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Revised 010/08

Efficacy of Drinking-Water Treatment Residual in Controlling Off-Site Phosphorus Losses: A Field Study in Florida

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Land application of drinking-water treatment residuals (WTR) has been shown to control excess soil soluble P and can reduce off-site P losses to surface and ground water. To our knowledge, no field study has directly evaluated the impacts of land application of WTRs on ground water quality. We monitored the effects of three organic sources of P (poultry manure, Boca Raton biosolids, Pompano biosolids) or triple superphosphate co-applied with an aluminum-based WTR (Al-WTR) on soil and ground water P and Al concentrations under natural field conditions for 20 mo in a soil with limited P sorption capacity. The P sources were applied at two rates (based on P or nitrogen [N] requirement of bahiagrass) with or without Al-WTR amendment and replicated three times. Without WTR application, applied P sources increased surface soil soluble P concentrations regardless of the P source or application rate. Co-applying the P sources with Al-WTR prevented increases in surface soil soluble P concentrations and reduced P losses to shallow ground water. Total dissolved P and orthophosphate concentrations of shallow well ground water of the N-based treatments were greater (>0.9 and 0.3 mg L^{-1}, respectively) in the absence than in the presence ($\sim$0.6 and 0.2 mg L^{-1}, respectively) of Al-WTR. The P-based application rate did not increase ground water P concentrations relative to background concentrations. Notwithstanding, Al-WTR amendment decreased ground water P concentrations from soil receiving treatments with P-based application rates. Ground water total dissolved Al concentrations were unaffected by soil Al-WTR application. We conclude that, at least for the study period, Al-WTR can be safely used to reduce P leaching into ground water without increasing the Al concentration of ground water.

OFF-SITE phosphorus (P) losses to water bodies is a major concern because P is a limiting nutrient for eutrophication of most freshwaters (Sims et al., 1998). Phosphorus movement into water bodies can occur as lateral (surface water) or vertical (ground water) transport. Most Florida soils are characterized by coarse-textured surface horizons with a very limited capacity to retain P (Elliott et al., 2002a). Thus, in these soils there is concern that large P inputs increase the risk of ground water nonpoint-source pollution. A P chemical fractionation of some Florida soils indicated that about 80% of the total P in the A-horizon was leachable (Graetz and Nair, 1995), and P leaching is likely to occur in such soils. Phosphorus leaching is exacerbated in soils where contaminated water is intercepted by drains or shallow water tables that join surface waters and feature relatively short ground water flow distances (Burgoa et al., 1991; Sims et al., 1998).

One suggested way to mitigate P leaching to ground water from soils with limited P sorption capacity is to amend the soil with drinking-water treatment residuals (WTR) (O'Connor et al., 2002; Elliott et al., 2002b; Ippolito et al., 2003; Elliott et al., 2005; Novak and Watts, 2005). Application of aluminum-based WTRs (Al-WTR) to P-impacted soils could serve as a practical chemical-based best management practice (BMP) to reduce off-site P movement from fields via runoff and leaching. Reducing off-site P transport can reduce P loads into nutrient-sensitive surface water systems and consequently minimize the risk of eutrophication (Dayton and Basta, 2005a). Studies have shown reductions in P concentrations of runoff (Dayton et al., 2003) and leachate (Elliott et al., 2002b; O'Connor et al., 2002; Novak and Watts, 2004; Dayton and Basta, 2005a) after amendment of P-impacted soils with WTR.

Underlying mechanisms behind WTR P adsorption have been studied. Ippolito et al. (2003) suggested that P is adsorbed by WTR initially as an outer-sphere complex or found in the diffuse ion swarm near individual WTR particles. With time, P becomes

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Abbreviations: BMP, best management practice; DPS, degree of phosphorus saturation; ICP–AES, inductively coupled plasma–atomic emission spectroscopy; PAN, plant-available nitrogen; TDP, total dissolved phosphorus; TSP, triple superphosphate; WEP, water-extractable phosphorus; WTR, drinking-water treatment residual; Al-WTR, aluminum-based WTR.

more strongly adsorbed as an inner-sphere complex onto WTR (Ippolito et al., 2003). Based on spectroscopic measurements, Makris and O'Connor (2007) showed that adsorption of P by WTRs was strongly hysteretic. The authors observed P diffusion into WTR bottleneck-shaped micropores, favoring long-term stability of sorbed P by WTRs (Makris and O'Connor, 2007). Agyin-Birikorang and O'Connor (2007) demonstrated, using an isotopic dilution technique coupled with a stepwise acidification procedure, that within the commonly encountered pH range of most agricultural soils (pH 4–7), WTR immobilized P is stable and essentially nonlabile.

Most studies that evaluated the impacts of P-source and WTR additions on soil soluble P concentrations have focused on laboratory incubations (Dayton et al., 2003; Novak and Watts, 2004; Agyin-Birikorang and O'Connor, 2007), column leaching studies (Elliott et al., 2002b; O'Connor et al., 2002), or indoor rainfall simulation studies with packed boxes (Agyin-Birikorang et al., 2007). Some studies have evaluated WTR effectiveness in controlling excess soil soluble P under field conditions (Dayton and Basta, 2005a; Novak and Watts, 2005; Agyin-Birikorang et al., 2007; Bayley et al., 2008). However, these studies examined the extent of soluble P reductions in soils, and no attempt was made to directly assess P concentrations of ground water due to WTR land application. To our knowledge, no field study has directly evaluated the impacts of land application of Al-WTRs on ground water quality.

Although land application of Al-WTR has been shown to effectively control excess soil soluble P and can reduce off-site P losses to surface and ground water, there is a concern that Al-WTR particle dissolution in acid soils or aqueous suspensions could release significant quantities of Al to the environment. Studies have shown that Al-WTR amendment significantly increased Al concentrations of a surface soil (Agyin-Birikorang et al., 2007). Excess Al in drinking water is of concern because of the suspected Al connection with Alzheimer's disease or dialysis encephalopathy (Carol and Arnold, 1990; Driscoll and Driscoll, 2005). Thus, in field experiments that involve Al-WTR land application, it is expedient to investigate Al losses to water bodies.

The objectives of this study were to evaluate (i) WTR effectiveness in reducing P losses to ground water under natural field conditions from a typical Florida Spodosol amended with P sources of different solubilities and (ii) the effects of surface-applied WTR on ground water Al concentration. Based on the effectiveness of Al-WTRs in reducing soluble P concentrations in soils, we hypothesized that land application of Al-WTRs would significantly reduce P leaching to ground water. Due to the circumneutral pH of most Al-WTRs, we hypothesized that land application of Al-WTR will not result in Al losses from the Al-WTR to ground water.

Materials and Methods

Study Site

The study was conducted at a cattle pasture located on the eastern border of Okeechobee County in southern Florida, 7 mi (11.3 km) northeast of Okeechobee and north of Lake Okeecho-

bee (27°N, 80.9°W). The soil at the study site is an Immokalee fine sand (sandy, siliceous, hyperthermic Arenic Alaquods). The soil series contains distinct A, E, and Bh horizons. Previous studies have shown that soil at the study site has a limited P sorption capacity (Graetz and Nair, 1995) and that P leaching is the dominant P loss mechanism (O'Connor et al., 2005).

Soil Characterization

The field was divided into 50 subunits, from which a composite soil sample (formed by mixing 20 2.5-cm-diam. core samples) was collected from each subunit to characterize the initial soil conditions. Samples were taken from the top 5 cm of the A horizon, the center of the E horizon (approximately 45–55 cm from the surface), and the top 10 cm of the Bh horizon (approximately 110–120 cm).

Air-dried soil samples (<2 mm) were analyzed for total P, Fe, and Al by inductively coupled plasma–atomic emission spectroscopy (ICP–AES) (PerkinElmer Plasma 3200; PerkinElmer, Wellesley, MA) after acid-peroxide digestion according to the USEPA Method 3050A (USEPA, 1986a). Oxalate- (200 mmol L^{-1}) extractable P, Fe, and Al were determined by ICP–AES after extraction at a 1:60 solid/solution ratio (Schoumans, 2000). Water-extractable P (WEP) was determined by reacting soil with deionized water at a ratio of 1:10 soil/solution ratio for 1 h (Kuo, 1996), with P concentration analyzed colorimetrically (Murphy and Riley, 1962). Degree of soil phosphorus saturation (DPS) was calculated as moles of oxalate- (200 mmol L^{-1}) extractable P divided by the moles of oxalate- (200 mmol L^{-1}) extractable Fe + Al (Schoumans, 2000) and assuming a saturation factor (α) of 0.55 as suggested by Nair and Graetz (2002) for Florida soils. The DPS of surface soils is a measure of P loss potential from a soil, and a critical value of 0.25 (corresponding to ~10 mg kg^{-1} WEP) has been suggested for FL soils (Nair et al., 2004).

Characterization of Amendments

Portions of the applied amendments (P sources and WTR) were air-dried and ball-milled for analysis. The amendments were analyzed for total- and oxalate-extractable P, Fe, and Al as described previously. The P saturation index for the amendments was calculated as moles of oxalate-extractable P divided by the moles of oxalate-extractable Fe + Al (Elliott et al., 2002b). Amendments for WEP concentrations were determined at a 1:200 amendment/solution ratio (Sharpley and Moyer, 2000). The percentage of total P that was water extractable was then calculated. Total carbon (C) and N concentrations were determined in the amendments by combustion at 1010°C using a Carlo Erba analyzer (NA-1500 CNS; Carlo Erba, Milan, Italy) as outlined in Nelson and Sommers (1996), and pH measurements were performed on the materials (1:2 solid/solution ratio) (Thomas, 1996). Percent solids were determined by drying materials at 105°C (Gardner, 1986).

Experimental Setup

The study was a 4 × 2 × 2 factorial experiment (four P sources and two application rates with or without WTR amendment),

with an individual plot size of 20.7 × 95 m. Each treatment was replicated three times in a randomized, complete block design. An extra plot per block was left untreated as a control; thus, 51 plots (17 plots/block and 3 blocks) were established. The field was fenced to prevent cattle from entering the plots, and individual plots were bermed to avoid cross-contamination of treatments from adjacent plots.

To assess the extent of P and Al leaching, two wells were installed in each plot at approximately 0.9 m and approximately 3 m, representing depths above and below the spodic horizon. The wells, constructed by Southern DataStream, Inc. (Labele, FL), consisted of a piezometer probe, a PVC covering pipe, and a sampling tube. The piezometer probe was made of a 15-cm stainless steel tube, and the sampling pipe was surrounded by PVC casing to reduce cross-contamination of well samples. A plastic collar, positioned above the sand-pack in the spodic horizon section of the deep well, and other engineering precautions ensured that samples collected represented ground water at the intended depth. Piezometers were purged slowly at least three times after installation and before initiating the study to minimize the effects of installation disturbance. The two wells were installed approximately 1.2 m apart in each plot (each located approximately 0.6 m away from the center of each plot at opposite sides).

Three organic sources (two biosolids [one from Pompano Beach, FL, and the other from Boca Raton, FL] and poultry manure [not treated with alum] collected from Indiantown, FL) and triple superphosphate (TSP) were used. The Boca Raton biosolids was an anaerobically digested (biological P removal–like) material, whereas the Pompano biosolids was produced via anaerobic digestion and stabilized with Al and Fe to reduce P solubility of the end product. The biosolids were selected based on differences in WEP content as a result of different processing procedure. The Boca Raton biosolids WEP content was approximately 5.5 g P kg^{-1}, whereas the Pompano biosolids contained ~1.2 g WEP kg^{-1} (O'Connor et al., 2000).

Two application rates of the P sources, based on the plant-available N (PAN) (179 kg PAN ha^{-1}) and P (39.6 kg total P ha^{-1}) requirements of bahiagrass (*Paspalum notatum*) (Hanlon, 1995), were used to attain N-based and P-based nutrient management. A preliminary study suggested ~40% organic N mineralization rate for the biosolids used for the study; therefore, PAN was calculated based on 40% organic N mineralization rate. Applying the organic fertilizer sources based on the N requirements of the crop (N-based rates) resulted in excessive P additions (~128 kg P ha^{-1}) to the soil. A TSP fertilizer dose of 128 kg P ha^{-1} was therefore selected as a high rate to approximate the rate of P applied when biosolids or manure are applied at an N-based rate. Ammonium nitrate was applied to plots receiving a P-based application rate of the P sources (and the "N-based" TSP treatment) to equalize the N supplied by the N-based application rates of the organic sources of P, based on the calculated PAN levels.

The P sources were surface applied with or without Al-WTR amendment obtained from the Manatee Co. Water Treatment Plant in Bradenton, FL. The Al-WTR was generated from the use of aluminum salts [Al$_2$(SO$_4$)$_3$·14H$_2$O] as coagulants to re-move turbidity, color, taste, and odor from raw water and to speed particulate matter removal from surface water obtained mostly from the Manatee River, which flows through south Florida. Dewatered Al-WTR was surface applied (22.4 dry Mg ha^{-1}, based on preliminary laboratory studies) to plots using a Knight ProTwin Slinger, model 8030 V-box spreader by making three passes on each side of the plot or three round trips. The Al-WTR was applied to the plots first, and the P sources were added to the field 72 h later. Application of all amendments (WTR, P sources, and ammonium nitrate) was completed in March 2003.

Soil Sampling and Analysis

Composite soil samples (formed by mixing 20 2.5-cm-diam. core samples) were obtained from the surface (0–5 cm) of each plot to monitor changes in the soil properties after amendment application. Similarly, samples were collected from the middle of the E horizon (~45–55 cm from the surface) and the top 10 cm of the Bh horizon (~110–120 cm) of each plot. Soil sampling was performed in April 2003, ~1 mo after amendment application. Studies evaluating WTR effectiveness (Elliott et al., 2002b; Agyin-Birikorang et al., 2007) have shown that significant WTR-P reactions are detectable >1 mo after WTR application. Soil sampling was then performed at 6, 12, and 18 mo after the first sampling. All holes were plugged to ensure that normal soil water infiltration occurred. Soil samples were air-dried and analyzed for total and oxalate-extractable P, Fe, and Al; WEP; and DPS as described previously. The study was terminated on 30 Sept. 2004 (~20 mo after commencement) because of hurricane-induced flooding of the field (Hurricanes Ivan on 19 Sept. 2004 and Jeanne on 25 Sept. 2004).

Ground Water Sampling and Analysis

Ground water samples were collected by pumping water slowly from the piezometers using the method of prolonged slow pumping at a steady rate (Backhus et al., 1993). A mini-suction lift pump was used to withdraw ground water samples. The ground water was pumped directly into a glass container. Ground water samples from the deep wells were collected monthly throughout the study period (March 2003–September 2004). Samples were obtained from the shallow wells monthly only during the rainy season (between June and October in 2003 and between July and September 2004; Fig. 1). Thus, a total of 20 and 8 sampling times were obtained for the deep and shallow wells, respectively.

Ground water samples were analyzed for pH, PO$_4$–P, total dissolved P (TDP), and total dissolved Al concentrations. The pH levels of the water samples were measured as described in Bates (1978). The PO$_4$–P measurement was performed on a subsample filtered through a 0.45-μm syringe filter, with concentrations determined within 24 h after sample collection using ion chromatography (DX 500; Dionex Corporation, Sunnyvale, CA). Total dissolved P and Al concentrations were measured in the filtered water samples after digesting 10 mL of the samples with 0.5 mL 6 nmol L^{-1} H$_2$SO$_4$ and 0.15 g of potassium persulfate in an autoclave for 1 h (Pote and Daniel, 2000). Digested samples were analyzed for P and Al concentra-
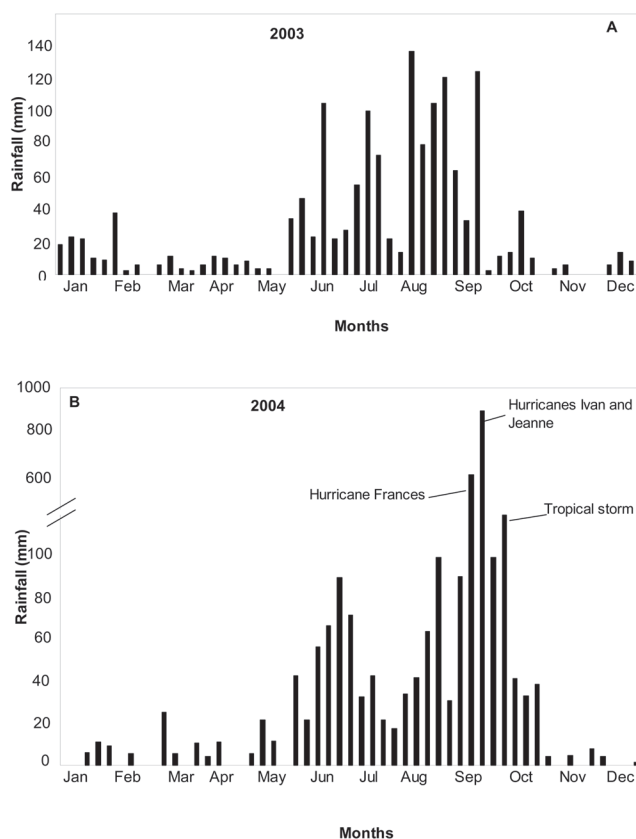
Fig. 1. Weekly total rainfall distribution at the research site in (A) 2003 and (B) 2004 (Source: South Florida Water Management District, Okeechobee County office). Note the differences in scales of the *y* axes of (A) and (B) after 100 mm rainfall.

tions using ICP–AES (PerkinElmer Plasma 3200; PerkinElmer, Wellesley, MA). All sampling and analyses were performed in accordance with the Florida Dep. of Environmental Protection's standard operating procedures (Florida Department of Environmental Protection, 2002) to minimize sampling and handling contamination.

Statistical Analysis

Soil chemical data were analyzed using the mixed procedure (PROC MIXED), and ground water P and Al concentrations data were analyzed using the repeated measures option of PROC MIXED in SAS version 9.0 (SAS Institute, 2002). Ground water P and Al concentrations were logarithmically transformed to conform to the normality and homogeneity assumptions of PROC MIXED. Because the total dissolved Al, PO_4–P, and most of the TDP concentration values of the ground water samples were <1 mg L^{-1}, we further transformed the values by adding 1 to the values and then determined the logarithm of the sum to avoid negative values (Neter et al., 1996). Data were back transformed for all discussions in this article. Treatment effects were evaluated using the LSMEANS statement of PROC MIXED along with adjusted Tukey's means separation (SAS Institute, 2002). Treatment differences were considered significant at $P \leq 0.05$.

Results and Discussion

Characteristics of Amendments and Pre-amended Soil

The pH values of the P sources ranged from 5.9 for the TSP to 8.2 for the Boca Raton biosolids (Table 1). The percent solids also varied with amendment; the biosolids (Boca Raton and Pompano biosolids) had ~140 g kg^{-1} solid contents, whereas the manure had ~270 g kg^{-1} solid contents. Boca Raton biosolids had the greatest amount of total P (39 g kg^{-1}), followed by the Pompano biosolids (24.1 g kg^{-1}); these values are consistent with values reported for most biosolids (~3–40 g kg^{-1}) (Elliott et al., 2002a, 2002b; O'Connor et al., 2005). The poultry manure had the least total P content, but the observed value (18.9 g kg^{-1}) was close to the average value (20 g kg^{-1}) reported for chicken manure total P by Barnett (1994). Of the organic sources of P, Boca Raton biosolids had the greatest "available" P (i.e., WEP) concentration.

The WTR pH was 5.6 (Table 1), which was slightly below the range of pH values reported for Al-WTRs (6.0–8.4) (Makris and O'Connor, 2007). The total P concentration was typical of Al-WTRs (0.3–4.0 g P kg^{-1}) (Dayton et al., 2003; Makris and O'Connor, 2007). The total Al concentration (107 g Al kg^{-1}) was within the range reported by others (15–177 g Al kg^{-1}) (Dayton et al., 2003; Makris and O'Connor, 2007). Oxalate-extractable Al was approximately 80% of the total, which is suggestive of WTRs amorphous nature. Phosphorus retention of WTRs is strongly related to amorphous Fe and Al concentrations (Elliott et al., 2005; Agyin-Birikorang et al., 2007). Gallimore et al. (1999), Dayton et al. (2003), and Dayton and Basta (2005a, 2005b) concluded that the amorphous (oxalate-extractable), rather than the total, Al content of WTR determines WTR effectiveness in reducing runoff P. Therefore, the high percentage of amorphous Al content portends that the Al-WTR will be an effective P sorbent.

Before amendments application, the surface soil at the study site was acidic (pH 5.5) and low in total and oxalate-extractable P, Fe, and Al (Table 2). The values of the chemical characteristics of the soil used for the study were consistent with values observed for Florida Spodosols (Graetz and Nair, 1995).

Amendments Effects on Soil

Analysis of variance indicated significant effects of P sources, P application rates, and WTR amendment as well as their interaction on WEP concentrations and the P-related measurements (e.g., DPS and oxalate-extractable Fe and Al) of the surface soil. There were no time effects on soil properties resulting from amendment with P sources and WTR.

Without WTR amendment, soil Fe+Al concentrations of the poultry manure- and TSP-treated plots were low (~5 mmol kg^{-1}) and not different from the background soil (control) concentrations (Fig. 2). The biosolids treatments, on the other hand, significantly increased the Fe + Al concentrations of the native soil. Amending the soil with WTR significantly ($p \leq 0.004$) increased the oxalate-extractable Fe + Al content of the soil (Fig. 2). Elliott et al. (2002b) reported that

Table 1. Chemical properties of amendments (oven dry basis) used in the study. Numbers are mean values of six replicates ± 1 SD.

Chemical characteristics	Poultry manure	Boca Raton biosolids	Pompano biosolids	TSP†	Al-WTR
pH	6.8 ± 0.1	8.2 ± 0.2	7.9 ± 0.2	5.9 ± 0.1	5.6 ± 0.2
Total C, g kg^{-1}	320 ± 42.9	347 ± 33.8	366 ± 26.8	ND	139 ± 10.9
Total N, g kg^{-1}	84.4 ± 1.06	80.6 ± 7.84	70.3 ± 9.21	ND	7.24 ± 1.06
Solids, g kg^{-1}	270 ± 42.8	134 ± 16.9	154 ± 34.7	996 ± 4.00	406 ± 62.4
Total P, g kg^{-1}	18.9 ± 3.8	38.7 ± 2.3	24.1 ± 4.2	209 ± 1.79	4.69 ± 0.7
Total Al, g kg^{-1}	0.94 ± 0.1	9.37 ± 0.4	9.26 ± 1.4	10.0 ± 0.42	107 ± 8.3
Total Fe, g kg^{-1}	1.53 ± 0.3	24.3 ± 3.1	32.8 ± 6.4	15.7 ± 0.64	6.08 ± 0.4
Oxalate P, g kg^{-1}	10.4 ± 1.2	26.4 ± 3.9	28.5 ± 3.2	186 ± 3.21	4.33 ± 0.8
Oxalate Al, g kg^{-1}	0.79 ± 0.1	6.50 ± 0.9	7.41 ± 0.6	6.90 ± 0.51	84.3 ± 6.2
Oxalate Fe, g kg^{-1}	0.82 ± 0.3	19.4 ± 2.3	24.7 ± 4.2	11.0 ± 0.43	5.16 ± 1.0
WEP, g kg^{-1}	0.85 ± 0.1	2.59 ± 0.6	0.34 ± 0.1	178 ± 3.25	ND
PWEP, %	4.49 ± 0.3	6.69 ± 0.5	1.41 ± 0.1	85.2 ± 0.9	NA
PSI	NA†	1.44	0.7	NA	0.02

† Al-WTR, aluminum-based water treatment residuals; NA, not applicable; ND, not determined; PSI, phosphorus saturation index (PSI = [oxalate-P/oxalate-Fe + oxalate-Al (in moles)]); PWEP, percent water-extractable phosphorus; TSP, triple super phosphate; WEP, water-extractable phosphorus.

the P sorbing ability of WTRs, when added to a sandy soil amended with biosolids, could be predicted based on the amorphous metal oxide content of the WTRs. Thus, an increase in oxalate-extractable Fe + Al content of the WTR-amended soil was expected to increase the P sorption capacity of the soil.

Application of the P sources alone (without WTR amendment) significantly ($p < 0.001$) increased the DPS values of the soil relative to the control (Fig. 3). The magnitude of DPS increase was a function of the type and the application rate of the P source. For example, at the higher rate (N-based rate) of application, soils treated with poultry manure and TSP had DPS values of 0.84 and 0.93, respectively; these values suggest that soils receiving these treatments could contribute significant amounts of P in runoff and/or leachate. The biosolids-amended soils (without WTR amendments) had DPS values significantly less than those of the TSP- and poultry manure–amended soils. The biosolids contained appreciable amounts of Fe and Al (Table 1), hence the lower DPS values. However, DPS values of the biosolids-amended soils were greater than the critical value for Florida soils (Fig. 3), suggesting that biosolids application alone to Florida sandy soils could result in off-site P losses.

Amendment with WTR significantly ($p < 0.001$) reduced the DPS of surface soil irrespective of the type and application rate of the P source (Fig. 3). The reduction in DPS persisted throughout the sampling period, suggesting a long-term increase in the P sorptive capacity of the system. Degree of P saturation has been shown to correlate positively with P loss potentials. Pautler and Sims (2000) found that P solubility increased significantly ($r^2 = 0.70$) as soil P saturation increased in 41 agricultural soils in Delaware, USA. Hooda et al. (2000) found that soil DPS was significantly related to soil P desorption. The critical DPS value for Florida soils is suggested to be 0.25, which corresponds to ∼10 mg WEP kg^{-1}. Thus, soils with DPS values >0.25 are expected to release significant amounts of P to surface runoff or leaching (Nair et al., 2004). In the presence of WTR amendment, soil DPS values were similar for the different P sources and the two P-source application rates of (Fig. 3). Irrespective of P source or P-source application rates, the soil DPS values were reduced below the 0.25 critical value by the added WTR; this suggests the ability of WTR to reduce P loss potential of the soil.

Table 2. Characteristics of the Immokalee soil used in the field study. Numbers are mean values of 100 replicates (50 subunits analyzed in duplicates) ±1 SD.

Chemical characteristics	Horizons		
	A	E	Bh
pH	5.5	5.9	5.1
Total P, mg kg^{-1}	24.5 ± 5.46	7.9 ± 4.56	24.5 ± 11.3
Total Al, mg kg^{-1}	72 ± 23.3	33.6 ± 7.55	1280 ± 170
Total Fe, mg kg^{-1}	101 ± 39.6	38.5 ± 6.21	94.8 ± 23.2
Oxalate P, mg kg^{-1}	10.0 ± 2.95	3.76 ± 2.76	23.8 ± 13.7
Oxalate Al, mg kg^{-1}	54.8 ± 6.74	15.8 ± 5.35	970 ± 418
Oxalate Fe, mg kg^{-1}	61.7 ± 8.52	13.0 ± 5.98	39.0 ± 5.08
WEP†, mg kg^{-1}	4.07 ± 0.78	0.77 ± 0.32	3.31 ± 0.54
DPS	0.20	0.14	0.02

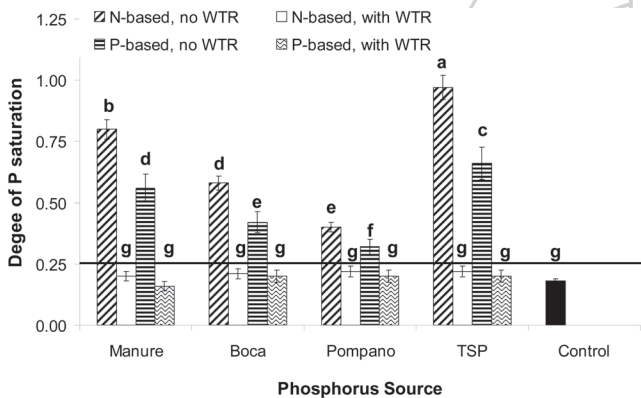† DPS, degree of phosphorus saturation; WEP, water-extractable phosphorus.

Consistent with the DPS values, soils receiving P sources only (without WTR amendments) had significantly greater WEP concentrations (>10 mg kg^{-1}) than the control and the WTR-amended soils (Fig. 4). In the absence of WTR, there were significant ($p \leq 0.001$) differences among the WEP values of the treatments having different P sources and between the two rates of P application. However, with WTR amendment, these differences were no longer significant (Fig. 4). The results of the soil DPS and WEP concentrations were consistent with findings from other studies that concluded that WTR can effectively immobilize and reduce soil-soluble P (Ippolito et al., 2003; Elliott et al., 2002b; Makris et al., 2005; Dayton and Basta, 2005a; Agyin-Birikorang et al., 2007).

Effects of Drinking-Water Treatment Residuals on Ground Water Phosphorus Concentrations

The TDP concentrations of the shallow wells ranged between 0.6 and 1.2 mg L^{-1}, and the PO$_4$–P concentrations of the shallow ground water ranged between 0.2 and 0.95 mg L^{-1}. Despite the differences in the TDP and PO$_4$–P values of the shallow wells, data for PO$_4$–P and TDP concentrations of samples followed similar trends (Fig. 5 and 6). The observed background (control) TDP concentration of the shallow wells of the study site (0.6–0.8 mg TDP mg L^{-1}) exceeded the critical P solution concentration (0.10 mg L^{-1}) guidelines estab-
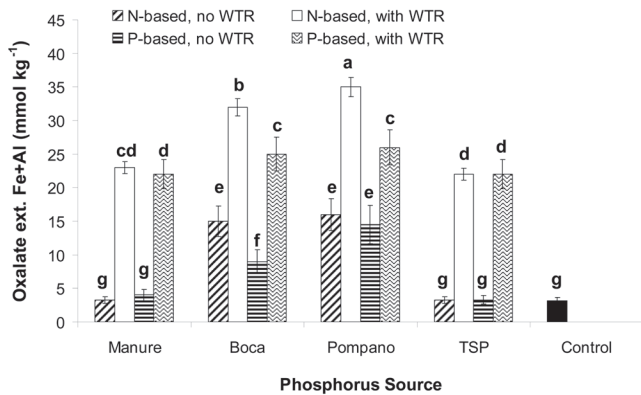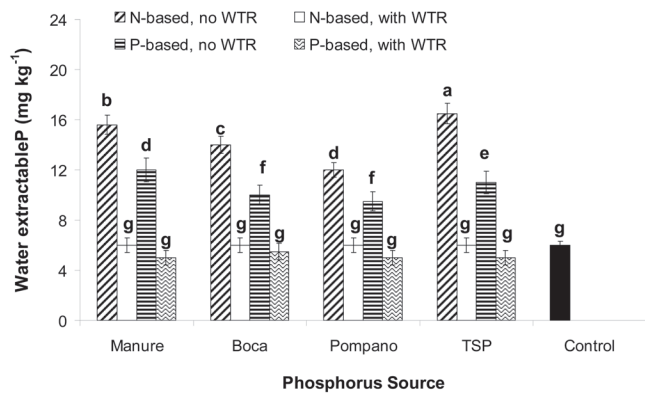
Fig. 2. Effect of phosphorus sources (applied at N-based and P-based rates) and drinking-water treatment residuals (WTR) amendment on mean oxalate-extractable Fe+Al concentrations of the A horizon (0–5 cm depth) soil samples collected throughout the soil sampling period (April 2003–September 2004). Error bars denote 1 SD of the mean. Treatments having the same letter are not significantly different according to the adjusted Tukey multiple comparison test at a significance level (α) of 0.05. Boca, Boca Raton biosolids; Manure, poultry manure; N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass; Pompano, Pompano biosolids; TSP, triple superphosphate.

Fig. 3. Effect of phosphorus sources (applied at N-based and P-based rates) and drinking-water treatment residuals (WTR) amendment on degree of phosphorus saturation (DPS) of the A horizon (0–5 cm depth) soil samples collected throughout the soil sampling period (April 2003–September 2004). Horizontal line represents critical DPS value for Florida sandy soils. Error bars denote 1 SD of the mean. Treatments having the same letter are not significantly different according to the adjusted Tukey multiple comparison test at a significance level (α) of 0.05. Boca, Boca Raton biosolids; Manure, poultry manure; N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass; Pompano, Pompano biosolids; TSP, triple superphosphate.

Fig. 4. Effect of phosphorus sources (applied at N-based and P-based rates) and drinking-water treatment residuals (WTR) amendment on mean water-extractable P of the A horizon (0–5 cm depth) soil samples collected throughout the soil sampling period (April 2003–September 2004). Error bars denote 1 SD of the mean. Treatments having the same letter are not significantly different according to the adjusted Tukey multiple comparison test at a significance level (α) of 0.05. Boca, Boca Raton biosolids; Manure, poultry manure; N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass; Pompano, Pompano biosolids; TSP, triple superphosphate.

lished for streams (USEPA, 1986b). However, the ground water P concentrations were similar to the upper limits of ground water P concentrations observed by others in Florida (0.1–0.7 mg TDP mg L^{-1}) (Woodard et al., 2007) and elsewhere (0.05–0.6 mg TDP mg L^{-1}) (Nelson et al., 2005). The elevated ground water P concentration possibly resulted from decades of intensive dairy cattle production at the site (Kenneth R. Woodard, personal communication, 2007). High ground water P concentrations are not uncommon from fields subjected to long-term disposal of manure (0.4–0.9 mg L^{-1}) (Breeuwsma et al., 1995) and sewage (0.3–1.5 mg L^{-1}) (Corbett et al., 2001). Despite the high ground water background P concentration of our study site, treatment effects of the soil surface applied amendments were observed.

Although the shallow ground water P concentrations showed significant temporal variability over the experimental period, treatment effects on shallow ground water P concentrations were still obvious (Fig. 5 and 6A–6D). The observed temporal variability of the ground water P concentrations could be attributed to rainfall variability over the sampling period (Fig. 1), which possibly resulted in differential P leaching over time.

Without WTR application, the greatest ground water TDP and PO_4–P concentrations during the first 7 mo of the study period were found with the TSP "N-based" treatment, followed by soils treated with poultry manure, Boca Raton biosolids, and Pompano biosolids (N-based). However, from 7 to 20 mo, the ground water TDP and PO_4–P concentrations of the TSP N-based treatment significantly decreased and stabilized at ~0.8 and 0.60 mg L^{-1}, respectively (Fig. 5D and 6D). The WEP values (Table 1) showed that TSP was >85% water soluble, suggesting that much of the excess P in the TSP (N-based) treated soil leached out during the first year of application. Despite the temporal variability of the ground water TDP and PO_4–P concentrations, P leaching from soils treated with the organic sources of P (poultry manure and biosolids) was apparently consistent throughout the sampling period (Fig. 5 and 6A–6C). A low percentage of the total P of the organic sources of P (~1.5–7%; Table 1) was water soluble and could have resulted in slow P release from the soils treated with the organic sources of P. Application of WTR prevented an increase in ground water (shallow well) P concentrations from the treatments with the P sources applied at the N-based rates.
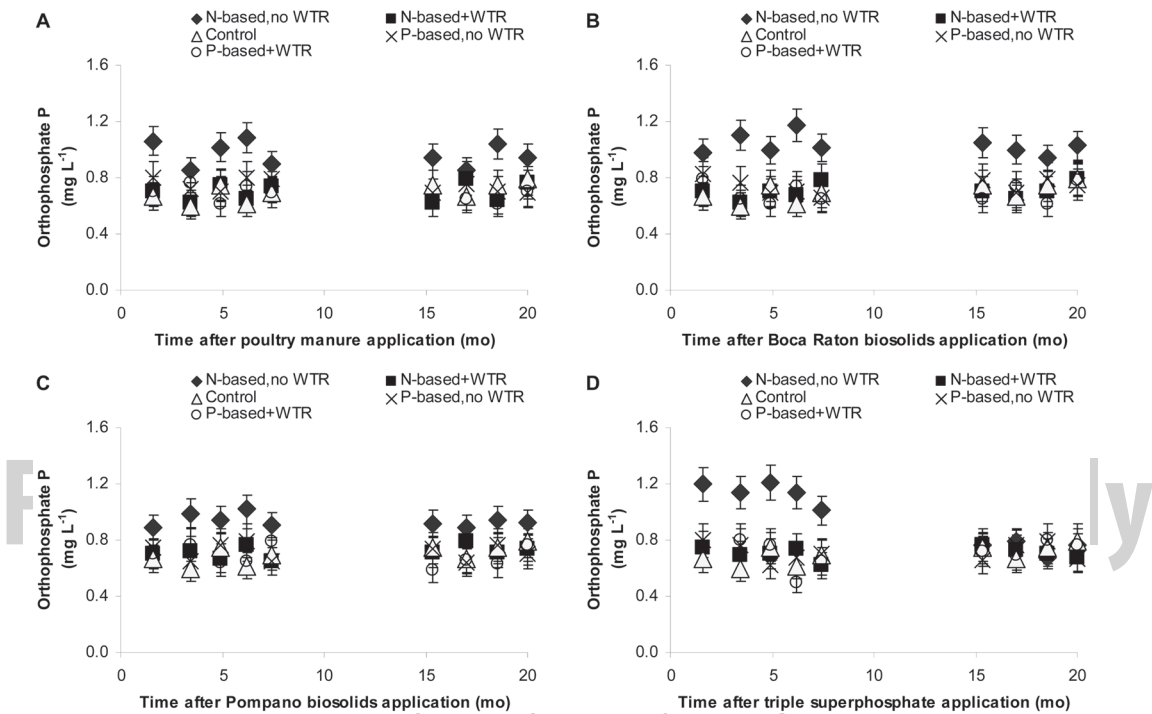
Fig. 5. Trends of total dissolved phosphorus concentrations of shallow ground water samples collected throughout the sampling period (July–October 2003 and June–September 2004) from plots amended with (A) poultry manure, (B) Boca Raton biosolids, (C) Pompano biosolids, and (D) triple superphosphate. A total of nine samplings from the shallow well of each plot were used to plot trends. Control, no P source and drinking-water treatment residuals (WTR) application; N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass.
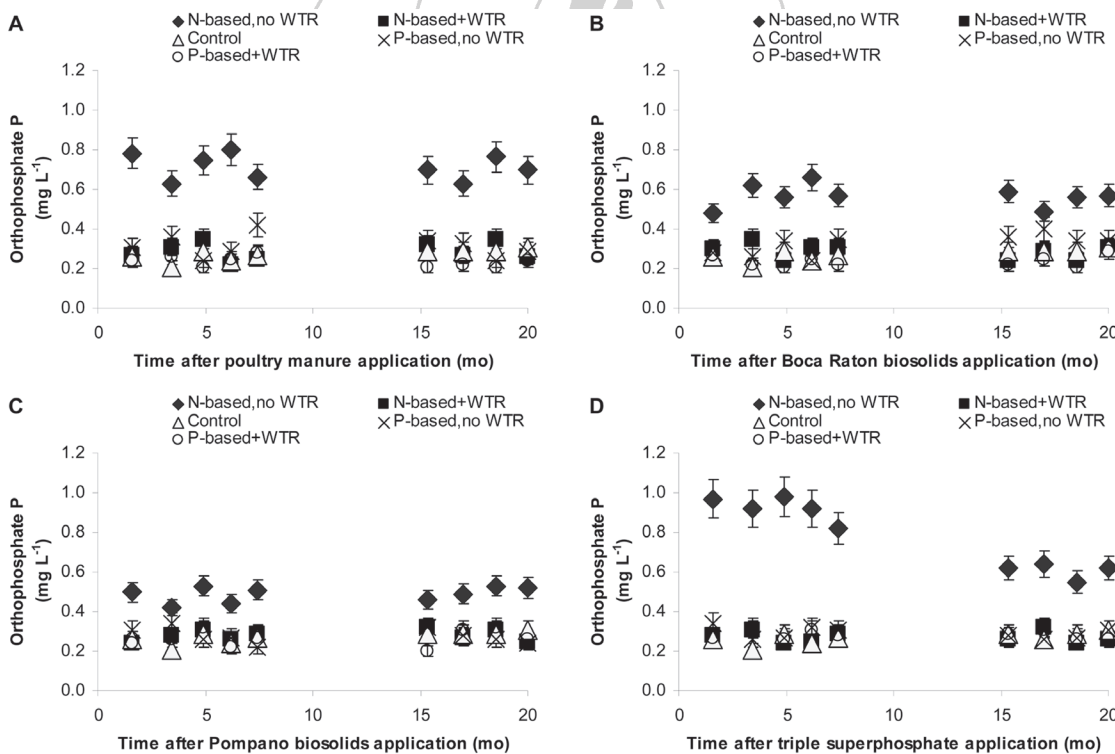

Fig. 6. Trends of orthophosphate concentrations of shallow ground water samples collected throughout the sampling period (July–October 2003 and June–September 2004) from plots amended with (A) poultry manure, (B) Boca Raton biosolids, (C) Pompano biosolids, and (D) triple superphosphate. A total of nine samplings from the shallow well of each plot were used to plot trends. Control, no P source and drinking-water treatment residuals (WTR) application; N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass; WTR, drinking-water treatment residuals.
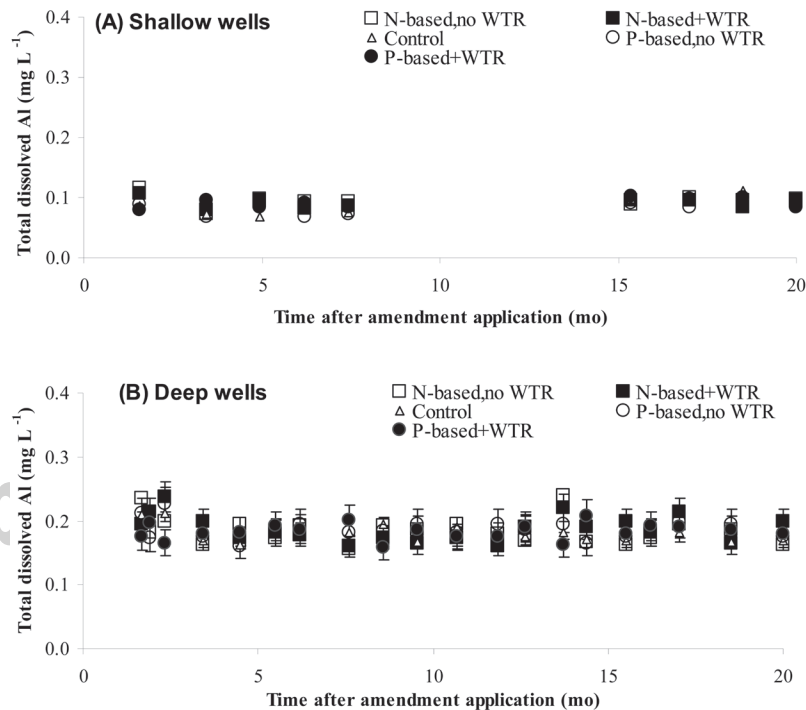
Fig. 7. Trends of total dissolved Al concentrations in water samples collected from (A) shallow and (B) deep wells throughout the sampling period (July–October 2003 and June–September 2004 for the shallow wells and March 2003–September 2004 or the deep wells). Nine samplings of the shallow wells and 20 deep well samplings from all the plots were used to plot trends. N-based, application rate based on the nitrogen requirements of bahiagrass; P-based, application rate based on the phosphorus requirements of bahiagrass; WTR, drinking-water treatment residuals.

In the presence of WTR, the ground water TDP and PO_4–P concentrations were, respectively, similar among both the plots treated with different P sources and to background concentrations (Fig. 5 and 6).

The water samples collected from the shallow wells of the plots having the P sources applied at P-based rates (without WTR amendments) had ground water TDP and PO_4–P concentrations similar to those of the control plots (Fig. 5 and 6). Thus, any P leached from the treatments with P sources applied at P-based rates would have been too little to significantly pollute the percolating water. The data suggest that application of P sources at P-based rates, without any P-binding soil amendment, could be a BMP to prevent off-site P losses from agricultural fields.

Drinking-Water Treatment Residual Effects on Ground Water Aluminum Concentrations

Land application of Al-WTR significantly increased oxalate-extractable Al content of the surface soil, which is consistent with the trends observed by others (Dayton and Basta, 2005a; Agyin-Birikorang et al., 2007). Although most of the WTR influence would occur initially near the soil surface, there is a concern among regulators that Al from the surface-applied Al-WTR could contaminate ground water. Therefore, total dissolved Al concentrations in shallow and deep ground water were monitored after surface application of Al-WTR.

Ground water total dissolved Al concentrations were unaffected by the Al-WTR application. Total dissolved Al concentrations of the samples obtained after amendments application

and throughout the sampling period from the shallow wells ranged from 0.07 to 0.12 mg Al L^{-1}, and concentrations in deep wells ranged from 0.14 to 0.25 mg Al L^{-1} (Fig. 7). The ranges in ground water Al concentrations of the WTR-amended treatments compared well with the controls (0.07–0.11 Al mg L^{-1} for shallow wells and 0.17–0.21 mg Al L^{-1} for deep wells). Total dissolved Al concentrations were within (or close to) the Florida Groundwater Guidance Concentration value of ≤0.2 mg L^{-1} (Merchant, 1989; Jain et al., 2005) and less than ground water Al concentrations observed by Nilsson and Bergkvist (1983). Generally, the Al concentrations were greater in the deep wells than in the shallow wells, which may reflect contributions of organically complexed Al species from the spodic horizon but not from the surface-applied treatments. Nilsson and Bergkvist (1983) studied Al chemistry in Swedish podzols and reported greater total Al concentrations (2.6–3.1 mg L^{-1}) in leachate samples below the Bh horizon than in water samples taken above the Bh horizon (0.09–1.3 mg L^{-1}).

The results suggest that Al-WTR can be safely used to enhance the P-sorption capacity of Florida sandy soils and reduce soluble P losses to ground water without increasing total dissolved Al concentrations in ground water. Several studies have shown that pH control of soluble Al concentrations dominates Al ecological risks (Lindsay, 1979; Sloan et al., 1995; Fest et al., 2007). The pH values of the Al-WTR, the WTR-amended soil, and the ground water were 5.6, 5.5, and 5.9, respectively. At such pH values, Al species are likely to be dominated by hydrolytes of Al (Lindsay, 1979; Sloan et al., 1995; Lindsay and Walthall, 1996) and other organically complexed Al forms

(Lindsay and Walthall, 1996; Fest et al., 2007) rather than free Al^{3+}. Hence, there is little concern that free Al^{3+} will leach from the soil surface–applied Al-WTR to contaminate water bodies except under adverse conditions (e.g., pH <4).

Summary and Conclusions

This study was conducted on a typical Florida Spodosol amended with different P sources (biosolids, manure, and inorganic fertilizer) applied at P- or N-based rates and with or without WTR application. Surface-applied WTR reduced P leaching from the soil treated with different P sources applied at the N-based rates. Although the native ground water P concentrations at the site were generally high, the results suggest that WTR could be used to prevent further deterioration of the ground water when P sources are applied to the site at the N-based rate. Minimal or no increases in ground water P concentrations occurred when the P sources were applied at P-based rates (without WTR amendment) relative to the background (control) P concentration. Nevertheless, WTR application reduced the ground water P concentrations resulting from the P-based treated plots. The increase in Al concentration of the surface soils after WTR application was not accompanied by increases in total dissolved Al concentrations in the ground water. Thus, at least for the study period, WTR can be safely used to enhance P sorption capacity of Florida Spodosols and prevent P leaching into the ground water without increasing ground water Al concentration. Although WTR adsorbs tremendous amount of P and does not readily release P back into the environment, P loadings in excess of WTR P retention capacity could result in off-site P losses from WTR-amended soils. Thus, BMPs that limit excessive P loadings must be adopted, in addition to WTR amendment, to ensure long-term P control. Further studies are needed to determine the impact of heavy rainfall events (e.g., hurricanes) on P and Al losses to surface runoff and to determine setback distances from drainage lines to minimize this impact.

Acknowledgments

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