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Long-term performance of a constructed wetland/filter basin system treating wastewater, Central Florida

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ABSTRACT

This study investigated the efficiency of a constructed wetland/filter basin (CW/FB) treatment system to improve the chemical composition of waste and surface waters. The system was constructed in closed phosphate mines used for clay settling and sand tailings. Monitoring was carried out for 18 months to evaluate the CW/FB performance under a variety of climatic conditions. Water samples were taken bi-monthly. To evaluate possible groundwater input into and water leaking out of the wetland 6 monitor wells were installed along the flow path and sampled monthly. In order to estimate the change of water chemistry along the wetland flow path, water samples along a transect were taken during the dry and rainy seasons. The samples were analyzed for pH, *T*, oxidation–reduction potential (ORP), conductivity, total dissolved solids (TDS), dissolved oxygen (DO), Fe(II), H_2S, major anions, major cations, arsenic, fecal and total coliform. The study showed the following changes in water quality between the input and output: (1) Substantial decrease of water temperature (up to 10 °C); (2) Significant change in pH from about 9 to 6.5–7; (3) Negative ORP confirming the reducing conditions of the treatment system; (4) Substantial increase of H_2S (up to 1060 µg/L); (5) Reduction of As from 5 to <2 µg/L (mostly <0.5); (6) Substantial reduction of SO_4, F, Cl, NO_3, NO_2, Br, Na, K, Ca, and Mg; (7) Reduction of fecal and total coliform from 30–730 and 1000–7000 to <2 and <100 count/100 mL, respectively. In general, the performance of the CW/FB treatment system showed great potential to improve the water quality of industrial and municipal wastewater. Despite significant seasonal variations with respect to temperature, rainfall and humidity, the chemical/microbiological composition of the wetland output remained relatively constant.

1. Introduction

Constructed wetlands (CWs) are artificial wastewater treatment systems composed of a shallow basin filled with substrate, such as soil or gravel, and planted with vegetation tolerant of saturated soil conditions (Davis, 1995; EPA, 2000). The use of CWs for wastewater or stormwater treatment was initiated in 1904 in Australia and became prevalent in the United States during the early 1970s (Cole, 1998). Natural processes in the wetland remove organic, inorganic and microbiological contaminants (Carruthers, 2002; Vrhovsek et al., 1996). Vascular plants play a particularly important role, stabilizing substrates while enhancing permeability, reducing water velocities and thus allowing the settling of suspended solids, using nutrients, carbon, and trace elements for plant stem and root systems, and transporting gases between the sediments and atmosphere (Butler and Dewedar, 1991; Liu et al., 2007). In addition, photosynthesis by algae increases the concentration of dissolved oxygen affecting nutrient and metal reactions (Davis, 1995). But more importantly, plants' stem and root systems provide the necessary surface area for growth and adhesion of microorganisms, which facilitate the decomposition of organic material and the recycling of nutrients (Martin and Moshiri, 1994).

Previous studies demonstrated that CWs were advantageous treatment systems for the remediation of acid mine drainage (AMD) due to comparably low cost and maintenance (Braun et al., 2003; Woulds and Ngwenya, 2004; Stottmeister et al., 2006). In general, contaminated mine water is highly acidic and rich in sulfate and metals such as Al, As, Cd, Cu, Fe, Pb, Mn, Ni and Zn (Ritcey, 1989; Stottmeister et al., 2006). The AMD treatment mechanisms in CWs may include metal adsorption on soil matter, accumulation into below- and above-ground plant tissues, and microbial mediated co-precipitation or volatilization of particular metalloids (Jacob and Otte, 2003; Stottmeister et al., 2006).

The reclamation of wastewater and phosphate mining lands using CW technology is very important in Florida, providing an excellent opportunity for environmental improvement and restoration (EPA, 1993). Phosphate mining in Central Florida is broadly distributed and annually, disturbs about 15–25 km^2 of land through generation of clay settling areas, open mine pits, and tailing sand deposits (FIPR). Florida law requires reclamation of previously mined phosphate lands into lakes, wetlands, pasture, and agricultural lands. Thus, constructed

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wetlands on clay settling areas could become a new option for reducing environmental pollution and reclamation through (1) wastewater treatment; (2) water quality improvement and encouragement of water reuse; (3) fish- and wildlife habitat; (4) passive recreation, such as bird watching and photography; (5) active recreation such as hunting; (6) flood storage, and (7) resynchronization of storm rainfall and surface runoff (Davis, 1995). Increased pumping from the Floridan Aquifer caused saltwater intrusion along the coast, and lowering of groundwater levels, thus affecting spring flows and lake levels (Peck et al., 2005). Therefore, a significant purpose of CWs in Florida metropolitan areas could be the generation of water that meets drinking water standards (DWS) to supplement rivers and streams and to satisfy public, industrial, and agricultural water demands. Moreover, this type of water could be important to the future of aquifer storage and recovery (ASR) as a means of water management in Florida and potentially worldwide. The principle behind ASR is the storage of treated surplus surface water in a confined aquifer followed by its recovery during times of need (Arthur et al., 2005). Unfortunately, the injection of treated surface water causes the dissolution of pyrite (FeS_2) and mobilization of arsenic (As) from the Floridan carbonate with As values in recovered water of up to 130 µg/L (Arthur et al., 2005; Price and Pichler, 2006). The dissolution of pyrite is a result of its oxidation due to the much higher oxygen content and thus, much higher redox potential of the injected surface water (Jones and Pichler, 2007). A water composition closer to the native groundwater would therefore be a much better choice for ASR. Discharge from a wetland could be the ASR water of choice, because it is often more reducing than other surface water and contains sulfide, which is favorable for the stability of pyrite (e.g., Jones and Pichler, 2007).

The present paper summarizes an 18-month performance study of a wetland in an area used during phosphate mining for clay settling in Polk County, Central Florida. The major objective of this study was to investigate the possibility to use wetlands in closed phosphate mines to improve the chemical composition of waste and surface waters.

1.1. Study area

The constructed wetland/filter basin (CW/FB) treatment system was located on a clay settling area and tailing sand deposits at the Hines Energy Complex, Polk County, Florida (27°48′N latitude and 81°52′W longitude) (Fig. 1). The CW/FB system was established in 1999 and used for the experimental treatment of industrial wastewater from the Hines Energy electric power generating plant (cooling water), tertiary treated effluent from the city of Bartow, as well as rain and excess surface water. The surface flow wetland was approximately 1500 m long, 10 m wide, ranged in water depth from 0.5 to 2 m and was constructed in a U-shape (Fig. 1). The length–width ratio of the wetland was higher than maximum suggested value of 1:1 to 1:2 (EPA, 2000) due to specific topographic settings of the study area. The area of the wetland was about 12,250 m^2. The wetland was not lined and the substrate consisted of clay matrix with the decomposing organic matter. Wetland vegetation was allowed to evolve naturally (i.e., not planned and planted) due to comparably high costs and maintenance. It consisted of both native Floridian and non-native species such as water lettuce (*Pistia stratiotes*), carolina willow (*Salix caroliniana*), Brazilian pepper (*Schinus terebinthifolius*), water fern (*Salvinia*), baby's tears (*Micranthemum umbrosum*), cattail (*Typha* spp.), willow (*Salix* spp.), and common duckweed (*Lemna minor*). The 6000 m^2 filter basin (FB) constructed on tailing sands was used to improve the efficiency and reliability of the treatment system. The depth of the FB (i.e., sand bed) was 4 m and the walls and bottom of the FB were lined with a polyethylene cover. The depth of the ponded water discharged on the FB surface was <10 cm. The most important feature of the FB was the development of a biological active layer called the "schmutzdecke" on the tailing sand surface (the top 0.5–2 cm). This reddish-brown slimy biofilm acted as a fine filter of solid particles (mechanical filtration) and a zone of biological action providing the degradation of soluble organics and the potential

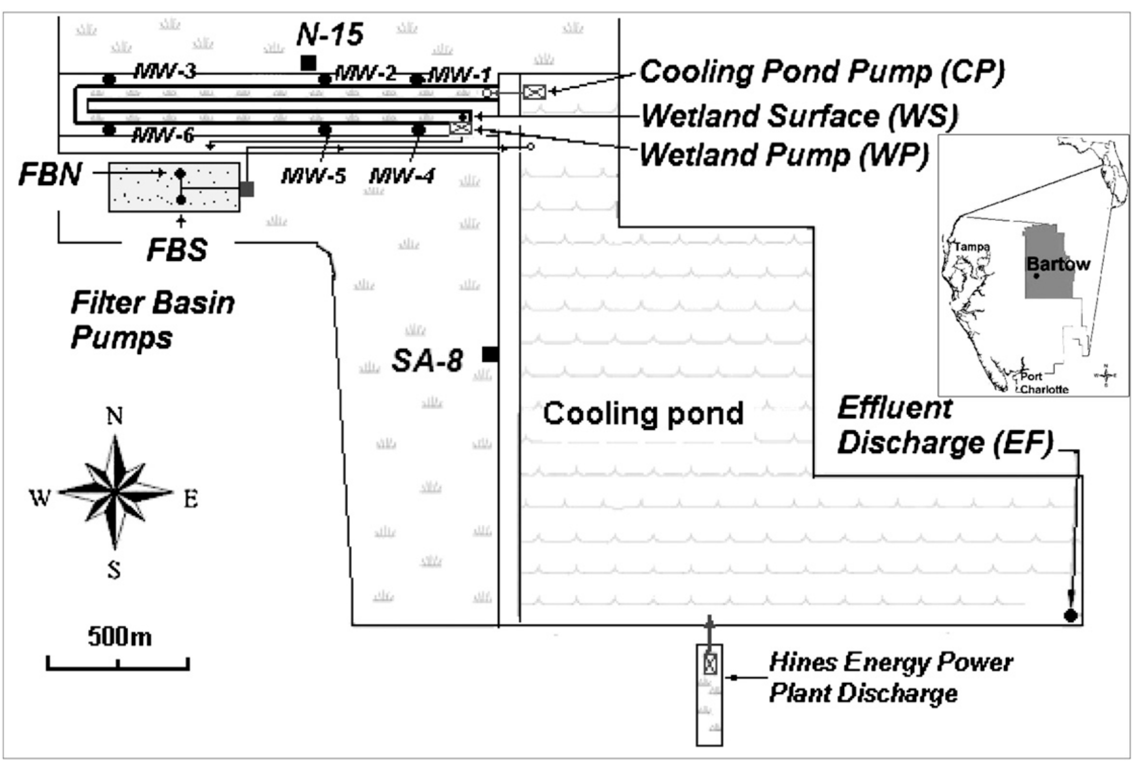

Fig. 1. Map of the study area located at the Hines Energy complex (Polk County, Central Florida) including water transfer system from the cooling pond to the U-shaped constructed wetland and filter basin treatment system. Note water sampling locations: MW-1 to MW-6 — monitor wells; CP — cooling pond pump; WP — wetland pump; water bodies to the north and south of the wetland — N-15 and SA-8 (surface water); WS — wetland surface; EF — effluent discharge (tertiary-treated effluent from Bartow); FBN and FBS — filter basin north and south pumps.

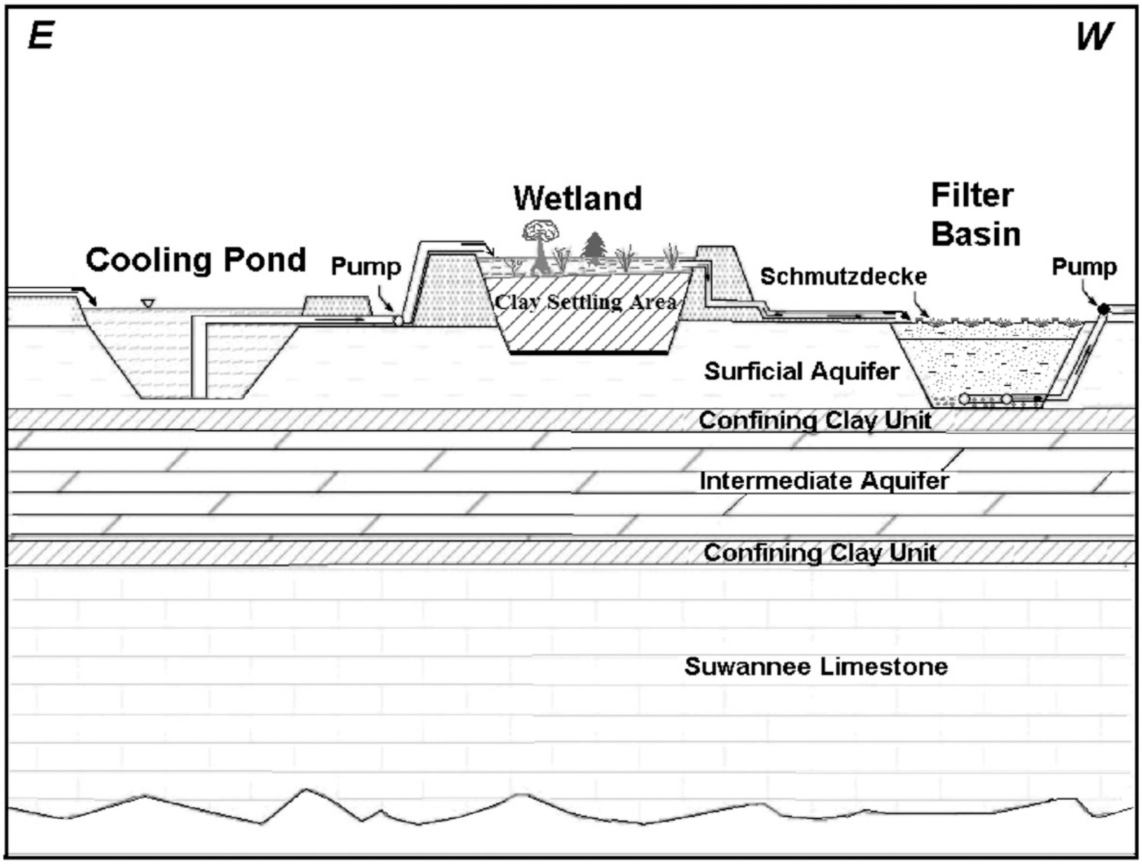

Fig. 2. Schematic concept diagram of the constructed wetland/filter basin treatment system. Water from the cooling pond was pumped into the wetland, discharged onto the filter basin sand surface, collected in a series of pipes on the bottom of the filter basin, and pumped out of the sand bed with the filter basin pumps. Note: Schmutzdecke — biological film formed on the sand surface (the top 0.5–2 cm).

elimination of pathogens, color and odor contaminants in water (Huisman and Wood, 1974; Muhammad et al., 1997).

Water from the Hines Energy Complex cooling pond (CP) was pumped into the wetland at different rates depending on the season (Fig. 1). During the rainy seasons of 2006 and 2007, the hydraulic loading rates into the wetland were around 0.33 and 0.45 L/day/m^2, respectively. During the dry season 2006, it was about 0.61 L/day/m^2. The residence time of water in the wetland was approximately 120 days. At the end of the flow path the water was either pumped back into the CP to control water levels in the wetland, or discharged onto the filter basin surface. Following filtration through the fine sand the water was collected in a series of pipes (Fig. 2).

2. Methods and sampling procedure

Monitoring was carried out for 18 months in order to evaluate wetland performance under a variety of climatic conditions. Water sampling began in April 2006 and finished in October 2007. Bi-monthly water samples were obtained from the effluent discharge (EF), cooling pond (CP), wetland pumping station (WP) and wetland

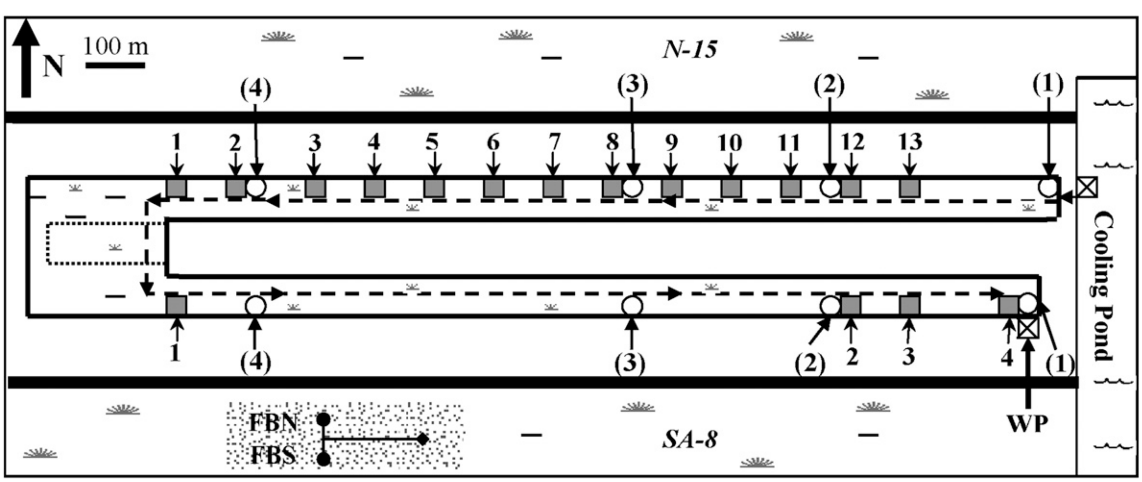

Fig. 3. Evaluation of water quality along the wetland flow path. Note: WP — wetland pump; FBN and FBS — filter basin north and south pumps; Dashed arrow — wetland flow path; Solid squares and empty circles — wetland water samples taken on March 2007 and September 2007, respectively.

Table 1
Maximum, minimum, arithmetic mean and standard deviation of analyzed parameters in the cooling pond (CP), wetland pump (WP) and surface (WS), filter basin south (FBS) and north (FBN) pumps.

		Max	Min	Med	AM	STD	*N*	Max	Min	Med	AM	STD	*N*
		Cooling pond (CP)						Wetland pump (WP)					
T	°C	38.3	19.4	30.3	30.0	5.4	38	30.3	13.9	24.0	23.2	4.3	36
pH		9.1	8.1	8.6	8.6	0.3	38	7.4	6.4	7.0	6.9	0.2	36
ORP	mV	200	122	23	18	85	38	4	176	85	80	39	36
Cond	S/cm	1122	766	850	878	84	24	1000	273	704	638	214	22
H_2S	g/L	51.0	0.0	3.5	10.2	13.9	37	1060.0	0.0	752.5	619.3	323.3	35
As	g/L	5.1	1.3	2.5	2.7	0.9	37	6.6	0.1	0.5	0.7	1.1	36
DO	mg/L	15.0	3.6	9.3	9.3	2.3	38	7.4	1.6	5.4	4.9	1.4	36
F		4.5	1.5	3.4	3.4	0.6	38	4.4	0.7	2.5	2.4	0.9	36
Cl		150.8	107.6	128.1	128.9	9.1	38	145.7	26.5	110.0	96.7	35.8	36
NO_2		0.7	0.0	0.0	0.0	0.1	38	0.1	0.0	0.0	0.0	0.0	36
Br		2.5	0.7	1.6	1.6	0.3	38	2.2	0.0	1.2	1.0	0.6	36
NO_3		3.5	0.0	0.0	0.1	0.6	38	2.1	0.0	0.0	0.1	0.4	36
PO_4		8.4	0.6	2.3	2.9	1.7	38	8.5	0.0	3.9	4.1	2.1	36
SO_4		115.7	69.5	83.5	86.6	9.9	38	81.5	1.3	44.7	40.5	26.1	36
K		15.3	8.8	10.9	11.0	1.5	38	16.3	0.8	9.5	8.8	3.9	36
Na		87.4	56.4	75.3	76.3	7.5	38	95.4	17.3	67.6	59.4	23.1	36
Ca		74.9	45.2	54.7	53.9	5.4	38	62.1	17.2	47.2	44.1	11.7	36
Mg		39.6	28.7	33.5	33.6	3.2	38	39.5	8.5	28.7	25.3	9.6	36
Fe(II)		0.1	0.0	0.0	0.0	0.0	38	1.0	0.0	0.2	0.3	0.2	34
Fe		0.3	0.0	0.0	0.0	0.0	38	0.6	0.0	0.1	0.2	0.1	36
Mn		0.0	0.0	0.0	0.0	0.0	38	0.1	0.0	0.0	0.0	0.0	36
Si		1.2	0.1	0.2	0.3	0.2	38	2.0	0.0	1.2	1.2	0.5	36
Sr		0.6	0.5	0.5	0.5	0.0	38	0.5	0.1	0.4	0.3	0.1	36

surface (WS), tailing sand filter basin northern (FBN) and southern (FBS) pumping stations, and water bodies to the north and south of the wetland, N-15 and SA-8 (surface water) (Fig. 1). The WP sample was collected from a submerged sump located at depth of 2 m and the WS sample — surface water from the same location. Overall, 244 samples were collected and analyzed.

To investigate and evaluate the change of water chemistry along the wetland flow path samples were collected within the surface water at depths of 0 and 0.5 m using a peristaltic pump. During the dry season (March 19–20, 2007), water samples were collected at 17 stations and during the rainy season (September 24–25, 2007) — at 11 stations along the flow path of the wetland (Fig. 3).

To evaluate possible groundwater input into and water leaking out of the wetland 6 monitor wells were installed along the flow path and sampled monthly. This was necessary to separate wetland induced changes in water chemistry from those due to dilution by Floridan groundwater or seepage from the water bodies to the north and south, N-15 and SA-8 (Figs. 1 and 3). In total, 121 samples from MWs were collected and analyzed.

2.1. Field and laboratory analysis

The samples were analyzed immediately in the field for pH, temperature (*T*), oxidation–reduction potential (ORP) and conductivity using a Myron-L Ultrameter^{TM}. The meter was calibrated in the field with known buffer solutions. The dissolved oxygen (DO) concentration was determined using a HACH HQ40d Meter with a LDO (luminescent dissolved oxygen) probe. The LDO sensor was calibrated following the manufacturer's specifications. The concentration of ferrous iron (Fe(II)) was determined with a CHEMets Colorimetric field kit with a color chart, and sulfide (H_2S) was analyzed using a Methylene Blue Method on a HACH DR 2400 portable photospectrometer. The water samples were filtered through a 0.45 m membrane and separated into two 30 mL HDPE bottles. One aliquot was used for the determination of major anion concentrations. The other sample was acidified to 1% HNO_3 for the analysis of major cations and arsenic. In addition, the set of unfiltered and unacidified samples from the CP, WP and FBS/FBN was collected for fecal and total coliform bacteria from September, 2006 to September, 2007.

The samples were stored in a cooler and transported to the Center for Water and Environmental Analysis, University of South Florida, Tampa. Calcium (Ca), iron (Fe), magnesium (Mg), manganese (Mn), silica (Si), sodium (Na), potassium (K), and strontium (Sr) were measured on a Perkin Elmer Optima 2000 DV inductively coupled plasma-optical emission spectrometer (ICP-OES). Fluoride (F), chloride (Cl), sulfate (SO_4), nitrate (NO_3), nitrite (NO_2), bromide (Br), and phosphate (PO_4) were determined by ion chromatography (IC) using a Dionex ICS 2000 system. Total arsenic (As) concentrations were measured by hydride generation-atomic fluorescence spectrometry (HG-AFS) on a PSA 10.055 Millenium Excalibur instrument. The accuracy and precision of the measurements were verified through the use of internal and external standards, indicating a precision and accuracy better than 5%. Measurements of fecal and total coliform bacteria were carried out at the Southern Analytical Laboratory in Florida. Precipitation measurements were performed by the staff from the Hines Energy Complex weather station. Meteorological data was available from the nearby Frostproof Station of the Florida Automated Weather Network (FAWN).

3. Results

3.1. Monitor wells

The lithology of monitor well (MW) cores along the wetland was uniform and sediments were composed of light-tan to brown poorly to well-sorted fine sands or silts with occasional grey clay nodules. The brown color was due to the presence of organic material. After installation, each MW was developed using a small submersible pump to recharge debris free water and sampled monthly for the duration of the study.

The chemical composition of water collected from the MWs was different from the cooling pond (CP), wetland pump (WP), and the wetland surface (WS), with values in the MWs either higher or lower (Tables 1 and S1). The chemical composition of the MWs was much closer in composition to sites SA-8 and N-15, and groundwater from the Surficial Aquifer System (GW) (Sacks and Tihansky, 1996). In Fig. 4A the average concentrations of SO_4, F, As, and Fe in monitor wells (MW-1 to MW-6) are compared. MWs were arranged according to the wetland flow path. MW-6 had the highest As (up to 9 μg/L) and Fe

Max	Min	Med	AM	STD	*N*	Max	Min	Med	AM	STD	*N*	Max	Min	Med	AM	STD	*N*
Wetland surface (WS)						Filter basin south pump (FBS)						Filter basin north pump (FBN)					
31.1	16.0	25.2	24.0	4.3	24	28.2	15.9	24.7	23.7	3.7	30	30.1	16.8	24.5	23.8	3.8	25
7.3	6.1	6.9	6.9	0.3	24	7.3	6.3	6.8	6.8	0.3	30	7.5	6.3	6.9	6.8	0.3	25
101	166	38	31	69	24	2	100	55	53	33	30	87	108	9	16	51	25
1002	248	771	690	254	16	932	250	732	688	199	19	925	275	741	730	163	16
900.0	0.0	17.0	247.8	373.0	24	160.0	0.0	15.5	23.8	32.5	30	40.0	0.0	6.0	8.8	11.4	25
13.1	0.2	0.9	1.8	3.2	24	2.7	0.5	1.0	1.2	0.6	29	2.2	0.5	0.9	1.3	0.6	25
5.4	0.1	1.5	1.9	1.4	24	6.2	0.9	2.3	2.6	1.3	30	6.5	1.2	5.0	4.6	1.7	25
4.3	0.2	2.9	2.6	1.1	24	3.4	1.0	2.1	2.2	0.6	29	3.4	0.3	2.2	2.3	0.8	25
157.1	5.6	116.6	100.4	49.2	24	158.6	10.1	104.4	92.5	39.3	29	158.4	23.5	111.1	103.1	38.8	25
0.0	0.0	0.0	0.0	0.0	24	0.2	0.0	0.0	0.0	0.1	29	0.2	0.0	0.0	0.0	0.1	25
2.0	0.0	1.3	1.2	0.6	24	2.0	0.0	1.2	1.0	0.5	29	2.7	0.3	1.3	1.3	0.6	25
0.1	0.0	0.0	0.0	0.0	24	4.0	0.0	0.2	0.4	0.7	29	5.8	0.1	0.4	0.8	1.2	25
8.2	0.0	3.6	3.8	2.3	24	4.3	0.0	1.7	1.6	1.0	29	5.1	0.0	2.6	2.4	1.1	25
79.8	0.6	44.8	40.8	27.4	24	81.8	0.6	36.9	37.2	21.6	29	76.6	5.6	46.1	43.9	21.1	25
14.6	3.0	11.1	10.4	3.4	24	13.8	3.9	9.7	9.7	3.0	29	13.9	4.1	10.5	10.3	2.6	25
79.2	17.0	55.0	54.1	15.7	24	75.5	28.5	52.2	52.5	12.4	29	74.8	30.4	54.4	55.0	9.9	25
63.3	17.0	44.6	44.9	12.9	24	57.4	25.6	43.0	42.5	8.8	29	56.5	22.0	43.9	43.5	8.1	25
39.5	8.9	28.6	27.4	8.7	24	35.1	10.5	26.0	25.3	7.3	29	35.7	8.9	27.6	26.9	6.8	25
0.8	0.0	0.2	0.3	0.2	24	3.0	0.3	1.5	1.4	0.7	30	1.5	0.1	0.4	0.5	0.4	25
0.5	0.0	0.0	0.1	0.1	24	2.3	0.1	0.7	0.9	0.7	29	1.3	0.0	0.2	0.3	0.3	25
0.1	0.0	0.0	0.0	0.0	24	0.1	0.0	0.0	0.0	0.0	29	0.1	0.0	0.0	0.0	0.0	25
0.5	0.1	0.4	0.4	0.1	24	0.4	0.2	0.3	0.3	0.1	29	0.4	0.2	0.3	0.3	0.1	25
3.8	0.0	1.1	1.2	0.8	24	2.5	0.5	1.4	1.4	0.5	29	2.0	0.0	1.3	1.3	0.5	25

(up to 50 mg/L) concentrations among all MWs. The analyses of water samples from the MWs showed that there was little to no leakage from the sites N-15 and SA-8 into the wetland treatment system. The analysis of variance (ANOVA) of the conservative tracer Na was applied to examine the difference in water chemistry between MWs, SA-8 and N-15 (Fig. 4B). For the first group of samples (MW-4 to MW-6 and SA-8), the F ratio (30.0) was significantly larger than the F critical value (2.7) indicating a statistically significant difference within the data set. For the second group of samples (MW-1 to MW-3 and N-15), the F ratio and F critical value were 7.5 and 2.7, respectively. The variance for MW-3 had the highest value (361.0) potentially demonstrating the highest influence from N-15.

3.2. Precipitation measurements

Due to periodic variations in precipitation and temperature in the study area two different seasons can be distinguished: the dry season from November to April, and the rainy or wet season from May to October. Daily precipitation was recorded at the Hines Energy Complex weather station from May 1, 2006 to October 31, 2007, covering one dry and two rainy seasons (Fig. 5). The highest levels of rainfall (up to 123 mm) were detected during the major hurricane events (Ernesto and Alberto) in June and August 2006. The mean monthly rainfall ranged from 0 to 11 mm. Total seasonal precipitation during the dry season of 2006 was 324 mm, while the rainy seasons of 2006 and 2007 had between 917 and 782 mm, respectively.

3.3. Evaluation of water quality along the ow path

Evaluation of the wetland during the dry (March 19–20, 2007) and rainy (September 24–25, 2007) seasons was important to understand the consistency and reliability of the treatment system in time and space. The wetland transect showed a distinct pattern of change in water chemistry along the flow path from the input – cooling pond (CP) to the output – wetland water from pump (WP) (Table 2, Fig. 6A–B).

3.3.1. Field measurements

3.3.1.1. Temperature (T). Generally, during the dry and rainy seasons temperature in the wetland was <20 and 25 °C, respectively. However during the dry season, it reached up to 24.8 °C (500–800 m) on the wetland surface.

3.3.1.2. pH. The pH values were reduced along the wetland flow path from 8.9 to about 7.0, but reached up to 8.7 on the wetland surface (500–700 m and 1400 m) during the dry season. The formation of organic acids in the wetland was likely responsible for this drastic change in pH.

3.3.1.3. Oxidation reduction potential (ORP). Generally, ORP was negative in the wetland indicating the reducing conditions. At the same time during the dry season, the ORP level at 300–700 m on the wetland surface and at a depth of 0.5 m reached +254 and +217 mV, respectively.

3.3.1.4. Sul de (H_2S). During the dry season the concentration of H_2S was up to 900 µg/L and did not show the trend along the wetland. In contrast, during the rainy season H_2S reached up to 2545 µg/L increasing along the wetland flow path.

3.3.1.5. Dissolved oxygen (DO). After entering the wetland, DO along the wetland flow path was generally reduced to <0.5 mg/L. During the dry season it reached up to 8.7 mg/L on wetland surface (200–700 m of the wetland flow path). The concentration of DO at the WP during the dry and rainy seasons was up to 6.0 and 3.3 mg/L, respectively. This was due to trapping of oxygen during the pumping procedure.

3.3.1.6. Ferrous iron (Fe(II)). Concentration of Fe(II) at the WP was up to 0.2 mg/L but was not present in the CP, which was likely due to high DO levels.

3.3.1.7. Anions. During the dry and rainy seasons the concentrations of most anions decreased along the wetland flow path. The F, Cl, and SO_4 levels gradually decreased from 3.1 to 2.8 mg/L, 118 to 107 mg/L, and 58 to 25 mg/L, respectively. The concentration of NO_2 and NO_3 was mostly below detection at the WP and the CP. During the dry season the concentration of Br and PO_4 at the WP was close to the CP. In the contrast, during the rainy season the level of Br was 1.5 times lower but PO_4 was 1.7 times higher in the wetland.

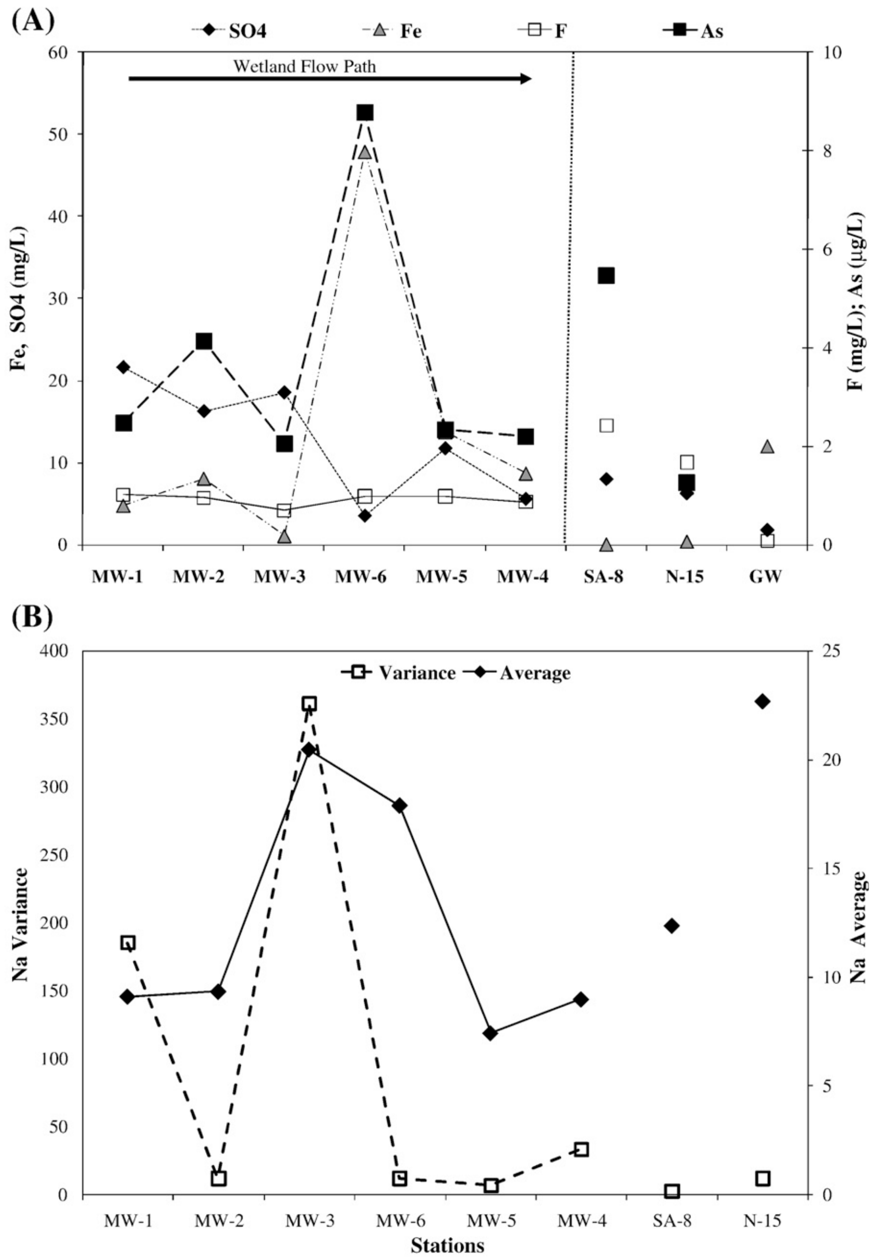

Fig. 4. (A) Average values of SO_4, Fe, F, and As and (B) Average and variance of Na estimated by ANOVA at MW-1 to MW-6, N-15 and SA-8. Note: MW-1, 2, 3, 6, 5 to MW-4 — monitor wells arranged according to the wetland flow path; GW — groundwater from the Surficial Aquifer System, Polk County (well ROMP 57A; data adapted from Sacks and Tihansky, 1996).

3.3.1.8. Cations. Generally, the behavior of most cations had a comparable pattern. During the dry season, the concentration of Na, Mg, K, Ca, and Sr in the wetland was close to the CP. In contrast, during the rainy season the concentration of these cations in the wetland was around 1.5 times less than in the CP. This behavior could be caused by a dilution effect or due to minor groundwater inflow. The concentration of Mn and Fe (total) was mostly below detection. The CP water was virtually Si-free. At the same time, during the dry and rainy seasons Si at the WP was 0.5 and 2 mg/L, respectively.

3.3.1.9. Arsenic (As). During both seasons the concentration of As significantly decreased along the wetland flow path from 2.3 to <0.2 µg/L.

The concentration of conservative tracer elements Na and Cl at the monitor wells (MWs), sampled on April 3 and October 3, 2007, was considerably lower than in the wetland water (Fig. 6B).

3.4. Wetland/ lter basin water quality monitoring

The major purpose of this study was to investigate the possibility of using a constructed wetland for the wastewater treatment in an area used during phosphate mining for clay settling. Summarized below are results derived from the field and laboratory analyses (Table S1, Fig. 7). The performance of the wetland was evaluated by mass flux of analytes removed from or contributed to the wetland water. The mass fluxes were calculated as *concentration ow rate = mass/time*. The flow rates used for calculation varied depending on the season between 5012, 6757 (rainy seasons 2006 and 2007, respectively) and 9255 L/day (dry season 2006). The percent removal of each parameter was calculated as ((*mass ux*$_{CP}$ *mass ux*$_{WP}$)/*mass ux*$_{CP}$) 100 with and without the rainfall dilution factor over the study area (Table 3). The average amount of rainfall over the wetland area varied from 1995, 1697 (rainy seasons 2006 and 2007, respectively) to 745 L/day (dry season 2006).

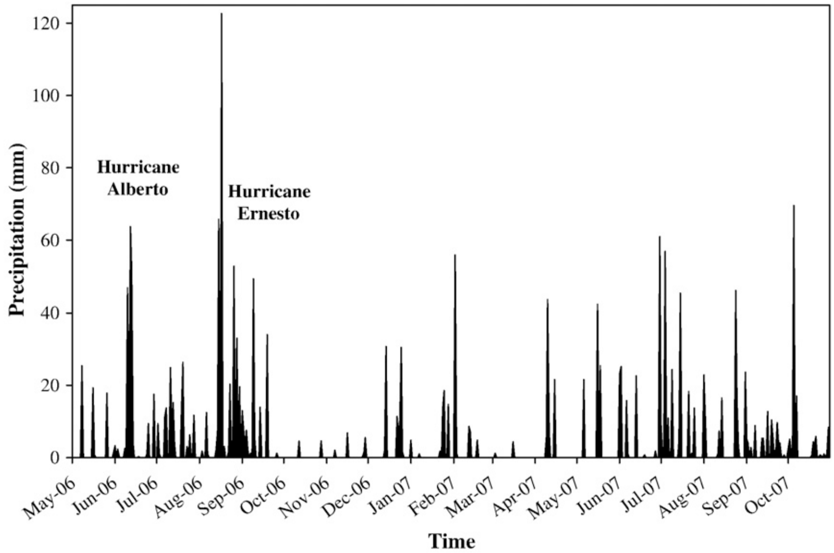

Fig. 5. Precipitation hydrograph recorded at the study area from May 1, 2006 to October 31, 2007, covering one dry and two rainy seasons. Note: Major hurricane events (Ernesto and Alberto) in June and August 2006.

The corrected concentration with the rainfall dilution factor was calculated as *concentration* (*ow rate*/(*ow rate rainfall*)) and subsequently used for the mass fluxes and percent removal.

Along the wetland flow path most of the monitored constituents were removed from the CP with the exception of Si, PO_4, and Fe. During the rainy season 2007 (May–July), however, the wetland contributed mass to the water due to technical difficulties. In addition, during the dry season, the wetland contributed mass in form of K (up to 40%), likely from plant stem and root systems (Fisher, 1971).

3.4.1. Fecal and total coliform

Previous studies demonstrated the capability of constructed wetlands to reduce pathogenic microorganisms in wastewater (e.g., Neralla and Weaver, 2000; Hill and Sobsey, 2001). Removal efficiency was >90% for total coliform and >80% for fecal streptococcus (Kadlec and Knight, 1996).

Table 2
Change in water composition from the cooling pond (CP) to the wetland from pump (WP) during the dry (March 2007) and rainy (September 2007) seasons.

		Dry season		Rainy season	
		CP-input	WP-output	CP-input	WP-output
DO	mg/L	9.4	6.0	9.4	3.3
T	°C	23.1	16.5	32.1	25.0
pH		8.7	7.0	8.9	7.0
ORP	mV	63	84	116	171
Cond	µS/cm	865	767	958	1056
Fe^{2+}	mg/L	n/d	0.1	n/d	0.2
H_2S		30	731	10	1175
F		3.8	3.1	3.9	2.8
Cl		129.8	118.5	137.6	107.3
NO_2		n/d	n/d	n/d	n/d
NO_3		n/d	n/d	1.5	n/d
Br		1.5	1.3	2.2	1.5
PO_4^3		3.9	3.9	1.7	3.0
SO_4^2		89.1	58.1	92.1	25.0
Na		56.2	56.6	71.6	52.3
Mg		30.1	29.0	40.4	27.1
K		10.3	10.8	14.4	9.3
Ca		47.5	45.9	60.8	40.6
Sr		0.4	0.4	0.5	0.3
Mn		0.0	0.0	0.0	0.0
Fe		0.0	0.1	0.0	0.0
Si		0.0	0.5	0.1	2.0
As	µg/L	2.0	0.2	2.3	0.2

Fecal and total coliform in the CP ranged between <1–370 count/100 mL and <1–2000 count/100 mL, respectively (Fig. 8A–B). The maximum contaminant level (MCL) for total coliform (including fecal coliform) is that no more than 5.0% of the samples should detect total coliform in one month (EPA DWS). Therefore, in this study, coliform bacteria must be <1 count/100 mL. The highest fecal and total coliform levels were detected in those samples collected at the end of the wetland flow path (WP). Levels were 30–730 count/100 mL fecal and 1000–7000 count/100 mL for total coliform bacteria. This may be caused by feces from fish, reptiles, amphibians, insects, and birds which were abundant in the wetland. In contrast, fecal and total coliform at the FBS/FBN were generally <2 count/100 mL (except one sample of 29 count/100 mL) and <100 count/100 mL, respectively. These results clearly demonstrate the crucial role of the biofilm “schmutzdecke”. It formed on the tailing sand surface providing mechanical filtration, degradation of soluble organics and elimination of pathogens, color and odor contaminants (Huisman and Wood, 1974; Muhammad et al., 1997).

4. Discussion

During the 18-month period of monitoring, the constructed wetland (CW) demonstrated promising treatment efficiency for the remediation of wastewater. The study showed a significant wetland cooling effect on water temperature with up to a 10 °C difference between the input – cooling pond (CP) and output – wetland water from pump (WP) (Fig. 7). Seasonal fluctuations of the wetland water temperature could influence the processes of microbial transformation (Kadlec, 1999). Kadlec (2006) studied the surface flow wetlands (Tres Rios, Arizona) for temperature and energy flows. The author reported that wetland water temperature had a tendency to approach the mean air temperature depending on humidity. In this study, temperature cycles for WP and air demonstrated comparable distribution with the correlation coefficient R^2 of 0.83 (Fig. 7). Treatment surface flow CW can have one or two thermal regions depending on the water residence time (Kadlec, 2006). The residence time in this study was approximately 14 days. When the residence time is >5 days, the wetland can be divided into 2 zones: accommodation and balance zones (Kadlec, 2006). In an accommodation region, the temperature and evapotranspiration profiles are initially steep due to adjustments to weather conditions. In a balance zone, temperature in the wetland approaches to the balance level, i.e.

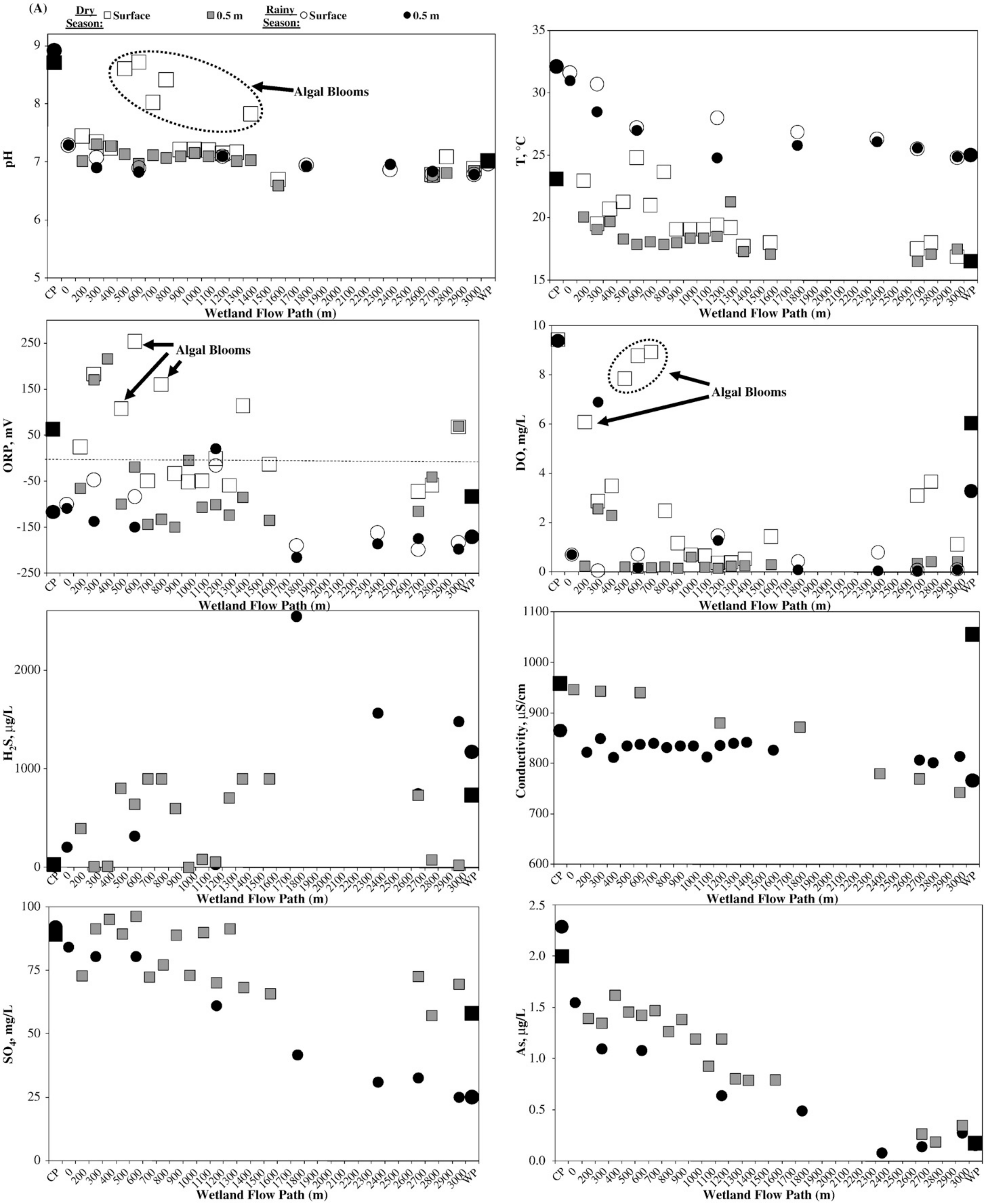

Fig. 6. A: Evaluation of *T*, pH, ORP, DO, H_2S, SO_4, conductivity, and As along the wetland flow path during the dry (March 2007) and rainy (September 2007) seasons; B: Distribution of Na and Cl along the wetland flow path and at the MWs during the dry (March 2007) and rainy (September 2007) seasons; Note: CP — cooling pond pump; WP — wetland pump; MW — monitor wells arranged according to the wetland flow path; Elevated pH, DO, *T*, positive ORP, and low H_2S during the dry season were caused by algal blooms.

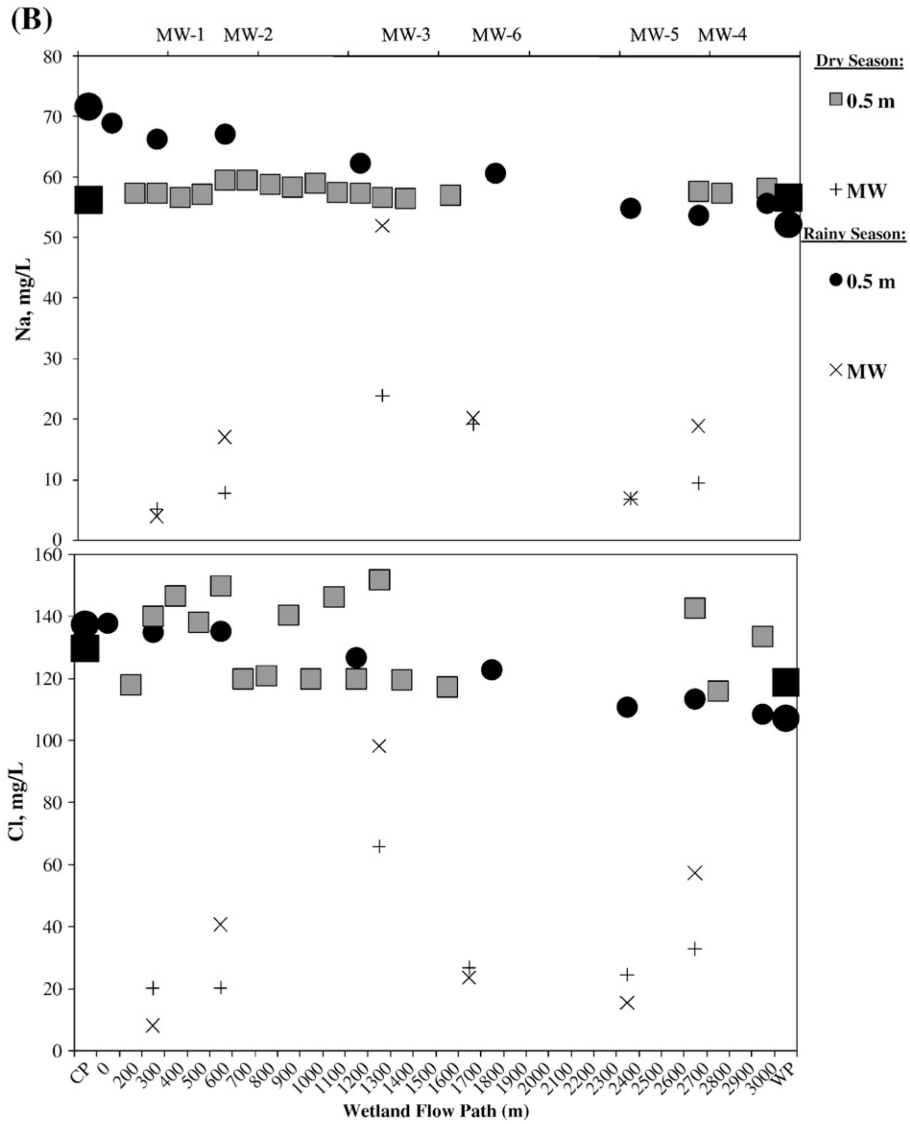

Fig. 6 (*continued*).

adjusts to the existing meteorological conditions (Kadlec, 2006). This finding was very useful for evaluating temperature along the CW flow path. Generally, the water temperature was gradually reduced along the wetland and could be divided into 2 zones with the boundary line around 1300 m (Fig. 6A). The cooling effect of the wetland could probably reduce evaporation losses.

Meteorological monitoring conducted during the study demonstrated the dominance of 3 tropical seasons: one dry and two rainy seasons (Fig. 5). Therefore, it is very important to evaluate the performance of the treatment system in response to seasonal changes such as heavy rainfall and drought events. Generally, the behavior of most cations had a comparable pattern. During the dry season, the concentration of Na, Mg, K, Ca, and Sr in the wetland was close to the CP. In contrast, during the rainy season the concentration of these cations in the wetland was around 1.5 times less than in the CP. This behavior could be caused by dilution of the wetland water by rainfall. Elevated pH, DO, *T*, positive ORP of the wetland surface and low H_2S during the dry season were generally caused by algal blooms during spring time (Fig. 6A). Sawyer and McCarty (1978) reported that in shallow ponds during daylight algae use CO_2 for photosynthesis and release O_2 increasing the pH and DO levels as the carbonate–bicarbonate equilibrium is destabilized. During night time hours this process is generally reversed when algae and plants stop producing O_2 but start using the available oxygen.

In order to quantitatively establish the efficiency and the possible groundwater input into the wetland, the Na and Cl mass fluxes in the CP and WP were examined (Table 3, Fig. 9). Both curves showed a comparable distribution as well as a significant change throughout the duration of the study. During the first rainy season (April–October 2006), the mass fluxes of Na and Cl at the WP were around 60% less than in the CP. These reduced mass fluxes were due to heavy rainfall during two hurricanes, which added low conductivity water directly and triggered enhanced groundwater input into the wetland (Criss and Winston, 2003). During the dry season (November 2006–April 2007), the percentage removal of Na and Cl from the wetland was close to 0% indicating the normal operation of the wetland treatment system, i.e. the balance between input and output. This allowed to estimate the residence time of water in the wetland to about 120 days. During the second rainy season (April–October 2007), the percent removal of Na and Cl from the wetland varied significantly from 52 to 4%. At the end of April, the CP pump was turned off and the wetland surface was lowered approximately 1 m for maintenance purposes

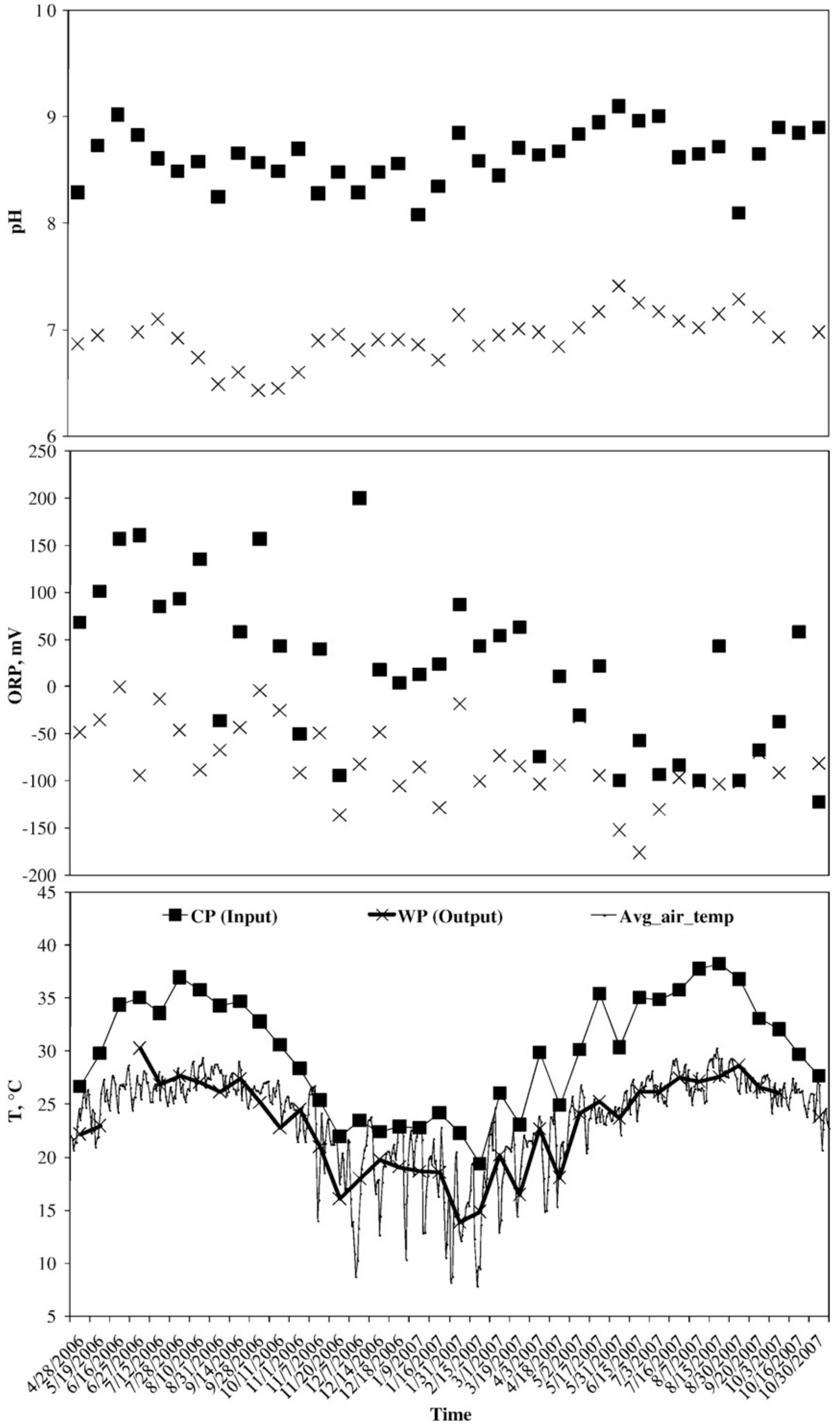

Fig. 7. Distribution of pH, ORP, and temperature (T) in the CP — input and WP — output waters. Note: CP — cooling pond pump; WP — wetland pump; Average air *T* measured at elevation of 0.6 m was obtained from the Frostproof Station in Polk County (FAWN).

(Fig. 9). From May 2007, the wetland treatment system became again operational. Therefore, during this time a considerable mass flux of Na and Cl accumulated in the wetland could be caused by mechanical issues as well as high evaporation. As soon as the maintenance problems were fixed, the percentage removal of Na and Cl from the wetland was close to 0%.

Fig. 10 was plotted to assess the variation of a weight concentration SO_4/Cl in the CP and WP as a function of time. The concentration of SO_4 changed through microbiological reduction which is greatly influenced by seasonal temperature (Urban et al., 1994). High temperatures during summer and fall accelerate microbial activity in decomposing organic material, thus producing higher levels of H_2S (Armannsson, 1999). The plot demonstrated a variability of SO_4/Cl ratio in the wetland throughout the period the study with the lower levels during the rainy seasons 2006 and 2007 (<0.15 and <0.3, respectively) and the highest (up to 0.6) — during the dry season 2006 (Fig. 10).

The monitoring of the treatment system frequently showed higher concentrations of total Fe at the filter basin south (FBS) and north (FBN) pumps compared to the WP. During the dry season the average Fe level at the FBS was 0.34 mg/L, while at the WP and FBN it was 0.26

Table 3
The wetland performance evaluated with the percent removal of each analyzed parameter.

	Date	As	As*	K	K*	Na	Na*	Ca	Ca*	Mg	Mg*	Sr	Sr*	S^T	S^{T*}	SO_4	SO_4*	F	F*	Cl	Cl*	Br	Br*	Si	Si*	PO_4	PO_4*
Rainy season 2006	4/28			70	50	74	58	14	42	68	47	70	50	97	96	98	97	56	26	78	63	100	100	536	957	323	602
	5/19	69	48	56	26	70	51	5	74	64	39	54	23	96	94	98	97	58	29	73	55	100	100	610	1080	229	446
	6/27	71	52	45	9	63	39	51	18	60	33	64	41	91	86	92	87	51	19	64	40	83	72	593	1052	221	434
	7/12	79	65	50	17	63	39	27	22	58	30	58	31	94	89	94	90	49	15	64	40	72	53	619	1094	275	523
	7/28	89	82	44	7	55	26	36	6	57	28	60	34	94	91	94	91	51	18	54	24	75	58	808	1408	268	512
	8/10	94	90	29	17	46	10	21	32	46	10	51	18	91	85	94	90	42	3	46	10	66	44	1015	1753	169	347
	8/31	53	22	59	32	65	42	57	28	67	45	80	67	90	83	92	86	42	3	66	43	60	34	1162	1996	103	237
	9/14	74	57	68	47	70	49	62	37	70	51	74	56	95	91	95	92	60	34	69	49	87	78	348	645	650	1146
	9/28	79	65	72	53	73	55	61	35	71	52	85	75	95	92	96	93	69	48	74	57	90	83	374	687	68	179
	10/11	96	94	66	44	61	35	48	13	61	36	68	46	91	86	92	87	48	14	61	36	73	55	920	1594	99	231
Dry season 2006–2007	11/1	85	84	3	5	11	3	4	4	16	9	34	28	39	34	43	38	100	100	11	3	100	100	358	398	7	2
	11/7	88	87	4	4	10	2	3	5	15	7	21	15	23	16	26	19	10	19	8	0	11	3	107	125	110	128
	11/20	65	62	8	17	2	7	17	27	2	7	13	5	29	22	32	26	3	12	7	2	7	1	12	22	125	145
	12/7	83	82	93	93	8	0	6	2	11	3	30	24	24	17	26	20	25	18	6	2	19	12	113	114	23	34
	12/14	79	78	14	25	2	6	2	6	8	0	17	10	15	8	18	11	5	14	0	9	2	11	60	75	61	75
	12/18	82	81	25	36	1	7	10	19	5	14	6	2	24	17	27	21	29	23	0	8	26	19	234	263	9	18
	1/9	89	88	29	40	5	3	20	12	11	3	20	12	43	38	47	42	5	14	10	2	10	20	125	144	103	121
	1/16	55	51	39	51	6	2	10	3	11	4	24	17	36	30	39	34	28	22	11	3	26	20	595	656	53	67
	1/31	55	51	4	14	6	2	13	6	13	5	17	10	19	12	19	12	8	17	10	2	12	22	96	113	93	110
	2/15	67	64	5	14	11	3	13	6	14	6	25	18	25	19	28	21	14	6	13	6	32	43	419	464	24	35
	3/1	73	71	3	12	9	1	6	3	11	4	24	17	33	27	36	30	29	22	9	1	30	23	3	12	27	21
	3/19	92	91	2	6	3	5	7	1	8	0	13	5	28	22	30	24	18	10	4	4	12	4	247	277	1	9
	4/3	87	86	9	1	9	1	1	10	0	8	4	13	29	22	31	25	10	19	8	0	18	28	92	92	52	66
	4/18	88	87	5	14	5	4	11	3	10	2	12	4	32	26	34	28	0	8	5	3	24	35	1545	1689	43	55
Rainy season 2007	5/2	80	74	8	45	0	34	4	29	3	29	4	29	28	4	28	5	8	23	4	28	9	21	1168	1593	53	104
	5/17	76	68	16	54	9	46	3	37	4	40	0	34	18	10	20	7	38	18	7	42	4	28	1620	2197	100	100
	5/31	77	69	11	48	14	52	5	41	9	46	2	36	16	12	18	10	82	76	9	45	46	28	496	696	100	100
	6/15	62	49	4	39	0	34	6	25	3	29	11	19	25	0	28	3	26	68	3	29	83	77	1000	1369	55	40
	7/3	30	74	7	24	10	20	6	26	3	30	9	22	32	10	35	14	14	53	10	20	8	23	665	921	394	559
	7/16	67	55	33	10	24	1	31	8	30	7	37	15	52	36	54	38	23	3	25	0	31	8	816	1124	96	162
	8/7	79	72	44	26	25	0	29	5	28	4	35	13	65	54	68	58	48	30	27	3	55	40	947	1298	76	69
	8/15	84	78	44	25	26	1	29	5	30	6	35	13	68	57	71	61	37	16	26	1	45	27	647	898	53	37
	8/30	88	84	33	10	22	4	25	1	25	0	32	10	60	47	63	50	24	2	23	3	31	8	466	656	46	95
	9/20	95	93	39	19	28	4	34	12	34	11	42	22	66	55	69	58	33	10	28	4	43	24	925	1269	58	44
	10/3	92	90	23	3	17	10	25	0	25	1	31	8	59	45	62	49	26	1	21	5	29	5	1065	1456	83	144
	10/30	90	86	10	20	17	11	17	11	20	7	26	1	58	45	61	49	28	4	19	8	25	1	2074	2804	40	20

Note: Positive values — wetland removes mass and negative — wetland contributes mass to the water that flows through it; percent removal of total S was calculated from a sum of S(VI) and S(II); Most of NO_2 and NO_3 was not detected in input and output waters; Fe was not detected in the CP.

* Percentage included the rainfall dilution factor over the site area.

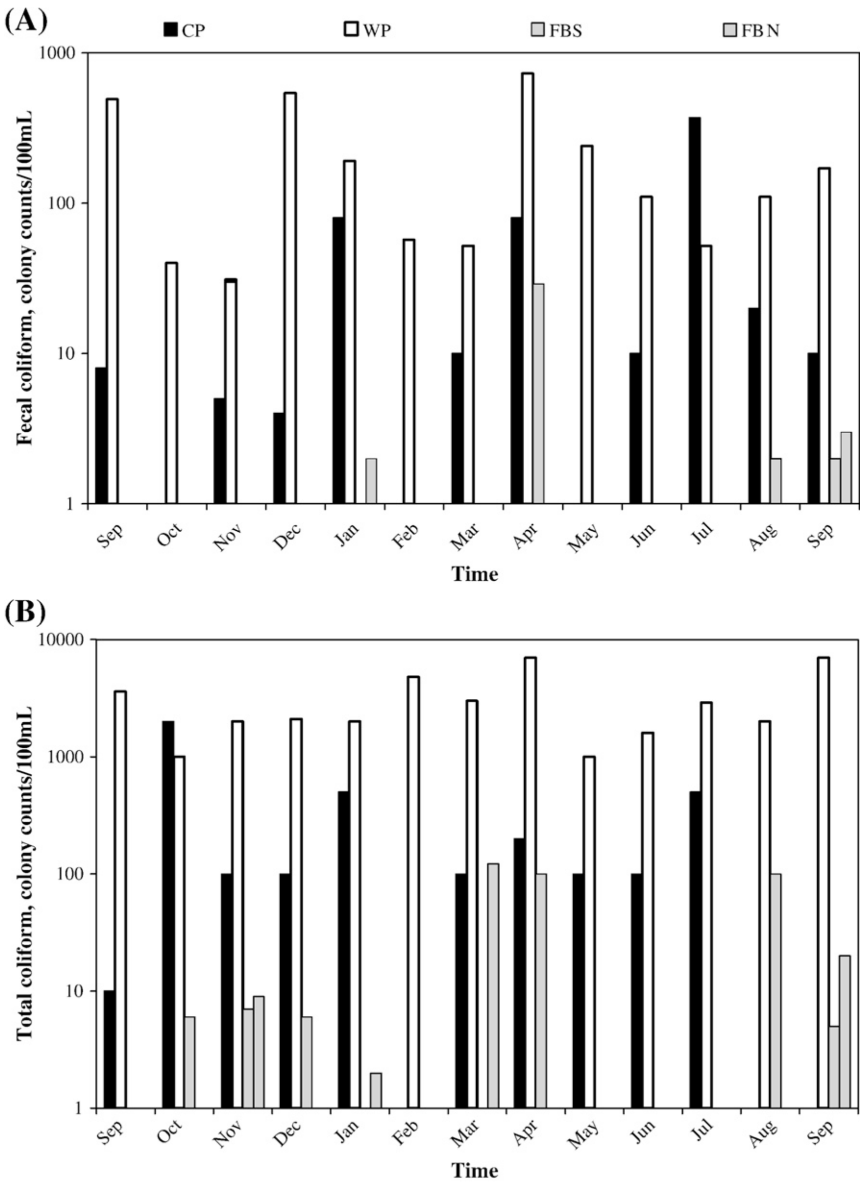

Fig. 8. Fecal (A) and total (B) coliform detected at the CP, WP, FBS and at the FBN. FBS/FBN had the lowest fecal and total coliform confirming the crucial role of the biofilm "schmutzdecke". Note: CP — cooling pond pump; WP — wetland pump; FBS and FBN — filter basin south and north pumps; Concentrations displayed in logarithmic scale; Time ranged from Sept., 2006 to Sept., 2007.

and 0.14 mg/L, respectively. In contrast, during the rainy seasons of 2006 and 2007, the average Fe at the FBS was up to 1.20 mg/L, while at the WP and FBN it was 0.20 and 0.42 mg/L, respectively. The distribution of Fe concentration at the WP, FBS and FBN with time demonstrated lower Fe at the WP and FBN compared to the FBS. At the same time, concentrations of Fe at the WP, which was being applied to the surface of the filter basin (FB), remained relatively constant, while Fe at the FBS fluctuated quite significantly (Fig. 11). Moreover, substantially higher Fe levels at the FBS were detected particularly during the rainy seasons potentially indicating an outside source of water to the FB containing high Fe. This is possible due to the migration of the surficial groundwater into the FB from the surrounding area. Previous studies reported that the concentration of Fe in the surficial groundwater of the study area could be about 12 mg/L (ROMP 57A; Sacks and Tihansky, 1996). Field observations confirmed that the levels of the surficial groundwater outside the FB increased during heavy rainfall therefore increasing the hydraulic gradient around the FB. This fact was supported by visual observation of iron mud buildup clogging flow meters and pipes of the FB, especially at the FBS.

4.1. Behavior of As

Arsenic (As) is an element of great interest in Florida principally to aquifer storage and recovery (ASR) (Arthur et al., 2005). Studies showed that As was found in the carbonate Floridan Aquifer mostly associated with pyrite (Price and Pichler, 2006; Lazareva and Pichler, 2007). Pyrite is thought to dissolve and release As during ASR due to the injection of ozone-treated oxidizing surface waters into reducing native groundwater. As a result, concentrations of As in recovered water were up to 130 µg/L (Arthur et al., 2005), far above the 10 µg/L drinking water standard (DWS) for As (EPA DWS). The type of constructed wetland/filter basin treatment system studied here could become a new alternative to treat wastewater in Florida and beyond. Water treatment processes in the wetland may consist of metal accumulation into vegetation, adsorption on soil particles, precipitation or co-precipitation caused by microbial activity (Dushenko et al., 1995; Stolz and Greger, 2002; Jacob and Otte, 2003; Stottmeister et al., 2006). Following after treatment through a CW the water would be in reducing conditions with high sulfide and low oxygen levels which are

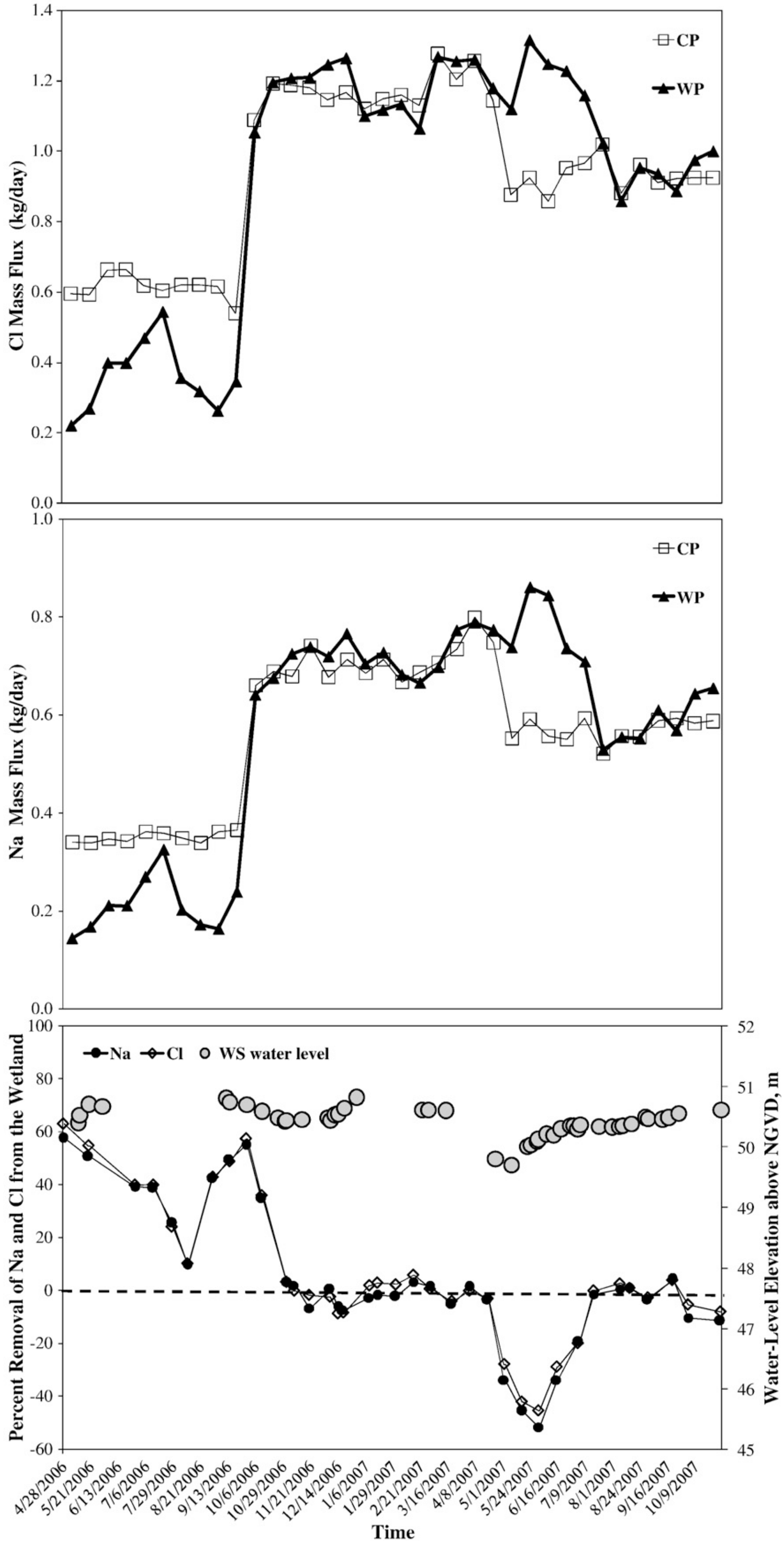

Fig. 9. Mass fluxes of Na and Cl in the CP and WP, and the calculated percent removal of Na and Cl from the wetland. Note: CP — cooling pond pump; WP — wetland pump; WS water level — wetland surface water level; NGVD — National Geodetic Vertical Datum; Calculation included the rainfall dilution factor over the site area; Positive values — wetland removes mass and negative — wetland contributes mass to the water that flows through it.

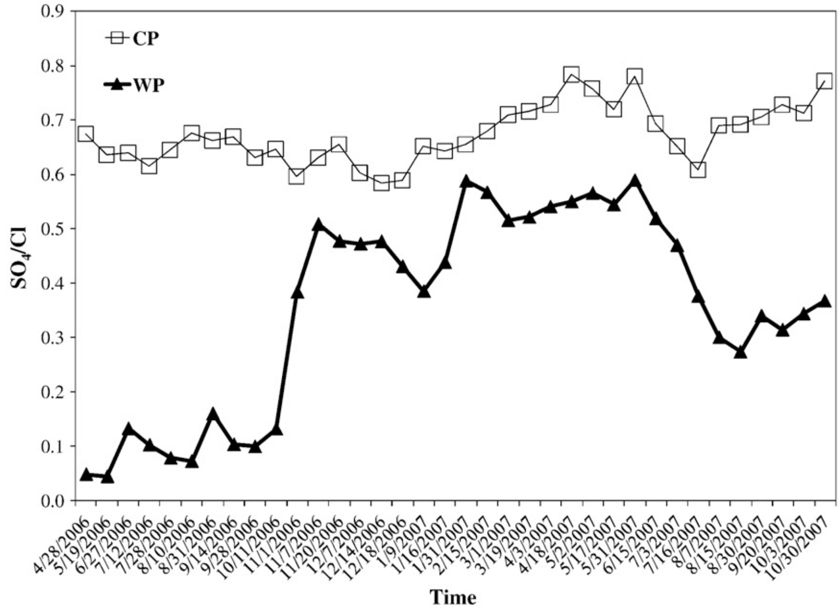

Fig. 10. Variation of SO_4/Cl in the cooling pond pump (CP) and the wetland pump (WP) as a function of time. Note: Values are in mg/L.

favorable for the stability of pyrite (e.g., Jones and Pichler, 2007). Water of this composition would be physically and chemically similar to native groundwater and should not cause the dissolution of pyrite and leaching of As during ASR.

The pH, DO and temperature gradient from the CP to the wetland could be important for the behavior of heavy metals such the redox-sensitive element As (Stottmeister et al., 2006). Concentrations of As in the CP (up to 5 µg/L) were considerably reduced at the WP and the FBS/FBN (<2 µg/L) (Table 1). The mass loading of As in the CP and the wetland was calculated to understand the removal effectiveness of the wetland over time (Fig. 12). The calculation was done with the rainfall dilution factor over the wetland study area. Generally, As in the wetland was reduced to 40–95% except on 07/03/08 it was 2 times higher than in the CP, which could be due to As-containing herbicides and insecticides used at the Hines Energy Complex. Seasonal variations of As in the treatment system showed lower concentrations during summer and fall and higher during winter and early spring. In the summer–fall seasons, when plants and algae grow they uptake and immobilize nutrients such as phosphorous and nitrogen into their biomass. Because of the high affinity between arsenate [As(V)] and phosphate, plants easily incorporate As(V) into their cells (Catarecha et al., 2007). In contrast, the reduced photosynthesis and decay of plants during winter – early spring facilitates the decrease of DO in water and releases nutrients back to the water column such as P and As. During certain seasons, plants and other wetland species could convert inorganic nutrients to organic compounds resulting in a net export of nutrients from the wetland (Devito and Dillon, 1993).

In addition to plant accumulation, the sandy clay-based sediment rich in organic material could be an important sink for As retention. Buddhawong et al. (2005) constructed a bench-scale experimental wetland in Grosskaya–Beuna area (Germany) to simulate the

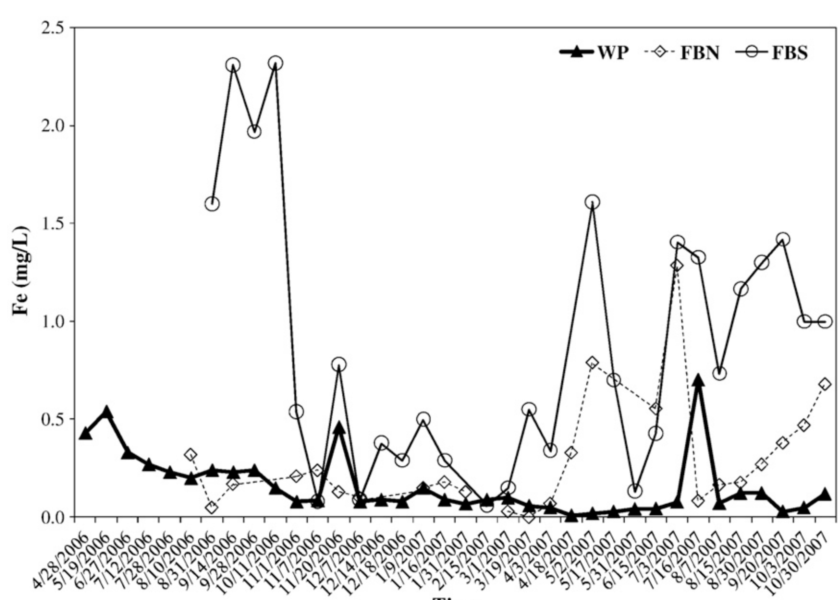

Fig. 11. Distribution of Fe at the WP, FBS and FBN with time. Note: WP — wetland pump; FBS and FBN — filter basin south and north pumps.

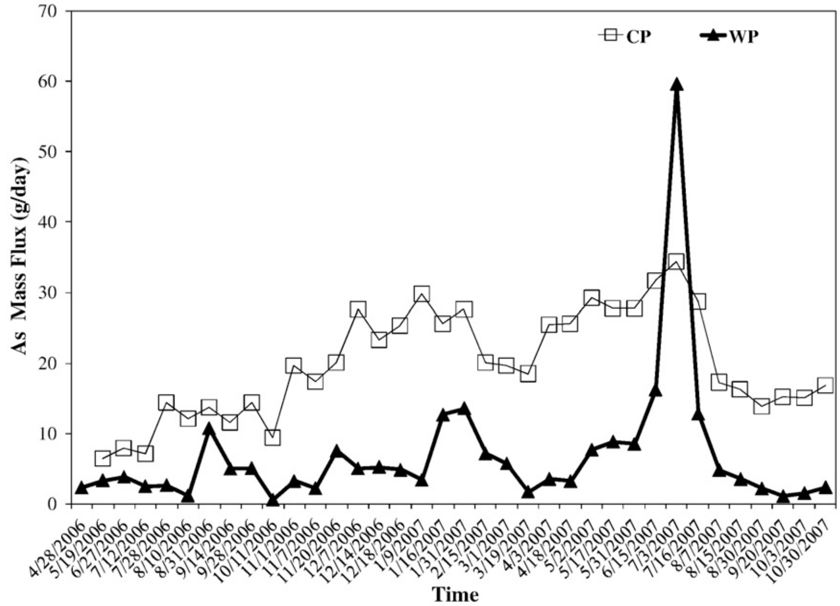

Fig. 12. Mass fluxes of As in the CP and WP. Note: CP — cooling pond pump; WP — wetland pump; Calculation included the rainfall dilution factor over the site area.

treatment of acid mine drainage. The authors reported that the highest efficiency of water treatment was achieved in the constructed wetland with the combined planted gravel/soil system. This type of wetland maintained the essential conditions (i.e., pH, redox, surface area) for As binding rather than those wetlands which were constructed exclusively using vegetation or soil matrix (Stottmeister et al., 2006). At the same time, As can be co-precipitated with iron oxides caused by root oxygen transport into rhizosphere. This transfer stimulates a significant metal buildup at the sediment–root boundary forming iron plaques on the plants roots (Colmer, 2003). In addition, elevated concentrations of S^{2} in the constructed wetland could develop favorable conditions for coagulation or coprecipitation of As with S^{2} . Langner et al. (1999) suggested that the formation of As(III)–S^{2} phases could be an important sink of As(III) under reduced conditions such as wetlands. Also, the authors reported rapid reduction of As(V) and SO_4^{2} to As(III) and S^{2} using controlled wetland chambers. Additional contributions of As(V) and SO_4^{2} could cause the formation of an amorphous As_2S_3 or Fe–AsS phases.

5. Conclusions

1 Evaluation of the wetland performance during dry and rainy seasons was important to assess the reliability of the treatment system in time and space. The wetland transect showed a distinct pattern of change in water chemistry along the flow path from the input – cooling pond (CP) to output – wetland water from pump (WP).

2) Chemical composition of monitor wells (MWs) was different from the CP and WP, but less different to sites N-15 and SA-8. Concentration of constituents in MWs was always higher or lower compared to the above, indicating little to no leakage from the sites N-15 and SA-8 into the wetland treatment system.

3) The mass fluxes of Na and Cl were used to evaluate groundwater input into the wetland and showed a comparable distribution as well as a significant change throughout the duration of the study. During the first rainy season the water quality was impacted by two hurricanes and inconsistent pumping operations due to maintenance or power issues. Once pumping operations stabilized and without the influence of hurricanes, the mass fluxes into and out of the wetland were relatively constant.

4) The study showed the following changes in water quality from the cooling pond (CP) to the constructed wetland/filter basin treatment system (CW/FB):

    1. Substantial decrease of water temperature (up to 10 °C);
    2. Significant change in pH from about 9 to 6.5–7;
    3. Negative ORP confirming the reducing conditions of the treatment system;
    4. Substantial increase of H_2S (up to 1060 µg/L).
    5. Reduction of As from up to 5 µg/L to <2 µg/L (mostly <0.5). Seasonal variations of As in the wetland showed lower concentrations during summer–fall and higher during winter–early spring.
    6. Substantial reduction of SO_4, F, Cl, NO_3, NO_2, Br, Na, K, Ca, and Mg.
    7. Reduction of fecal and total coliform at the FBS/FBN from 30–730 and 1000–7000 count/100 mL to <2 and <100 count/100 mL, respectively. These results clearly demonstrate a crucial role of a biofilm “schmutzdecke”.

Overall, the performance of the wetland/filter basin treatment system showed a great potential to improve the water quality of industrial and municipal wastewater. Despite of significant seasonal variations with respect to temperature, rainfall and humidity, the chemical/microbiological composition of treated water remained relatively constant.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemgeo.2009.06.006.

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