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Relationships of surface water, pore water, and sediment chemistry in wetlands adjacent to Great Salt Lake, Utah, and potential impacts on plant community health

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HIGHLIGHTS

- ► We characterize surface water, pore water, and sediment chemistry in GSL wetlands.
- ▶ We compare chemistry from each compartment with plant community health metrics.
- ▶ Plant health is inversely correlated with trace elements (sed) and nutrients (sw).
- ► Remediation efforts should consider pore water and sediment chemistry.

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ABSTRACT

We collected surface water, pore water, and sediment samples at five impounded wetlands adjacent to Great Salt Lake, Utah, during 2010 and 2011 in order to characterize pond chemistry and to compare chemistry with plant community health metrics. We also collected pore water and sediment samples along multiple transects at two sheet flow wetlands during 2011 to investigate a potential link between wetland chemistry and encroachment of invasive emergent plant species. Samples were analyzed for a suite of trace and major elements, nutrients, and relevant field parameters. The extensive sampling campaign provides a broad assessment of Great Salt Lake wetlands, including a range of conditions from reference to highly degraded. We used nonmetric multidimensional scaling (NMS) to characterize the wetland sites based on the multiple parameters measured in surface water, pore water, and sediment. NMS results showed that the impounded wetlands fall along a gradient of high salinity/low trace element concentrations to low salinity/high trace element concentrations, whereas the sheet flow wetlands have both elevated salinity and high trace element concentrations, reflecting either different sources of element loading or different biogeochemical/hydrological processes operating within the wetlands. Other geochemical distinctions were found among the wetlands, including Fe-reducing conditions at two sites and sulfate-reducing conditions at the remaining sites. Plant community health metrics in the impounded wetlands showed negative correlations with specific metal concentrations in sediment (THg, Cu, Zn, Cd, Sb, Pb, Ag, Tl), and negative correlations with nutrient concentrations in surface water (nitrite, phosphate, nitrate). In the sheet flow wetlands, invasive plant species were inversely correlated with pore water salinity. These results indicate that sediment and pore water chemistry play an important role in wetland plant community health, and that monitoring and remediation efforts should consider pore water and sediment chemistry in addition to surface water chemistry.

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1. Introduction

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A number of studies investigating wetland chemistry have primarily focused on surface water quality and the removal of nutrients and

uality and the removal of nutrients and their roots (Weis and Weis, 20 pore water may also be import

0048-9697/\$ – see front matter © 2012 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.scitotenv.2012.11.063 contaminants from surface water systems without considering the role of sediment and pore water (e.g. Goulet and Pick, 2001; Kadlec et al., 2010; Kropfelova et al., 2009; Wood and Rubec, 1989). Given that many wetland plants primarily uptake and store toxic metals in their roots (Weis and Weis, 2004), the geochemistry of sediment and pore water may also be important towards understanding impacts to wetland biota, including submerged aquatic vegetation (SAV) and emergent plants. These three compartments (surface water, pore water, and sediment) are interdependent and connected by a variety

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of physical, hydrological, biological, and chemical processes. For example, trace element and nutrient fluxes from pore water to surface water have been shown to be an important process in several studies (Beck et al., 2008; Covelli et al., 2008; Gill et al., 1999; McCaffrey et al., 1980). Other studies have shown that sediments are a source of trace elements and nutrients to overlying water due to diffusion, dissolution, or bioturbation (Benoit et al., 2009; Delongchamp et al., 2010).

The need to consider pore water and sediment chemistry, in addition to surface water, is amplified by the fact that surface water chemistry is temporally variable, with diurnal, seasonal, and annual variations in concentrations of some elements being driven by biological, hydrological, and chemical processes. Relating plant community health to site chemistry requires determining which compartment (surface water, pore water, sediment) is most relevant, but only a handful of studies have comprehensively examined the three compartments in wetlands, and a majority have focused on only a few elements in a single wetland pond or estuary (Beck et al., 2008; Benoit et al., 2009; Covelli et al., 2008; Delongchamp et al., 2010; Gill et al., 1999; McCaffrey et al., 1980).

Most previous characterizations for contaminants in freshwater wetlands of Great Salt Lake (GSL), Utah, have focused on surface water chemistry, with lesser attention given to pore water and sediment chemistry (Carling et al., 2011; Dicataldo et al., 2011; Naftz et al., 2011). GSL wetlands are critical habitat for millions of migratory and resident waterfowl and shorebirds and receive nutrient-rich treated wastewater effluent and other runoff from a rapidly increasing urban population of >2 million people. The system includes both man-made impounded wetlands and free-flowing sheet flow wetlands. Due to recently observed degradation of wetland plant communities, including early senescence of SAV (Hoven and Miller, 2009), the Utah Department of Environmental Quality-Division of Water Quality (DWQ) is developing wetland assessment criteria that will include an evaluation of the adverse effects due to elevated nutrient concentrations in the surface water of GSL wetlands. The criteria could ultimately lead to further limitations on nutrient effluent from waste water treatment plants (WWTPs), for which compliance costs are estimated at up to \$1,000,000,000 (J. Ostermiller, personal communication). A potential metric for DWQ's assessment criteria is the percent coverage of filamentous algae (Cladophora glomerata) or duck weed (Lemna minor) surface mats. The rationale is that excessive water column nutrients contribute to surface mat growth on some of the ponds, which in turn are thought to negatively impact SAV health due to shading or other water quality alterations. SAV drupelets and tubers are a primary food source for millions of migratory birds that utilize the GSL wetlands each year, thus unhealthy SAV communities could result in diminished food resources. Excessive nutrients are also postulated to promote the spread of emergent invasive species in Great Salt Lake wetlands, particularly phragmites (Phragmites australis), leading to further degradation of bird habitat. Given the expense associated with reduction of nutrient concentrations in WWTP effluent, it is important to understand the relative nutrient contributions to surface water from sediment/pore water versus tributary/WWTP inputs. Furthermore, it is necessary to understand the ecological effects of legacy nutrients and contaminants that have accumulated in these wetlands over the past century. It is possible that trace elements and nutrients in sediment and pore water exert equal or greater influences on the system relative to surface water loads from the WWTPs.

The purpose of this study is to characterize trace element and nutrient concentrations in surface water, pore water, and sediment across the GSL wetlands to determine which compartments represent wetland chemistry in a way that is relevant to understanding early SAV senescence in impounded wetlands, as well as to understand factors driving encroachment of invasive emergent vegetation in sheet flow wetlands. Specific objectives include: 1) determine which characteristics define wetland chemistry in the three primary compartments: surface water, pore water, and sediment; 2) examine spatial and temporal variability in surface water, pore water, and sediment chemistry; 3) compare the geochemistry of wetlands influenced by different hydrologic regimes (impounded versus sheet flow wetlands); and 4) compare chemical parameters measured in surface water, pore water, and sediment with plant health metrics. We hypothesize that chemical signatures at each wetland site will be defined by proximity to highly-saline GSL environment, nutrient-laden WWTP effluent/urban runoff, and legacy/contemporary sources of trace element-rich runoff, and therefore within-site variability will be less important than across-site variability. We also hypothesize that the state of a site from year-to-year, including the accumulation of legacy contaminants, will be reflected more accurately by pore water and sediment chemistry than surface water chemistry, and thus plant health metrics will align more closely with sediment and pore water. The wetlands on the eastern fringe of GSL were selected as a study area because they include impounded wetlands and sheet flow wetlands that span the range from fresh to brackish water that have both healthy and unhealthy SAV and emergent plant communities and because of their importance to wetland bird communities. Whereas development and refinement of chemical data are described in detail in this paper, the development and refinement of plant health data are described in another manuscript (Hoven et al., in preparation).

2. Methods

2.1. Sample sites

Surface water, pore water, and sediment samples were collected during 2010 and 2011 at the following impounded wetlands: Ambassador Duck Club pond W-1 (AM), New State Duck Club pond 47 (NS), Farmington Bay Wildlife Management Area Units 1 and 2 (FB1 and FB2), and Public Shooting Grounds Pintail pond (PN) (Fig. 1). Additional pore water and sediment samples were collected during 2011 at the Central Davis outfall (CD) and Kays Creek (KC) sheet flow wetlands (Fig. 1). Surface water was not collected at the CD or KC sheet flow sites because in most cases the water was very shallow (a few cm depth). The set of sites includes subsets that represent a range of: a) groundwater- versus surface water-fed; b) contributions from metropolitan or industrial effluents; and c) periodic flooding by GSL saline water versus no direct GSL influence.

The wetlands selected for this study reflect the range in nutrient and salinity gradients found in GSL wetlands. The AM, NS, FB1 and FB2 impounded wetlands are primarily fed by nutrient-rich water from the Jordan River, which receives treated sewage effluent and urban runoff as it flows through the metropolitan Salt Lake Valley, whereas the PN pond is primarily groundwater-fed. The CD sheet flow wetland receives water from Central Davis WWTP discharge, whereas KC is fed by a stream that follows a short reach from the Wasatch Mountains, through several km of urban development, to GSL. The impounded wetlands and sheet flow wetlands also represent the range of hydrologic and biologic conditions found in GSL wetlands. The surface water at the impounded wetlands is deeper (0.5 to 1.5 m) and has longer residence times (up to several days) relative to the shallow (<0.2 m) flowing surface water at the sheet flow wetlands. The impounded wetlands are managed to remain flooded throughout much of the year (depending on water supply), whereas some of the sheet flow wetlands tend to go dry during summer. The plant communities reflect the hydrology of each wetland type: SAV (Ruppia cirrhosa and Stuckenia sp.) and emergent plants (Schoenoplectus maritimus) are the most common native vegetation at impounded and sheet flow wetlands, respectively. However, in spite of different hydrologic and biologic conditions, the impounded and sheet flow wetlands are geochemically similar in that when flooded, both tend to display anaerobic conditions in bottom sediment.

2.2. Sample collection

The intensive sampling campaign resulted in a large data set that included measurements of trace elements, major elements, nutrients, and field parameters on 39 surface water samples, 45 sediment

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Fig. 1. Map of Great Salt Lake (Utah) showing the location of wetland sampling sites. Sheet flow wetlands and impounded wetlands are labeled with open and closed circles, respectively. Exact locations are provided on the associated KML file.

samples, and 90 pore water samples from impounded and sheet flow wetlands of GSL. Sampling at the impounded wetlands was conducted monthly from June through September 2010, and again during August and September 2011. Whereas pore water samples were collected on all occasions, surface water samples were not collected during August 2011 and sediment samples were not collected during June or July 2010. The five ponds were sampled during each month over a 2–3 day period by multiple teams of workers. Each team was given specific instructions and training on sampling procedures. Sampling at the sheet flow wetlands was conducted over a one week period between 28 June and 5 July 2011.

2.3. Surface water, pore water, and sediment sampling methods

Surface water was collected at or near the outlet of each impounded wetland to obtain a representative sample. During June, July, and August 2010, samples were collected during early morning (pre-dawn) and afternoon to assess diel variability in water chemistry. Samples for total and methyl mercury (THg and MeHg) were collected in 250 mL FLPE bottles (pre-cleaned by triple rinsing with Milli-Q water) and acidified to 1% v/v HCl. Samples for trace elements/major cations were collected in 30 mL LDPE bottles (pre-cleaned by soaking in 10% v/v HCl at 60 °C followed by triple rinsing with Milli-Q water and drying in a laminar flow hood) and acidified to 2.4% v/v HNO₃. Samples for major anions were collected in serially rinsed HDPE bottles, and sulfide, ammonia, and nitrite were measured immediately upon collection using a CHEMetrics spectrophotometer and ampoules.

"Clean hands, dirty hands" protocols (USEPA, 1996) were used during all steps of the sampling process, and new vinyl gloves were used for each set of water samples. The samples were filtered by collecting water in a 60 mL polyethylene syringe and forcing it through a 0.45 µm PES syringe filter. New filtering equipment was used for each sample set. Syringes were pre-cleaned by filling with 10% v/v HCl, placing in an oven at 65 °C for 2-3 days, triple rinsing with Milli-Q water, and drying in a laminar flow hood, and syringe filters were cleaned by forcing 50 mL 10% v/v HCl through the filter membrane followed by rinsing with 150 mL Milli-Q water. Field blanks for all constituents were processed by each team to determine potential contamination in the sampling process. Other surface water measurements included dissolved oxygen (DO), conductivity, pH, temperature (T), dissolved organic carbon (DOC), and alkalinity. Field parameters were measured using YSI Professional Series Quatro probes. The probes were calibrated for conductivity, pH, and DO prior to each sampling day. DOC samples were filtered within 12 h in the laboratory using a glass filtration apparatus and glass fiber filters (Advantec Grade GF75). Alkalinity samples were collected in 125 mL HDPE bottles leaving no head space.

Sediment cores were collected in acrylic tubes (5 cm diameter) that contained pre-drilled ports to allow for pore water sampling. Cores were collected at least 30 m from the shore at each pond, but from a different location each time; thus samples represent potential spatial and temporal variability within each pond. Pore water was collected by mechanically squeezing the sediment using pistons inserted on either end of the acrylic tube, as described in Chin et al. (1998). Sediment was squeezed on-shore out of direct sunlight immediately after the core was collected; pore water was filtered using a 5 cm long Porex rod (30–70 µm pore size) inserted through sample ports (spaced vertically 2–3 cm apart) and connected to a 0.45 µm PES syringe filter by a luer adapter, and water was collected at each port in a 30 mL plastic syringe. Care was taken to avoid collection of oxidized pore water; the

syringes were not attached to the syringe filter until after the first few drops of water had already passed through the saturated filter. All materials that came in direct contact with the pore water were acid cleaned prior to use. Porex rods and luer adapters were cleaned by soaking in 10% v/v HCl at 65 °C for 2-3 days, rinsing with Milli-Q water, and drying in a laminar flow hood. Syringes and syringe filters were cleaned as described above for surface water sampling. During the 2010 sampling, pore water from 0 to 6 cm ("top") and from 6 to 14 cm ("bottom"), respectively, below the sediment-water interface were composited from the corresponding syringes and subdivided in separate containers for analyses of THg and MeHg (50-100 mL), trace elements/major cations (10 mL), and sulfide (10-25 mL). During 2011, all pore water from 0 to 14 cm was composited in order to obtain sufficient volume for the above samples plus additional analyses, including DOC (10-20 mL), major anions (12 mL), ammonia (10-25 mL), alkalinity and field parameters (40 mL), and nitrite (10-25 mL; in a limited number of cores).

Samples for THg and MeHg analyses were collected in 125 mL FLPE bottles (pre-cleaned by triple rinsing with Milli-Q water and drying in a laminar flow hood) and immediately acidified to 1% v/v HCl. Samples for trace element/major cation analyses were collected in 15 mL LDPE centrifuge tubes (pre-cleaned by soaking in 10% v/v HCl at 65 $^\circ\text{C}$ followed by triple rinsing with Milli-Q water and drying in a laminar flow hood) and immediately acidified to 2.4% v/v HNO₃. Samples for major anions were collected in 12 mL HDPE bottles (leaving no head space), and samples for DOC were collected in 30 mL amber glass bottles. A field blank was collected for the above measurements to assess potential contamination from the filters, syringes, and sample bottles. Samples for alkalinity were collected in 40 mL HDPE bottles (leaving no head space) using the same aliquot that had previously been used to measure field parameters with the multiparameter probe since volume limitations did not allow for separate aliquots for each measurement, and alkalinity was the least likely of all measured parameters to be contaminated by the probe sensors. Sulfide, ammonia, and nitrite were analyzed immediately using a CHEMetrics field spectrophotometer and ampoules.

Sediment samples (top ~10 cm) were collected by vertically inserting a 50 mL polyethylene centrifuge tube into the sediment core after squeezing was completed. Sediment samples were capped, sealed with electrical tape, and stored frozen until subsectioning for analyses of THg (1 g), MeHg (1 g), trace elements (1 g), and dry weight/total volatile solids (TVS) (~50 g).

2.4. Sample analyses

Surface water and pore water samples for THg and MeHg were analyzed using a Brooks Rand Model III CVAFS according to EPA Methods 1631e (USEPA, 2002) and 1630 (USEPA, 2001a), respectively. Acidified samples were analyzed within one to six months of collection, which is an acceptable holding time for THg and MeHg (Parker and Bloom, 2005). THg samples were analyzed after a minimum 24 h in-bottle BrCl oxidation, and MeHg was analyzed after distillation with APDC. At a minimum, matrix spike recoveries and replicates were analyzed for every 10 samples. For the sample run to be accepted, matrix spike recoveries had to fall within 75-125% of the original sample run and replicate analyses had to fall within \pm 10%. Method blanks were analyzed at the beginning of each run in order to calculate a daily detection limit (D.L.). The accepted value for D.L. is 0.4 ng/L and 0.02 ng/L for THg and MeHg, respectively, although lower detection limits are often achieved in our laboratory. In order to test accuracy of laboratory results, USGS standard reference samples (SRS) for THg were analyzed during each sample run, and results within ~10% of the accepted value were consistently obtained. Currently no SRS exist for MeHg. However, intercomparison tests with over 40 other laboratories (in a round-robin conducted by Brooks Rand Labs) showed that results from our lab fell within $\pm 20\%$ of the most probable value.

Pore water and surface water samples for trace element/major cations were analyzed using an Agilent 7500ce quadrupole inductively coupled plasma mass spectrometer (ICP-MS) with a collision cell, a double-pass spray chamber with perfluoroalkoxy fluorocarbon (PFA) nebulizer (0.1 mL/min), a quartz torch, and platinum cones. Acidified samples were analyzed within one to six months of collection, which is an acceptable holding time for trace elements (USEPA, 1994), although most of the samples were run within three months of collection. Regardless of the amount of time between sample collection and analysis, data clustered according to site (as discussed in Section 3), not according to holding time. Concentrations were measured for the following elements: Ag, Al, As, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, Sr, Ti, Tl, U, V, and Zn. A calibration solution containing all the elements reported was prepared gravimetrically using 1000 mg/L single-element standards (Inorganic Ventures, Inc.). This solution was used to prepare a calibration curve with six points plus a blank for each sample run. Ca, Cr, Fe, K, Mn, Na, and V were determined using 4 mL He/min in the collision cell, and As and Se were determined using 4 mL He/min plus 2.5 mL H₂/min. D.L. was determined as three times the standard deviation of the 23 blanks analyzed throughout the run. A USGS standard reference sample (T-205) was analyzed multiple times in each run together with the samples as a continuing calibration verification. The long term reproducibility for T-205 and differences relative to the accepted values obtained using our method indicate that the elemental concentrations reported here are accurate within 10% for most elements.

Major anion samples for pore water and surface water were analyzed within two weeks of collection by ion chromatography (IC), and alkalinity samples were analyzed on the day of collection by titration. DOC samples were analyzed within one week of collection using a Shimadzu TOC-5000A.

Sediment samples for THg were analyzed according to EPA method 1631-appendix (USEPA, 2001b). Briefly, the 1 g sediment subsample was digested in a 7:3 HNO₃/H₂SO₄ solution, and the digestate was diluted in a 1% BrCl solution to destroy any remaining material. The sample was then analyzed as described above for surface water and pore water samples. MeHg was extracted from sediment following methods described in Bloom et al. (1997). Briefly, MeHg was leached from the sediment via addition of 5 mL of 18% (w/v) KBr + 5% (v/v) H₂SO₄ and 1 mL 1 M CuSO₄ to the 1 g sediment subsamples, followed by methylene chloride extraction of MeHg from the leachate. A 2 mL aliquot of the methylene chloride was pipetted to a Teflon tube containing 50 mL Milli-Q water. The methylene chloride was evaporated at 70 °C, leaving MeHg in a pure water solution that was analyzed as described above for surface water and pore water samples.

Trace elements were measured on sediments by leaching the 1 g sediment subsample in 20 mL 5% (v/v) trace metal grade HCl for three days at room temperature, followed by centrifugation and analysis of supernatant as a water sample via ICP-MS (described above). This was not a total digestion of the sediment, but rather a weak acid leach meant to recover the bioavailable trace element fraction (i.e. available for plant uptake). Sediment dry weight and total volatile solids (TVS) were determined according to EPA Method 1684 (USEPA, 2001c). Briefly, sediment was dried at 105 °C for a minimum of 24 h, followed by ignition at 550 °C for 2 h (TVS was calculated as % loss on ignition).

2.5. Data quality control

Field blanks for pore water and surface water showed low levels of contamination for all measured elements except for Zn, which had high concentrations in the field blanks. Thus Zn was removed from the surface water and pore water data sets for further analysis. Be, Ag, Tl, Cd, and Mo were removed because concentrations in pore water and surface water samples were either below D.L. or similar (within a factor of 2) for the majority of samples. For other elements, values < D.L. were set as 1/2 D.L. for all further analysis.

Major ion charge balances were calculated for surface water and pore water samples using ICP-MS cation data (Na⁺, K⁺, Ca²⁺, Mg²⁺), IC anion data (F⁻, Cl⁻, HPO₄⁻, NO₃⁻, SO₄²⁻), and alkalinity (HCO₃⁻). Surface water samples and a majority of pore water samples showed acceptable charge balances errors of \pm 5%. However, a number of pore water samples showed charge balance errors of \pm 15%, likely because subsamples for anions, cations, and alkalinity were collected at different depths in each sediment core in order to obtain sufficient volume for the measurements.

2.6. Data ordination (NMS)

Ordination was used to explore multivariate relationships of surface water, pore water, and sediment data. For exploratory, visual analyses, ordination techniques are often superior for explaining relationships of assemblages and communities relative to hypothesis testing approaches (McCune and Grace, 2002). Basically, ordination is the ordering of objects along axes according to their similarities. The main objective of ordination is data reduction and expressing many-dimensional relationships into a small number of easily interpretable dimensions (axes on a plot). The strongest correlation structure in the data is extracted and is then used to position objects in ordination space. Objects that are close in the ordination space are generally more similar than objects distant in the ordination space (McCune and Mefford, 1999).

Several types of ordination exist; non-metric multidimensional scaling (NMS) was used for these data sets. NMS has been shown to be robust for ordination of species composition and is often more useful than other ordination techniques because, among other things, it avoids the assumption of normal distributions and linear relationships among variables (Peck, 2010). NMS is now the most widely accepted ordination technique used in ecology community (Peck, 2010).

The computer program PC-ORD (McCune and Mefford, 1999) was used for the NMS ordination. The data were log generalized prior to conducting NMS using the equation: $b = log(x + x_{min}) - log(x_{min})$, where x_{min} is the smallest positive value in the data set (column). We used Euclidean distance measures and ran the analyses 250 times with the real data and compared these results with 250 randomized data Monte Carlo simulations. We chose final models that had stable solutions and provided the greatest reduction in the amount of stress. Stress is a type of goodness of fit measure; it maximizes the rank correlation between the calculated dissimilarities/distances and the plotted distances in the model (Mather, 1976). We then conducted a post hoc analysis of coefficients of determination for the correlations between ordination distances and distances in the original n-dimensional space to estimate the amount of variability in the data explained by each of the ordination axes. Because ordination results are best visually interpreted, we provide our data as graphs of axes with each wetland group of data contained by convex hulls (i.e. the outer points in the group are connected by a closed polygon). Details defining labels and raw data associated with each sample shown in NMS plots are provided in the Supporting information.

2.7. Geochemical modeling

Aqueous speciation and saturation indices (SI) were calculated with PHREEQC using the Minteq.v4 thermodynamic database (Parkhurst and Appelo, 1999). Positive SI values, calculated as log (IAP/K_{sp}), indicate oversaturation and thermodynamic potential for mineral precipitation. PHREEQC uses ion-association and Debye Hückel expressions to account for the non-ideality of aqueous solutions. Distribution of elements among their valence states was estimated based on a specified range of pH and pe values. Average surface water and pore water concentrations of major elements, trace elements, and nutrients were used in the simulations. Redox condition (pe) was calculated using the nitrite/nitrate redox couple for surface water, and either the

sulfide/sulfate or Fe^{2+}/Fe^{3+} redox couples for pore water (depending on the predominant condition at the site).

3. Results

3.1. NMS results for surface water chemistry of impounded wetlands

The surface water data (Fig. 2) were best described using a two dimensional NMS model (final stress = 0.09), where axis 1 explains 82.1% of the variability and axis 2 explains an additional 9.5%. The individual samples from a given site were grouped in polygons referred to as "convex hulls"; the data associated with each sample are provided in the Supporting information. For surface water chemistry, which was analyzed only for the impounded wetlands, the sites fell along a spectrum from high salinity/low trace element concentrations (PN) to low salinity/high trace element concentrations (AM and NS). The NS and AM samples plot together on the positive end of axis 1 (with overlapping convex hulls), defined by elevated trace element concentrations (Se, As, Ni, Sb, Mn, and U). The PN samples plot at the negative end of axis 1, defined by elevated salinity (Na, Cl, K, and Li). Notably, the nutrients phosphate, nitrate, and nitrite were elevated at AM and NS, whereas DOC was elevated at PN. FB1 and FB2 group together between PN and AM/NS, reflecting moderate concentrations of trace elements, major elements, and nutrients. Axis 2 shows relationships of the sites based primarily on differences in Sb, V, U, THg, and Pb concentrations; these differences reflect within-site rather than across-site variability.

3.2. NMS results for pore water chemistry of impounded wetlands and sheet flow wetlands

The pore water data (Fig. 3), including both impounded wetlands and sheet flow wetlands, were also described using a two dimensional NMS model (final stress = 0.12), where axis 1 explains 70.0% of the variability and axis 2 explains an additional 23.7%. The impounded wetlands were distinguished along axis 2 on the basis of elevated Fe and Mn at NS, and elevated K and sulfide at PN, with intermediate concentrations of these elements at FB1, FB2, and AM (although there was substantial overlap in the convex hulls for AM, FB1, FB2, and PN). This is similar to what was shown in the surface water NMS plot (Fig. 2). The sheet flow sites were also primarily distinguished from one another along axis 2, with elevated Fe and Mn concentrations at KC1 and elevated K and sulfide concentrations at KC3 and CD1-2-3. The impounded wetlands and sheet flow wetlands plot on opposite ends of axis 1, demonstrating that pore water from the sheet flow wetlands contains both elevated trace element concentrations (U, As, MeHg, Ni, THg, Se, and Cu) and elevated salinity (Mg, Na, K, and Li) relative to the impounded wetlands. Considering only pore water collected during 2011 (Supporting information), which included additional nutrients not analyzed during 2010 (i.e. nitrate, phosphate, sulfate, and ammonia), similar groupings were found: the impounded wetlands plot separately from the sheet flow wetlands, but the distinction between high salinity/ low trace elements (i.e. PN) and low salinity/high trace elements (i.e. NS) is more clearly defined for the impounded wetlands.

3.3. NMS results for sediment chemistry of impounded wetlands and sheet flow wetlands

The sediment leach data (Fig. 4) were described using a three dimensional NMS model (final stress = 0.03), where axis 1 explains 78.4% of the variability, axis 2 explains 12.8%, and axis 3 explains an additional 8.0%. Similar to results for surface water (Fig. 2) and pore water (Fig. 3), sediments in the impounded wetlands were distinguished along a spectrum of high salinity/low trace elements (PN) to low salinity/high trace elements (NS), with intermediate concentrations of these elements at FB1, FB2, and AM. As was the case for pore

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Fig. 2. Surface water NMS results for 2010 and 2011 data from the impounded wetlands. Axis 1 explains 82.1% of the variability and axis 2 explains an additional 9.5%. Final stress = 0.09. The r values for factors describing each axis are plotted on the horizontal line (axis 1) and vertical line (axis 2).

water (Fig. 3), sediments in the sheet flow wetlands were distinguished by low salinity/high trace elements at KC1, and high salinity/high trace elements at KC3 and CD1-2-3. Axis 3 (Supporting information) shows that sediments in KC1 are also unique from the other sites based on elevated MeHg concentrations and low Sr and Ca concentrations.

3.4. Relationships between plant health metrics with surface water, pore water, and sediment chemistry

The controls on SAV health in the impounded wetlands, including *R. cirrhosa* and *Stuckenia sp.*, are the subject of a separate manuscript (Hoven et al., in preparation). Here, we summarize some of the correlations of surface water, pore water, and sediment chemistry with plant community health metrics. As an example of the impact sediment and pore water chemistry apparently have on SAV health, we found very strong inverse correlations between SAV drupelet mass and sediment trace element concentrations (Fig. 5a). Drupelets are the reproductive fruiting body of SAV, and thus drupelet biomass is indicative of overall plant health. Additionally, drupelets are an important source of fat and protein for many migratory waterfowl during the fall (Anderson and Low, 1976). Strong negative correlations ($R^2 > 0.7$; based on either linear or logarithmic fits) were found between drupelet mass and trace element concentrations in sediment, including THg, Cu, Zn, Cd, Sb, and Pb

(Fig. 5a), as well as Ag and Tl (Supporting information). Metals are known to be toxic to plants, since plant roots can create localized oxidizing conditions that mobilize metals from the sediment and make them available for uptake (Dunbabin et al., 1988; Pahlsson, 1989).

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Significant negative correlations were also found between SAV drupelet mass and nutrient concentrations in surface water, including nitrite, nitrate, and phosphate (Fig. 5b). Thus excessive nutrient concentrations in surface water may have a negative impact on plant health. However, the relationships between drupelets and surface water nutrient concentrations ($0.51 < R^2 < 0.66$) were not as strong as the relationships between drupelets and sediment trace element concentrations ($R^2 > 0.7$), and trace element concentrations in surface water were not correlated with drupelets.

None of the parameters measured in pore water were inversely correlated with plant health metrics, which may be surprising given the strong correlations with trace element concentrations in sediment. The trace elements that affect plant health (Pb, Cu, Cd, etc.) may not be bioavailable in pore water due to complexation with, for example, organic matter or sulfide. Complexation with DOC would be indicated by direct correlation between the dissolved *fraction* of elemental mass (ratio of filtered to non-filtered mass) to DOC. However, because only filtered pore water was collected, no such comparison could be made. Notably, DOC was positively correlated ($R^2 > 0.5$) with dissolved Ba,

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Fig. 3. Pore water NMS results for 2010 and 2011 data from the impounded wetlands and 2011 data from the sheet flow wetlands. Axis 1 explains 70.0% of the variability and axis 2 explains an additional 23.7%. Final stress = 0.12. The r values for factors describing each axis are plotted on the horizontal line (axis 1) and vertical line (axis 2).

Ca, Sc, Se, Sr, and Ni in pore water (Supporting information), but there was no correlation between DOC and other measured dissolved elements in pore water. With respect to complexation with sulfide, PHREEQC-calculated saturation indices indicated thermodynamic potential for precipitation of a number of sulfide-bearing minerals in pore water, including cinnabar (PbS) and covellite (CuS).

For the sheet flow wetlands, which generally do not contain SAV due to shallow surface water, comparisons were made between pore water and sediment chemistry with the fraction of invasive emergent vegetation (e.g. *Phragmites autstralis* and *Typha latifolia*). Invasive plant species are hypothesized to out-compete native vegetation after disturbance in Great Salt Lake wetlands; thus, locations with elevated nutrients and trace elements may also contain a greater fraction of invasive species. However, rather than showing a positive correlation between nutrient/ trace element concentrations and invasive species distribution, our data indicate that pore water salinity is the most important factor: site KC3 had the highest salinity (Cl, Li, sulfate, etc.) and the smallest fraction of invasive species (Fig. 6). These results demonstrate that further work is needed to investigate the relationships between salinity, nutrients, and trace elements with invasive species distribution.

3.5. Spatial and temporal relationships between surface water and pore water at the impounded wetlands

A number of elements showed significant (p<0.01) positive correlations between surface water and pore water at the impounded wetlands, including Ca concentrations relative to Sr, Ba, and Ni (Fig. 7a) and Na concentrations relative to Mg, K, and Li (Fig. 7b). This indicates that these elements are closely related between surface water and pore water. However, some of the trends show differences between PN and the other impounded wetlands. PN surface water and pore water show elevated Sr:Ca ratios relative to the Jordan River-fed impounded wetlands (Fig. 7a), likely reflecting equilibration of source water with different geologic substrates. PN surface water, but not pore water, shows different trends for Mg:Na and K:Na relative to surface water/pore water from all other ponds (Fig. 7b), indicating

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Fig. 4. Sediment NMS results for 2010 and 2011 data from the impounded wetlands and 2011 data from the sheet flow wetlands. Axis 1 explains 78.4% of the variability, axis 2 explains 12.8%, and axis 3 (shown in the Supporting information) explains an additional 8.0%. Final stress = 0.03. The r values for factors describing each axis are plotted on the horizontal line (axis 1) and vertical line (axis 2).

depletion in Mg and K with increasing Na (salinity). This may reflect differences in surface water inputs to PN or site-specific differences in geochemical processes affecting Mg and K concentrations in the surface water.

Surface water salinity increased during the summer months at all of the impounded wetlands (Fig. 8), in contrast to pore water, which showed no seasonal trend (Supporting information). Na and Li were used to represent salinity because they showed the strongest correlations of all the combinations of elements that were measured in both surface water and pore water. Na and Cl were also strongly correlated, but Cl was not measured on all the pore water samples. Na-Li data for all impounded wetlands are shown in a single plot in Fig. 7b, but to investigate temporal trends, Na-Li data from 2010 are separated by pond in Fig. 8. During 2010, surface water chemistry evolved from lower to higher salinity from early summer (June) to late summer/fall (July-August-September) (Fig. 8). Since pore water did not trend with time, pore water data are not differentiated by month in Fig. 8. Instead, the pore water data are differentiated by depth below the surface watersediment interface (0-6 cm and 6-14 cm). The differences in pore water chemistry likely reflect spatial variability (cores were collected from a different location within each pond during each month) rather than temporal trends. Notably, surface water chemistry evolves towards the higher salinity of the deeper pore water throughout the summer which could reflect either a) evaporation of surface water or b) increasing groundwater component relative to surface water inputs.

In addition to seasonal variability in surface water chemistry, diel variations were discernible for a subset of trace elements, including Se, Sb, V, and Mn, which is consistent with previous findings from these wetlands (Carling et al., 2011). However, these variations yielded <20% differences around the mean for all parameters except pH, DO, and T, and hence were not as significant as seasonal variations.

In contrast to the major elements (Na, K, Mg, Cl, etc.), which showed similar concentrations (within a factor of two) in surface water and pore water at each wetland, a subset of trace elements and nutrients showed large differences (factor of five or greater) between the two compartments. Mn, Al, Ti, DOC, sulfide, ammonia, alkalinity, and phosphate concentrations were consistently higher in pore water relative to surface water at all sites, whereas pH, Pb, U, Se, Sb, sulfate, and nitrate concentrations were higher in surface water (Fig. 9). Arsenic was elevated in surface water relative to pore water at all sites except NS, where concentrations were similar (Fig. 9).

3.6. Pore water chemistry of impounded wetlands versus sheet flow wetlands

Just as Na was significantly (p<0.01) correlated with K, Mg, and Li between pore water and surface water in the impounded wetlands, these elements were also correlated in pore water across both impounded and sheet flow wetlands (compare Figs. 7 and 10). These relationships indicate a similar source of salinity to pore water across all

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Fig. 5. Negative correlation of drupelets with trace element concentrations in sediment leachate (A) and nutrient concentrations in surface water (B). Data from each wetland are indicated by different shapes and shading. A logarithmic fit was used when warranted by a substantial increase in R² value relative to a linear fit.

sites and possibly reflect aqueous transport through evaporite sediments surrounding GSL. However, the Sr–Ca trends show that PN has a uniquely high Sr:Ca ratio relative to the other impounded wetlands (as described above) and relative to the sheet flow wetlands (Fig. 10). KC1 has a low Sr:Ca ratio relative to all other sites, likely indicating differences in flow paths for surface water and groundwater feeding each of these locations. Ni and Ca were also positively correlated across pore water of impounded wetlands and sheet flow wetlands (Fig. 10), as it was for surface water and pore water of impounded wetlands (Fig. 7), possibly reflecting dissolution of Ni-containing carbonate minerals. Interestingly, V and Al were strongly correlated in pore water, although the reasons for this correlation are not well understood. Sulfide and Fe



Fig. 6. Negative correlation of % invasive species with Na, Li, and sulfate concentrations in pore water from the sheet flow wetlands.



Fig. 7. Scatter plots of surface water and pore water data from the impounded wetlands. Closed circles and squares are surface water and pore water data, respectively, from AM, NS, FB1, and FB2, and open circles and squares are surface water and pore water data, respectively, from PN. Panels A and B show elements that are significantly (p<0.01) correlated with Ca and Na, respectively. In panel A, separate trendlines are shown for Ca–Sr data from PN and the other wetlands. The trendline for Ca–Ba data excludes two samples (circled) that had anomalously high Ba concentrations. In panel B, separate trendlines are shown for PN surface water data and for data from all other wetlands (for both surface water and pore water).



Fig. 8. Scatter plots of surface water and pore water Na and Li concentrations for samples collected during 2010, separated by wetland. A plot showing Na–Li data from all sites together is provided in Fig. 7. Surface water data are shown as circles, where the shading indicates the timing of sample collection. Pore water data are identified by depth, where squares represent shallow pore water (0–6 cm below the sediment-water interface) and crosses represent deeper pore water (6–14 cm). A separate plot showing pore water data differentiated by month is provided in the Supporting information.

concentrations were inversely related in pore water, with elevated Fe concentrations at KC1 and NS and elevated sulfide at the other sites (Fig. 10). KC1 and NS also had elevated Mn and Pb concentrations, respectively (Fig. 10).

4. Discussion

4.1. Relationships between surface water and pore water geochemistry at impounded wetlands

Concentration differences in surface water relative to pore water at the impounded wetlands for As, U, Se, Sb, and Pb (higher concentrations

in surface water), and Mn, Ti, and Al (higher concentrations in pore water) (Fig. 9) were driven by differences in pH and redox conditions. PHREEQC-predicted species for the anion-forming elements As, U, Se, and Sb favored the oxidized form in surface water: $HASO_4^{-2}$ (+V oxidation state), $UO_2(CO_3)_3^{4-}$ (+Vl oxidation state), $Sb(OH)_6^-$ (+V oxidation state), and SeO_4^{2-} (+Vl oxidation state). PHREEQC-predicted species favored reduced forms of these anions in the pore water: H_3ASO_3 (+III oxidation state), $U(OH)_5^-$ (+IV oxidation state), $Sb_2S_4^{2-}$ (+III oxidation state), and HSe⁻ (-II oxidation state). In contrast to the anion-forming elements, Mn and Ti were predicted as divalent cations in both surface water and pore water. Thus, the reducing conditions and circumneutral pH of pore water most likely inhibit sorption of the cations



Fig. 9. Average surface water and pore water concentrations of trace elements, nutrients, and pH measured at each wetland. Error bars indicate ± 1 standard deviation. Averages of all available data collected during 2010 and 2011 are shown. Pore water DOC, sulfide, ammonia, alkalinity, phosphate, sulfate, nitrate, and pH were only measured during 2011.

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Fig. 10. Scatter plots of pore water data from impounded wetlands (collected during 2010 and 2011) and sheet flow wetlands (collected during 2011). Closed blue squares represent pore water data from AM, FB1, FB2, and PN impounded ponds; open blue squares are data from NS. Closed red circles represent pore water data from CD1, CD2, CD3, and KC3 sheet flow wetland sites; open red circles are data from KC1. The top three panels show elements that are inversely related to sulfide. The middle three panels show elements that are positively correlated with Na. The bottom three panels show correlations of Ca with Sr and Ni, and correlation of Al with V. The regression line for Ca–Sr data excludes data from PN and KC1.

Mn and Ti to sediment—due to reductive dissolution of metal oxyhydroxide coatings (e.g. Grantham et al., 1997), as well as positively charged sorption sites (e.g. Fuller and Davis, 1989)—and result in elevated Mn and Ti concentrations in pore water relative to surface water. The same conditions that inhibit cation sorption enhance anion sorption, leading to relatively reduced concentrations of As, Se, Sb, and U in pore water (e.g. Carling et al., 2011). Depletion of Pb in pore water relative to surface water, and depletion of Al in surface water relative to pore water, are explained by precipitation of galena (PbS) and diaspore (AlOOH) in pore water and surface water, respectively, since PHREEQC predicted saturation indices >1 for these minerals in those two compartments.

For the nutrients, the differences between surface water and pore water concentrations are largely driven by redox conditions, especially the tradeoff of N and S species between oxic surface water (nitrate and sulfate, respectively) and anoxic pore water (ammonia and sulfide, respectively). Sulfide and ammonia are commonly found in higher concentrations in pore water relative to surface water (Frazier et al., 1996; Wang and Chapman, 1999). Elevated DOC and phosphate in pore water indicate that the sediment is a primary source for these compounds owing to equilibration of bottom-accumulated decomposing organic matter (e.g. O'Loughlin and Chin, 2004) and phosphorus (e.g. Sundby et al., 1993) between the water and sediment phases. Elevated alkalinity in pore water likely reflects a longer period of carbonate mineral dissolution (longer residence time) as well as greater solution:solid ratio (Hu and Burdige, 2007).

4.2. Pore water geochemistry at impounded wetlands and sheet flow wetlands

Elevated Fe concentrations in pore water at NS and KC1, and corresponding lack of sulfide, reflect Fe-reduction as the dominant anaerobic process at these sites (Chapelle et al., 2009) (Fig. 10). Mn concentrations were also very high at KC1, and slightly elevated at NS, suggesting that Mn reduction is also active (Fig. 10). In contrast, all other sites showed elevated sulfide concentrations and low Fe/Mn concentrations, indicating that sulfate-reduction is the dominant anaerobic process. Sulfide also showed a negative relationship with Pb, likely reflecting precipitation of lead sulfide complexes in the sulfide-rich pore water (Fig. 10). As described above, galena precipitation was thermodynamically favorable in pore water on the basis of positive PHREEQC-calculated SI.

The reasons why Fe-reduction predominates over sulfate reduction at NS and KC1 sites are not well understood since Fe concentrations in sediment are similar across all of the sites. Although the two sites did not show any striking similarities in sediment leach chemistry according to the 3-axis NMS model (Fig. 4), a 2-axis NMS model for sediment shows NS and KC1 plotting together (with overlapping convex hulls) due to low Na, Li, and Mg concentrations and elevated Cu, Cd, Pb, Tl, and Zn concentrations (Supporting information). Further work is needed to understand if this chemical signature contributes to inhibition of sulfate reduction or stimulation of iron reduction in the wetland sediment. For example, Chapelle and Lovley (1992) found that Fe(III)-reducing bacteria could exclude sulfate-reducing bacteria by maintaining low concentrations of dissolved hydrogen, formate, and acetate, leading to elevated dissolved iron concentrations in groundwater. We did not measure organic electron donors, so we can only speculate that such a condition exists at NS and KC1.

4.3. Sources of trace and major elements to impounded wetlands and sheet flow wetlands

The NMS results indicate that the impounded wetlands fall along a spectrum from high salinity/low trace element concentrations (defined by PN) to low salinity/high trace element concentrations (defined by NS), whereas the sheet flow wetlands contain both high salinity and high trace element concentrations. The elevated concentrations of anthropogenic-derived trace elements and nutrients in surface water, pore water, and sediment at AM and NS may reflect input from the Jordan River, which contains urban runoff and WWTP effluent. However, FB1 and FB2 are also fed by the Jordan River and contain only intermediate concentrations of nutrients and trace elements. The reasons for lower trace element and nutrient concentrations at FB1 and FB2 relative to NS and AM are not well understood, but indicate that upstream loading may not be the primary control on concentrations. These relationships also illustrate the complex biogeochemical and hydrological processes occurring in the wetlands: a number of factors may play a role in trace element and nutrient concentrations, including (but not limited to) pond size, water depth and residence time, or even plant communities within the ponds. For example, the high volume and short residence times of water flowing through FB1 and FB2 may flush nutrients and trace elements through these ponds. In contrast, organic matter (and associated nutrients and trace elements) may settle out in the NS and AM ponds due to longer residence times. Additionally, the different plant communities among these wetlands may sequester and recycle these elements between sediment and surface water at different rates. More research is needed to test these possibilities.

Low concentrations of trace elements at PN reflect lack of anthropogenic influence, whereas elevated salinity reflects the predominance of groundwater as the primary source of water. Elevated Sr:Ca ratios at PN show that the groundwater inputs to PN have a unique chemistry relative to groundwater feeding other ponds, and also indicate that groundwater is the primary source of water to PN, as has been concluded at other settings on the basis of these elemental ratios (Land et al., 2000). Further work is needed to quantify the relative amount of groundwater input relative to surface water runoff at PN and the other ponds.

Elevated salinity in the sheet flow wetlands reflects periodic flooding by hypersaline GSL water (multi-decadal frequency; USGS, 2012). Na, Mg, K, and Li concentrations in pore water are on average about five times higher at the sheet flow wetlands relative to the impounded wetlands. Trace element concentrations are also elevated in the sheet flow wetlands. For example, THg, MeHg, As, and U concentrations in pore water are on average an order of magnitude higher at the sheet flow wetlands relative to the impounded wetlands. Other element concentrations in pore water, including Al, V, Ni, Cu, and Se, are factors of two to five times higher at the sheet flow wetlands relative to the impounded wetlands. Elevated trace element concentrations may be the result of a number of factors, including differences in water sources, hydrology, geochemistry, and plant type in the sheet flow wetlands relative to the impounded wetlands. The sheet flow wetlands receive water directly WWTP effluent (CD) and urban runoff (KC), with no pre-treatment from upstream ponds. The high salinity of sheet flow wetland pore water may act to increase the solubility of trace elements, or the continual wetting/drying cycles in the sheet flow wetlands may oxidize metals in the sediment and increase their solubility. Differences in plant communities and plant distribution among the sheet flow wetlands and impounded wetlands may also play a role in trace element concentrations and speciation, especially for MeHg. Whereas SAV and floating surface mats are the predominant plant types in the impounded wetlands, the sheet flow wetlands are dominated by invasive emergent vegetation (e.g. P. australis). Windham-Myers et al. (2009) found that plants can enhance microbial activity in the rhizosphere by exuding organic carbon, which leads to higher MeHg concentrations in sediment. Thus the plant community and plant density can have a direct effect on MeHg concentrations, suggesting that the dense emergent vegetation at the sheet flow wetlands may be creating conditions favorable for MeHg production.

4.4. Which compartment best characterizes a site in a manner that is consistent over time?

The wetland sites were similarly grouped on the basis of surface water, pore water, and sediment chemistry, indicating that surface water chemistry characterizes the sites as a first-order approach. However, surface water chemistry was found to vary over time, with diel (albeit relatively minor), seasonal, and interannual variability in trace and major element concentrations (Fig. 8). In contrast, parameters measured in pore water and sediment did not show obvious temporal variability over the course of this study and thus are better indicators of the long-term condition of a site. Surface water chemistry cannot be characterized without considering potential sediment and pore water fluxes over time, especially for the elements that were found with much higher concentrations in pore water (e.g. Mn, Ti, sulfide, ammonia, and phosphate). Other characteristics of pore water, such prevalence of sulfate- versus iron-reducing conditions, may also have important implications on the health of SAV and emergent vegetation.

4.5. Will reduced nutrient loads to GSL wetlands help improve ecosystem health?

The negative correlations observed between plant health metrics and surface water nutrient concentrations and sediment trace element concentrations (Fig. 5) suggest that both nutrients and trace elements in the water column and the sediment potentially act as stressors to the system. While the relative importance of the water column versus sediment as stressors remains unclear, the observed relationships indicate that improvement of plant health may require far more than reduction of nutrients in effluents to surface water, and may play out over timescales far longer than are required to simply reduce effluent concentrations. Elevated phosphate and ammonia concentrations in pore water relative to surface water (Fig. 9) indicate that pore water and sediment may act as long-term sources of nutrients to the water column and illustrate the need for additional research to understand the relative contribution of WWTP discharges, urban runoff, and accumulated organic matter and nutrients in sediment. A key factor in this process is the degree of internal cycling within the wetland ponds. For example, Thiebaut and Muller (2003) reported that aquatic macrophytes can derive all of their N and P from sediments. Because fall senescence includes the complete die-off of above ground plant material, the nutrients accumulated in the biomass can return to the sediment or can be released back to the water column as decomposition proceeds (Davis et al., 2006). Therefore, the various sources of nutrients must

be quantified prior to making decisions of whether reductions in nutrient concentrations in WWTP effluent can be expected to improve wetland plant health. In addition, or alternatively, to surface water effluent reduction, direct restoration efforts within the ponds may include periodic drying of the sediments to facilitate oxidation of organic matter, sulfide and associated trace elements, and nutrients, which may serve to enhance their flushing from the system (e.g. Gambrell, 1994; Litaor et al., 2004). Preliminary studies are underway to assess whether this treatment may lead to improved plant health as well as contribute to greater biomass and diversity in the macroinvertebrate community.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx. doi.org/10.1016/j.scitotenv.2012.11.063. These data include Google maps of the most important areas described in this article.

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