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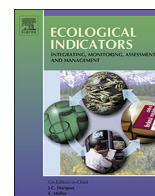
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Original Articles

Multi-element fingerprinting of waters to evaluate connectivity among depressional wetlands

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ABSTRACT

Establishing the connectivity among depressional wetlands is important for their proper management, conservation and restoration. In this study, the concentrations of 38 elements in surface water and porewater of depressional wetlands were investigated to determine chemical and hydrological connectivity of three hydrological types: recharge, flow-through, and discharge, in the Prairie Pothole Region of North America. Most element concentrations of porewater varied significantly by wetland hydrologic type ($p < 0.05$), and increased along a recharge to discharge hydrologic gradient. Significant spatial variation of element concentrations in surface water was observed in discharge wetlands. Generally, higher element concentrations occurred in natural wetlands compared to wetlands with known disturbances (previous drainage and grazing). Electrical conductivity explained 42.3% and 30.5% of the variation of all element concentrations in porewater and surface water. Non-metric multidimensional scaling analysis showed that the similarity decreased from recharge to flow-through to discharge wetland in each sampling site. Cluster analysis confirmed that element compositions in porewater of interconnected wetlands were more similar to each other than to those of wetlands located farther away. Porewater and surface water in a restored wetland showed similar multi-element characteristics to natural wetlands. In contrast, depressional wetlands connected by seeps along a deactivated drain-tile path and a grazed wetland showed distinctly different multi-element characteristics compared to other wetlands sampled. Our findings confirm that the multi-element fingerprinting method can be useful for assessing hydro-chemical connectivity across the landscape, and indicate that element concentrations are not only affected by land use, but also by hydrological characteristics.

1. Introduction

Prairie Pothole Region (PPR) wetlands are located on the glaciated landscape of central North America in depressions formed during the Wisconsin glacial episode (Bluemle, 1991). These depressional wetlands are important components of the global environment, performing ecosystem services such as supporting carbon cycling and provisioning wildlife habitat (Mitsch and Gosselink, 2000; Niemuth et al., 2010). However, more than 50% of the depressional wetlands within the PPR have been lost, mainly due to agricultural activities (Euliss et al., 2006; Dahl, 2014).

Multi-element fingerprinting is based on quadrupole Inductively Coupled Plasma Mass Spectrometry (ICP-MS) analysis for determining

the composition of influential chemical elements, and has been used for identification of water and sediment sources (Stutter et al., 2009; Rauch et al., 2010). The element composition (so-called multi-element fingerprint, see Markert et al., 2015) of soils and waters reflects the variation in biogeochemistry between wetlands in terms of environmental conditions such as hydrology, position in the landscape, and disturbances (Jacob et al., 2013; Kissoon et al., 2013, 2015; Sullivan et al., 2014; Yellick et al., 2016). Knowledge about the element composition of wetland soils and waters is important for our understanding of their biogeochemistry, and for monitoring and restoration (Moreno-Mateos et al., 2012).

Depressional wetlands of the PPR, often referred to as prairie pothole wetlands, are mostly rain and meltwater fed. Seasonal and yearly

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Table 1

Coordinates, elevation and condition of sampling sites in depressional wetlands in the Prairie Pothole Region, see also Fig. 1.

Site	Coordinate	Elevation (ASL, m)	Land use
1	47°20'26.1924"N, 99° 15'52.6428"W	540–546	natural wetland
2	47°20'27.3372"N, 99° 15'44.0532"W	527–543	natural wetland
3	47°18'45.0936"N, 99° 17'3.0408"W	567–578	restored wetland (previously drained and restored 27 years)
4	47°9'14.166"N, 99° 7'2.8668"W	547–552	natural wetland
5	47°7'42.0668"N, 99° 15'29.8836"W	570–577	natural wetland
6	47° 5'55.0356"N, 99° 5'40.1268"W	532–541	natural wetland
7	47° 4'31.6344"N, 99° 2'2.8644"W	513–525	natural wetland
8	46° 58'37.3044"N, 99°4'21.3528"W	526–530	grazed wetland (previously drained and restored more than 40 years)
9	46° 56'42.702"N, 99°6'39.336"W	532–565	wetland with seeps (disturbed by tile-drainage 27 years ago)
10	47° 12'59.8968"N, 99°11'39.2892"W	561–571	natural wetland

variation in evapotranspiration and precipitation are the dominant factors influencing variation in water levels of prairie pothole wetlands (Winter and Rosenberry, 1998; Johnson et al., 2005). In general, under anticipated influences of a changing climate, prairie pothole wetlands in the US are projected to become drier in western parts of the PPR, mainly caused by an increasing precipitation/evapotranspiration deficit in addition to increasing variability in the intensity and frequency of summer storms and winter snowfall (Johnson et al., 2010). If these projections materialize, they will have major effects on the hydrology and nutrient cycling of prairie pothole wetlands.

Hydrological connectivity plays a critical role in the functioning and sustainability of wetlands (Leibowitz and Vining, 2003), which is important for maintenance of wildlife habitat and global biogeochemical cycling. However, depressional wetlands in the PPR are often incorrectly referred to as “isolated wetlands” (Mushet et al., 2015). The debate of over use of the term “geographically isolated” to describe depressional wetlands highlights our poor understanding of how these wetlands are connected to each other physically, chemically and ecologically (Leibowitz, 2015; Mushet et al., 2015; Calhoun et al., 2017). Depressional wetlands in the PPR can be intermittently connected via surface water (Leibowitz and Vining, 2003; Leibowitz et al., 2016), and their linkage via groundwater is generally accepted (Euliss et al., 2004; Neff and Rosenberry, 2018). The importance of knowledge of the biogeochemical connectivity of so called “isolated wetlands” to the surrounding ecosystems was also highlighted by Marton et al. (2015). Such information is critical in relation to proper protection and restoration, particularly in relation to the expected effects of climate change and associated changes in land use.

Depending on hydrologic and topographic landscape position, depressional wetlands in the PPR can be categorized as discharge, flow-through or recharge wetlands (Euliss et al., 2004). Previous multi-element fingerprinting studies focused on relationships between environmental variables and element composition of individual wetlands: the shallow lakes of Minnesota (Kissoon et al., 2013, 2015) and seasonal recharge wetlands of North Dakota (Yellick et al., 2016). However, it is unclear how wetlands are connected across the landscape. In addition, if climate change leads to overall drier conditions, many seasonal wetlands may dry up permanently. A decrease in the permanency of ponded water in wetlands in the PPR will make these wetlands more vulnerable to disturbance by agricultural activities, affect their biogeochemistry and the movement of elements through the landscape, and drastically alter element cycling and hydrological relationships.

The main aim of our study was to assess the element composition of surface water and porewater in depressional wetlands in clusters with known linkages through groundwater along a recharge to discharge gradient. We hypothesized that: (1) because of downward movement of surface water and groundwater between interconnected wetlands, the concentration of many elements in discharge wetlands would be higher compared to those in flow-through and recharge wetlands; (2) the element composition of porewater of interconnected wetlands would be more similar to each other than to those located farther away; and (3)

the element composition of water in wetlands across the study area would show spatial variations.

2. Materials and methods

2.1. Study area

Depressional wetlands in the PPR receive precipitation as rain or snow meltwater, and include wet meadows and shallow-water ponds, saline lakes, marshes, and fens. Their density varies across the PPR from less than 1 to greater than 40 wetlands/km² (Cowardin et al., 1979; Kantrud et al., 1989). The climate of the study area is continental and semi-arid, with mean temperatures ranging from –15 °C to 20.5 °C. Average annual precipitation is 440 mm (Rosenberry, 2003).

Surface water and porewater samples were collected from ten sites based on geomorphic feature and the potential of hydrological connectivity among wetlands in North Dakota (46°56'–47°21'N, 99°01'–99°18'E) (Table 1, Fig. 1) during June and July 2017. Each site included at least one recharge, one flow-through, and one discharge wetland located along a subsurface hydrologic flow-path. In total, we sampled 11 recharge wetlands, 14 flow-through wetlands, and 11 discharge wetlands.

2.2. Sampling and physicochemical analysis

Porewater samples were collected in situ using ‘peepers’ (Teasedale et al., 1995) placed in each sampling location during June 19–23, 2017. Peepers were retrieved one month later after installation (during July 23–27, 2017). In recharge and flow-through wetlands, five peepers were placed in a circle around the center of each wetland, within vegetation that was considered representative of most such wetlands in the area. Discharge wetlands were too large to be sampled in this way. Instead, the five replicate peepers were installed along the shoreline nearest the upstream wetlands, i.e., the area of potential discharge of water from the flow-through wetlands, at 10-meter intervals (Fig. 2). Each peeper was constructed using a 125 mL polyethylene bottle with a hole in the top of the lid, and a 21 µm nylon-mesh filter secured between the top of the bottle and the cap. Peepers were filled with deionized water before placement into the soil, and assembled under water to ensure no air bubbles were trapped. The peepers were buried 10–20 cm under the soil surface at each sampling location and attached to a magnet buried just below the surface for later retrieval. The magnets were later detected using a GA-52Cx Magnetic Locator (Schonstedt, United States). Upon retrieval, peepers were sealed with mud to prevent gas exchange. To prevent contamination with surface sediments, 125 mL surface water samples were collected in polyethylene bottles before commencement of any other activities in the water at each site (EPA, 1983). These samples were collected from within one meter of the porewater samples. Unfortunately, we were unable to obtain surface water samples from all recharge and flow-through wetlands in all sites, because of a lack of water that caused by

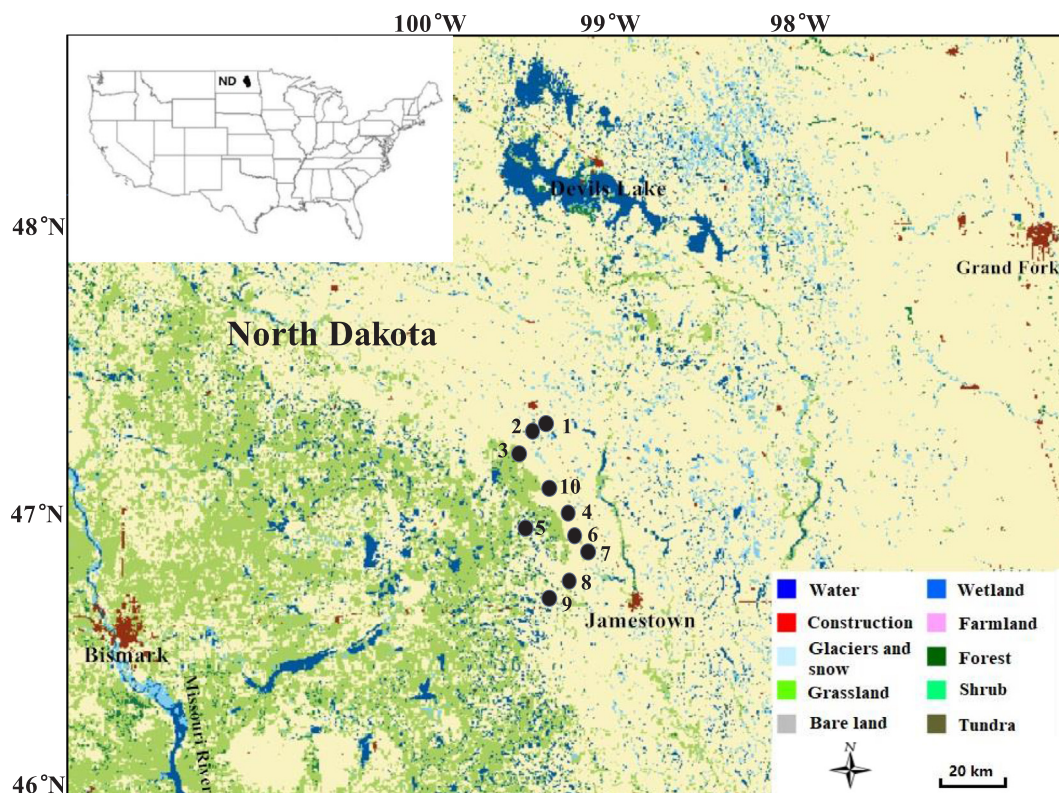


Fig. 1. Location of sampling sites in Prairie Pothole Region wetlands in North Dakota. (Data source: GlobeLand30-2010, 91 Satellite Image Assistant).

low precipitation and high evapotranspiration during the summer of 2017. Electrical conductivity (EC) and pH of the surface water were measured in situ using a SympHony pH probe (14002–860, VWR, United States) and SympHony pH meter (SP70P, VWR, United States). All samples were stored on ice in a cooler and then transported to the laboratory for further processing. To minimize systematic errors, all samples were randomized before processing and chemical analysis. As soon as possible after collection, surface water and porewater samples were filtered through syringe-mounted 0.45 μm filters (25 mm diameter membrane filter, PALL GN-6, United States) in sufficient amounts for further analyses (100 mL). EC and pH of porewater were determined in our laboratory using the same probe and pH meter as used for surface water. Samples for ICP-MS analysis were acidified with a drop of

concentrated HNO₃ after filtering. ICP-MS analysis was performed by Elan 9000 ICP-MS (Perkin Elmer Sciex, United States) in Activation Laboratories (Ontario, Canada). See [Supplementary Materials Table S1](#) for raw data and detection limits.

Soil samples were collected as well, but are presented and discussed elsewhere (Zhu et al., submitted for publication). However, the organic matter content (OM) of the wetland soils was tested as one of the environmental variables potentially affecting the composition of waters, and so the sampling and analysis of soils as relevant to the analysis of water samples in this paper are presented here. Soil samples were collected at a depth of about 10 cm using a spade and, to prevent contamination from the spade or surface of the substrate, subsamples of about 125 cm³ were taken by hand with an inverted zip-lock bag from

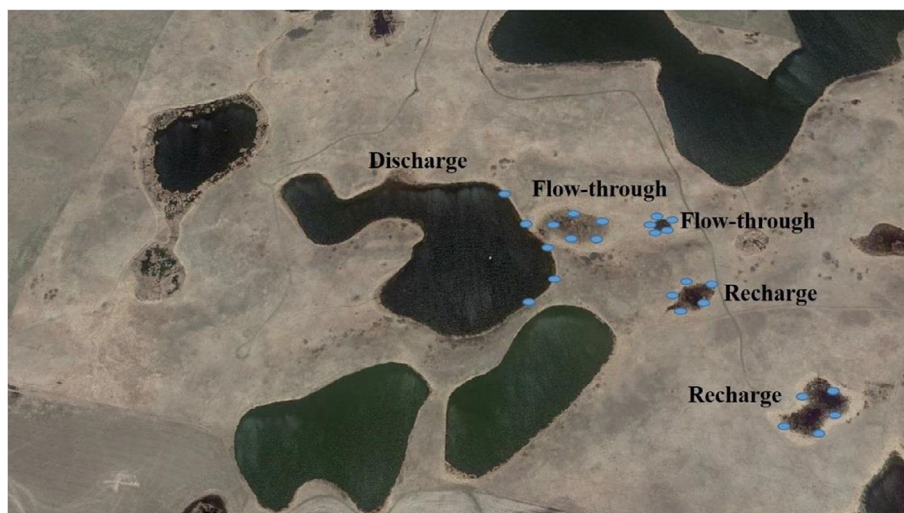


Fig. 2. Location of sampling locations within each depressional wetland in a cluster (Site 6, Cottonwood Lake Study Area).

within the center of those samples. Samples were stored in polyethylene bags on ice in a cooler after collection. To minimize systematic errors, all samples were randomized before processing and chemical analysis. In the laboratory, samples were dried at 60 °C until reaching a constant weight, and then crushed and homogenized using a ceramic mortar and pestle. Subsequently, samples were sieved through a 2-mm-mesh screen (USA Standard Testing Sieve, VWR, United States). Organic matter was determined by loss-on-ignition (LOI). Samples, approximately 5 g each, were first dried at 105 °C for 2 h, then heated at 360 °C for another 2 h (Combs and Nathan, 2012).

2.3. Statistical analysis

Data on element concentrations were deemed non-detectable overall and excluded from further consideration if more than half of the values of all samples were below the detection limit. For the remaining elements, values below the detection limit were replaced with the detection limit.

The similarities of element compositions among different wetlands were calculated by non-metric multidimensional scaling (NMDS) based on Bray-Curtis distance using CANOCO for Windows (Version 5.0). Similar element compositions were grouped together on a two-dimensional NMDS diagram. Stress values were used to determine the number of dimensions. A stress value less than 0.1 is considered a good ordination of the data (Stauffer et al., 2014). In addition, hierarchical cluster analysis was employed to group and classify wetland characteristics using JMP software from SAS (version 13). Cluster analysis can divide objects (e.g., wetlands) into different subgroups in such a way that objects in the same group are more similar with each other than with those in other groups (Richarme, 2002; Sheela et al., 2015).

Preliminary Detrended Correspondence Analysis (DCA) indicated that linear gradient analysis was appropriate for analysis of environmental variables and multi-element data, as the gradient lengths were < 3.0 standard deviations (ter Braak and Šmilauer, 2002). The relationships between the environmental variables, EC, pH and soil OM, and element concentrations were determined using redundancy analysis (RDA) within CANOCO for Windows (Version 5.0). We normalized the data using the logarithmic transformation ($\log_{10}x$) before the analyses to minimize the influence of extreme values on ordination scores. The RDA models were determined by manual forward selection with Monte Carlo test (499 permutations).

To compare the variance of element concentrations and environmental variables among depressional wetlands, non-parametric analysis by Kruskal-Wallis test, Friedman test, and Spearman Rank Correlation Analysis were performed using Minitab statistical software (version 18). Differences and correlations were considered statistically significant when $p < 0.05$.

3. Results

At the time of sampling, all recharge and flow-through wetlands were dry, so that only porewater could be collected from those. All raw data are presented as Online Supplementary Materials (Table S1). Consistent with Werkmeister et al. (2018), elements with more than 50% of values below the detection limit were excluded from further analysis and discussion. The excluded elements were Be, Al, Sc, Ti, Cr, Se, Nb, Mo, Ag, Cd, In, Sn, Te, Eu, Tb, Ho, Tm, Ta, W, Hg and Bi.

3.1. Element properties of porewater and surface water

The element concentrations of porewater are shown in Table 2. Because the data were not normally distributed, and no suitable transformation could be identified, non-parametric tests were used for comparisons between groups of data. Friedman test results showed that concentrations of most of the elements in porewater: Li, Na, Mg, Si, K, Ca, Mn, Cu, Ga, As, Sr, Zr, Cs, Ba, Ce, Pr, Pb and U, varied significantly

Table 2
The element concentrations of porewater (mean value \pm standard deviation) in recharge, flow-through and discharge wetlands in the Prairie Pothole Region. *p*-values are from Friedman test, bolded results indicate a significant difference ($p < 0.05$). Differences between different sites were represented by site, differences between recharge, flow-through, and discharge wetlands were represented by type.

Variable (mg/L)	Recharge		Flow-through		Discharge		Friedman test		Friedman test			
	Mean	SD	Mean	SD	Mean	SD	Site	Type	Site	Type		
Li	0.0097 \pm 0.0095	0.061 \pm 0.057	0.18 \pm 0.11	0.205	0.000	0.000	Zr	0.00022 \pm 0.00021	0.00028 \pm 0.00009	0.00039 \pm 0.00021	0.112	0.020
Na	7.42 \pm 8.83	30.7 \pm 30.2	87.6 \pm 83.5	0.307	0.000	0.000	Sb	0.00038 \pm 0.00037	0.00049 \pm 0.00041	0.00053 \pm 0.00022	0.511	0.061
Mg	8.32 \pm 10.84	50.4 \pm 39.5	136.6 \pm 93.5	0.044	0.000	0.000	Cs	0.00020 \pm 0.00019	0.00014 \pm 0.00012	0.00062 \pm 0.00001	0.378	0.007
Si	3.23 \pm 2.24	9.66 \pm 5.66	16.79 \pm 6.20	0.081	0.000	0.000	Ba	0.02 \pm 0.01	0.04 \pm 0.02	0.10 \pm 0.03	0.155	0.000
K	5.24 \pm 1.92	8.91 \pm 7.90	13.26 \pm 10.78	0.126	0.006	0.000	La	0.00054 \pm 0.00038	0.00032 \pm 0.00033	0.00019 \pm 0.00010	0.323	0.122
Ca	20.7 \pm 26.0	99.2 \pm 81.5	131.1 \pm 46.4	0.044	0.001	0.001	Ce	0.0010 \pm 0.0008	0.0005 \pm 0.0007	0.0002 \pm 0.0001	0.108	0.007
V	0.0017 \pm 0.0009	0.0021 \pm 0.0016	0.0014 \pm 0.0005	0.224	0.273	0.000	Pr	0.00013 \pm 0.00011	0.00014 \pm 0.00011	0.00011 \pm 0.00009	0.126	0.004
Mn	0.14 \pm 0.29	0.29 \pm 0.47	1.37 \pm 1.14	0.071	0.007	0.000	Nd	0.00013 \pm 0.00010	0.00014 \pm 0.00012	0.00001 \pm 0.00001	0.139	0.061
Fe	0.72 \pm 0.66	0.31 \pm 0.28	0.60 \pm 1.06	0.078	0.497	0.000	Sm	0.00013 \pm 0.00012	0.00013 \pm 0.00012	0.00001 \pm 0.00001	0.113	0.061
Co	0.00079 \pm 0.00071	0.00073 \pm 0.00062	0.0012 \pm 0.0013	0.252	0.122	0.000	Gd	0.00013 \pm 0.00011	0.00012 \pm 0.00012	0.00001 \pm 0.00001	0.123	0.061
Ni	0.0067 \pm 0.0096	0.0040 \pm 0.018	0.0047 \pm 0.024	0.286	0.407	0.000	Dy	0.00013 \pm 0.00011	0.00012 \pm 0.00012	0.00001 \pm 0.00001	0.099	0.061
Cu	0.0098 \pm 0.0048	0.0083 \pm 0.0044	0.0035 \pm 0.015	0.181	0.002	0.000	Er	0.00013 \pm 0.00012	0.00001 \pm 0.00001	0.00001 \pm 0.00001	0.073	0.076
Zn	0.05 \pm 0.05	0.03 \pm 0.02	0.02 \pm 0.01	0.370	0.202	0.000	Yb	0.000012 \pm 0.000011	0.000013 \pm 0.000010	0.000014 \pm 0.000011	0.140	0.122
Ga	0.00038 \pm 0.00032	0.00022 \pm 0.00020	0.00010 \pm 0.00001	0.101	0.014	0.000	Lu	0.000011 \pm 0.000010	0.000012 \pm 0.000011	0.000014 \pm 0.000011	0.454	0.241
Ge	0.00010 \pm 0.00001	0.00010 \pm 0.00001	0.00010 \pm 0.00001	0.116	0.067	0.000	Hf	0.000010 \pm 0.000009	0.000012 \pm 0.000011	0.000013 \pm 0.000011	0.100	0.839
As	0.0015 \pm 0.0010	0.0023 \pm 0.0013	0.0051 \pm 0.0035	0.047	0.006	0.000	Tl	0.000010 \pm 0.000009	0.000012 \pm 0.000011	0.000014 \pm 0.000011	0.538	0.341
Rb	0.0023 \pm 0.0010	0.0022 \pm 0.0008	0.0020 \pm 0.0008	0.690	0.670	0.000	Pb	0.00083 \pm 0.00051	0.0011 \pm 0.0019	0.00032 \pm 0.00048	0.225	0.025
Sr	0.05 \pm 0.06	0.31 \pm 0.27	0.63 \pm 0.25	0.242	0.000	0.000	Th	0.000010 \pm 0.000011	0.000013 \pm 0.000011	0.000014 \pm 0.000012	0.697	0.273
Y	0.00049 \pm 0.00044	0.00032 \pm 0.00038	0.00010 \pm 0.00001	0.212	0.061	0.000	U	0.00020 \pm 0.00020	0.0035 \pm 0.0037	0.0036 \pm 0.0031	0.441	0.002

Table 3

The variations of surface water element concentrations among sampling sites, and the correlation of element concentrations between different water types (surface water and porewater) in discharge wetlands in the Prairie Pothole Region. *p*-values are from Kruskal-Wallis test of differences between different sites and Spearman correlation analysis of correlations between different types, bolded results indicate a significant difference ($p < 0.05$).

Variable	Surface water	Variation	Correlation	Variable	Surface water	Variation	Correlation
Li (mg/L)	0.31 ± 0.21	0.001	0.003	Zr ($\times 10^{-3}$ mg/L)	0.44 ± 0.37	0.005	0.144
Na (mg/L)	158.9 ± 143.2	0.007	0.000	Sb ($\times 10^{-3}$ mg/L)	0.93 ± 0.71	0.003	0.445
Mg (mg/L)	241.5 ± 190.7	0.003	0.000	Cs ($\times 10^{-5}$ mg/L)	0.77 ± 0.01	0.002	0.000
Si (mg/L)	16.92 ± 8.84	0.000	0.170	Ba (mg/L)	0.07 ± 0.04	0.000	0.101
K (mg/L)	34.5 ± 17.8	0.000	0.000	La ($\times 10^{-4}$ mg/L)	0.82 ± 0.50	0.001	0.290
Ca (mg/L)	103.8 ± 65.6	0.001	0.000	Ce ($\times 10^{-4}$ mg/L)	0.96 ± 0.64	0.001	0.088
V ($\times 10^{-2}$ mg/L)	0.20 ± 0.08	0.000	0.159	Pr ($\times 10^{-4}$ mg/L)	0.14 ± 0.11	0.003	0.001
Mn (mg/L)	0.04 ± 0.04	0.012	0.573	Nd ($\times 10^{-4}$ mg/L)	0.47 ± 0.49	0.014	0.031
Fe (mg/L)	0.08 ± 0.05	0.001	0.003	Sm ($\times 10^{-4}$ mg/L)	0.13 ± 0.11	0.004	0.000
Co ($\times 10^{-3}$ mg/L)	1.18 ± 0.01	0.000	0.000	Gd ($\times 10^{-4}$ mg/L)	0.14 ± 0.12	0.004	0.000
Ni ($\times 10^{-2}$ mg/L)	0.47 ± 0.25	0.000	0.506	Dy ($\times 10^{-4}$ mg/L)	0.13 ± 0.10	0.003	0.000
Cu ($\times 10^{-2}$ mg/L)	1.06 ± 0.50	0.000	0.550	Er ($\times 10^{-4}$ mg/L)	0.10 ± 0.01	0.001	0.001
Zn (mg/L)	0.03 ± 0.01	0.000	0.768	Yb ($\times 10^{-5}$ mg/L)	0.98 ± 0.01	0.001	0.000
Ga ($\times 10^{-3}$ mg/L)	0.08 ± 0.05	0.001	0.002	Lu ($\times 10^{-5}$ mg/L)	0.77 ± 0.01	0.002	0.000
Ge ($\times 10^{-3}$ mg/L)	0.10 ± 0.06	0.001	0.417	Hf ($\times 10^{-4}$ mg/L)	0.11 ± 0.01	0.002	0.044
As ($\times 10^{-2}$ mg/L)	1.23 ± 1.04	0.005	0.366	Tl ($\times 10^{-5}$ mg/L)	0.82 ± 0.01	0.003	0.216
Rb ($\times 10^{-2}$ mg/L)	0.39 ± 0.17	0.000	0.024	Pb ($\times 10^{-3}$ mg/L)	0.14 ± 0.10	0.001	0.466
Sr (mg/L)	0.61 ± 0.43	0.001	0.031	Th ($\times 10^{-4}$ mg/L)	0.21 ± 0.15	0.002	0.493
Y ($\times 10^{-4}$ mg/L)	0.70 ± 0.62	0.006	0.009	U ($\times 10^{-2}$ mg/L)	0.29 ± 0.14	0.000	0.174

among recharge, flow-through, and discharge wetlands, when grouped for all sites ($p < 0.05$). In addition, Mg, Ca and As varied significantly ($p < 0.05$) among sites, when grouped by type (recharge, flow-through and discharge wetlands). The concentrations of most elements in porewater that varied significantly among wetland types increased from recharge to discharge wetlands, except for Cu, Cs, Ce, Pr and Pb, which showed the reverse pattern.

Significant spatial differences between sites were evident for concentrations of all elements in the surface water of the discharge wetlands ($p < 0.05$) (Table 3). For example, the concentrations of Na (453 ± 33 mg/L) and Ca (225 ± 8 mg/L) were higher in site 9 compared to other sites. Besides, many elements showed significant correlations between porewater and surface water, for instance, Li, Na, Mg

and K (Fig. 3), indicating strong vertical connection of waters.

3.2. Variations of environmental variables of porewater and surface water

The properties of pH, EC and OM in depressional wetlands are shown in Table 4. There were no significant differences in pH of porewater or OM content among wetland sites or types. However, EC of porewater showed significant differences among both wetland sites and types ($p < 0.05$). pH and EC of surface water of discharge wetlands also varied significantly among sites ($p < 0.01$). In addition, EC of porewater increased from recharge wetlands to discharge wetlands (Table 4).

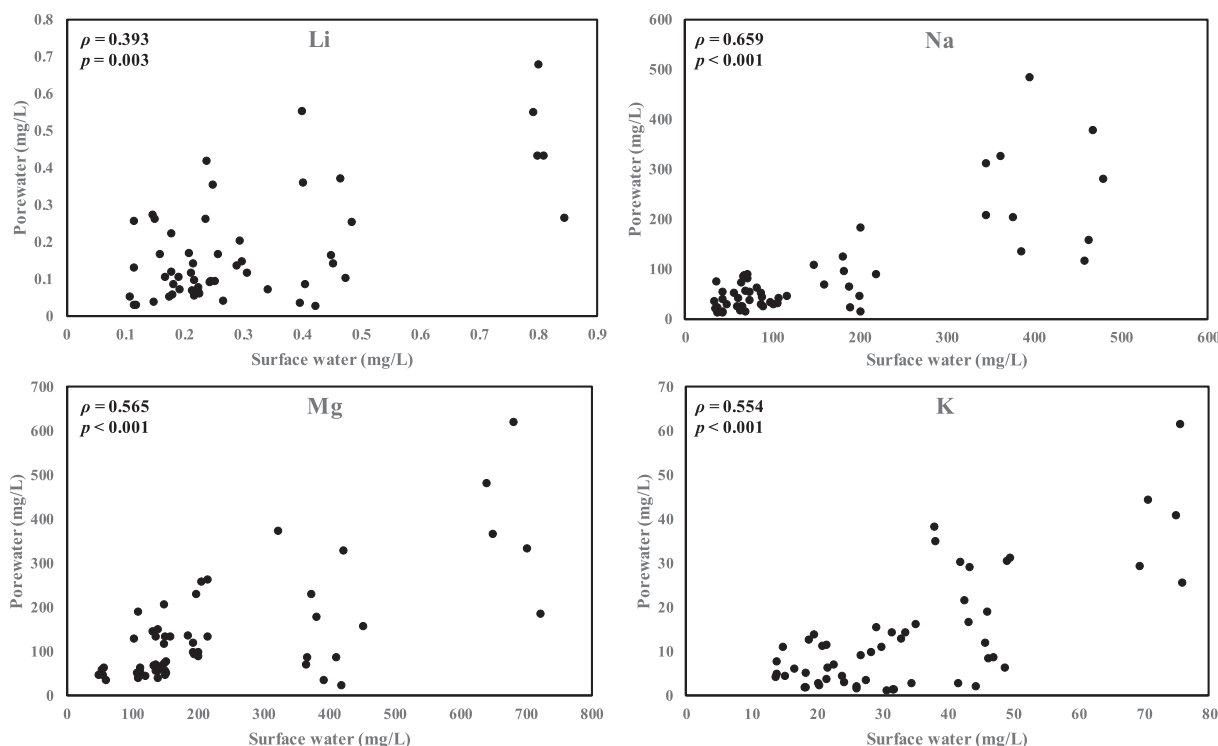


Fig. 3. Correlations between element concentrations of porewater and surface water in discharge wetlands.

Table 4

The variations of soil organic matter (OM), pH and electrical conductivity (EC) in depositional wetlands in the Prairie Pothole Region with Friedman test, and their contributions in explaining the variations in all elements concentrations with Monte Carlo permutation test. Friedman test is used to compare the difference between sites and types in porewater. Kruskal-Wallis test is used to compare the difference between sites in surface water. The bold value in the bracket means significance ($p < 0.05$).

Variable	Recharge	Flow-through	Discharge	Friedman test		Kruskal-Wallis test
				Site	Type	Site
OM (%)	11.9 ± 8.7	10.3 ± 4.2	6.5 ± 3.5	0.089	0.061	
Porewater pH	6.5 ± 0.5	6.7 ± 0.4	6.7 ± 0.2	0.065	0.407	
Porewater EC (ms/cm)	0.22 ± 0.20	0.81 ± 0.59	1.53 ± 0.80	0.034	0.000	
Surface water pH			8.0 ± 0.6			0.000
Surface water EC (ms/cm)			2.26 ± 1.36			0.000

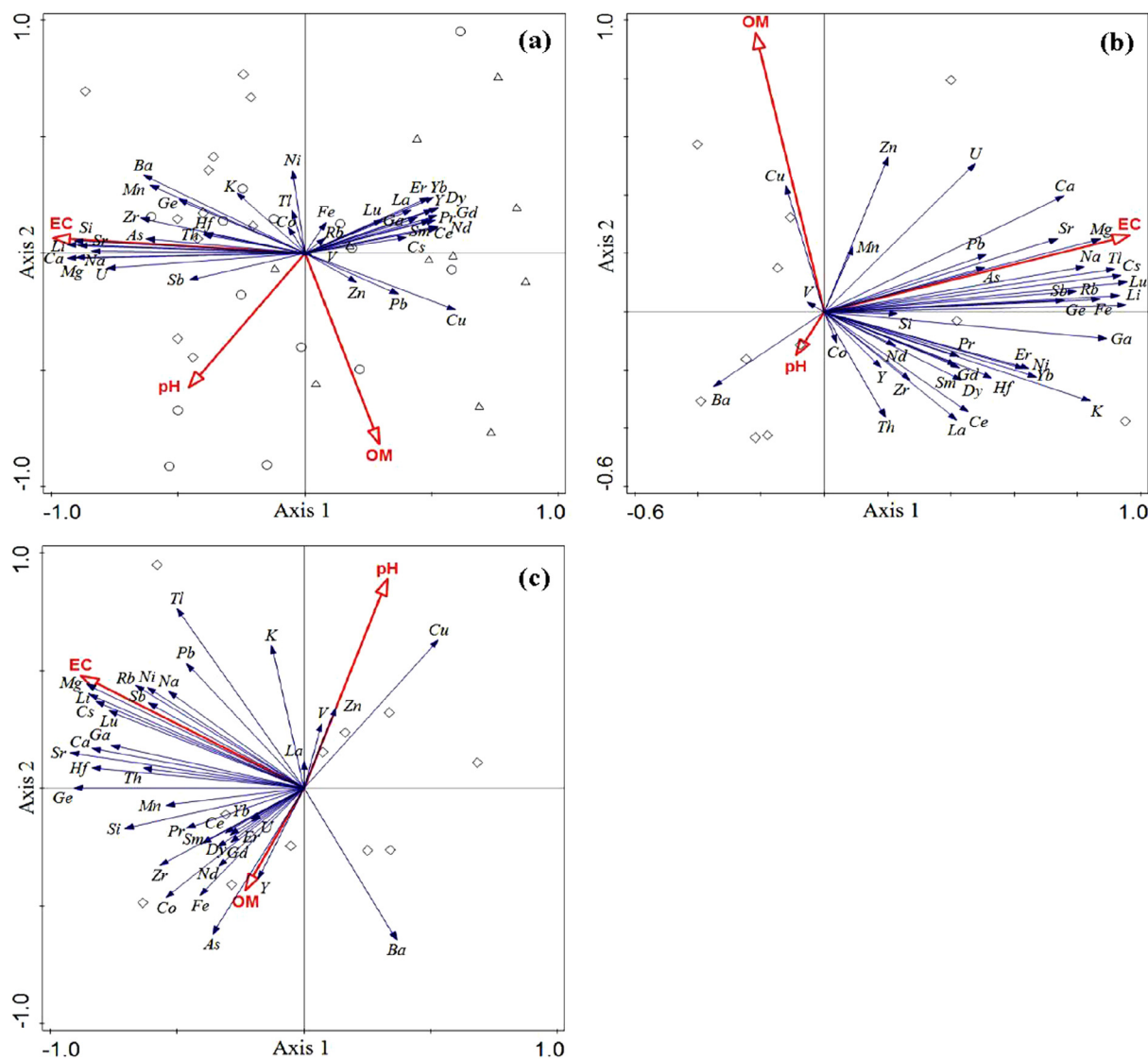


Fig. 4. Redundancy analysis (RDA) ordination diagrams of the relationships between porewater element concentrations and environmental variables for all wetlands (a), surface water (b), and porewater of discharge wetlands (c). Environmental variables (displayed as red lines with arrows) were pH, electrical conductivity (EC), and organic matter content (OM). Elements are represented by blue lines with arrows. Recharge wetlands are represented by triangles “△”, flow-through wetlands are represented by circles “○”, and discharge wetlands are represented by diamonds “◇”. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.3. Relationships between environmental variables and element concentrations

Statistical relationships between environmental variables (OM, pH and EC) and multi-element concentrations were explored using RDA.

The RDA ordination (Fig. 4a) of porewater element concentrations and environmental variables for all wetlands combined showed that EC had a strongly negative correlation with the first RDA axis ($r = -0.896$), while OM was negatively correlated with the second axis ($r = -0.342$). The Monte-Carlo test confirmed the significance of the relationship

model ($p < 0.01$). EC explained most variation (42.3%, $p = 0.002$) of all element concentrations, while pH explained 1.5% and OM explained 1.8%. OM was negatively correlated with EC ($r = -0.304$). pH showed a positive correlation with EC ($r = 0.361$). EC was the primary variable associated with porewater element concentrations in depressional wetlands. U, Sb, As, Zr, Ge, Mn, Ba, K, Li, Na, Sr, Mg, Si and Ca were associated with high EC and high pH, and such elements were usually higher in discharge wetlands. In contrast, the lanthanides and several other metals (Cu, Ga, and Y) were negatively correlated with pH and EC, which were mostly observed in recharge wetlands. Most of elements showed negative correlations with OM, except Cu, Pb, Zn and Rb.

Results from RDA ordination (Fig. 4b) of surface water element concentrations and environmental variables for discharge wetlands only (because we were unable to collect surface water from flow-through and recharge wetlands) revealed that EC showed strongly positive correlation with the first RDA axis ($r = 0.820$), while OM was negatively correlated with the second axis ($r = 0.569$). The Monte-Carlo test confirmed the significance of the relationship model ($p < 0.01$). EC explained most variance (30.5%, $p = 0.02$) of all element concentrations, while pH explained 1.9% and OM explained 7.1%. OM showed a negative correlation with pH ($r = -0.314$), and pH showed a negative correlation with EC ($r = -0.128$). EC and OM were the primary variables associated with surface water element concentrations in depressional wetlands. The elements U, Sb, As, Rb, Ge, Zn, Ga, La, Ce, Ni, K, Li, Na, Fe, Sr, Mg, Si and Ca were positively correlated with EC. In contrast, Ba and Cu were negatively correlated with EC. Most elements showed a negative correlation with OM and pH.

We further analyzed the relationships between environmental variables and element concentrations in porewater of discharge wetlands by RDA (Fig. 4c). The results again showed that EC has a strong negative correlation with the first RDA axis ($r = -0.786$), while pH was positively correlated with the second axis ($r = 0.809$). The Monte-Carlo test confirmed the significance of the relationship model ($p < 0.01$). EC explained most variance (23.0%, $p = 0.006$) of all element concentrations, while pH explained 15.9% and OM explained 6.5%. EC and pH were the primary variables associated with surface water element concentrations in depressional wetlands. Most element concentrations were positively correlated with EC, except Cu and Ba (Fig. 4c). The elements Mg, Li, Cs, Lu, Ga, Ca, Sr, Hf, Th, Ge, Mn, Si, Yb, Ce, Pr, Sm, Zr, Dy, U, Co, Er, Gd, Nd, Fe, As, Y and Ba all showed a positive correlation with OM. Other element concentrations were positively correlated with pH.

The relationships between environmental variables and element concentrations in porewater and surface water were further investigated by Spearman correlation analysis (Table 5). The correlations of Li and Na concentrations with EC are illustrated in Fig. 5 as an example. Li and Na showed positive correlations with EC.

3.4. Similarity in depressional wetlands

Similarity in concentrations of porewater elements in recharge, flow-through and recharge wetlands was explored using NMDS (Fig. 6). Almost all recharge wetlands were located at the right quadrant of the NMDS ordination diagram (except 2R), while all discharge wetlands were located at the left quadrant. Flow-through wetlands spanned across both the right and left portions of the ordination (Fig. 6). Moreover, the similarity of wetlands in each sampling site decreased from recharge to flow-through to discharge wetlands. Cluster analysis further suggests that the wetlands can be divided into three groups, in which the similarity between discharge and flow-through wetlands, and between recharge and flow-through wetlands were higher than that between discharge and recharge wetlands (Fig. 7).

Cluster analysis was also used to distinguish the differences of element composition of porewater between sites, with discharge, flow-through and recharge wetlands at the same site considered as a group. The results showed that the sampling sites could be divided into four

groups (Fig. 8a), in which two adjacent wetlands (site 1 and 2) with unknown history of disturbance were grouped separate from other natural wetlands. The restored wetlands showed similar multi-element characteristics with natural wetlands sampled. Concentrations of Pr ($p < 0.01$), Gd ($p < 0.01$), Ce ($p < 0.01$) and Nd ($p < 0.01$) in the site with wetlands connected by seeps (site 9) were higher than in other sites, resulting in discriminating element compositions in the spatial distribution. In addition, concentrations of Ni ($p < 0.01$), Er ($p < 0.01$) and Tl ($p < 0.01$) were higher in grazed wetlands (site 8), which had distinctive element characteristic compared to other sites.

The cluster analysis results of surface water revealed that the sampling sites could be divided into three groups (Fig. 8b). The site with wetlands connected by seeps (site 9) and the natural wetlands in site 6 were grouped separately. This may be because they had high concentrations of Li, Na, Sr and Sb. However, grazed wetlands (site 8) and natural wetlands in site 4 had similar element characteristic, which might be because they had high concentrations of Y, Zr and Nd. In contrast, element composition of the restored wetlands in site 3 was similar with the other natural wetlands.

The cluster analysis results of porewater of discharge wetlands revealed that the sampling sites could be divided into three groups (Fig. 8c). The discharge wetland in site 6 that connected with two recharge and two flow-through wetlands (Fig. 2) showed distinct element characteristic compared to the other wetlands. This may be the result of high concentrations of Li, Mg, K, Sr, Hf and Pb in the porewater. The grazed wetland (site 8) and the natural wetland connected by seeps (site 9) showed similar element composition with natural wetlands in two sites (site 1 and 2). Element composition of wetlands in restored wetland (site 3) was similar with the other natural wetlands.

4. Discussion

4.1. Variation in element concentrations of porewater and surface water in depressional wetlands

Both hydrogeological characteristics of the soil material and landscape position influence water flow and elements transfer (Sophocleous, 2002), leading to variation in element concentrations among wetlands. Consistent with Arndt and Richardson (1989), the concentrations of most elements in our study were higher in discharge wetlands than in flow-through and recharge wetlands, most likely because of downward movement of surface water and groundwater from adjacent uplands caused by hydrological connectivity (Arndt and Richardson, 1988). Element mobility and accumulation in these wetlands are influenced by precipitation, landscape position and land use. The chemistry of discharge wetlands is complex and depends on many hydrological processes including precipitation, overland flow, groundwater input, open-water evaporation, and mineral precipitation and dissolution (Mills et al., 2011), while recharge wetlands in particular depend solely on precipitation and meltwater as inputs for water. This leads to different element signatures in discharge, flow-through and recharge wetlands. In addition, the rate of element transfer with water movement depends on soil texture and connected pathways (Winter and LaBaugh, 2003), that can be considered as another reason of spatial variation in multi-element composition. Our findings confirmed our first hypothesis that water movement leads to most element concentrations being higher in discharge wetlands.

Consistent with Hummel (2016), we observed that element concentrations were higher in surface water compared to those in porewater. Although the cluster analysis showed that the characteristic element compositions were different between porewater and surface water in discharge wetlands, their concentrations correlated strongly. Surface water is mainly transferred rapidly across the land surface and via groundwater. Porewater is the most available fraction of the overall soil matrix for plant uptake (Marschner, 1995), and it moves at low velocities from recharge wetland to discharge wetland through the

Table 5

Relationships between environmental variables (soil organic matter (OM), pH, electrical conductivity (EC)) and element concentrations in porewater and surface water in prairie pothole wetlands. Spearman correlation coefficients are shown. Asterisks indicate significant differences: * $p < 0.05$, ** $p < 0.01$.

Element	Porewater			Surface water			Porewater of discharge wetlands				
	OM	pH	EC	Element	OM	pH	EC	Element	OM	pH	EC
Li		0.345*	0.916**	Li			0.814**	Li			0.827**
Na		0.346*	0.872**	Na			0.669**	Na			0.643**
Mg		0.385*	0.911**	Mg			0.911**	Mg			0.850**
Si			0.893**	K	-0.361**		0.557**	Si	0.433**		0.596**
Ca		0.367*	0.925**	Ca			0.743**	Ca			0.767**
Mn	-0.355*		0.658**	V		0.367**		V	0.279*	0.404**	0.286*
Cu	0.412*		-0.582**	Fe			0.732**	Mn		-0.368**	0.298*
Ga			-0.362*	Co		0.412**		Fe		-0.313*	0.295*
Ge			0.568**	Ni			0.440**	Co	0.316*	-0.588**	
As			0.648**	Cu	0.364**			Ni			0.472**
Sr		0.348*	0.897**	Zn	0.324*		0.273*	Cu		0.562**	
Y		-0.301*	-0.424*	Ga			0.671**	Zn		0.377**	
Zr			0.669**	Ge			0.532**	Ga			0.571**
Sb		0.385*	0.553**	As			0.284*	Ge			0.703**
Cs			-0.341*	Rb			0.667**	As	0.328*	-0.285*	
Ba	-0.439**		0.685**	Sr			0.668**	Rb			0.511**
La			-0.386*	Sb			0.387**	Sr			0.799**
Ce		-0.390*	-0.469**	Cs			0.794**	Zr			0.526**
Pr		-0.347*	-0.469**	Ba			-0.631**	Sb			0.484**
Nd		-0.349*	-0.456**	La	-0.283*			Cs			0.718**
Sm		-0.351*	-0.426**	Pr			0.345**	Ba		-0.308*	
Gd		-0.331*	-0.427**	Sm			0.316*	Er			0.370**
Dy		-0.334*	-0.423**	Gd			0.308*	Yb			0.381**
Er			-0.406*	Dy			0.320*	Lu			0.648**
Yb			-0.393*	Er			0.424**	Hf			0.673**
Hf			0.403*	Yb			0.413**	Tl		0.440**	0.546**
Pb			-0.364*	Lu			0.849**	Pb			0.469**
Tl			0.459**	Hf			0.355**	Th			0.547**
U		0.437**	0.763**	Tl			0.837**	U			0.277*
				Pb			0.335*				
				U			0.702**				

fractured till compared to surface water (Winter and Rosenberry, 1995). The different transfer patterns influence element concentrations in surface water and porewater (Winter and Rosenberry, 1995). In addition, disturbance and chemical processes can lead to changes of element concentrations in porewater and surface water (Miao et al., 2006; Polizzotto et al., 2008). Most element concentrations further showed positive correlations with EC in both porewater and surface water in discharge wetlands, indicating similar correlations of EC with element concentrations in waters. However, EC was higher in surface water (2.26 ± 1.36 ms/cm) compared to porewater (1.53 ± 0.80 ms/cm), which can be explained by the higher element concentrations in surface water.

Element concentrations are related to environmental variables such as OM, EC, and pH (Otero and Macias, 2002; Jacob and Otte, 2003; Jiménez-Cárceles et al., 2008). Consistent with Clemente (2008) and Beesley (2010), OM of the wetland soil was an important factor

influencing Cu and Pb in porewater, while Cu, As, Zn and Sb were affected more by variation in pH. Consistent with Yellick et al. (2016), OM content was higher in undisturbed wetlands compared to disturbed (grazed and restored) wetlands, most likely due to the deterioration of soil and vegetation that resulted in decreased OM content in disturbed wetlands (Schmidt et al., 2011). The seasonal wet-dry cycles can increase OM formation in wetland soils (Van Gestel et al., 1991), possibly explaining higher OM in the soils of recharge wetlands. Moreover, OM binds some metals, such as Hg, As, Cd and Zn (Haldar and Mandal, 1979; Benedetti et al., 1996; Kalbitz and Wennrich, 1998). This can be one reason for these concentrations being lower in recharge wetlands compared to flow-through and discharge wetlands.

Dynamics of element concentrations are strongly influenced by pH because hydrogen and hydroxide ions can bind with trace elements or cause redox reactions in waters (Otero and Macias, 2002; Reddy and DeLaune, 2008). Consistent with Wang et al. (2014), many elements,

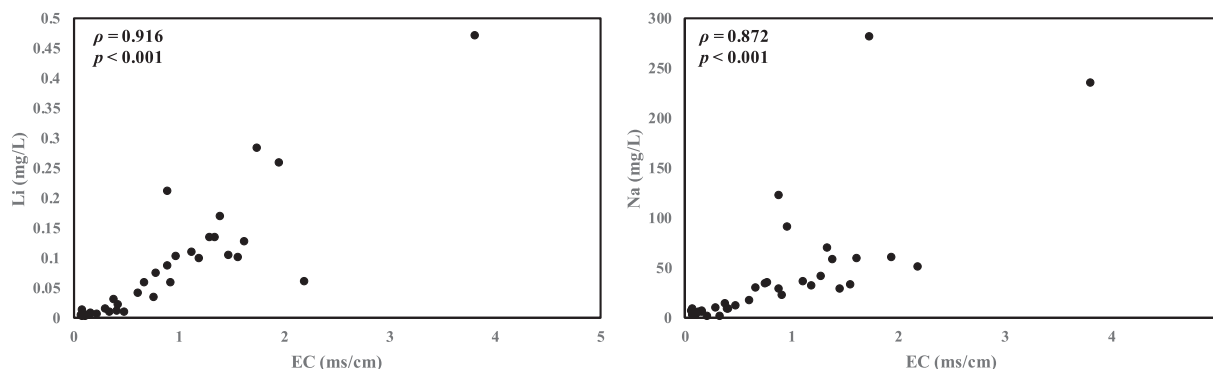


Fig. 5. Correlations between electrical conductivity (EC) and concentrations of Li and Na. Element concentrations are averages of five samples in each wetland.

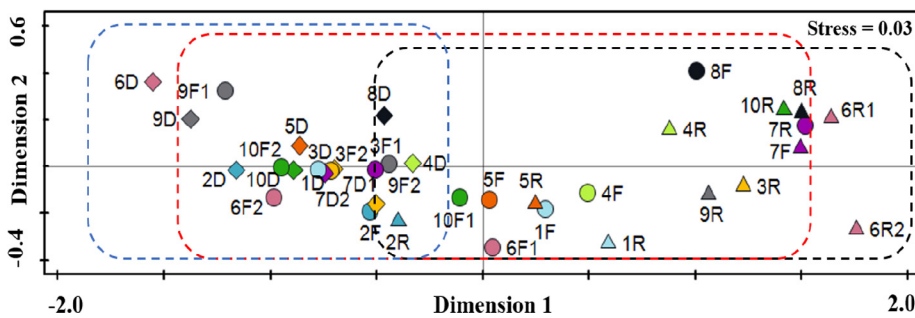


Fig. 6. Ordination diagrams for non-metric multi-dimensional scaling (NMDS) analysis of porewater elements based on Bray-Curtis distance among different depressional wetlands. The distance between points represents the similarity degree in multi-element composition. Recharge, flow-through, and discharge wetlands are represented by triangles “Δ”, circles “○”, and diamonds “◊”, and are enclosed by black, orange, and blue dotted lines. The sampling sites are represented by different colors. The presentations of each site are shown in Table 1. Stress = 0.03. (For interpretation of the references to colour in this figure legend, the reader is referred

to the web version of this article.)

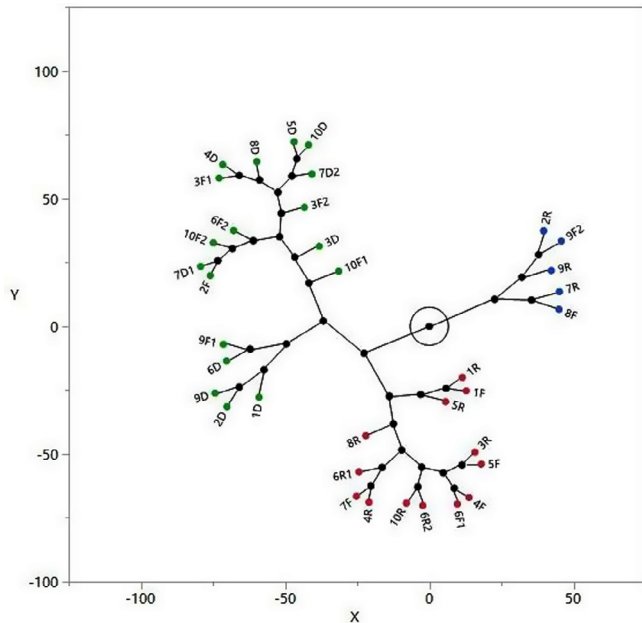


Fig. 7. Hierarchical cluster analysis of porewater elements in different types of wetland in all sites (n = 36). Groups are represented by different colors. The presentations of each site are shown in Table 1.

such as Co, Fe and Pb showed inverse correlations with pH in waters.

Electrical conductivity was an important indicator of salinity, explaining 69.5% and 58.7% variance of element concentrations in porewater and surface water in wetlands. This was consistent with Richardson and Bigler (1984). The increase in EC from recharge to discharge wetlands was also reported by Arndt and Richardson (1988). The increase in porewater element concentrations from recharge to discharge wetlands was strongly associated with an increase in EC, such as Li, Na, Mg, Si, Ca, Ba, As, Sr and U. The spatial variation of EC may be due to variation in the intensity and location of recharge and drawdown episodes (Arndt and Richardson, 1993). In addition, EC of porewater and surface water in the undisturbed wetlands sampled was generally higher than that in the disturbed wetlands, possibly associated with deterioration in vegetation and soil that can increase the movement of salts from wetlands to surrounding land (Arndt and Richardson, 1988).

4.2. Connectivity between wetlands

We observed evidence of chemical connectivity in porewater. For instance, cluster analysis indicated similarity of element composition between discharge and nearby flow-through wetlands was higher than between discharge and flow-through wetlands located farther away. Element composition in discharge wetlands showed the lowest similarity with that in recharge wetland at site 10. Moreover, many element

concentrations in porewater increased from recharge to flow-through to discharge wetlands, such as Li, Na, Mg, K, Ca, As and Pb, indicating connectivity of porewater between recharge, flow-through and discharge wetlands through downward flow. This is consistent with our second hypothesis that element composition in porewater of interconnected wetlands are more similar than those farther away.

We were unable to assess connectivity of surface water in this study, because of lack of data of element concentrations in flow-through and recharge wetlands. Water storage capacity in recharge wetlands can be exceeded during periods of high precipitation, creating temporary surface water connections between recharge and flow-through wetlands when spilling occurs (Shaw et al., 2012). Although recharge, flow-through and discharge wetlands are not always connected through surface water, many studies have noted the occurrence of intermittent surface water connections between recharge, flow-through and discharge wetlands (Winter, 1989; Winter and Rosenberry, 1998; Leibowitz and Vining, 2003; Leibowitz et al., 2016). Temporal connectivity can cause similar water level and specific conductance between depressional wetlands (Leibowitz and Vining, 2003; Leibowitz et al., 2016), influencing the transfer and accumulation of elements. Furthermore, depressional wetlands being connected via groundwater was observed by Euliss et al. (2004), and groundwater input influences element concentrations.

Consistent with Yellick et al. (2016) for soils, we observed that the element composition of waters in a disturbed (grazed) wetland were different from undisturbed (natural) wetlands, most likely due to the deterioration of vegetation and soil. The surrounding landscape and land use practices can impact chemistry and element transfer in waters (Kissoon et al., 2015; Hummel, 2016), which lead to changes of element composition. Moreno-Mateos et al. (2012) observed that biogeochemical functioning of restored wetlands was still lower than in reference wetlands even a century after restoration. However, in our study, we found that element composition of surface water and porewater in a restored wetland (site 3, about 25 years after restoration) was similar with that in natural wetlands, meaning that element composition can be recovered under certain conditions. This finding can provide a useful method to assess wetland restoration progress and effort.

Wet-dry cycles driven by climate change are a major factor influencing connectivity among wetlands (Leibowitz and Vining, 2003) via altering water input and output. Wetland drainage also can have a mixed effect on connectivity, including decreased wetland area, vegetation loss, and soil erosion (Leibowitz and Vining, 2003). In our study, wetlands connected by seeps showed distinct element composition with other wetlands, indicating that changes in connectivity are an important factor influencing hydro-chemical connectivity between depressional wetlands. This was also observed by Yuan et al. (2018) in different wetland ecosystems. These findings confirmed our third hypothesis that element composition of wetlands showed spatial variations across the study area.

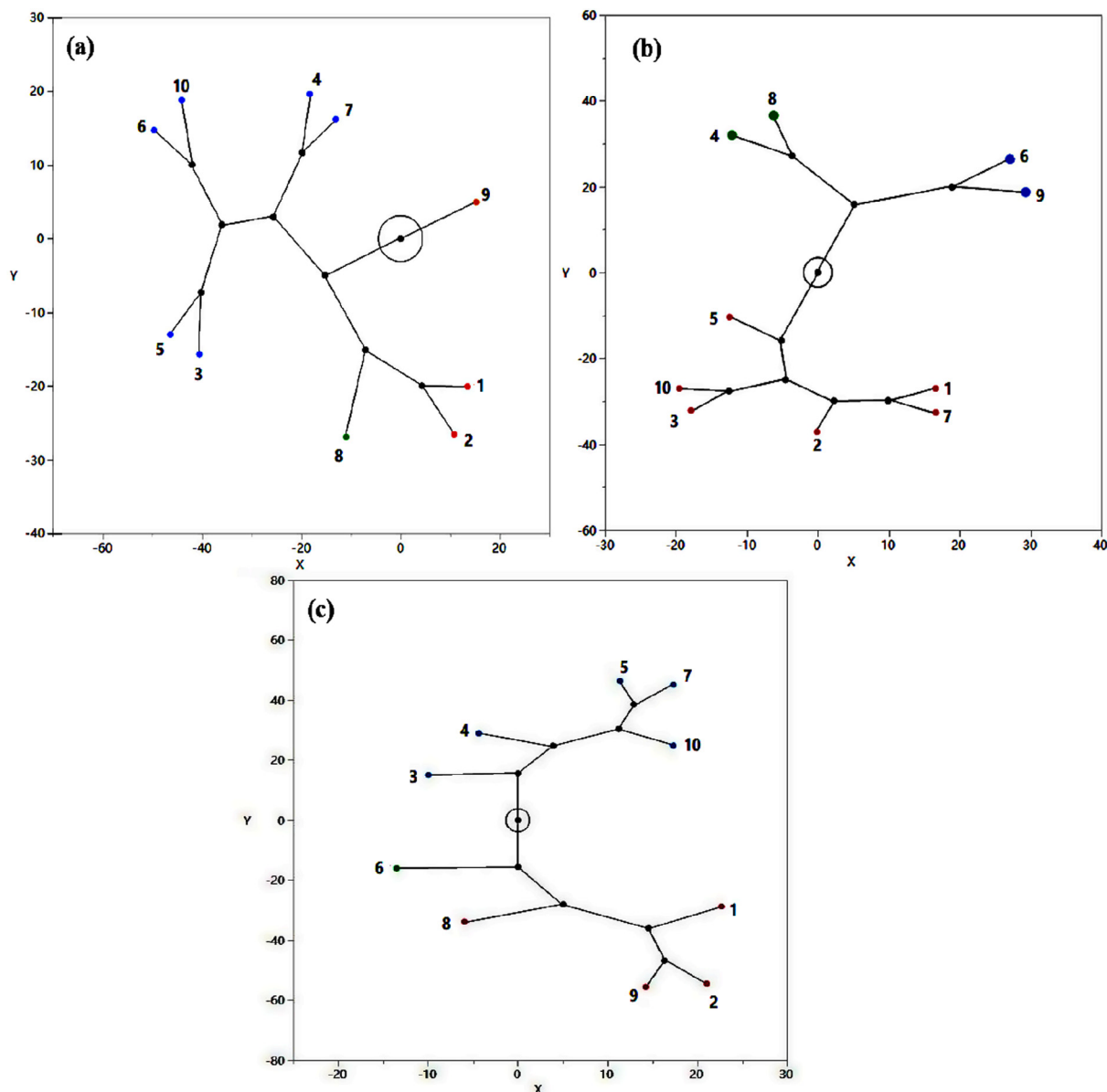


Fig. 8. Hierarchical cluster analysis of porewater of all wetlands together (a), surface water of discharge wetlands only (b), and porewater of discharge wetlands only (c) in all sampling sites ($n = 10$). Groups are represented by different colors. The presentations of each site are shown in Table 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

5. Conclusions

This study highlights the importance of understanding hydrological connectivity among depression wetlands in terms of multi-element and environmental variables. Both porewater and surface water element compositions showed spatial variations. Porewater element concentrations were enriched in discharge wetland. Most element concentrations were lower in porewater than those in surface water. Electrical conductivity was the primary factor associated with porewater and surface water element concentrations. The determination of element concentrations through depression wetlands has shown that disturbances lead to changes in element composition in disturbed wetlands compared to natural wetlands. Element compositions in porewater of interconnected wetlands were more similar than those farther away. This study confirms that multi-element fingerprinting can be a useful tool for assessing hydro-chemical connectivity across the landscape. Our findings further the understanding of connectivity effects on depression wetlands, and have potential to facilitate wetland

management and restoration efforts.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ecolind.2018.10.033>.

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