PHOSPHORUS RETENTION AND SOIL ORGANIC CARBON IN RESTORED AND NATURAL FRESHWATER WETLANDS

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Abstract: Nutrient (e.g., phosphorus) retention is an important function of wetlands that can improve water quality. We examined soil physical and chemical characteristics and phosphorus (P) sorption capacities in three recently restored herbaceous wetlands (RWs) on previously cultivated soils and three adjacent natural forested wetlands (NWs) on Kent Island, Maryland, USA. Our objective was to compare P retention in these two wetland types. As hypothesized, NW soils differed fundamentally in soil chemistry and had significantly higher total organic carbon (TOC) contents than RW soils ($5.7 \pm 1.7\%$ vs. $1.2 \pm 0.1\%$, respectively, $p < 0.05$). A number of soil properties (bulk density, pH, labile organic and microbial P, total N, and total N: total P ratios) differed between natural and restored wetlands, as expected from the differences in TOC. Concentrations of pyrophosphate-extractable (organically-bound) Al (Al_p) were an order of magnitude larger in NW than in RW soils (2099.1 ± 365.5 vs. 767.0 ± 194.7 kg/ha, respectively). Although past studies have suggested that higher concentrations of organically-bound Al can enhance P sorption, P-sorption capacities were significantly greater in the RW soils, likely due to differences in soil chemistry. In the RWs, 15 soil chemical parameters were significantly correlated with P sorption (based on single factor regression), including residual Al, oxalate-extractable Al and Fe, clay, HCl-extractable Fe and pyrophosphate-extractable Fe ($r^2 = 0.90, 0.89, 0.87, 0.85, 0.83$ and 0.82, respectively). In contrast, P sorption in the NWs was correlated only with Al_p ($r^2 = 0.68$). As restored wetland soils are likely in transition from a non-hydric to a hydric state, they should be reevaluated periodically to determine the ultimate effects of this transition on their capacity to retain P.

Key Words: aluminum, iron, phosphorus, phosphorus sorption, restored wetlands, total organic carbon

INTRODUCTION

Wetlands are often constructed or restored to replace the functions of natural wetlands lost to drainage for agriculture and/or development (Confer and Niering 1992, Mitsch 1994, Wang and Mitsch 1998). Constructed or restored wetlands can differ significantly in classification (as defined by Cowardin et al. 1979 or Brinson 1993) from the natural wetlands they were meant to replace, and thus, due to the inferred link between wetland structure and function, may not replace lost wetland functions (Craft et al. 1991, Brinson 1993, Mitsch 1994, Niswander and Mitsch 1995, Gwin et al. 1999, Shaffer and Ernst 1999).

Phosphorus (P) retention and transformation are important biogeochemical functions of wetlands that can improve the quality of surface and subsurface waters (Walbridge 1993, Mitsch 1994, Walbridge and Lockaby 1994, Weller et al. 1996, Lockaby and Walbridge 1998, Novak et al. 2004). Significant quantities of water-borne P can be removed by wetlands through the processes of sedimentation, plant uptake, microbial immobilization, and adsorption and precipitation reactions with clay, Al, Fe, and Ca minerals (Gale et al. 1994, Walbridge and Lockaby 1994, Vymazal 1995, Reddy et al. 1999). A number of human activities (e.g., agriculture, residential development, industry, municipal waste treatment) can produce both point and non-point source inputs of P to aquatic ecosystems (Walbridge and Struthers 1993, Mitsch 1994, Carpenter et al. 1998). Our study focused on non-point source inputs resulting from agriculture. Wetlands play a role in improving water quality because they often occupy

transition zones between uplands and aquatic ecosystems.

Soil sorption processes have been shown to be an important component of P retention in freshwater wetlands (Richardson 1985, Walbridge and Struthers 1993, Walbridge and Lockaby 1994). A number of studies have shown that the P sorption potential of a wetland soil can be predicted from its oxalate-extractable (amorphous, non-crystalline) Al (Al_o) and Fe (Fe_o) content (Richardson 1985, Freese et al. 1992, Walbridge and Struthers 1993, Yuan and Lavkulich 1994, Lockaby and Walbridge 1998, Bruland and Richardson 2004). Aluminum may be more important than Fe in controlling P sorption in wetland soils because Fe-bound P is subject to release under anoxic conditions (Richardson 1985, Walbridge and Struthers 1993, Vymazal 1995). Strong correlative relationships observed between soil organic matter (OM) content and Al_o concentration in forested wetlands in the southern United States suggest that the active form of Al in these soils may be Al complexed with OM rather than amorphous inorganic Al (Lockaby and Walbridge 1998, Axt and Walbridge 1999, Darke and Walbridge 2000). If so, the importance of OM as a factor controlling P sorption in some wetlands, through its interaction with Al, has been previously unrecognized.

Anaerobic conditions, frequently present in wetland systems, slow decomposition, allowing OM to accumulate (Paludan 1995, Mitsch and Gosselink 2000). Pools of OM may be smaller in recently restored wetlands because of the brief length of time enhanced OM accumulation has been occurring and the level of soil inundation, as well as the hydrogeomorphic class (Shaffer and Ernst 1999). Sodium pyrophosphate can be used to estimate Al and Fe associated with OM (Al_p and Fe_p); however, because Fe_p can include oxyhydroxide Fe in close association with OM and Al_p may also include Al hydroxides associated with or adsorbed to OM, the specificity of this extraction has been questioned (Schuppli et al. 1983, Parfitt and Childs 1988, Kaiser and Zech 1996).

We examined soil physical and chemical characteristics and phosphorus (P) sorption capacities in three recently restored herbaceous wetlands (RWs) and three adjacent natural forested wetlands (NWs) developed over similar substrates on Kent Island, Maryland, USA. Our objective was to determine how these two types of wetlands compared with respect to P retention. Because the RWs originated from previously cultivated soils, we hypothesized that they would differ fundamentally in soil chemistry from the NWs, particularly with respect to the accumulation of OM. We further hypothesized that higher soil OM concentrations in the NWs would favor the formation of organically-bound forms of Al (i.e., OM-Al complexes). Fi-

nally, because soil OM-Al complexes have been suggested as important agents of P retention in riparian wetlands in the southeastern US, we hypothesized that the NWs, with their greater OM accumulations, would have greater P sorption capacities, and thus be more effective at P retention, than the RWs.

MATERIALS AND METHODS

Study Sites

Study sites were located on Kent Island, Maryland, adjacent to the Eastern Shore of the Chesapeake Bay (Figure 1). Annual precipitation in the area is 105 cm (yearly snowfall is 54 cm). Mean summer air temperature is 23.4°C; mean winter air temperature is 2.1°C (Queen Anne's County Office of Economic Development 1998). The Braun study site was located at the Braun farm; Barnstable 1 and Barnstable 10 were located at the Barnstable farm. Each site consisted of a natural forested wetland (NW), a restored herbaceous wetland (RW), and an upland grass buffer strip (UP) composed of a mixture of native grasses. The Barnstable 10 NW was approximately 2 ha in area; the Braun and Barnstable 1 NWs were each approximately 4 ha in area. The RWs were approximately 4 ha (Barnstable 10), 1.6 ha (Braun), and 0.4 ha (Barnstable 1) in area, respectively (Whigham et al. 2002).

Prior to their conversion to ditched agricultural fields, the RW sites were likely forested depressional or slope wetlands *sensu* Brinson (1993) located in topographically low areas (Whigham et al. 2002). The RWs were restored in 1986 (Barnstable 1) and 1992 (Braun and Barnstable 10) by Chesapeake Wildlife Heritage to provide wildlife habitat and improve water quality (Pepin 1998, Jordan et al. 1999, Whigham et al. 2002). The RWs were restored to a hydrology similar to depressional wetlands as described by Brinson (1993), characterized by the highest water levels in winter, with a growing season drawdown (Pepin 1998). The RWs may also be classified as palustrine, persistent, emergent wetlands as described by Cowardin et al. (1979). The UP grass strips were created as buffers between the fertilized agricultural fields and the NWs and were maintained by Chesapeake Wildlife Heritage. The NWs were chosen for their proximity to the RWs and their location on similar soil types. In contrast to the open water herbaceous RWs, the NWs were characterized by closed canopy palustrine forest as described by Cowardin et al. (1979), with a seasonally flooded hydroperiod.

Our study focused on comparing forested natural systems, like those that were historically lost to agriculture and development, with the open water systems being constructed to restore lost wetland area in the
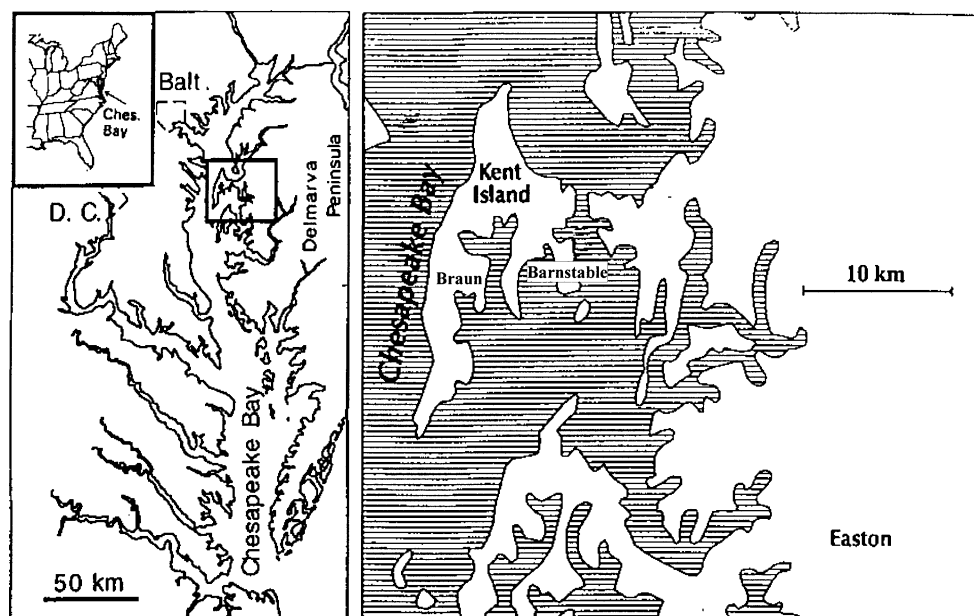
Figure 1. Location of study sites. One study site was located at the Braun farm; two study sites were located at the Barnstable farm. Each site included a natural forested wetland, a restored herbaceous wetland, and an upland grass buffer strip.

region. All wetlands studied (both RW and NW) receive agricultural drainage as bulk surface flow via channels that have been constructed in the agricultural fields surrounding each wetland. A detailed study of nutrient and sediment removal in the Barnstable 1 RW can be found in Jordan et al. (2003), and an analysis of aboveground biomass and nutrient dynamics in these and other RWs can be found in Whigham et al. (2002).

Restored wetland soils have been identified as Mattapex (Braun) or Mattapex/Elkton (Barnstable 1 and 10) series (Pepin 1998, Whigham et al. 2002); NW soils are mapped as Mattapex/Butlertown series; UP soils are mapped as Mattapex (Queen Anne's County, Maryland 1997). Mattapex soils are moderately well-drained, fine-silty, mixed, mesic Aquic Hapludults. Elkton soils are poorly drained, hydric, fine-silty, mixed, mesic Typic Endoaquults (National Cooperative Soil Survey 1998). Butlertown soils are similar to Mattapex but may include soils that are poorly drained or contain a perched water table on lower landscape positions (Queen Anne's County, Maryland 1997).

Soil Collection

Soil cores (0–13 cm) were collected randomly from each RW, NW, and UP site on July 15 (Braun) and August 4 (Barnstable 1, 10), 1997 by driving a 4-cm-diameter, 13-cm-long aluminum pipe with a sharpened edge into the ground after brushing away surface litter. Twelve soil samples were collected in each RW, each a composite of four randomly located cores collected

from previously established 2×2 m plots (n = 12). For the NWs and UPs, four 5×5 m plots were established at each site, and four soil cores were collected from each plot and composited to one sample per plot (n = 4 per UP or NW site). Soils were collected in polyethylene bags, kept on ice in the field, and stored at 4°C (field moist) upon returning to the laboratory. Soils were homogenized by hand, removing coarse roots and rocks when present. Phosphorus, Al, and Fe extractions and pH measurements were all begun within 24 h of soil collection.

General Soil Characteristics

All soil analyses were done using two replicates per soil sample. Soil pH was determined using a 1:2 soil: deionized water slurry (5 g soil:10 ml water) following a 10 min equilibrium period. Soil texture was determined by the Bouyoucos (1962) method. Moisture content was determined by oven drying soil subsamples at 90°C to a constant mass after samples had been composited and rocks and roots removed. Bulk density was calculated as the dry mass per unit volume of soil collected. Soil organic matter (OM) was estimated by mass loss on ignition (LOI) in a muffle furnace for 20 h at both 430°C and 550°C (Nelson and Sommers 1982, Carter 1993). Because Grewal et al. (1991) reported that using a dry combustion method such as LOI to estimate OM can lead to overestimation of soil OM content due to weight changes in thermally reactive clay minerals, total carbon (TC) was also determined using a Perkin-Elmer 2400 CHN Analyzer,

following determination of the presence of inorganic C (Nelson and Sommers 1996). When inorganic C concentrations are insignificant, total C is essentially equal to total organic carbon (TOC) (Grewal et al. 1991, Nelson and Sommers 1996). Total organic carbon was used as an index of OM in these soils (Nelson and Sommers 1996).

Soil P Fractions

Soil P fractions were characterized using a modification of the sequential extraction procedure of Paludan and Jensen (1995) (Axt 1997, Hogan 2000). This procedure identifies nine inorganic (P_i) and organic (P_o) soil P fractions and was chosen specifically for its ability to isolate P associated with Al and Fe in humic acid (HA) complexes. The nine P fractions are water-soluble (H_2O -P_i and -P_o), bicarbonate dithionite (BD-P_i and $-P_o$), sodium hydroxide P (NaOH-P_i and $-P_o$), humic acid P (HA-P), hydrochloric acid P (HCl-P_i), and residual P (Res-P). The water-soluble P (H_2O–P) fraction is an estimate of porewater and loosely adsorbed P. The bicarbonate dithionite (BD) fraction estimates P adsorbed onto the surfaces of oxidized Fe and Mn. Sodium hydroxide-extractable P (NaOH-P) includes P sorbed onto the surfaces of clay and Al oxides, as well as the majority of sediment organic P. Humic acid P (HA-P) estimates P in association with humic acids and is isolated via acidification of the NaOH fraction and filtration of the organic matter precipitate. Hydrochloric acid P (HCl-P) estimates calcium-bound P_i, and finally, residual P (Res-P) estimates refractory organic and some inorganic P. Extracts were centrifuged at 3400 rpm for 20 minutes and then filtered through Gelman Supor-450 0.45-um membrane filters. Humic acid extracts were filtered through Whatman GF/F glass fiber filters.

Soils were also analyzed for 0.5 M NaHCO$_3$-extractable (labile, available) and CHCl$_3$-labile (microbial biomass) P by the method of Hedley et al. (1982). Microbial biomass P (Mic-P) was estimated as the difference in 0.5 M NaHCO$_3$-extractable P from the CHCl$_3$-fumigated minus the non-fumigated soils. A correction factor (k_p) of 0.4 was used to compensate for microbial P not converted to phosphate during CHCl$_3$ fumigation (Hedley et al. 1982, Walbridge 1991, Qualls and Richardson 1995, Axt and Walbridge 1999).

The inorganic P_i concentrations of all extracts were analyzed colorimetrically as orthophosphate by the method of Murphy and Riley (1962), using a Technicon II Autoanalyzer (Bran and Luebbe Inc. 1989). Bicarbonate, CHCl$_3$-Bic, H_2O, BD, and NaOH extracts were also digested by wet oxidation with potassium persulfate in an autoclave (Koroleff 1983) to estimate total P (TP) concentrations. The organic P (P_o) concentrations of these extracts were estimated by difference (TP minus P_i). For each soil sample, total P was estimated by summing the TP concentrations of the nine extracts in the Paludan and Jensen (1995) sequential extraction scheme–H_2O-P_i, BD-P_i, NaOH-P_i, HA-P_i, HCl-P_i, Res-P_i, H_2O-P_o, BD-P_o, and NaOH-P_o. Total organic P (sum P_o) was defined as the sum of H_2O-P_o, BD-P_o and NaOH-P_o. Although HA and residual fractions may also contain some organic P, they are considered separately in this paper. Total P and N were also determined separately by digesting soils with sulfuric acid and potassium sulfate in a block digester using a mercuric oxide catalyst (Walbridge 1991).

P Sorption Capacity

Phosphorus sorption capacity was estimated in a 24-hour equilibration of 2.0 g dry weight equivalent fresh soil with 25 ml of 0.01 M CaCl$_2$ solution containing 16, 33, 130, or 260 mg P/L as KH$_2$PO$_4$ (Bache and Williams 1971) using non-sterilized soils (Darke and Walbridge 2000). The capacity of the soil to remove dissolved phosphate from solution was estimated by examining the decrease in orthophosphate concentration of each solution. Phosphorus concentrations were analyzed by the method of Murphy and Riley (1962) using a Technicon Autoanalyzer II (Bran and Luebbe Inc. 1989). Phosphorus sorption isotherms were constructed using the methods of Bache and Williams (1971) as modified by Richardson (1985) and Walbridge and Struthers (1993). A single point P sorption index (PSI) was also calculated as x/log c, where x = P sorbed in mg P/100g soil, and c = the equilibrium P concentration in μM/L at the 130 mg P/L level (Bache and Williams 1971). Previously sorbed P was estimated as the concentration of exchangeable or labile P present in each soil, determined by summing H_2O-P_i, BD-Pi and NaOH-P_i (Bache and Williams 1971, Menzies et al. 1999, Darke and Walbridge 2000). Phosphorus desorption was not examined in this study.

Soil Al, Fe, and Ca

Non-crystalline (amorphous) and crystalline Al and Fe were estimated by the method of Darke and Walbridge (1994). A sequential 0.2M acid ammonium oxalate (pH = 3.0) / 0.1 M NaOH extraction using fresh soil was used to estimate non-crystalline Al and Fe (Al$_o$ and Fe$_o$) and crystalline Al (Al$_c$), respectively. Non-crystalline plus crystalline Fe was estimated by a separate 16 h dithionite-citrate-bicarbonate (DCB) extraction. Crystalline Fe (Fe$_c$) was estimated as the difference between DCB Fe and Fe$_o$. Extractable Ca (Ca$_e$)

was estimated by the method of Thomas 1982. All extracts were clarified by centrifugation (20 min, 3400 rpm).

Organically bound (pyrophosphate-extractable) Al and Fe (Al_p and Fe_p) were estimated using a 0.1 M pyrophosphate (pH = 10) extraction (Parfitt and Childs 1988), with superfloc (0.5 ml of a 1.0g/kg solution) added to facilitate sedimentation (Schuppli et al. 1983), and was followed by a sequential 0.2 M acid ammonium oxalate extraction. Concentrations of Al and Fe extracted by acid ammonium oxalate following the 0.1M pyrophosphate extraction were compared with Al and Fe extracted by separate oxalate extraction to estimate organically bound (pyrophosphate-extractable) Al and Fe extracted during oxalate extraction.

Extracts from each step of the Paludan and Jensen (1995) sequential P fractionation were also analyzed for Al and Fe to estimate the concentrations of Al and Fe associated with each P fraction (not necessarily bound to P). Molar concentrations of metals in each Paludan and Jensen (1995) fraction were compared with molar P concentrations to identify potentially important metal-P associations. An association between HA-P and HA-Al or -Fe may suggest that P in the HA fraction exists as complexes of OM with oxides of Al or Fe, the metal oxide serving as a link between the HA and P (Paludan and Jensen 1995, Dolfing et al. 1999). All Al, Fe, and Ca concentrations were determined using a Perkin Elmer Model 5100 Atomic Absorption Spectrophotometer (Perkin Elmer 1982).

Statistical Analyses

Landforms (RWs, NWs, and UPs) were compared using analysis of variance (ANOVA) within each study site (Braun, Barnstable 1, and Barnstable 10, see Hogan 2000) and as an average of the three study sites (n = 3) to determine significant differences in soil chemistry. When ANOVA indicated statistical significance, differences among the means were analyzed using the Student-Newman Keuls multiple comparison test (Sokal and Rohlf 1995). Relationships between the P sorption index and 19 different soil characteristics that might be useful predictors of soil P sorption capacity were analyzed by both single factor and stepwise multiple regression. All statistical analyses were performed using SAS and PROC GLM when data were unbalanced (SAS 1996) or Microsoft Excel 97 SR-1 (Microsoft Corporation 1997). Significant differences were considered at p < 0.05, with the exception of the stepwise multiple regression where significant differences were considered at p < 0.15; SAS (1996) recommends use of a moderate significance level in stepwise multiple regression to avoid inclusion of var-

iables that do not contribute to model predictive power.

RESULTS AND DISCUSSION

Soil Characteristics and Nutrient Concentrations

As hypothesized, RW and NW landforms differed fundamentally in soil chemistry, most likely because the RWs were restored on previously drained and cultivated soils while the NWs remained forested. Differences included significantly higher TOC concentrations in NW than in RW or UP soils (5.7, 1.2, and 1.0%, respectively) and significantly lower soil pH and bulk densities in the NW soils (Table 1). Both soil pH and bulk density are known to co-vary with OM content in wetland soils (Mitsch and Gosselink 2000).

Labile organic P, microbial P, total N, and soil N:P ratios were all significantly higher in NW than in RW or UP soils (Table 1). Microbial P accounted for 21% of total P in NW soils, versus 6% in RW and UP soils (Table 1), consistent with Axt and Walbridge (1999) and Wright et al. (2001), who also found significantly higher concentrations of microbial P in forested riparian wetland vs. adjacent upland soils in VA and GA, respectively. However, percentages of microbial P in NW soils (21%) were slightly lower than those reported for bay forest soils in North Carolina (37.4%) or floodplain forest soils in Georgia (29.2%) (Walbridge 1991 and Wright et al. 2001, respectively). Surface soil N:P ratios in the NWs (3.2 to 8.5) (Table 1) are at the low end of the range reported for forested wetlands (swamps) in temperate North America (4.6 to 25.8) (Bedford et al. 1999).

Maximum concentrations of $NaOH-P_o$ and HA-P also occurred in NW soils, where organic P comprised the largest percentage (69.7 ± 11.9 %) of total P extracted by the Paludan and Jensen (1995) procedure (Figure 2a). Concentrations of residual P were generally lower in RW than in NW soils and significantly lower than in UP soils (Figure 2a). Concentrations of HA-P (12.5% of TP in NWs, 7.1% in RWs, and 7.8% in UPs) (Figure 2a) are comparable to those reported for forested riparian wetlands in VA (13 to 19% of TP; Axt 1997) and drained and recently reflooded Histosols in the Everglades (12% of TP; Ivanoff et al. 1998). However, they are lower than those reported for a freshwater marsh in South Carolina (24–51% of TP) (Paludan and Morris 1999) and both non-enriched (23.6%) and enriched (39.9%) soils in the northern Everglades (Qualls and Richardson 1995).

Organic matter accumulation is generally favored in wetland soils due to the inhibition of decomposition by periodic or continual flooding (Mitsch and Gosselink 2000). In contrast, soils of newly constructed or

Table 1. Soil properties, percent P_o, bicarbonate-extractable and microbial ($CHCl_3$-labile) P, soil N:P ratios, and total N and P for the average of the natural wetlands (NWs), restored wetlands (RWs), and uplands (UPs). Data are means $\pm$ standard errors except for pH, which are means with ranges in parentheses. The F-value for the ANOVA is shown and the lower case letters x, y, and z indicate significant differences at p < 0.05 between the averages of the NWs, the RWs, and the UPs to a soil depth of 13 cm.

	Averages			
	NWs (n = 3)	RWs (n = 3)	UPs (n = 3)	F-Value ($F_{2,6}$)
pH	5.0 (4.4–5.7) x	6.4 (6.3–6.6) y	6.6 (6.2–7.1) y	9.4
Bulk density (g/cm³)	0.9 $\pm$ 0.1 x	1.1 $\pm$ 0.1 y	1.5 $\pm$ 0.0 z	39.6
% Total Organic Carbon	5.7 $\pm$ 1.7 y	1.2 $\pm$ 0.1 x	1.0 $\pm$ 0.1 x	7.2
% Sand	24.5 $\pm$ 10.0	29.0 $\pm$ 2.5	31.5 $\pm$ 5.3	0.3
% Clay	34.8 $\pm$ 7.1	22.2 $\pm$ 2.4	17.3 $\pm$ 2.1	4.0
% Silt	40.8 $\pm$ 4.1	48.8 $\pm$ 3.5	51.3 $\pm$ 3.2	2.3
% P_o[1]	69.7 $\pm$ 11.9	52.6 $\pm$ 1.8	59.7 $\pm$ 11.7	0.8
Bicarb-P_i[2] (kg/ha)	59.0 $\pm$ 14.3	107.0 $\pm$ 13.2	81.2 $\pm$ 24.7	1.7
Bicarb-P_o[3] (kg/ha)	54.4 $\pm$ 7.6 y	24.6 $\pm$ 2.4 x	30.1 $\pm$ 3.1 x	10.2
Available P[4] (kg/ha)	114.0 $\pm$ 6.7	131.6 $\pm$ 13.1	111.2 $\pm$ 23.4	0.5
Microbial P[5] (kg/ha)	101.8 $\pm$ 21.7 y	26.4 $\pm$ 12.1 x	39.1 $\pm$ 10.7 x	6.7
N:P ratio	5.4 $\pm$ 1.6 y	2.4 $\pm$ 0.3 x	1.9 $\pm$ 0.2 x	28.0
Total N[6] (kg/ha)	3650.6 $\pm$ 266.0 y	1549.0 $\pm$ 118.7 x	1948.6 $\pm$ 104.7 x	39.0
Total P[6] (kg/ha)	783.1 $\pm$ 149.7	689.7 $\pm$ 113.2	1070.1 $\pm$ 43.9	3.2

[1] Total P_o/Total P.
[2] Inorganic P extracted with 0.5M sodium bicarbonate.
[3] Organic P extracted with 0.5M sodium bicarbonate.
[4] Bicarbonate P_i + bicarbonate P_o.
[5] Increase in total 0.5M sodium bicarbonate-extractable P following an 18 hour chloroform fumigation.
[6] Total N or P as found by sulfuric acid, potassium sulfate and mercury oxide block digestion procedure.

restored wetlands may retain properties more typical of the terrestrial soils from which they were created, including lower soil OM concentrations and higher concentrations of weatherable minerals such as Fe and Mn (Craft et al. 1991).

Al and Fe Fractionation

In all landforms, residual-Al accounted for more than 80% of the Al extracted by the Paludan and Jensen (1995) sequential fraction technique (Figure 2b). A structural component of clay, residual-Al was significantly positively correlated with soil clay content (r^2 = 0.76, 0.84, and 0.48 in NW, RW and UP soils, respectively) (Hogan 2000). Sodium hydroxide-extractable Al comprised approximately 10% of total Al; all other fractions each accounted for less than 4% of total Al (Figure 2b). Residual and BD-extractable Fe were the two largest Fe fractions, accounting for $\geq$50% and $\geq$30% of the total Fe present in all landforms (Figure 2c). Hydrochloric acid-extractable Fe comprised approximately 5 to 6.5% of total Fe, while each of the other fractions accounted for less than 5% of the total Fe present (Figure 2c). There were no significant differences in the total amounts of either Al or Fe between landforms (Figure 2b,c).

More Al than Fe was associated with P in both HA and Res fractions at all sites except for the HA fraction at the Braun site (Figures 3 and 4). This is consistent with the generally higher total Fe concentrations observed at the Braun site (Hogan 2000). The predominance of Al relative to Fe in HA and Res P fractions appears to be a common feature in many types of wetland soils and may reflect the greater tendency of Al (vs. Fe) to form associations with OM (Borggard et al. 1990, Axt 1997, Paludan and Morris 1999). Molar ratios of HA-Al:HA-Fe (8:1, 5:1 and 3:1 in NWs, RWs and UPs, respectively) (Hogan 2000), are comparable to those reported for South Carolina marshes (4:1 to 7:1) (Paludan and Morris 1999) but are low in comparison to a Danish fen surrounded by agricultural land (20:1) (Paludan 1995). Molar ratios of HA-Al:HA-P and HA-Fe:HA-P (19 and 5 in the NWs, 27 and 11 in the RWs, and 20 and 9 in the UPs, respectively), are similar to the values of 12 and 2.5 reported by Paludan and Jensen (1995) for a Danish freshwater wetland and acid-bog lake sediments, respectively. Molar ratios of Res-Al:Res-P were 6.5, 4.5, and 3.6 times higher than ratios of Res-Fe:Res-P in NWs, RWs, and UPs, respectively. Similarly, Axt (1997) found 5–7 times more Al than Fe in the HA and Res pools of forested riparian wetland soils in Virginia.

Al and Fe by Selective Dissolution

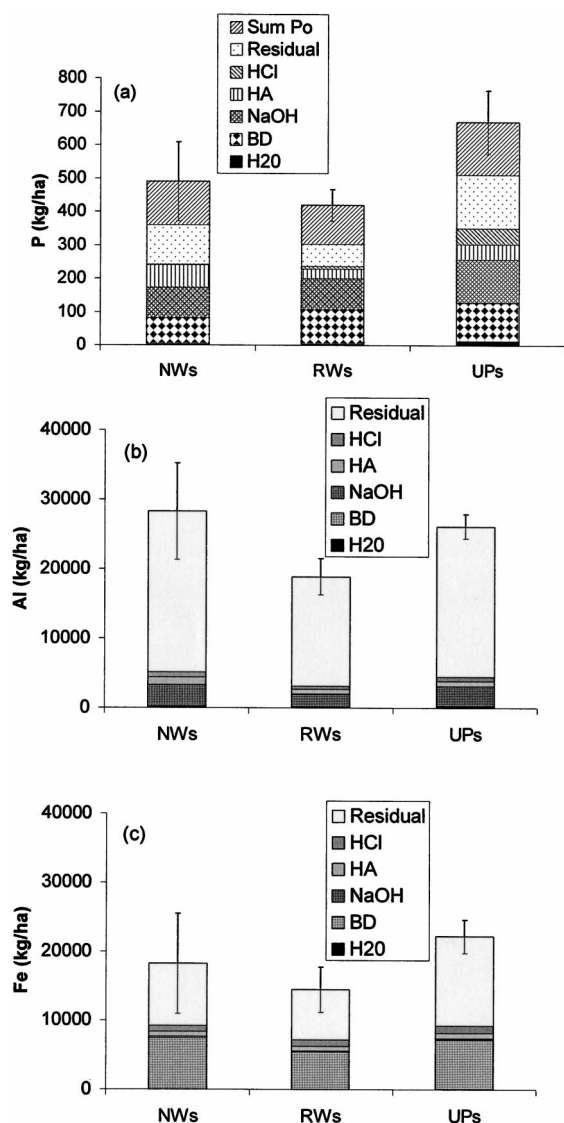Both Al_p and Al_o (by separate extraction) and Fe_p and Fe_o (by separate extraction) were significantly pos-

Figure 2. Total amounts of P (a), Al (b), and Fe (c) as extracted in the Paludan and Jensen (1995) sequential fraction technique in the natural wetlands (NWs), restored wetlands (RWs), and uplands (UPs) to a depth of 13 cm. Error bars indicate $\pm$ 1 SE and n = 3 for each landform. The ANOVA F-value $F_{(2,6\ \alpha=0.05)}$ = 1.83 for P (a), 1.22 for Al (b), and 0.65 for Fe (c). For P (a), Sum P_o is defined as the sum of WS-P_o + BD-P_o + NaOH-P_o; the HA and residual fractions are also considered to be organic P but are diagrammed separately in this figure.

itively correlated (r^2 = 0.70 and 0.80, respectively) (Figure 5). Concentrations of Al and Fe obtained by separate oxalate extraction were consistently greater than those obtained by either pyrophosphate extraction or by sequential oxalate extraction following pyrophosphate extraction (Table 2), suggesting that a considerable proportion of soil Al_o was organically-bound (i.e., pyrophosphate-extractable).
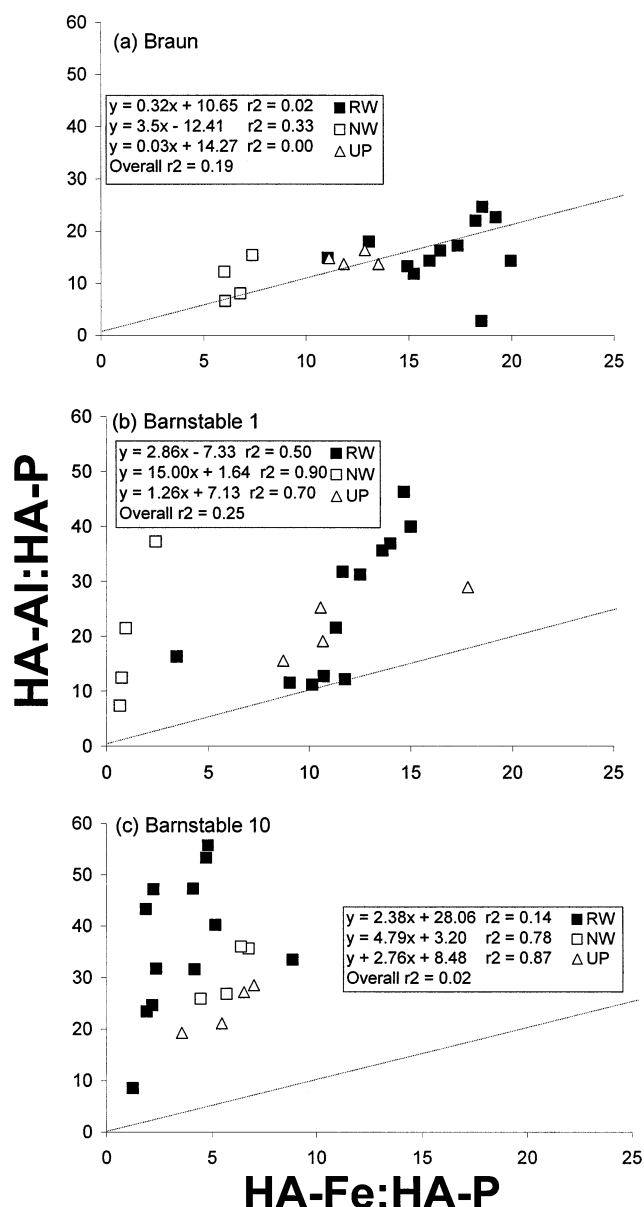
As hypothesized, concentrations of organically

Figure 3. Comparison of the molar ratios HA-Fe:HA-P vs. HA-Al:HA-P in restored wetlands (RWs), natural wetlands (NWs), and uplands (UPs) at the Braun (a), Barnstable 1 (b), and Barnstable 10 (c) study sites. Dotted lines represent [Al]/[Fe] = 1.

bound Al and Fe were significantly positively correlated with both soil organic matter (OM) and total organic carbon (TOC). Higher correlations were generally observed between Al_o and OM (r^2 = 0.84 and 0.41) and between Fe_o and OM (r^2 = 0.76 and 0.23) (Hogan 2000) than between Al_o and TOC (r^2 = 0.27 and 0.37) and Fe_o and TOC (r^2 = 0.24 and 0.39) (Figure 6a and c) in both RW and NW soils, respectively. Removal of a single outlier point (circled) improved the correlation between Al_o and TOC from r^2 = 0.37 to r^2 = 0.86 in NW soils (Figure 6a). A significant
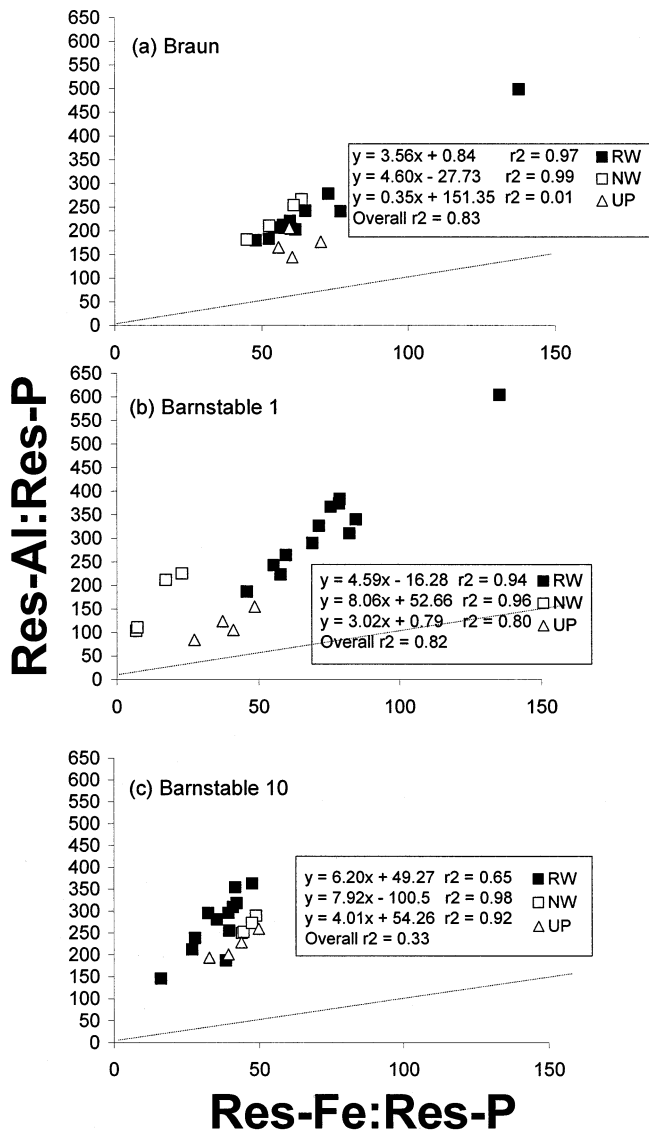
Figure 4. Comparison of the molar ratios Res-Fe:Res-P vs. Res-Al:Res-P in restored wetlands (RWs), natural wetlands (NWs), and uplands (UPs) at the Braun (a), Barnstable 1 (b), and Barnstable 10 (c) study sites. Dotted lines represent [Al]/[Fe] = 1.
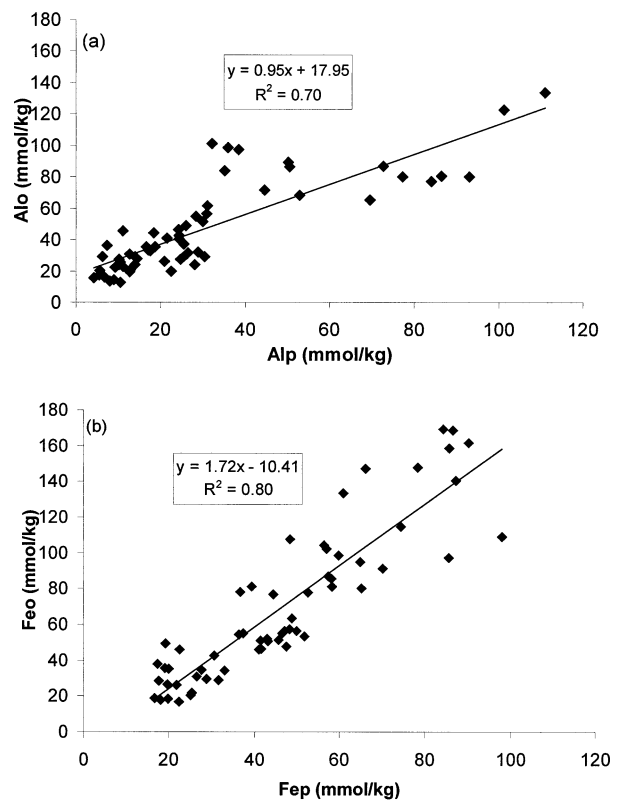

Figure 5. Relationships between organically-bound Al (Al_p) and oxalate-extractable Al (Al_o) (a) and between organically-bound Fe (Fe_p) and oxalate-extractable Fe (Fe_o) (b).

correlation was found between Al_p and TOC for the NWs ($r^2 = 0.46$) when a single outlier point (circled) was removed (Figure 6b). Small but significant correlations also occurred between Fe_p and TOC ($r^2 = 0.26$ and 0.38) in both RW and NW soils, respectively (Figure 6d). Darke and Walbridge (2000) observed that in GA floodplain soils, correlative relationships between Fe_o and soil OM appeared to be inhibited in frequently flooded swale microsites, similar to the patterns observed for NWs in Figure 6.

Table 2. Comparison of pyrophosphate-extractable Al and Fe, sequential oxalate (following pyrophosphate extraction) extractable Al and Fe, and a separate oxalate extraction of Al and Fe. Comparisons are made between the different extractions within each landform (natural wetlands, restored wetlands, or uplands). The F-value for the ANOVA is shown and the lower case letters x and y indicate significant differences at $p < 0.05$. Values are means $\pm$ standard errors in mmol/kg to a soil depth of 13 cm.

|	Al (mmol/kg)			Fe (mmol/kg)		
	Natural Wetlands (n = 3)	Restored Wetlands (n = 3)	Uplands (n = 3)	Natural Wetlands (n = 3)	Restored Wetlands (n = 3)	Uplands (n = 3)
Pyrophosphate extractable	71.3 ± 15.9 y	21.1 ± 5.8	15.8 ± 1.4	57.2 ± 16.9	50.8 ± 7.1	23.6 ± 3.4
Sequential oxalate extractable	28.8 ± 6.6 x	23.0 ± 4.6	25.8 ± 6.7	43.3 ± 20.3	32.5 ± 10.2	25.0 ± 4.1
Separate oxalate extractable	82.0 ± 12.3 y	38.8 ± 10.7	34.9 ± 9.5	84.0 ± 28.5	75.4 ± 16.0	38.7 ± 4.3
F-Value ($F_{2,6}$)	5.3	1.7	2.0	0.8	3.4	4.4
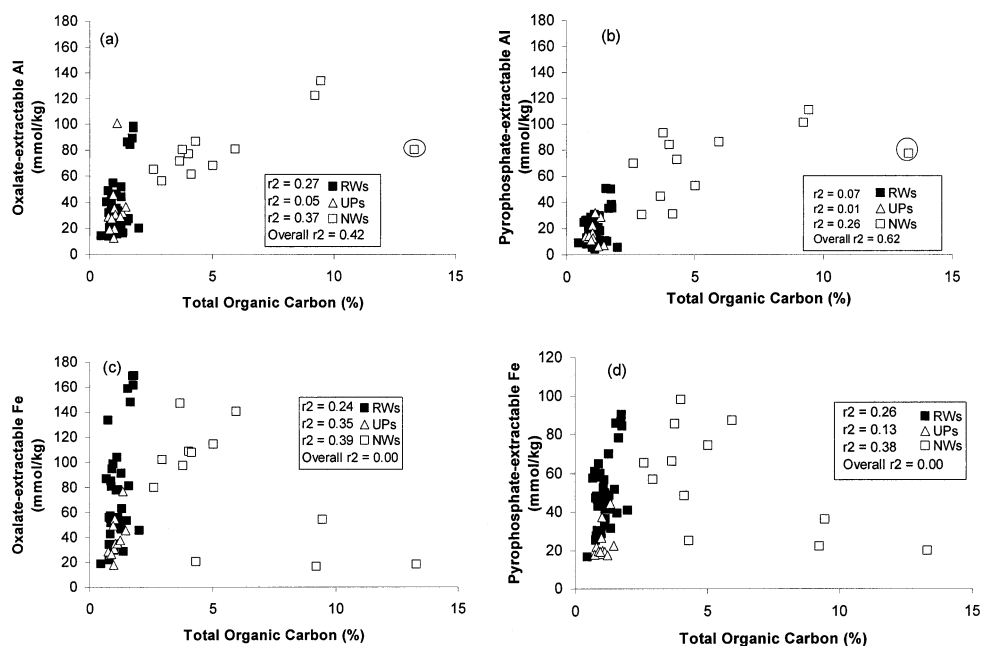
Figure 6. Relationships between oxalate-extractable Al (Al_o) and total organic carbon (a), between pyrophosphate-extractable Al (Al_p), and total organic carbon (b) between oxalate-extractable Fe (Fe_o) and total organic carbon (c) and between pyrophosphate-extractable Fe (Fe_p) and total organic carbon (d) in restored wetlands (RWs), natural wetlands (NWs), and uplands (UPs).

The higher concentrations of both Al_o and Al_p in NW as compared to RW or UP soils (Table 3), and the positive correlations between oxalate- or pyrophosphate-extractable Al and TOC (Figure 6) are consistent with our hypothesis that the higher soil TOC concentrations of the NWs would favor the formation of organically-bound forms of Al. Similar correlative relationships between Al_o and percent OM have been

Table 3. Soil aluminum and iron by selective dissolution and extractable calcium for the average of the natural wetlands (NWs), restored wetlands (RWs), and uplands (UPs). The F-value for the ANOVA is shown and the lower case letters x, y, and z indicate significant differences at $p < 0.05$ between the averages of the NWs, the RWs, and the UPs. Data are means $\pm$ standard errors and are expressed in kg/ha to a soil depth of 13 cm.

Al, Fe or Ca (kg/ha)	Soil Aluminum and Iron Averages			F-Value ($F_{2,6}$)
	NWs (n = 3)	RWs (n = 3)	UPs (n = 3)	
Crystalline Al[1] (Al_c)	3710.8 $\pm$ 235.1 z	2303.6 $\pm$ 8.3 y	1249.2 $\pm$ 206.9 x	46.6
Non-crystalline Al[2] (Al_o)	2440.0 $\pm$ 203.9	1373.0 $\pm$ 295.0	1821.5 $\pm$ 40.7	2.3
Non-crystalline Al[3]	898.6 $\pm$ 265.7	824.0 $\pm$ 121.4	1351.5 $\pm$ 344.8	1.2
Organically bound Al[4] (Al_p)	2099.1 $\pm$ 365.5 y	767.0 $\pm$ 194.7 x	826.3 $\pm$ 73.1 x	9.6
Total Al[5]	6150.8 $\pm$ 244.8 y	3676.6 $\pm$ 290.1 x	3070.8 $\pm$ 544.6 x	18.1
Crystalline Fe[6] (Fe_c)	5250.1 $\pm$ 2625.5	3926.8 $\pm$ 2044.5	10826.8 $\pm$ 1918.1	2.7
Non-crystalline Fe[2] (Fe_o)	5483.5 $\pm$ 2017.1	5549.8 $\pm$ 926.0	4178.3 $\pm$ 438.5	0.3
Non-crystalline Fe[3]	2854.1 $\pm$ 1385.3	2354.2 $\pm$ 558.8	2700.7 $\pm$ 428.8	0.1
Organically bound Fe[4] (Fe_p)	3680.8 $\pm$ 1127.7	3828.0 $\pm$ 410.0	2550.8 $\pm$ 368.2	0.9
Total Fe[7]	10558.4 $\pm$ 4784.6	9476.6 $\pm$ 2798.2	15005.1 $\pm$ 1932.6	0.7
Ca[8]	902.0 $\pm$ 374.6	606.1 $\pm$ 98.2	1478.4 $\pm$ 391.3	1.9

[1] 0.1M sodium hydroxide-extractable Al (sequential following oxalate extraction).
[2] Separate 0.2M acid ammonium oxalate-extractable Al or Fe.
[3] Oxalate-extractable Al or Fe following 0.1M sodium pyrophosphate extraction (sequential extraction).
[4] 0.1M sodium pyrophosphate-extractable Al or Fe.
[5] Oxalate-extractable Al plus NaOH extractable Al.
[6] DCB Fe minus oxalate extractable Fe.
[7] DCB Fe.
[8] 1.0 M ammonium acetate-extractable Ca.

Table 4. Single soil characteristics as predictors of P sorption capacity for the average of the restored wetlands (RWs), natural wetlands (NWs), RWs and NWs together, and the uplands (UPs). Shown are the r^2 and F values; asterisks indicate single significant predictors of P sorption capacity at $p < 0.05$.

	Landform Site Averages			
	RWs	NWs	RWs and NWs	UPs
Residual (Res) Al	$r^2 = 0.90$, $F_{(1,34)} = 292.02*$	$r^2 = 0.01$, $F_{(1,10)} = 0.05$	$r^2 = 0.35$, $F_{(1,46)} = 25.04*$	$r^2 = 0.15$, $F_{(1,10)} = 1.81$
Noncrystalline (Al_o)	$r^2 = 0.89$, $F_{(1,34)} = 279.81*$	$r^2 = 0.25$, $F_{(1,10)} = 3.35$	$r^2 = 0.29$, $F_{(1,46)} = 18.67*$	$r^2 = 0.15$, $F_{(1,10)} = 1.77$
Noncrystalline (Fe_o)	$r^2 = 0.87$, $F_{(1,34)} = 220.42*$	$r^2 = 0.00$, $F_{(1,10)} = 0.00$	$r^2 = 0.56$, $F_{(1,46)} = 58.75*$	$r^2 = 0.38$, $F_{(1,10)} = 6.12*$
% Clay	$r^2 = 0.85$, $F_{(1,34)} = 190.16*$	$r^2 = 0.05$, $F_{(1,10)} = 0.55$	$r^2 = 0.29$, $F_{(1,46)} = 19.23*$	$r^2 = 0.16$, $F_{(1,10)} = 1.89$
HCl-Fe	$r^2 = 0.83$, $F_{(1,34)} = 165.20*$	$r^2 = 0.01$, $F_{(1,10)} = 0.13$	$r^2 = 0.57$, $F_{(1,46)} = 60.54*$	$r^2 = 0.00$, $F_{(1,10)} = 0.03$
Pyrophosphate (organic) Fe_p	$r^2 = 0.82$, $F_{(1,34)} = 158.92*$	$r^2 = 0.19$, $F_{(1,10)} = 2.38$	$r^2 = 0.48$, $F_{(1,46)} = 42.60*$	$r^2 = 0.38$, $F_{(1,10)} = 6.02*$
HA-Al	$r^2 = 0.70$, $F_{(1,34)} = 80.59*$	$r^2 = 0.30$, $F_{(1,10)} = 4.35$	$r^2 = 0.26$, $F_{(1,46)} = 15.98*$	$r^2 = 0.29$, $F_{(1,10)} = 4.18$
Residual (Res) Fe	$r^2 = 0.69$, $F_{(1,34)} = 75.84*$	$r^2 = 0.07$, $F_{(1,10)} = 0.77$	$r^2 = 0.34$, $F_{(1,46)} = 24.21*$	$r^2 = 0.23$, $F_{(1,10)} = 2.99$
NaOH-Al	$r^2 = 0.67$, $F_{(1,34)} = 68.93*$	$r^2 = 0.08$, $F_{(1,10)} = 0.85$	$r^2 = 0.28$, $F_{(1,46)} = 18.21*$	$r^2 = 0.31$, $F_{(1,10)} = 4.59$
Crystalline (Al_c)	$r^2 = 0.63$, $F_{(1,34)} = 59.12*$	$r^2 = 0.08$, $F_{(1,10)} = 0.88$	$r^2 = 0.10$, $F_{(1,46)} = 4.99*$	$r^2 = 0.48$, $F_{(1,10)} = 9.33*$
Pyrophosphate (organic) Al_p	$r^2 = 0.56$, $F_{(1,34)} = 43.75*$	$r^2 = 0.68$, $F_{(1,10)} = 21.11*$	$r^2 = 0.04$, $F_{(1,46)} = 1.69$	$r^2 = 0.70$, $F_{(1,10)} = 23.19*$
BD-FE	$r^2 = 0.56$, $F_{(1,34)} = 43.83*$	$r^2 = 0.04$, $F_{(1,10)} = 0.40$	$r^2 = 0.15$, $F_{(1,46)} = 8.07*$	$r^2 = 0.02$, $F_{(1,10)} = 0.21$
% Total Organic Carbon (TOC)	$r^2 = 0.39$, $F_{(1,34)} = 21.77*$	$r^2 = 0.01$, $F_{(1,10)} = 0.05$	$r^2 = 0.00$, $F_{(1,46)} = 0.21$	$r^2 = 0.02$, $F_{(1,10)} = 0.20$
HA-Fe	$r^2 = 0.38$, $F_{(1,34)} = 20.66*$	$r^2 = 0.06$, $F_{(1,10)} = 0.66$	$r^2 = 0.28$, $F_{(1,46)} = 17.62*$	$r^2 = 0.21$, $F_{(1,10)} = 2.7$
Ca	$r^2 = 0.31$, $F_{(1,34)} = 14.96*$	$r^2 = 0.32$, $F_{(1,10)} = 4.80$	$r^2 = 0.01$, $F_{(1,46)} = 0.06$	$r^2 = 0.09$, $F_{(1,10)} = 0.95$

observed in a range of southern forested wetland soils (Lockaby and Walbridge 1998, Axt and Walbridge 1999, Darke and Walbridge 2000), suggesting that the formation of organically-bound Al forms is a common occurrence in soils of forested riparian wetlands in this region.

P Sorption

Single factor regression models identified 15 chemical parameters that were significantly correlated with P sorption capacity in RW soils, including Res-Al, Al_o, Fe_o, clay, HCl-Fe, and Fe_p ($r^2 = 0.90$, 0.89, 0.87, 0.85, 0.83, and 0.82, respectively) (Table 4). A 5-term stepwise multiple regression model using Res-Al, Fe_o, Res-Fe, Fe_p, and clay explained 96% of the variation in P sorption capacity in RW soils (Hogan 2000), just 6% more than was explained by Res-Al alone (Table 4).

In contrast, only one and four soil chemical parameters measured were significantly correlated with P sorption capacity in NW and UP soils, respectively (Table 4). Pyrophosphate-extractable (organically-bound) Al was the best single predictor of P sorption capacity in both NW and UP soils ($r^2 = 0.68$ and 0.70, respectively) (Table 4). A 3-term model of Al_p, clay and HA-Fe explained 84% of the variation in UP soils (Hogan 2000).

Because of recent evidence suggesting the potential importance of OM-Al complexes as agents of P retention in some riparian wetlands in the southeastern US (Lockaby and Walbridge 1998, Axt and Walbridge 1999, Darke and Walbridge 2000), we had hypothesized that the higher concentrations of TOC in NW soils would ultimately result in higher P sorption potentials in these soils. Phosphorus sorption capacity was significantly positively correlated with organically-bound Al (Al_p) in NW soils ($r^2 = 0.68$) (Table 4), suggesting that Al complexed with OM is an important
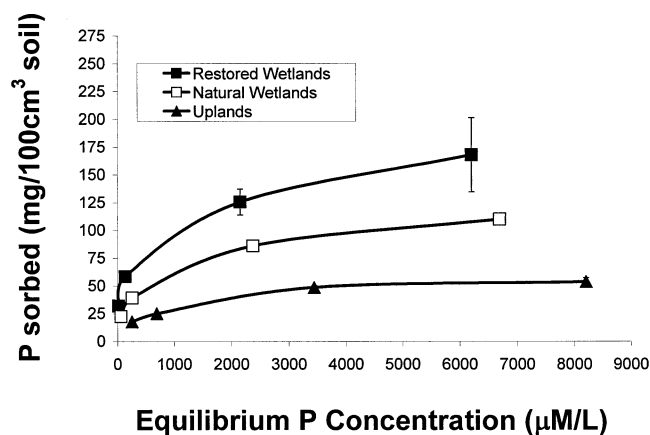
Figure 7. Average P sorption isotherms for restored wetlands, natural wetlands, and uplands (n = 3 per landform type) based on P sorbed during a 24-h equilibration, with initial P concentrations of 16, 33, 130, and 260 mg P/L plotted as a function of the P remaining in solution following equilibration. Error bars indicate ± 1 SE.

component of P sorption capacity. Humic Al complexes can represent important sites of P sorption in soils (cf., Gerke and Jungk 1991, Dolfing et al. 1999).

Despite the significantly higher concentrations of organically-bound Al (Table 3) and the strong relationship between Al_p and P sorption in the NWs (Table 4), RW soils had higher P sorption capacities than NW soils (Figure 7), even when the effects of previously sorbed P were considered (Hogan 2000). Although the RWs and NWs were developed over similar substrates, the RWs were restored on previously cultivated soils. The large number of factors significantly positively correlated with P sorption capacity in the RWs, compared to both NW and UP soils (Table 4), suggests important initial differences in soil chemical composition among the three landforms. These differences appear to have resulted in higher P sorption capacity in the RW soils. The RW soils are currently in transition to a hydric state. As these soils mature, OM should accumulate, favoring increased concentrations of organically bound Al. However, rates of OM accumulation in the RW soils could be slower than in forested wetlands due to differences in productivity, decomposition rates, and organic matter quality in comparison with the NWs.

As RW soils mature, chronic flooding is likely to cause a decrease in soil Fe concentrations, as Fe (III) is reduced to soluble Fe (II) and lost over time (Ponnamperuma 1972, Darke and Walbridge 2000). Iron losses might reduce RW P sorption capacities in the future but could be offset by flooding-induced changes in the chemistries of both Al and Fe through complexation of Al by OM (Lockaby and Walbridge 1998, Axt and Walbridge 1999, Darke and Walbridge 2000)

and decreases in the relative crystallinity of remaining Fe (Kuo and Mikkelsen 1979, Darke and Walbridge 2000).

Because Al is not converted to a lower, more soluble oxidation state during flooding, Al has been suggested as a more important component of P retention than Fe in wetland soils (Walbridge and Struthers 1993, Axt 1997, Darke and Walbridge 2000). Darke and Walbridge (2000) observed significant, flooding-induced decreases in soil Al concentrations in a GA floodplain forest soil, although concentrations rapidly returned to pre-flooding levels following drainage. Because of the potential for both Al and Fe losses to occur over the course of soil development, affecting P retention, the P sorption capacities of our RW soils should be reevaluated periodically, to determine their continued effectiveness in retaining P.

CONCLUSIONS

As hypothesized, the forested NWs we studied had higher soil TOC concentrations than recently restored herbaceous wetlands established nearby on similar parent materials. Soil physical and chemical properties in the NWs (bulk density, pH, labile organic and microbial P, total N, and soil N:P ratios) all varied in ways predictable from their higher soil TOC concentrations in comparison with the RWs. As predicted, concentrations of organically-bound Al (Al_p) were significantly higher in NW versus RW soils and were significantly positively correlated with P sorption capacity. However, despite higher Al_p concentrations in the NWs, P sorption capacities were significantly higher in the RWs.

We explain these findings based on initial differences in RW and NW soils (RWs having been restored on soils that were previously cultivated) and suggest that RW soils are in transition from a non-hydric to a hydric state. Soils that are saturated for shorter periods of time may have a greater capacity to sorb P due to higher Al and Fe concentrations; however, chronic flooding could result in a gradual loss of both Al and Fe over time, with a concomitant loss in the capacity to retain P. As soil OM accumulates in the RWs, organically-bound Al may become more important in binding P. Since RW soils are in transition to a more hydric state, P retention efficiencies should be reevaluated periodically to ensure that the P-retention functions of these soils remain comparable to those of the nearby NWs they were meant to replace.

ACKNOWLEDGMENTS

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