mg P/L)	DOP (mg P/L)	TDFe (mg Fe/L)
Abandoned field	2.59 (NA)	7.03 (NA)	0.31 (NA)	0.05 (NA)	0.87 (0.22)	0.29 <sup>a</sup> (0.13)
Ditch	9.71 <sup>b</sup> (0.98)	5.79 <sup>ab</sup> (0.17)	0.38 <sup>a</sup> (0.04)	0.33 <sup>c</sup> (0.04)	0.81 (0.09)	0.19 <sup>a</sup> (0.05)
Forest	2.15 <sup>a</sup> (0.36)	4.95 <sup>b</sup> (0.21)	0.40 <sup>a</sup> (0.12)	0.05 <sup>a</sup> (0.01)	0.49 (0.11)	5.61 <sup>a</sup> (2.74)
Intruded field	15.71 <sup>b</sup> (1.09)	5.18 <sup>ab</sup> (0.23)	0.39 <sup>a</sup> (0.05)	0.30 <sup>b</sup> (0.01)	0.81 (0.27)	1.56 <sup>a</sup> (0.89)
Intruded forest	9.84 <sup>b</sup> (0.36)	4.95 <sup>b</sup> (0.25)	0.37 <sup>a</sup> (0.04)	0.03 <sup>a</sup> (0.01)	0.66 (0.11)	18.08 <sup>b</sup> (5.23)
Tidal creek	9.62 (NA)	7.66 (NA)	0.11 (NA)	0.18 (NA)	0.46 (0.21)	0.14 <sup>a</sup> (0.02)
Marsh	16.58 <sup>c</sup> (0.84)	7.26 <sup>a</sup> (0.04)	2.29 <sup>b</sup> (0.2)	0.29 <sup>c</sup> (0.02)	0.81 (0.10)	0.74 <sup>a</sup> (0.27)

Note: Statistically significant differences in means within each variable are indicated by different letters at  $P < 0.01$ .

but there were no statistically significant differences in DOP concentrations across habitats (Fig. 7A). For the one sampling date selected in 2017, TDFe concentrations were significantly higher in the intruded forests than in the other habitats ( $P < 0.01$ ). Overall, TDFe concentrations ranged from 0.14 to 18.08 mg Fe/L (Table 4, Fig. 7B, C).

In order to determine whether the sources of NH<sub>4</sub>-N, SRP, and DOP were estuary-derived, we examined their values at four CBP monitoring stations. These nutrients were all two orders of

magnitude lower than the highest values we observed in our habitats (0.028, 0.005, and 0.010 mg/L, respectively).

## DISCUSSION

This research was conducted on privately owned land under a wide range of management practices rather than researcher-managed plots, and yet, we observed very strong trends in surface water and soil porewater chemistry. Overall, we saw significant differences in EC, NH<sub>4</sub>-N,



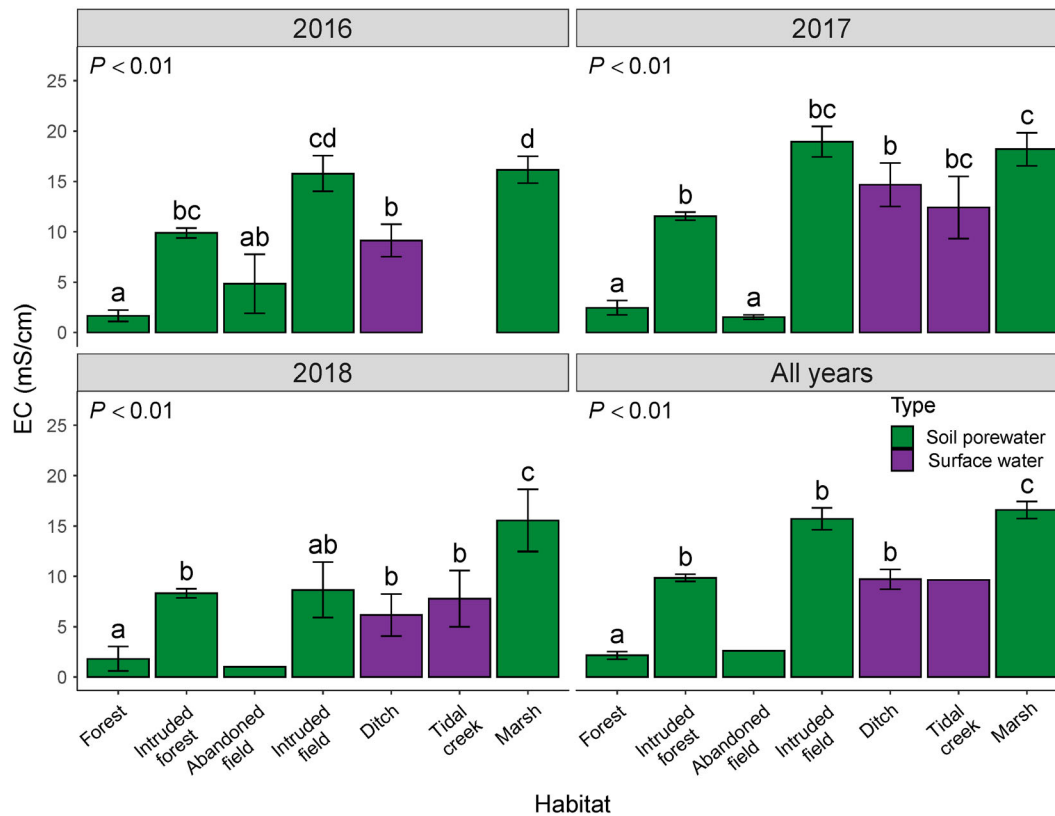


Fig. 3. Soil porewater and surface water conductivity (EC) by habitat for each year of the study. All years indicates average values taken across all three years. Statistically significant differences in means are indicated by different letters at  $P < 0.01$ . Error bars represent standard error of the mean. Tidal creeks were not monitored in 2016 but were added as a habitat in 2017 and 2018.

and SRP among habitats, with relatively consistent trends across years (Figs. 3, 5, 6;  $P < 0.01$ ).

The EC threshold tolerances for the survival of widely planted corn, soy, and wheat are 1.7, 5.0, and 6.8 mS/cm, respectively (Allison et al. 1954). We observed high EC on intruded fields across all years and sampling dates that were well above these thresholds for crop survival (Fig. 3). Though fertilizer application can affect EC levels, our previous research on these fields has shown that EC values are low in the center of the fields, where crops are still healthy, and increase dramatically toward the field edges, which are undergoing SWI (Tully et al. 2019b). Therefore, high EC levels were a direct result of SWI. We observed patches of bare soil on the intruded fields where plants did not grow. The formation of these patches was likely due to the combination of SWI and herbicide application that

farmers reported using on the edges of the fields to stave off the encroachment of invasive and native wetland plant species. In turn, plant die-off exposed patches of bare soil and effectively increased their evaporative surface, further increasing salt levels. In intruded fields, extensive evaporation can cause salts to move to the soil surface through capillarity (Fowler et al. 2014) and form a visible crust (Fig. 2B). Within these crusts, we measured EC as high as 60 mS/cm and it mostly ranged within 11–35 mS/cm (for reference, ocean water is ~55 mS/cm).

The pH of soil porewater on abandoned and intruded fields, and surface water in ditches and tidal creeks ranged from slightly acidic to slightly basic (pH of 5.18–7.66; Fig. 4). The soils across the intruded field habitats are Ultisols, which typically have a pH of  $< 5$  (Brady and Weil 2016). A common agricultural practice in this region is

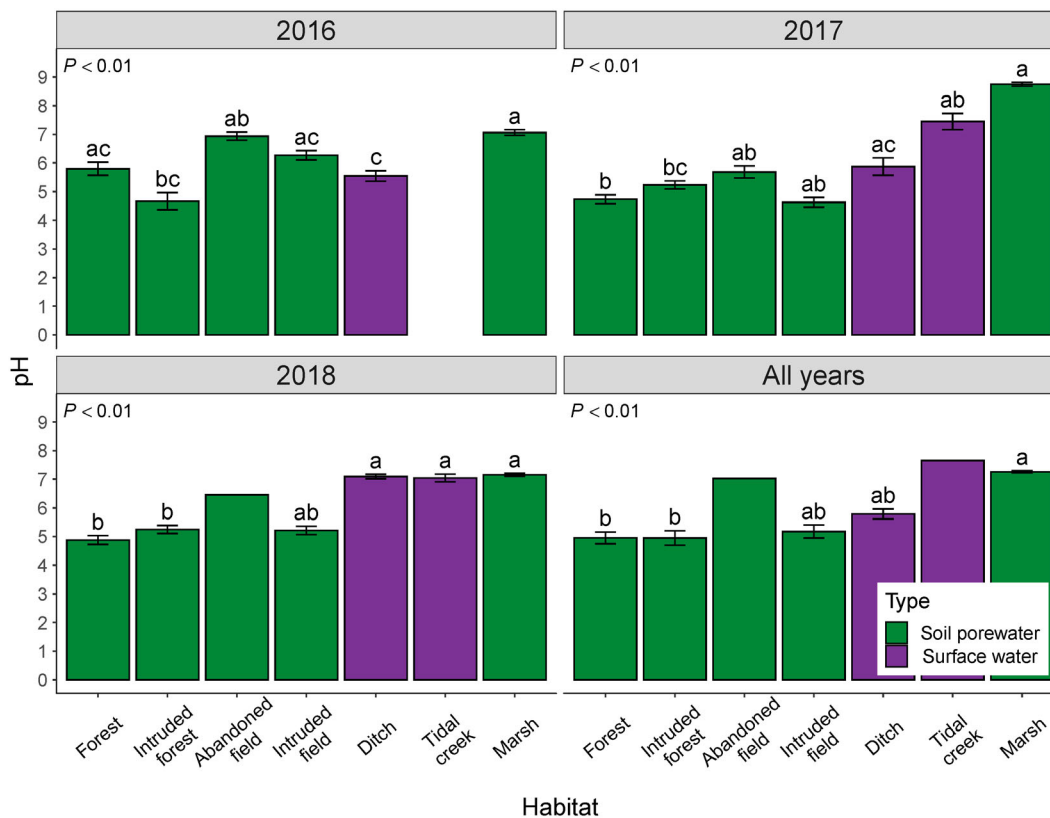


Fig. 4. Soil porewater and surface water pH by habitat for each year of the study. All years indicates average values taken across all three years. Statistically significant differences in means are indicated by different letters at  $P < 0.01$ . Error bars represent standard error of the mean. Tidal creeks were not monitored in 2016 but were added as a habitat in 2017 and 2018.

the addition of lime ( $\text{CaCO}_3$  or  $\text{CaMg}(\text{CO}_3)_2$ ) at 2.1–8.4 Mg/ha, to increase the pH of acidic topsoil (Gascho and Parker 2001). Alkaline saltwater inputs also increase porewater pH. From year to year, pH values may have differed within habitats due partly to varying freshwater and saltwater inputs.

### Nitrogen

Nitrogen cycling in soils is complex; therefore, there are several pathways through which N could have been transformed across the study habitats leading to the buildup of  $\text{NH}_4^+$  in marshes. Though we did not measure  $\text{NH}_4\text{-N}$  in reference salt marshes, other studies report concentrations in these habitats that range from 0.1 to 0.5 mg  $\text{NH}_4\text{-N/L}$  (Harvey and Hall 1992, Weston et al. 2011, Wilson and Morris 2012), an order of magnitude lower than the  $\text{NH}_4\text{-N}$

concentrations we observed in marshes receiving drainage from farm fields. We propose that this is due to a combination of effects including the following: leaching losses of  $\text{NO}_3^-$ ; denitrification; mineralization of organic marsh soils; dissimilatory  $\text{NO}_3^-$  reduction to  $\text{NH}_4^+$  (DNRA); and inputs of fertilizer N from agricultural fields upstream.

First,  $\text{NO}_3\text{-N}$  concentrations measured in our samples were mostly at or near the method detection limit ( $<0.025$  mg  $\text{NO}_3\text{-N/L}$ ). Since  $\text{NO}_3^-$  is prone to leaching, particularly in sandy soils (Clarke et al. 2002) like those of our study habitats (Table 2), some  $\text{NO}_3^-$  may have leached into deeper soil layers or groundwater below the depth of our porewater samples (Clarke et al. 2002, Tanner and Sukias 2011, Jessen et al. 2017). Second, as these systems are frequently inundated,  $\text{NO}_3^-$

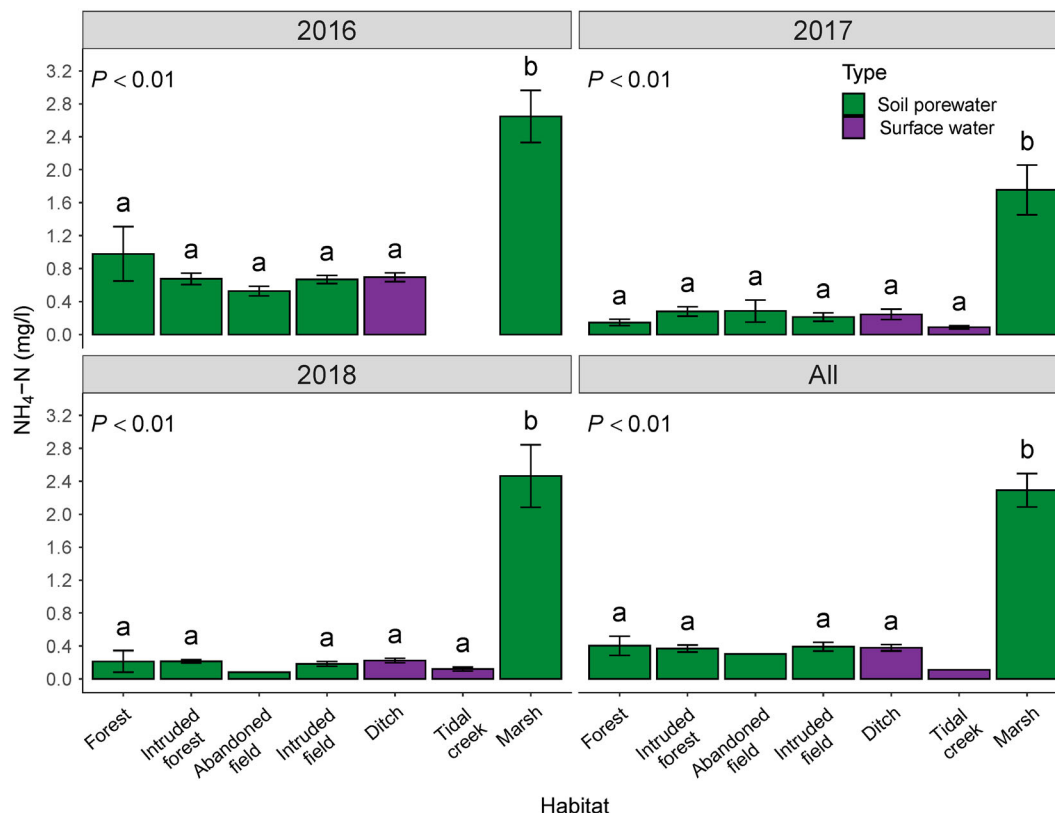


Fig. 5. Soil porewater and surface water ammonium nitrogen (NH<sub>4</sub>-N) concentrations by habitat for each year of the study. All years indicates average values taken across all three years. Statistically significant differences in means are indicated by different letters at  $P < 0.01$ . Error bars represent standard error of the mean. Tidal creeks were not monitored in 2016 but were added as a habitat in 2017 and 2018.

may also be converted to gaseous forms during denitrification (Burgin and Hamilton 2007, Helton et al. 2015). Third, in inundated marsh soils, mineralization of organic soils can lead to NH<sub>4</sub><sup>+</sup> release (Helton et al. 2015, Jia et al. 2017) as the reducing conditions prevent it from being oxidized to NO<sub>3</sub><sup>-</sup> (Jia et al. 2017). However, since we did not observe high DIN in the forests and intruded forests, which also contain soil high in organic matter, it is likely that this process does not account for all of the NH<sub>4</sub>-N measured in these habitats and in the marshes. Fourth, many studies have shown that while DNRA rates are low in freshwater wetlands, they can be much higher and comparable to rates of denitrification in salt marshes and anoxic marine sediments (Koop-jakobsen and Giblin 2010, Rütting et al. 2011, Giblin et al. 2013). For example, one N

tracer study in a salt marsh showed that DNRA converted 30% of the NO<sub>3</sub><sup>-</sup> to NH<sub>4</sub><sup>+</sup>, while 70% of the NO<sub>3</sub><sup>-</sup> was denitrified (Tobias et al. 2001). Finally, as farm fields receive influxes of saltwater, NH<sub>4</sub><sup>+</sup> ions can be released into soil porewater as they are replaced on negatively charged soil surfaces by base cations found in saltwater (Ardón et al. 2013). High NH<sub>4</sub><sup>+</sup> water may then be flushed downstream via tides or storm events through highly connected ditch networks. This mechanism may partially explain why we measured much higher NH<sub>4</sub>-N concentrations in the marshes than in any other habitat (Fig. 5). Previous studies also report high soil porewater concentrations of NH<sub>4</sub>-N (1–10 mg/L) in wetlands bordering agricultural areas (Koretsky et al. 2005, Ardón et al. 2013, 2017), which suggests that agricultural N inputs may

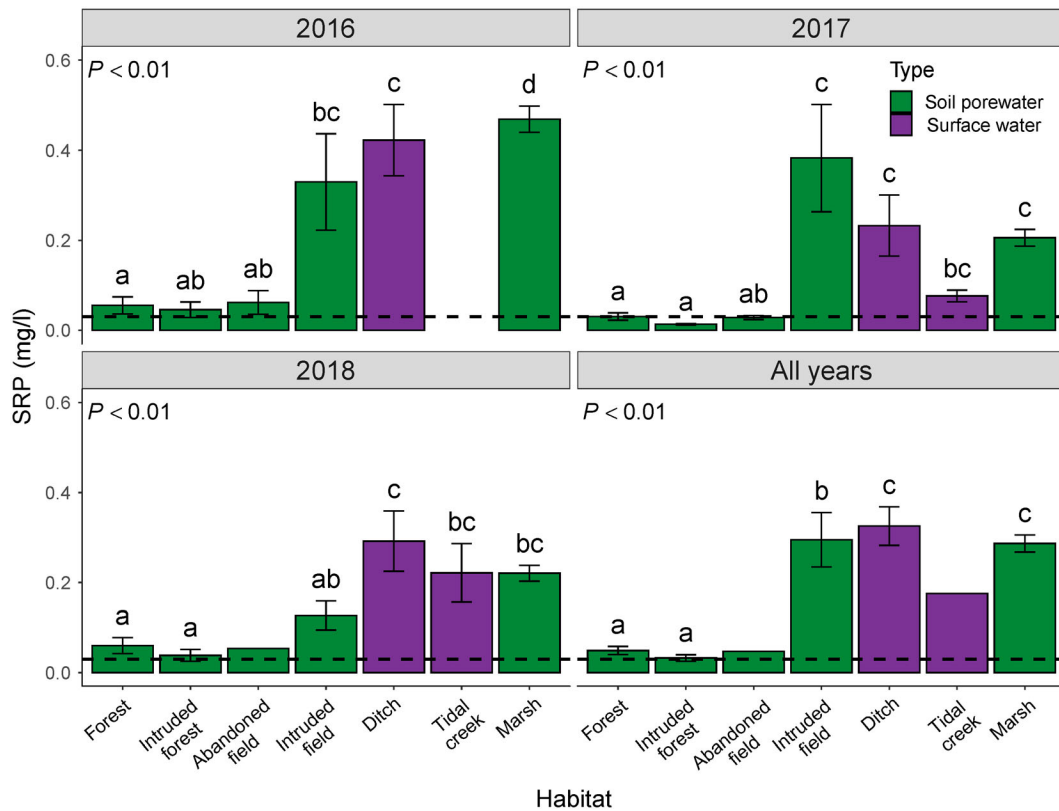


Fig. 6. Soil porewater and surface water-soluble reactive phosphorus (SRP) concentrations by habitat for each year of the study. All years indicates average values taken across all three years. Error bars represent standard error of the mean. Statistically significant differences in means are indicated by different letters at  $P < 0.01$ . Dashed line shows EPA-designated total phosphorus concentration threshold of 0.03 mg P/L for eutrophication of rivers, streams, and creeks of the Atlantic Coastal Plain (USEPA 2000). Tidal creeks were not monitored in 2016 but were added as a habitat in 2017 and 2018.

increase  $\text{NH}_4\text{-N}$  concentrations in wetlands downstream of SWI-affected farmland.

**Phosphorus**

The most striking observation during the study period was the magnitude of SRP concentrations in ditches, tidal creeks, and marshes, which suggests that dissolved P may be transported from the intruded fields into agricultural ditches and eventually downstream into tidal creeks and marshes (Figs. 6, 7A). Even though the intruded fields were not significantly higher in SRP than the intruded forests and abandoned fields in 2016, their mean concentration was much higher (0.29 mg P/L across all years vs. 0.03 and 0.05 mg P/L in the intruded forests and abandoned fields; Fig. 6). The large variability in SRP

concentrations in the intruded fields was due to differences in farmer management practices over the course of the study. Based on data from our previous study, intruded fields on Site 2 and Site 3 received less P than on Site 1 in 2017 (Tully et al. 2019b). In 2018, two of the farmers (Site 2 and Site 3) stopped fertilizing their intruded fields as a response to worsening SWI. However, the fact that we still observed high SRP concentrations in the ditches and marshes through all three years of this study (and tidal creeks in 2017 and 2018) indicates that legacy P may still be moving off these fields and serving as a persistent source of nutrient pollution downstream. For comparison, abandoned fields had low SRP concentrations despite a legacy of agriculture and fertilizer application (Fig. 6) due to a combination of a lack of new

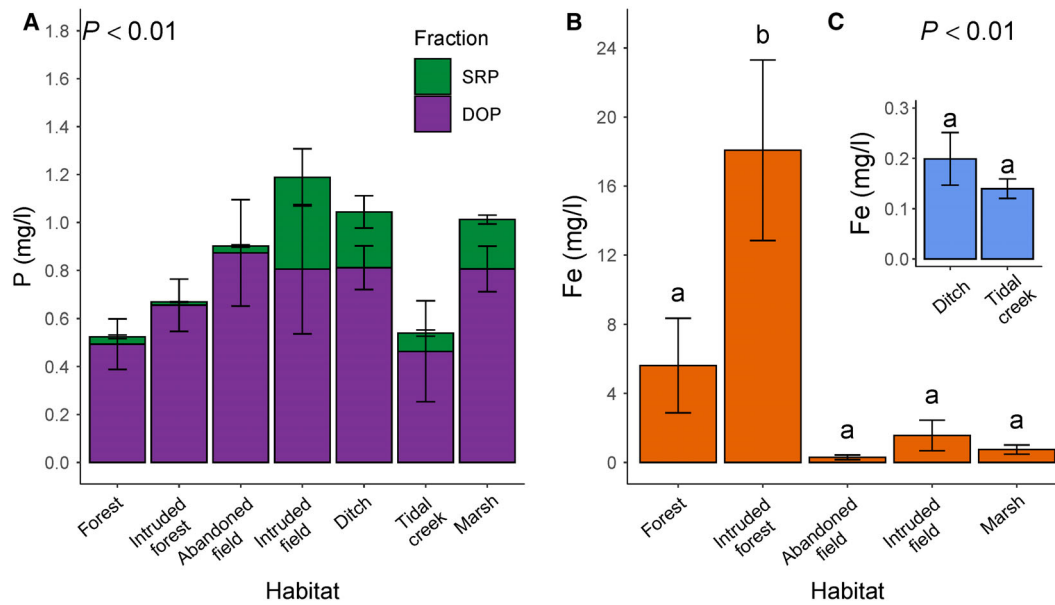


Fig. 7. (A) Soil porewater and surface water dissolved organic phosphorus (DOP) and soluble reactive phosphorus (SRP) concentrations in mg P/L by habitat in 2017. The sum of the means of the stacked bars equals the mean total dissolved phosphorus (TDP) for each habitat type. Error bars represent standard error of the mean. (B) Soil porewater total dissolved iron (TDFe) concentrations across habitats for the 5 July 2017 sampling date. Statistically significant differences in TDFe means are indicated by different letters at  $P < 0.01$ . Error bars represent standard error of the mean. (C) Surface water total dissolved iron (TDFe) concentrations across habitats for the 5 July 2017 sampling date. Statistically significant differences in TDFe means are indicated by different letters at  $P < 0.01$  and are comparable across Panel B and Panel C. Error bars represent standard error of the mean.

fertilizer inputs to these fields, consistent runoff of SRP downstream, and high biomass agricultural weeds and woody upland wetland plants (Gedan and Fernández-Pascual 2019) that may have assimilated some P into their tissues. In abandoned fields, soil porewater DOP concentrations were 17 times higher than SRP concentrations (Fig. 7A, Table 4), which is another indicator of legacy P. High DOP concentrations in the abandoned fields could be the result of plant residue decomposition and the formation of soil organic matter (Condon et al. 2005).

Dissolved organic P concentrations were not similar to SRP concentrations in water samples of each habitat. Instead, we observed very high DOP concentrations as compared to SRP concentrations across all habitats (Fig. 7A), which may be partially explained by a legacy of intensive agriculture in these areas. Most of the forested land cover on Maryland's Lower Eastern Shore is secondary growth and, at one

time, was under intensive agricultural use before it was converted back to forest (Lister et al. 2011). For comparison, DOP background concentrations in soil porewater of forests without a legacy of fertilizer inputs are often an order of magnitude lower than those measured in this study (0.02 mg P/L; Qualls and Haines 1991, Qualls et al. 2000, Lilienfein et al. 2004). High DOP concentrations in habitats not actively farmed may also be partially due to lateral movement of DOP through the soil or through surface water flow (Gburek et al. 2005). The high degree of hydrologic connectivity among habitats in this region raises the possibility that some of the N and P could be contributed by the saltwater itself. However, concentrations of N and P in the Chesapeake Bay mainstem were at least two orders of magnitude lower than concentrations measured in this study. This suggests that the high nutrient concentrations in water are the result of

agricultural practices and can cause localized water quality problems.

### Iron

Overall, the soil porewater and surface water of the habitats were high in TDFe, which has major implications for how P cycles through the system. We have previously shown that Fe changes from crystalline to poorly crystalline forms in the soil profile from the center of the SWI intruded fields to their border with tidal salt marshes (Tully et al. 2019b). Poorly crystalline Fe (Fe(III)) undergoes dissolution and is released as Fe(II) in soil porewater and surface water when the soils are inundated (Kemp et al. 2005, Musolf et al. 2017). Because wetting and drying cycles occur in the topsoil layers (<50 cm) on the intruded fields, Fe(II) in soil porewater can re-oxidize to Fe(III) when the soils dry and sorb to organic complexes, returning Fe to an oxidized, poorly crystalline form. Soil organic matter has been shown to have a high affinity for Fe and a tendency to sorb DIP to form Fe-P complexes (Giesler et al. 2005). Though we did not measure Fe(II) and Fe(III) speciation directly, the TDFe concentrations in soil porewater and surface water reflect the ability of TDFe to bind SRP under oxidizing conditions and release it under reducing conditions such as those in soils inundated with saltwater.

High TDFe concentrations in the forests and intruded forests reflect the effects of low soil porewater pH in these habitats (Figs. 4, 7C,  $P < 0.01$ ). Iron solubility has been shown to increase as pH decreases (Islam et al. 1980, Johnson and Loeppert 2006). Further, other studies have shown the stoichiometric relationship between dissolved Fe and SRP to be an important control on SRP bioavailability (Chambers

and Odum 1990, Hartzell and Jordan 2012). At 0.19 mg Fe/L (Fig. 7C), the concentration of TDFe measured in the ditch surface water was present at approximately a 1:1 (TDFe:SRP) molar ratio in 2017 (0.23 mg P/L; Fig. 6). This TDFe concentration is not high enough to bind all of the bioavailable SRP before it moves out to the tidal creeks, marshes, and larger tributaries, even if all of the TDFe were in oxidized form as Fe(III) compounds. Studies show that the molar ratio of TDFe:SRP in a water sample must be greater than 2:1 to allow all of the Fe to bind all of the SRP under completely oxidized conditions (Gächter and Müller 2003, Jordan et al. 2008). It is well documented that oxygen-depleted (reducing) conditions tend to form in the surface water and soil porewater in slow-moving water of agricultural ditches and connected creeks (King et al. 2015) and in marshes (Reddy and DeLaune 2008). Furthermore, in marshes, some of the Fe (II) may have been converted to insoluble iron sulfide (FeS) minerals under reducing conditions, reducing the ability of Fe to bind SRP, as has been documented in many other studies (Dierberg et al. 2011, Kraal et al. 2013, Hartzell et al. 2017). Therefore, TDFe is not present in high enough concentrations in the ditches, tidal creeks, and marshes to adequately buffer against SRP loss to downstream areas.

Each site is located along a minor tributary that drains into a major tributary, or a major tributary that drains into the Chesapeake Bay (Table 5). The distances between sites and their major tributaries range from just 0.15 to 4.4 km, and the route is hydrologically linked by ditch networks (Table 5). Instead of traveling through forested areas, agricultural fields on the sites drain into ditches that connect directly to tidal creeks. Therefore, forest habitats may not buffer

Table 5. Distance from center of study site to the closest major tributary in kilometers.

Agroecosystem site	Minor tributary	Major tributary	Site distance to major tributary (km)
1		Manokin River	0.2
2	Monie Creek	Monie Bay	4.4
3	Johnson Creek	Pocomoke Sound	2.7
4	Little Creek	Monie Bay	2.8
5	Back Creek	Manokin River	1.4
5 (Ditch)	Colburn Creek	Big Annemessex	0.7

Notes: Sites drain directly into minor tributary listed in this table. If no minor tributary is listed, the site drains directly into the major tributary. Site 5 had one ditch habitat that drained to separate major and minor tributaries from the rest of the site.

against SWI-related nutrient losses from farm fields. Even though SRP concentrations were low in forest soil porewater, forest soils were high in DOP likely as a result of the legacy of agricultural land use. As saltwater encroaches on forests, DOP can enter waterways, where it may be transformed to inorganic P. Current best management practices to curb P and N pollution in the Bay include creating vegetated buffers of grass, wetland plants, or trees (Sharpley et al. 2006). However, the fact that we found the highest inorganic N and P concentrations in marsh soil porewater suggests that tidal salt marshes may be less effective at reducing nutrient pollution downstream if nutrient fertilizer applications on upstream farm fields are not also reduced. This is due to the marshes' ability to convert N and P to bioavailable forms (i.e.,  $\text{NH}_4\text{-N}$  and SRP) that tend to remain solubilized in water bodies such as ditches, tidal creeks, and tidal salt marshes.

The excessively high concentrations of P in the focal habitats of this research suggest that coastal agroecosystems should be targeted for nutrient reduction initiatives to prevent nutrient release into waterways and consequent eutrophication (Paerl et al. 2014). Current models of nutrient loading in the Chesapeake Bay do not include potential contributions from legacy and SWI-derived nutrients. As we adapt to sea level rise and SWI, management approaches must take into consideration the potential release of nutrients from inundated soils to prevent problems associated with downstream pollution. Studies that measure legacy nutrient movement through ecosystems with SWI may help to explain why localized water quality issues persist. These findings can lead to the development of conservation practices adapted to meet water quality improvement goals in watersheds worldwide.

## CONCLUSION

As sea levels rise, SWI will continue to affect coastal agroecosystems. Biogeochemical changes in soil and water due to this process may lead to nutrient export from these areas. Here, we observed this effect on land with a legacy of heavy fertilizer applications. Remarkably, we found consistent patterns in soil porewater and surface water chemistry across study years. Both  $\text{NH}_4\text{-N}$  and SRP concentrations were very high

in the marshes as compared to reference conditions. This suggests that these nutrients are moving off farm fields and downstream through connected agricultural ditches and tidal creeks. Concentrations of TDFe support the idea that the habitats have a large capacity to release sorbed P from Fe-P complexes as dissolved oxygen levels decrease in soil porewater and surface water. However, TDFe concentrations are not high enough to rebind released P once it enters waterways with higher levels of dissolved oxygen. Therefore, we show N and P from coastal farmland may end up in high concentrations in nearby marshes and poised for export downstream where it may pose nutrient pollution issues in watersheds.

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