



RESEARCH ARTICLE

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Key Points:

- Pore water geochemistry relates to species-specific plant water use and aeration
- Sediment texture variations are a secondary influence on pore water patterns
- Pore water seepage was 41% of ebb solute export overall, and up to 87% for Ni

Supporting Information:

- Supporting Information S1

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Relating salt marsh pore water geochemistry patterns to vegetation zones and hydrologic influences

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Abstract Physical, chemical, and biological factors influence vegetation zonation in salt marshes and other wetlands, but connections among these factors could be better understood. If salt marsh vegetation and marsh pore water geochemistry coorganize, e.g., via continuous plant water uptake and persistently unsaturated sediments controlling vegetation zone-specific pore water geochemistry, this could complement known physical mechanisms of marsh self-organization. A high-resolution survey of pore water geochemistry was conducted among five salt marsh vegetation zones at the same intertidal elevation. Sampling transects were arrayed both parallel and perpendicular to tidal channels. Pore water geochemistry patterns were both horizontally differentiated, corresponding to vegetation zonation, and vertically differentiated, relating to root influences. The geochemical patterns across the site were less broadly related to marsh hydrology than to vegetation zonation. Mechanisms contributing to geochemical differentiation included: root-induced oxidation and nutrient (P) depletion, surface and creek-bank sediment flushing by rainfall or tides, evapotranspiration creating aerated pore space for partial sediment flushing in some areas while persistently saturated conditions hindered pore water renewal in others, and evapoconcentration of pore water solutes overall. The concentrated pore waters draining to the tidal creeks accounted for 41% of ebb tide solutes (median of 14 elements), including being a potentially toxic source of Ni but a slight sink for Zn, at least during the short, winter study period in southern San Francisco Bay. Heterogeneous vegetation effects on pore water geochemistry are not only significant locally within the marsh but may broadly influence marsh-estuary solute exchange and ecology.

1. Introduction

Spatially discrete vegetation zonation is a key characteristic of salt marshes and other wetland systems. This zonation is especially important as it has been linked to adaptive ecosystem resilience [Wiens, 1976; Holling, 1992; Scheffer *et al.*, 2001; Suding *et al.*, 2004; Rietkerk and van de Koppel, 2008]. In general, for marshes lacking much freshwater inflow, plant zonation among the lowest and highest elevations of the marsh is strongly influenced by environmental stress (flooding and salinity, respectively) and zonation among intermediate elevations by interspecific competition [Armstrong *et al.*, 1985; Bertness, 1991a, 1991b; Pennings and Callaway, 1992; Ward *et al.*, 2003; Pennings *et al.*, 2005]. Pattern outcomes are also influenced by nutrient availability [Hanson, 1977; Mendelsohn, 1979; Osgood and Zieman, 1998; Rogers *et al.*, 1998; Emery *et al.*, 2001], although even nitrogen fixation may be mediated by abiotic sediment properties [Moseman, 2007]. For salt marshes, a spatial correspondence between vegetation zones and discrete elevation platforms within the intertidal range has now been identified as a signature of the simultaneous presence of multiple stable biogeomorphic ecosystem states [Marani *et al.*, 2013]. These states are maintained by a positive feedback, promoting sedimentation among plant stems, and a linked negative feedback, limiting sedimentation or even causing erosion adjacent to plant patches, especially among salt marsh *Spartina* spp. grasses [Ward *et al.*, 2003; van de Koppel *et al.*, 2005; Fagherazzi *et al.*, 2006; Morris, 2006; Marani *et al.*, 2007; Temmerman *et al.*, 2007; van Wesenbeeck *et al.*, 2008a, 2008b; Bouma *et al.*, 2009]. Similar biogeomorphological concepts are beginning to be expanded to other intertidal systems such as coastal fluvial deltas [Smith, 2014].

Even as the links between vegetation and the physical organization of intertidal landscapes become better understood, there has been comparatively little documentation of potential links between vegetation and the geochemical organization of the intertidal zone. For the much-studied salt marsh grass *Spartina alterniflora*, a positive feedback between plant growth and sediment oxidation is well-known to exist [Howes *et al.*,

1981; Mendelsohn *et al.*, 1981], with consequent effects on sediment biogeochemistry. Many studies document correlations between sediment chemistry and salt marsh plant tissue chemistry, e.g., due to metal uptake by the plants (phytoremediation) in vegetated compared to unvegetated plots [Caçador *et al.*, 1996; Doyle and Otte, 1997; Kostka *et al.*, 2002; Koretsky *et al.*, 2003, 2005; Roychoudhury, 2007; Cambrollé *et al.*, 2008; Reboresda *et al.*, 2008]. Few studies have found differences in bulk sediment metal concentrations among multiple salt marsh vegetation zones [Caetano *et al.*, 2008; Cambrollé *et al.*, 2008; Caçador *et al.*, 2009], with two exceptions [Cambrollé *et al.*, 2008; Santos-Echeandía *et al.*, 2010].

The available data do not thoroughly test the potential for there to exist geochemical variations between multiple adjacent vegetation zones in a largely unpolluted salt marsh. Such variations might take the form of horizontal marsh pore water differentiation between vegetation zones or vertical geochemical differentiation within the root zone. The likelihood for such geochemical differentiation to exist is strongly suggested by empirical data and modeled values of the ecohydrological zonation of salt marshes [Moffett *et al.*, 2010a; Cao *et al.*, 2012; Xin *et al.*, 2013a; Wilson *et al.*, 2015] and of the role of both rooting depth and sediment hydraulic properties on the concentration of aqueous constituents such as methylmercury [Zhang *et al.*, 2014]. These studies also suggest that such geochemical differentiation, if present, may correspond to a combination of the spatial patterns of vegetation zonation and of hydrologic influences [Moffett *et al.*, 2012; Wilson *et al.*, 2015].

This study tested the hypotheses that the pore water geochemistry of the salt marsh root zone is: (a) horizontally differentiated in a manner strongly corresponding to the vegetation zonation, (b) vertically differentiated in a manner relating to root influences, and (c) influenced by marsh hydrological dynamics to a limited degree, less so than by vegetation zonation, especially given zone-specific plant water uptake. The study was conducted in a largely undisturbed and unpolluted environment with complex vegetation zonation on a nearly flat high marsh plain in South San Francisco Bay. The study undertook a multidepth, multi-constituent survey of pore water across five salt marsh vegetation zones at the same intertidal elevation. Transects were sampled both parallel to and perpendicular to marsh tidal channels to help test the potential influence of hydrologic regime on root zone pore water geochemistry in addition to the potential relationships to vegetation zonation.

2. Methods

2.1. Study Area and Vegetation Zones

The study area was an intertidal salt marsh in far southern San Francisco Bay, California (37°27'54" N, 122°6'58" W, Figure 1). The site covered 0.96 ha and was contained between levees, a bounding channel, and San Francisco Bay. Levee construction to reroute San Francisquito Creek adjacent to the site in the 1920s changed the site hydrology and allowed development of the site's bounding channel. It also resulted in the atrophy of a larger historical tidal channel, which became the smaller channel bisecting the modern site [Moffett *et al.*, 2012]. Since the borrow-ditch dredging methods of levee construction at the time were unlikely to impact the marsh more than a few meters from the levee, the site was minimally disturbed compared to most regional marshes. The site remained otherwise undisturbed and became part of the Palo Alto Baylands Nature Preserve as it was established between the 1920s and 1950s.

The climate of the site is Mediterranean, with October–April winter precipitation, dry May–September summers, and average evapotranspiration rate of 2.7 mm/d during daylight in the late summer, for a daily water loss of about 3 cm/d [Moffett *et al.*, 2010b]. The average elevation of the marsh plain is 1.02 ± 0.06 m above mean sea level (mean \pm 1 std. dev.). The marsh is typically flooded once-per-day during the spring portion of the mixed semidiurnal tidal regime, but can remain exposed for multiple days at a time during neap tides, with only the channels filled with bay water at neap high tides. The bay water is slightly fresher than average sea water and is well-mixed (bay salinity typically 25–30 PSU) [Conomos, 1979; USGS, 2007]. Some rainfall occurred just prior to the study period (supporting information Table S1), as measured at a weather station 200 m to the WNW of the site (supporting information Figure S1). Surface water levels, temperature, and salinity were continuously monitored at the site throughout the study interval, spanning before and after a few days of intensive pore water sampling in January 2008.

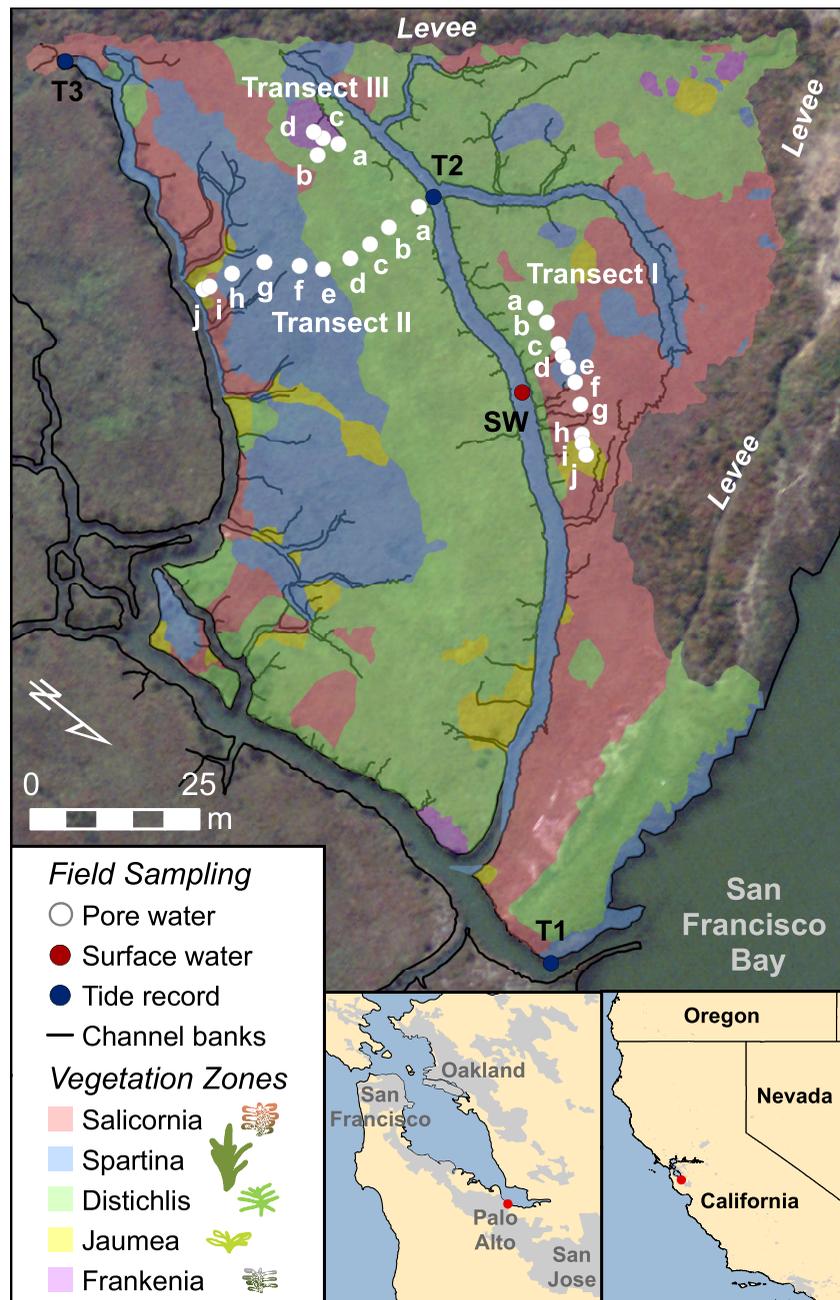


Figure 1. Map of the study area in far southern San Francisco Bay (red dot location in insets) and locations of the field samples collected from among the major vegetation zones of the site.

The patterns of vegetation zonation at the field site and the vegetation's relationship to site hydrology were previously described [Moffett et al., 2010a, 2012], as were specific aspects of plant water use, patterning, and marsh hydrology [Moffett et al., 2008, 2010b; Moffett and Gorelick, 2012]. Geochemistry at the site was not previously explored except as related to methylmercury [Zhang et al., 2014]. The plant species identified at the site were: *Spartina foliosa* (with some possible admixture/hybridization with *Spartina alterniflora*), *Salicornia pacifica* (*S. virginica*), *Distichlis spicata*, *Jaumea carnosa*, *Grindelia stricta*, *Frankenia salina*, *Salsola soda*, and *Atriplex prostrata*. In some years, the site was mildly affected by *Cuscuta salina*. The five major vegetation zones, delineated according to the dominant plant genus, were *Spartina*, *Salicornia*, *Distichlis*, *Jaumea*, and *Frankenia* (Figure 1) [Moffett et al., 2010a]. For the purposes of this study, predominant rooting depths within these vegetation zones were considered to be: 20–30 cm for *S. foliosa* [Mahall and

Park, 1976b; Boyer *et al.*, 2000], 10–25 cm for *S. pacifica* with a few deeper roots possible to >50 cm [Chapman, 1938; Purer, 1942; Mahall and Park, 1976a; Seliskar, 1983; Justin and Armstrong, 1987; Zedler and West, 2008], 25–30 cm for *D. spicata* [Purer, 1942; Seliskar, 1983], 40 cm for *J. carnososa* [Brown and Bledsoe, 1996], and 5–20 cm for *F. salina* with tap roots possible to nearly a meter [Purer, 1942; Brightmore, 1979].

2.2. Pore Water and Surface Water Sampling and Analysis

Sampling on 26–30 January 2008 during low tides mapped the variations in pore water geochemistry at high spatial resolution across known vegetation zone boundaries. As in Figure 1, transects are denoted by roman numerals (I, II, III) and sample locations within transects by lowercase letters (a, b, c, d, e, f, g, h, i, j); a sample location is uniquely identified by the combination, e.g., location Ia. Transect I spanned vegetation zones dominated by: *Distichlis* (locations I: a–b), *Spartina* (I: d–e), *Salicornia* (I: f–h), and *Jaumea* (I: i–j). Transect II spanned zones: *Distichlis* (locations II: a–d), *Spartina* (II: f–h), and *Salicornia* (II: i–j). Transect III spanned zones: *Distichlis* (locations III: a–b) and *Frankenia* (III: c–d).

The sampling locations within each transect were 1–5 m apart. At each location, pore water samples (~12 mL) were extracted under suction using a sipper (finely slotted stainless steel gas chromatograph tubing with crimped lower end and syringe attached to upper end). Sampled depths were 8, 14, 20, 26, 32, 38, and 44 cm below ground surface. Each pore water sample was immediately filtered into vials preacidified with trace-metal grade hydrochloric acid. The number of samples at each depth from each vegetation zone varied, with an average of about five samples from each depth per vegetation zone, overall (supporting information Table S2). Surface water samples were collected by filling 1 L bottles from the central channel (Figure 1) during flood and ebb tides. Ten well-mixed subsamples of these bulk surface water samples were filtered and acidified within a week of collection.

Dissolved element concentrations are denoted by their standard symbols. Total elemental concentrations of boron (B), barium (Ba), calcium (Ca), chromium (Cr), iron (Fe), mercury (Hg), potassium (K), lithium (Li), magnesium (Mg), manganese (Mn), nickel (Ni), phosphorus (P), sulfur (S), silicon (Si), strontium (Sr), and zinc (Zn) in pore water and surface water samples were measured using the Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES) of the Stanford Environmental Measurement I facility. Due to its high abundance in the salt marsh pore waters, sodium (Na) concentrations could not be accurately determined at dilutions suitable for measuring the other constituents. Concentrations of Cr and Hg were below detection and so are not reported. Values above known linear instrument calibration ranges achievable during the study period for some elements (> 300 mg/L Ca, > 250 mg/L K, > 500 mg/L S, > 5 mg/L Sr) were retained but are interpreted in a relative sense only. Three (of 20) flood tide Mn concentration measurement values (0.0168, 0.0173, 0.0174 mg/L) were below detection (0.018 mg/L) and three (of 163) pore water Zn values (0.0007, 0.0008, 0.001 mg/L) were below detection (0.001 mg/L), but these values were retained in the calculations (rather than omitting or counting as zero values) so as not to skew the means and medians. Pore water and surface water elemental concentrations are reported throughout this paper in mg/L, i.e., mg of total dissolved element per L solute.

2.3. Sediment and Evapotranspiration Variations Within Sampling Transects

The marsh soils were all estuarine clay (62% clay) [Moffett *et al.*, 2010a] but known to be heterogeneous with respect to their hydraulic properties [Moffett *et al.*, 2012]. All of Transects I and III and a portion of Transect II: a–f were considered to be within a zone of lower hydraulic conductivity sediments located within the central marsh. Only the portion of Transect II: g–j was considered to be among higher hydraulic conductivity sediments. However, boundaries between sediment zones were interpreted and interpolated, and so may be uncertain at a given location [Moffett *et al.*, 2012]. Overall, we consider Transect I, parallel to a tidal channel, an experimental treatment with uniform sediment properties and uniform external hydrological influences along its length. We consider Transect II, between two tidal channels, as spanning spatial heterogeneity in sediments and in external hydrological influences.

As a consequence of regular tidal inundation and a low permeability levee on the inland side of the marsh, groundwater in the marsh is known to be fully saline. For some distance inland (>1 km) and to depths much greater than the root zone sampled by this study (>10 m), there is no fresh groundwater influx or storage [Hamlin, 1983; Reymers and Hemmeter, 2001]. There is also no fresh surface water discharge to the

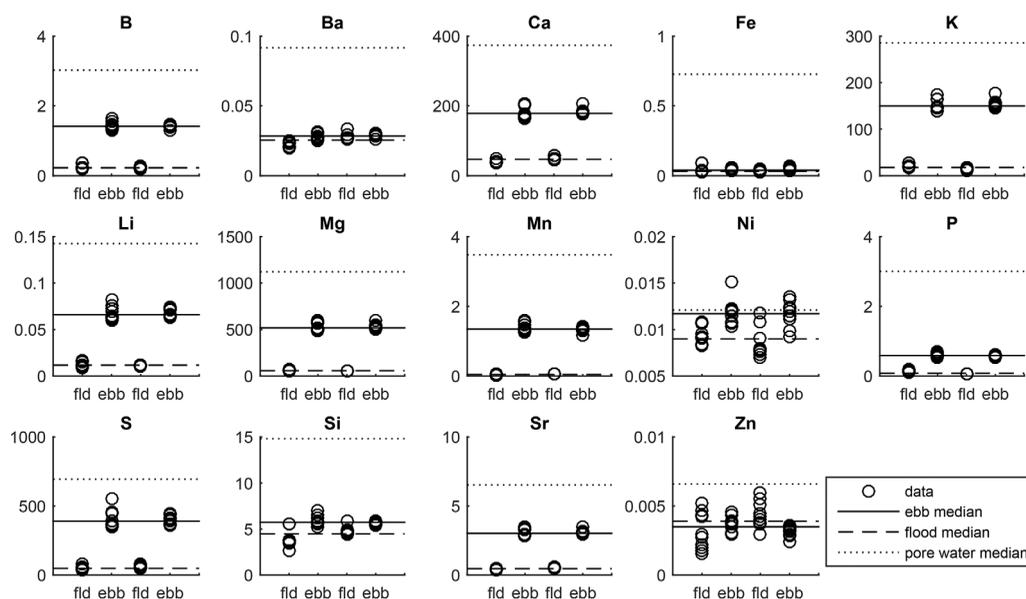


Figure 2. Surface water chemistry during flood and ebb of two tidal cycles. Element concentrations (y axes) in mg/L. Results of 10 subsamples of each tide (circles). Flood and ebb medians were significantly different ($p < 0.05$) except for Fe and Zn.

site. There was substantial rainfall (37 mm) on the day before the pore water sampling period (supporting information Table S1).

Superimposed on the pattern of sediment heterogeneity was a pattern of spatially variable plant evapotranspiration corresponding to the vegetation zones [Moffett *et al.*, 2012]. Prior study found that the continual withdrawal of water from low-permeability marsh sediments by plant root uptake and evapotranspiration would maintain persistent unsaturated conditions in the shallow root zone in some locations, even during tidal flooding [Moffett *et al.*, 2010b; Zhang *et al.*, 2014], consistent with known mechanisms [e.g. Dacey and Howes, 1984; Ursino, 2004; Li, 2005; Wilson and Gardner, 2005; Xin *et al.*, 2013a]. Based on the simulation results by Zhang *et al.* [Zhang *et al.*, 2014] for this field site, Transect I: a–e, Transect II: a–f, and all of Transect III were considered areas that maintained persistent unsaturated conditions in the root zone. In contrast, Transect I: f–j and Transect II: g–j were considered areas that remained saturated or became resaturated during each flooding tide.

3. Results

3.1. Surface Water Chemistry

Notable differences were observed between flood and ebb tide surface water chemistry (Figure 2). Flood tide surface water was sourced from far south San Francisco Bay and ebb tide surface water was influenced by drainage from the marsh site. The median concentrations in the waters collected from the tidal channel during ebb tide were significantly higher than the median flood tide concentrations for most elements (two-sample t test, $p < 0.05$). Only for Fe and Zn were the ebb and flood water concentrations insignificantly different. For almost every element, surface water concentrations were lower than the median pore water concentration (Figure 2 and Table 1).

3.2. Pore Water Geochemical Patterns

Both horizontal and vertical pore water geochemical differentiation were apparent in the data. Patterns varied by element: e.g., dissolved Fe notably accumulated at intermediate depths in the root zone while P was notably depleted at shallow depths. Pore water geochemical signatures strongly differed among vegetation zones. On average, pore waters beneath the Jaumea zone were the most concentrated in most elements at most depths and pore waters beneath Frankenia the least concentrated. Major biologically relevant cation results are presented in detail in Figure 3a and results for the additional analyzed elements in Figure 3b. All data are summarized in Figure 4.

Table 1. Pore Water and Surface Water Total Elemental Concentration Summary Statistics (mg/L)

Element	Pore Water (N = 163)				Surface Water Flood Tide (N = 20)				Surface Water Ebb Tide (N = 20)			
	Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
B	2.808	3.023	1.588	4.016	0.236	0.224	0.181	0.351	1.419	1.411	1.281	1.635
Ba	0.092	0.092	0.007	0.207	0.025	0.025	0.020	0.034	0.028	0.028	0.026	0.032
Ca	367.8	373.9	137.0	648.9	45.47	46.62	37.17	57.67	182.2	178.2	165.6	206.6
Fe ^a	4.397	0.727	0.033	41.91	0.039	0.033	0.025	0.091	0.043	0.038	0.035	0.066
K	292.6	285.5	141.7	524.7	18.00	17.30	12.37	27.54	153.0	149.6	139.5	178.1
Li	0.148	0.143	0.044	0.270	0.012	0.012	0.009	0.017	0.068	0.066	0.060	0.082
Mg	1104	1122	425.6	1931	58.41	58.90	50.81	69.43	529.5	517.9	485.8	599.7
Mn ^b	4.355	3.475	0.024	14.87	0.042	0.047	0.017	0.078	1.361	1.347	1.190	1.609
Ni ^c	0.015	0.012	0.006	0.146	0.009	0.009	0.007	0.012	0.012	0.012	0.009	0.015
P ^d	3.505	3.002	0.013	14.23	0.094	0.084	0.045	0.186	0.592	0.588	0.526	0.684
S	704.3	694.5	225.1	1420	53.02	48.94	41.09	76.99	404.6	390.5	350.4	550.0
Si	14.50	14.80	4.267	22.90	4.378	4.484	2.715	5.868	5.791	5.744	5.275	6.994
Sr	6.501	6.540	2.370	11.73	0.479	0.460	0.430	0.550	3.092	3.030	2.830	3.510
Zn ^e	0.009	0.007	0.001	0.032	0.004	0.004	0.002	0.006	0.004	0.004	0.002	0.005

^aThe pore water Fe mean was skewed relative to the median by high values in many specific sampling locations.

^bThree flood tide Mn values (0.0168, 0.0173, 0.0174 mg/L) were below detection (0.018 mg/L) but retained in the calculations (rather than omitting or counting as zero values) so as not to skew the mean and median toward higher values. Five additional flood values and one pore water value were above detection but less than twice the detection limit, so may have carried higher uncertainty but were retained as is.

^cTwo ebb tide Ni values, 15 flood tide values, and 46 pore water value were above detection (0.005 mg/L) but less than twice the detection limit.

^dOne pore water P value was above detection (0.007 mg/L) but less than twice the detection limit.

^eThree pore water Zn values (0.0007, 0.0008, 0.001 mg/L) were below detection (0.001 mg/L) but retained in the calculations. Three flood tide values and 21 additional pore water values were above detection but less than twice the detection limit.

Differentiation of pore water geochemical signatures among vegetation zones was particularly evident along Transect I, which was oriented parallel to the tidal channel, and Transect III, which spanned a *Distichlis*/*Frankenia* zone boundary (Figure 3). Within Transect I, horizontal pore water geochemistry beneath the *Jaumea* zone was more concentrated in B, Ca, K, Mg, S, Si, Sr than beneath other vegetation zones. Pore waters beneath the *Spartina* zone were more dilute in B, Ca, Fe, Li, Ni, and S. In general, pore water concentrations in Transect I followed the trend: *Jaumea* > *Salicornia* > *Distichlis* > *Spartina*. Transect III exhibited even clearer horizontal differentiation of pore waters, with concentrations among *Distichlis* almost uniformly greater than among *Frankenia*.

Along Transect II, which spanned between two tidal channels, horizontal patterns in pore water geochemistry were less starkly associated with plant zonation than in Transect I and III (Figure 3). Various elements (B, Ba, K, Li, P, Si) were generally more dilute in certain regions, such as within the *Spartina* and *Salicornia* zones close to the large tidal channel (locations II: g–j). Some elements (B, Ba, Ca, Mg, P, Si, Sr) were more concentrated at the inland edge of the *Spartina* zone, adjacent to the *Spartina*-*Jaumea*-*Distichlis* ecotone (location II: f).

In addition to the spatially explicit patterns captured in Figure 3, geochemical patterns associated with plant zonation appeared in median concentration–depth profiles (Figure 4). These aggregated profiles showed that the major differences in pore water geochemistry among vegetation zones were consistent across the field site, regardless of zone location. The distinct geochemical signatures of vegetation zones, visually apparent in Figure 4, were mostly significant according to pairwise Kolmogorov–Smirnov tests between profiles (supporting information Table S3). In particular, these tests confirmed that *Jaumea* and *Frankenia* root zone pore waters were geochemically distinct from the other vegetation zones with respect to most elements. In contrast, *Spartina* and *Distichlis* profiles were statistically almost indistinguishable, distinct only with respect to boron.

4. Discussion

4.1. Geochemical Signatures of Salt Marsh Vegetation Zones

Among many likely influences, near-surface oxidation was a dominant influence on the observed salt marsh root zone geochemistry. Given the high-resolution profile data, the oxic zone depth could be identified from the vertically variable pore water iron concentrations: *Blasco et al.* [2000] reported peak Fe as denoting the top of the anoxic zone. The profiles of Fe from our site highlighted different magnitudes and depths of

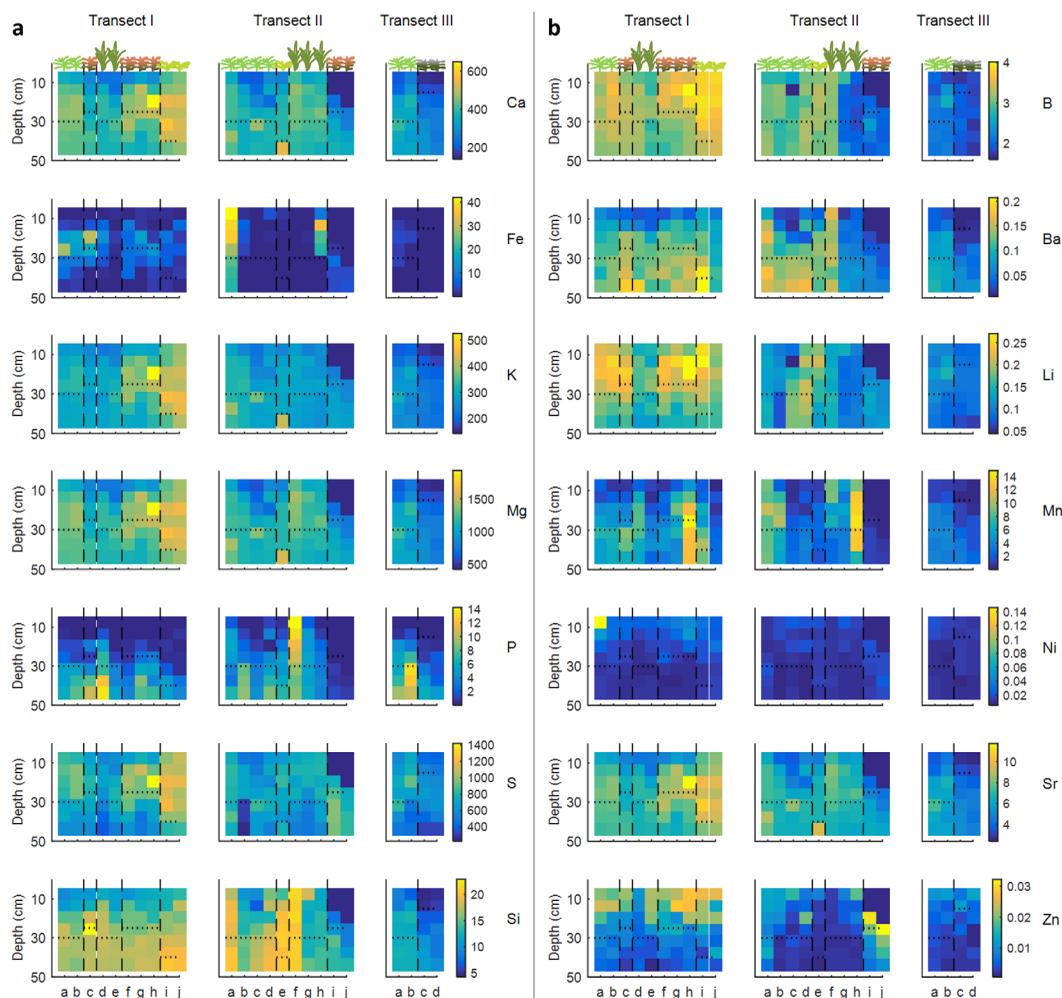


Figure 3. Pore water geochemistry patterns beneath Transects I, II, and III. Columns are: Transect I, parallel to a tidal creek; Transect II, perpendicular between two tidal creeks; and Transect III, spanning a patch boundary between *Distichlis* and *Frankenia* (see Figure 1). Colors and colorbars denote total pore water concentration in mg/L (at right). Y axes are depth below ground surface in cm (at left). X axes are location within the transect, not to scale (letters a–j at bottom). Vegetation zones are divided by vertical-dashed lines and distinguished by schematic symbols, as in legend of Figure 1. Zones, in order from left to right are: Transect I – *Distichlis*, *Salicornia*, *Spartina*, *Salicornia*, *Jaumea*; Transect II – *Distichlis*, *Jaumea*, *Spartina*, *Salicornia*; Transect III – *Distichlis*, *Frankenia*. Approximate depth of root influence indicated by horizontal dotted lines: 30 cm—*Distichlis*, 30 cm—*Spartina*, 25 cm—*Salicornia*, 15 cm—*Frankenia*, and 40 cm—*Jaumea*. (a) Concentrations of major biologically relevant cations. (b) Concentrations of other elements analyzed.

peak dissolved total Fe concentrations among vegetation zones, which suggested that the oxic zone depth differed markedly among vegetation zones (Figure 4). If the interpretation by Blasco et al. holds, then the *Salicornia*, *Jaumea*, and *Frankenia* zones had oxic conditions to 26 cm depth and the *Spartina* and *Distichlis* zones to 20 cm at this site.

Oxic conditions in the shallowest sediments were likely maintained both by diffusion and flushing. Any reduced Fe in solution within the shallowest (~0–17 cm) sediments was likely reprecipitated (oxidized) around plant roots, facilitated by oxygen diffusion out of the roots [Sundby et al., 1998, 2003] maintaining low pore water concentrations. Pore water Fe concentrations then peaked at intermediate depths (20–26 cm). Here, at the base of the oxic zone, Fe remained in solution at high concentration and so must have been reduced but not reoxidized/reprecipitated. The accumulation of dissolved Fe at these intermediate depths, compared to shallower depths, was likely due to the progressively reduced root mass and perhaps less active root-induced sediment oxygenation with depth. This effect is consistent with literature documenting more Fe-plaques around the older, upper portions of wetland roots than the deeper, younger root tips [Seyfferth et al., 2010]. However, the redox processes suggested by the pore water Fe

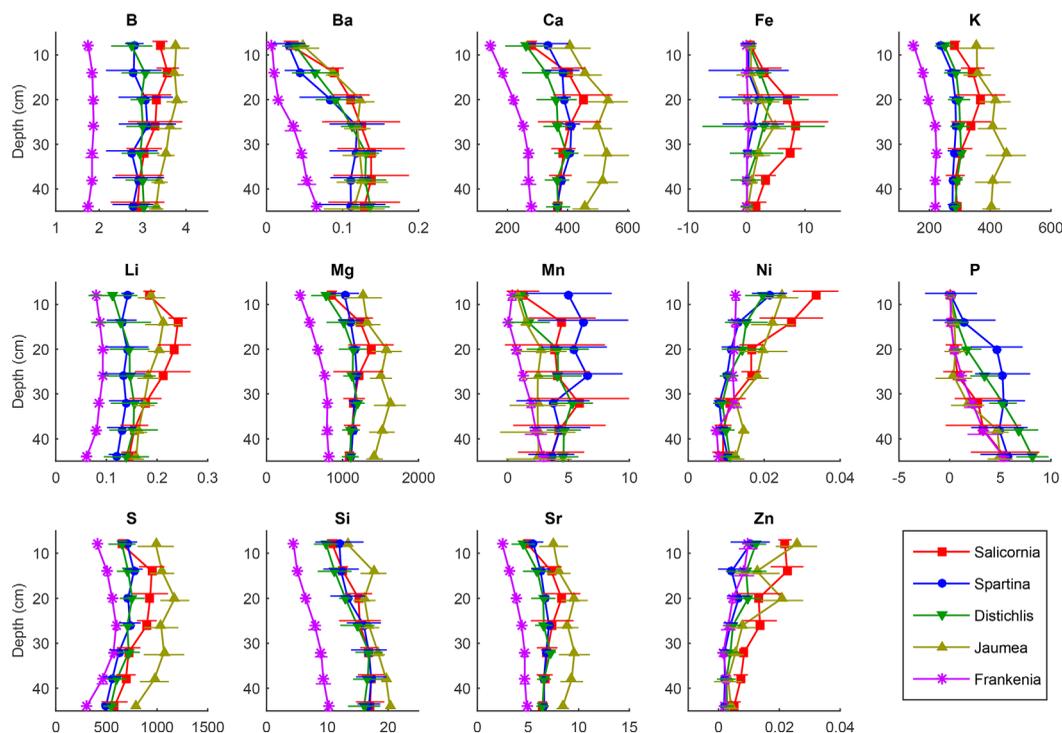


Figure 4. Vertical profiles of median pore water geochemistry, colored by vegetation zone. Profiles shown are median total element concentrations (x axis, in mg/L) at each depth (y axis, in cm) from all field locations sampled within each vegetation zone. Horizontal error bars illustrate the interquartile range (IQR) of the data around each median point. IQR bars are offset slightly vertically to avoid superposition. Note that profiles can be significantly different overall, even with partial overlap of their IQR bars at some depths (see Kolmogorov-Smirnov profile separation test results, supporting information Table S3).

concentrations were not necessarily good explanations for patterns in other elements, as Fe concentrations were only weakly correlated with other elements (supporting information Table S4).

Patterns of the primary plant macronutrient potassium appeared related to the vegetation zonation. Profiles of K were distinct among all zones except Spartina and Distichlis, with root zone pore water concentrations generally in the order Jaumea > Salicornia > Spartina and Distichlis > Frankenia. Incidentally, B and S exhibited almost the same pattern of pore water discrimination on average as K, in the same hierarchical order (Figure 4). These results suggest some species-specific and ion-specific salt management, perhaps more effective ion exclusion by Jaumea roots and less effective by Frankenia.

In contrast to clear zonal differentiation with respect to potassium, the primary plant macronutrient phosphorus exhibited more uniform patterns across the site. Pore water P concentrations increased with depth among all zones (Figure 4) and any apparent differences were not statistically significant according to the Kolmogorov-Smirnov tests (supporting information Table S3). The speciation of P within marsh sediments is known to covary with salinity, e.g., more inorganic P among salt marshes and more organic P among freshwater marshes [Paludan and Morris, 1999]. However, the salinity of our site was consistently high throughout. That P concentrations were much greater at depth across all vegetation zones, especially below 30 cm within Transects I and III (Figure 3), was likely due to high P utilization within the root zone and either underutilization at, or leaching to, deeper depths.

Patterns of secondary plant macronutrients Ca, Mg, and S had similar median profile shapes (Figure 4). All three elements were found in the highest concentration in the Jaumea zone, lowest in the Frankenia zone, and intermediate among Salicornia, Spartina, and Distichlis. For all three elements, median pore water concentrations increased somewhat with depth until about 26 cm and then remained constant or decreased slightly at greater depths. Prior studies reported distinct peaks in pore water sulfate or hydrogen sulfide with depth [Kostka et al., 2002; Koretsky et al., 2003; Roychoudhury, 2007], but we did not observe this. Our total-element analysis combined all dissolved S species, and so our diffuse peaks at intermediate depths

were likely the sum of a shallower sulfate peak and a deeper dissolved sulfide peak. Another factor potentially contributing to our smooth S profiles is that a temporally averaged concentration of total S in profile may be inversely related to the sediment sand/clay ratio due to the texture-associated organic carbon concentration [Gardner *et al.*, 1988], but our sediments were quite uniformly clay through the sampled depths.

Dissolved silicon can also be considered an important plant nutrient. In this study, Si exhibited median profile patterns that were a hybrid between those of P and of Ca/Mg/S. Like P, Si increased with depth among all vegetation zones but, like the other macronutrients, Si was most concentrated in pore waters in the Jaumea zone, least in the Frankenia zone, and intermediate among *Salicornia*, *Spartina*, and *Distichlis*.

Combining considerations of both vertical and horizontal pore water differentiation, as enabled by the high spatial resolution of data, yielded further interesting insight into potential geochemical “hot spots” in the marsh sediments. The overall concentration patterns in Ca, Mg, Sr, K, and S exhibited strikingly similar two-dimensional patterns within Transect I and to a lesser extent within Transect II. All five elements exhibited a two-dimensional “hot spot” of higher pore water concentrations at intermediate depths beneath *Salicornia* and *Jaumea* (Figure 3 Transect I), with high concentrations extending somewhat deeper beneath *Jaumea*. All five elements exhibited relatively lower concentrations (a “cold spot”) at shallow depths beneath *Spartina*, among other similarities (Figure 3 Transect I). This correspondence was borne out statistically by correlations among the elements (supporting information Table S4). Further study is required to determine whether these similarities in the patterns of Ca, Mg, Sr, K, and S pore water concentrations might have been caused by chemical interactions among the elements or by joint response to external influences such as sediment mineralogy, or ion-specific plant uptake or exclusion.

4.2. Temporal Considerations

Despite this remarkable spatial correspondence among elemental patterns, the geochemical results of this study provide temporally limited insight into what may be dynamic patterns also influenced by seasonality and phenology. Due to the low permeability of the clay marsh sediments [Moffett *et al.*, 2012], fast movements of relevant water volumes through the subsurface are not possible, especially in the marsh interior. Only in the creek banks is there evidence that the greater surface water-groundwater interface area (surface plus bank) and likely locally concentrated macroporosity may have enabled more rapid sediment drainage and flushing on tidal timescales, but only in the few meters or so closest to the tidal creeks [Gardner, 2005; Wilson and Gardner, 2006; Moffett *et al.*, 2012]. Pore water residence times in the marsh interior are on the order of tens of years, based on prior study at the site and modeling from other salt marsh environments [Wilson and Gardner, 2006; Moffett *et al.*, 2012]. It is likely, therefore, that the pore water geochemical patterns reported here were generally representative of at least the sampled season (winter of 2008).

That such distinct geochemical zonation with high correspondence to vegetation zonation and rooting depth was observed in the winter season is notable, as a substantial fraction of the aboveground macrophyte biomass had senesced, and root-associated microbial activity in the sediments was likely reduced due to the seasonally lower ambient temperatures. However, in the mild Mediterranean climate, some live macrophyte biomass was persistent across the marsh even throughout the winter season, including portions of perennial *Salicornia* and *Distichlis* stems and winter *Spartina* sprouts likely taking advantage of light afforded by the higher canopy's seasonal dieback. Therefore, although reduced, plant water uptake was not zero, and differential effects of each vegetation zone's canopy (live and/or dead-standing) on surface evaporation would also have persisted throughout the study period. The study by Gardner and Reeves [Gardner and Reeves, 2002] of the seasonal water balance of a marsh in another mild climate (South Carolina, USA) similarly reported persistent evapotranspiration, at a low but nonzero level, through the winter. Although results from other seasons outside the scope of the study would likely differ, we hypothesize that a stronger influence from differential plant water uptake and differential macrophyte solute management among the vegetation zones during the summer might actually strengthen many of the patterns observed in this winter-season data, rather than soften them.

Although our results indicate substantially elevated solute concentrations in marsh pore waters compared to the water sources, bay water (see Figure 2) and rainwater, it seems unlikely that marsh concentrations would increase indefinitely. This single field study was unable to determine possible longer term trends in solute concentrations in the marsh, but there are likely multiple mechanisms that balance a tendency toward continued pore water concentration over longer timescales. These include site-scale groundwater flow and

eventual discharge [e.g., as in *Robinson and Gallagher* [1999]; salt fingering, whereby if pore waters become sufficiently salty and dense as to be unstably stratified, they may locally overturn with underlying pore waters and so remove the salinity from the near-surface [e.g., as in *Shen et al.*, 2015]; surficial diffusion and flushing on the tidal timescale [e.g., *Moffett et al.*, 2010a]; plant uptake and then system loss as litter is washed away; and direct erosion of salinized sediments. Further research is required to understand the influences of such mechanisms on each other and on the spatiotemporal patterns of marsh pore water geochemistry over annual and longer timescales.

4.3. Pore Water Geochemical Patterns Among Different Hydrological Regimes

Prior study at the field site has documented spatially distinct ecohydrological zonation due to the combined influences of three hydrological factors: zonal plant water uptake, heterogeneous sediment hydraulic properties, and spatially variable hydrological flushing [*Moffett et al.*, 2010a; *Zhang et al.*, 2014]. The conclusions of those studies emphasized that the physical hydrology of the marsh, especially of the marsh plain away from the tidal channels, cannot be fully understood as a sum of just tidal and hydrogeological factors, without also accounting for a prominent role of the spatially variable evapotranspiration effects on the marsh water balance. Here we examined whether these three factors were also related to the observed spatial variations in pore water geochemistry. Transect I (and most of Transect III) was intentionally oriented parallel to a tidal creek, whereas Transect II was placed to span the marsh plain perpendicularly between two tidal creeks. Based on field observations during this study and on prior modeling results for the site [*Moffett et al.*, 2012; *Zhang et al.*, 2014], Transects I and III experienced surface water flooding at nearly the same times and to the same degrees throughout their lengths but were too distant from the tidal channel to experience tidally driven groundwater drainage and flushing. In contrast, Transect II experienced more rapid flooding and more groundwater drainage toward both ends of the transect.

4.3.1. Hydrological Influence on Transect I and III Geochemistry

Transects I and III exhibited stronger vertical pore water geochemical differentiation within the root zone and more clear horizontal differentiation among vegetation zones, compared to Transect II. Given the relative homogeneity of sediments and consistent distance-to-channel of Transects I and III, spatially variable evapotranspiration was the factor likely to have influenced the pore water geochemistry patterns most strongly. Withdrawal of pore water from the root zone to sustain daytime evapotranspiration, even during flooding tides, was a key influence of the vegetation [*Dacey and Howes*, 1984; *Ursino*, 2004; *Li*, 2005; *Wilson and Gardner*, 2005; *Moffett et al.*, 2012; *Xin et al.*, 2013a]. Across all three transects, most elements (Ca, K, Li, Mg, Mn, S, Sr, Zn) had lower average concentrations in areas that remained persistently unsaturated due to this plant water uptake (as simulated at this site by *Zhang et al.* [2014]) and higher concentrations in areas that were saturated during flood tides. Only for P was this trend reversed, with lower average concentrations in areas that remained saturated and higher concentrations in areas that were kept persistently unsaturated by plant water uptake.

The lower concentrations in areas that remained persistently unsaturated were likely due to partial flushing by tidal waters or rainfall and were enabled by prior evapotranspiration. These less concentrated water influxes would partially displace the airspace created by root water uptake. This pore-scale mixing would generally reduce the pore water element concentrations, even if the sediments did not become fully resaturated upon flushing [*Boaga et al.*, 2014]. Such partial flushing may be particularly active in the larger soil pores that are more likely to be sampled by the suction lysimeter methodology employed [*Harvey and Nuttle*, 1995; *Harvey et al.*, 1995]. This mechanism appeared particularly relevant for causing the less concentrated pore water in the surficial layer (~8 cm) of sediments (see Figure 3). Prior study of this field site has highlighted the ability for transient tidal flooding to influence near-surface saturation and salinity [*Moffett et al.*, 2010a].

The opposite patterns in P, with higher concentrations in areas maintained unsaturated by plant water uptake, may be explained by the partial flushing replenishing P that would otherwise be depleted by biotic uptake. This proposal of partial P replenishment by flood infiltration echoes that from a South Carolina marsh [*Dame and Gardner*, 1993]. It is also similar to an idea by *Gardner* [*Gardner*, 2005], who noted the possibility that surface water infiltration into the marsh root zone could supply sufficient oxygen and sulfate to support substantial organic matter decomposition. Indeed, in the areas that remained saturated, low P values observed at our site were likely due to the combination of biotic depletion and reduced flushing. In the study by *Zhang et al.* at this field site [*Zhang et al.*, 2014], the methylmercury concentration was also found

to be more elevated in persistently unsaturated areas of the marsh compared to areas that remained saturated. This was likely due to a different mechanism, however, whereby labile organic root exudates promote Hg methylation via changes to the local rhizospheric microbial metabolism [Windham-Myers *et al.*, 2009], which process was perhaps more active in the persistently unsaturated areas than in the saturated areas of the marsh.

In general, therefore, pore water geochemical differentiation within Transects I and III was more related to vegetation zonation and differential plant species influences on the chemistry, including via plant water uptake, rather than to externally imposed or preexisting hydrological influences such as tidal forcing or sediment heterogeneity. The scope of this study, of macroscopic pore water geochemistry patterns at the scales of vegetation patches to vegetation zones, did not resolve data potentially useful for diagnosing specific plant root influences on sediment chemistry. This limitation could be overcome in future work by activities including collection of root samples, collection and analysis of relevant compounds such as root exudates and organic acids, and measurement of geochemical gradients around roots at the micron-to-mm scale.

4.3.2. Hydrological Influence on Transect II Geochemistry

Transect II experienced notable creek bank drainage into the channels at its extreme ends after tidal flushing [Moffett *et al.*, 2012; Zhang *et al.*, 2014]. Lower concentrations of many elements (Ca, Mg, Mn, P, Si, Sr) are very likely attributable to this locally enhanced drainage and lateral flushing of the steeply cut bank [Jordan and Correll, 1985]. The flushing effect was mainly localized in the upper portion of the bank, within about 2 m of the channel (Figure 3 locations II: i–j). A related phenomenon was the accumulation of high concentrations of dissolved Fe and Mn just at the interior edge of the creek bank flushing zone (Figure 3 locations II: a and II: h), likely indicating the transition zone between more oxic, flushed creek-bank sediments, through a Fe and Mn reduction zone, into more anoxic conditions within the marsh interior.

Tidal creek bank flushing has previously been identified as a source of spatial variations in salt marsh sediment chemistry, but for much more highly bioturbated creek banks [Kostka *et al.*, 2002; Roychoudhury, 2007], wherein crab burrows can greatly enhance aeration and flushing [Xin *et al.*, 2009]. Xin *et al.* [2013a] demonstrated in a general numerical model that the marsh close to tidal creeks is drained regularly by semidiurnal tidal fluctuations, to the benefit of vegetation growth, that marsh areas slightly further from the creek may be drained fortnightly during neap tides, but that interior marsh saturation is controlled by evapotranspiration. (Also see Xin *et al.* [2010, 2013b].) Surface water-groundwater exchange on a tidal timescale is of course greater with higher permeability sediments surrounding or outcropping to the tidal creeks [Wilson and Gardner, 2006], but is still concentrated within just a few meters of the creek banks in homogenous-sediment simulations [Gardner, 2005]. Prior hydrogeological modeling of this field site likewise simulated that lateral groundwater drainage during ebb tide was limited to within a few meters around the creek banks [Moffett *et al.*, 2012].

Most of Transect II was away from the creek banks and so the pore water patterns could not be accounted for by creek bank flushing. For some elements (B, Ba, Li, P) low concentrations extended 11 m into the marsh interior (through locations II: g–j), much too far to be accounted for by lateral flushing through the low-permeability clay sediments. Instead, these sediments in the central portion of the field site (locations II: a–f and all of Transects I and III) had higher clay and lower sand contents on average (62% clay, 36% silt, 1% sand) compared to the major creek bank (54% clay, 39% silt, 7% sand, locations II: g–j) [Moffett *et al.*, 2010a]. This suggests that the creek bank may have experienced higher mixing of pore waters with overlying tidal surface waters (likely by diffusion and small-scale convective overturn in macropores) [Harvey *et al.*, 1995; Rocha, 2000] compared to the less permeable marsh interior (II: a–f).

Studies of marshes that overlie sandy aquifers have indeed demonstrated substantial surface water-groundwater exchange, e.g., driven by storms [Wilson *et al.*, 2011], and numerical modeling studies have repeatedly demonstrated the important influence of sediment permeability and heterogeneity on marsh groundwater movement [Gardner, 2005, 2007; Wilson and Gardner, 2006; Moffett *et al.*, 2012a; Xin *et al.*, 2012; Zhang *et al.*, 2014]. These permeability-related effects are likely to be even more important in other locations with much higher sand fractions, such as in eastern United States [Osgood and Zieman, 1998, and references therein] or Australian salt marshes [Hughes *et al.*, 1998]. Among higher-permeability sediments, fluid mixing due to overturning density gradients near the surface (salt fingering) is also possible, especially

in the marsh interior regions [Shen et al., 2015; Gardner, 2005, 2007; Wilson and Gardner, 2006; Moffett et al., 2012; Xin et al., 2012; Zhang et al., 2014].

The differential accumulation or dilution of many elements in the two portions of Transect II therefore seemed to be strongly influenced by the combination of subsurface (sediment hydraulics) and surface (flooding and flushing) hydrology, in addition to the strong vegetation influences inferred from Transects I and III. This conclusion is consistent with previous work in this salt marsh, which showed that the physical dynamics of near-surface soil saturation, draining by evapotranspiration and filling by tidal waters (i.e., shallow surficial flushing), and vertical shallow groundwater flow direction are jointly controlled by the combination of local plant water uptake rates and the local sediment texture/hydraulics in which the plants are rooted [Moffett et al., 2012]. This study adds additional geochemical understanding to our conceptual model of the physical, chemical, and biological coorganization of a salt marsh.

4.4. Pore Water Drainage as an Influence on Surface Water Chemistry

Even as tidal hydrology influenced pore water geochemistry, at least in Transect II, marsh pore water geochemistry also appeared to influence ebb tidal chemistry. The concentrations of elements in ebb tide channel waters were significantly ($p < 0.05$) greater than in flood tide waters, in general. The difference between the ebb and flood data was only insignificant for Fe and Zn. Median concentrations in marsh pore waters were generally much greater than in either ebb or flood waters (Figure 2). Only for Ni did the median pore water concentration overlap with measured surface water concentrations. Higher aqueous metal concentrations in salt marsh pore waters compared to surface waters have previously been shown to induce rapid and substantial temporary fluxes of trace metals out of marsh sediments and into overlying surface waters during the onset of tidal flooding [Santos-Echeandía et al., 2010].

A simple mixing model was developed based on median ebb tide and pore water concentrations.

Model:

$$ebb_concentration = pw_fraction * pw_concentration + (1 - pw_fraction) * flood_concentration \quad (1)$$

with variables:

<i>ebb_concentration</i>	median of 20 ebb tide surface water measurements for each element
<i>flood_concentration</i>	median of 20 flood tide surface water measurements for each element
<i>pw_concentration</i>	median of 163 pore water measurements for each element
<i>pw_fraction</i>	fraction of ebb tide waters sourced from pore water [unknown solved for]
$(1 - pw_fraction)$	fraction of ebb tide waters sourced from the bay flood tide waters

The model was solved for the fraction of the ebb tide water in the channel originating from marsh pore water (*pw_fraction*) that would have been required, if supplied to the tidal creek at the median measured pore water concentration (*pw_concentration*), to mix with the preexisting water in the channel originating from the prior flood tide ($1 - pw_fraction$) at the median flood tide concentration (*flood_concentration*) in order to achieve the measured ebb tide concentration (*ebb_concentration*). Based on this model, marsh pore water drainage (*pw_fraction*) contributed approximately 41% of the solutes in the ebb tide waters leaving the marsh (median across all elements, Table 2). The estimated contribution of marsh drainage to ebb tide chemistry was similar to the median for many elements (38–53% for Mn, Ca, Li, Sr, B, Mg, K, and S), but much less for other elements (1–17% for Fe, Ba, Si, P). The 17% contribution of pore water drainage to P export is at the low end of a previously estimated range for a salt marsh (19–40%) [Jordan and Correll, 1985]. The marsh may have been a slight sink for Zn, on average, although Zn concentrations were generally near the detection limit and so absolute results somewhat uncertain. The marsh appeared to be a strong source of Ni, potentially accounting for 87% of Ni in ebb tide waters. This drainage of dissolved Ni from the marsh appeared to raise the aqueous concentrations from the 0.009 mg/L flood tide influent level to 0.012 mg/L in marsh ebb tide effluent. This effluent level was 1.5 times the current U.S. Environmental Protection Agency Criterion Continuous Concentration (0.0082 mg/L) for negligible long term effects in exposed saltwater aquatic organisms [EPA, 2015].

Table 2. Results of Pore Water-and-Surface Water Mixing Model Analysis

Element	pw_fraction
Ni	0.871
S	0.529
K	0.493
Mg	0.432
B	0.424
Sr	0.423
Li	0.415
Ca	0.402
Mn	0.380
P	0.173
Si	0.122
Ba	0.045
Fe	0.008
Zn	-0.148
Overall median	0.408

These simple mixing-model results may be indicative of short-term winter fluxes from southern San Francisco Estuary marshes, but elemental balances are likely different at other locations and other spatial and temporal scales. For example, Gardner and others found net dispersive fluxes of salt, nitrogen, phosphorus, and carbon to be not significantly different from zero over a 9 year time frame in a South Carolina coastal marsh [Gardner and Kjerfve, 2006], but statistically significant dissolved oxygen export over 3 years at the same location [Gardner et al., 2006], with diurnal variations depending on the timing of the day's tides [Gardner and Gorman, 1984]. Also, the spatial source area and temporal residence time of the pore water-derived solutes discharged to the tidal creek were unknown for this study. Numerical modeling has demonstrated that long bank length and close bank proximity promote the collection of pore water drainage in tidal creeks, more so than the relative channel size, but most creek drainage is sourced locally [Xin et al., 2011]. For this field site, it was previously

determined that both groundwater seepage and drainage from shallow depressions on the marsh surface likely contribute to the composition of ebb tidal waters [Zhang et al., 2014]. Although long recognized [Miller and Gardner, 1981], tidal exchange occurring as nonchannelized sheet-flow is often poorly quantified. The diffusion of constituents from surficial pore waters into thin films of overlying surface water can in fact account for an important fraction of the measured tidal discharge [Gardner, 1975], and also may explain the somewhat lower concentrations in the top-most sediment layers for some elements (Figure 3). For the purposes of the above mixing analysis, water that may have drained from surficial ponds was not explicitly modeled; if it was in equilibrium with pore waters then it was implicitly included in the total fractional volume attributed to groundwater drainage; if its chemistry remained similar to that of flooding tidal waters then it was implicitly included in the total fractional volume attributed to surface water.

5. Conclusions

The evidence from this study supports the three hypotheses tested, that the pore water geochemistry of the salt marsh root zone was (a) horizontally differentiated in a manner strongly corresponding to the vegetation zonation, (b) vertically differentiated in a manner related to root influences, and (c) less affected by marsh hydrology (flooding, sediment hydraulics) than by vegetation influence (water uptake, root zone aeration, and associated geochemical influences) in the regions of the marsh more than a few meters from the creek banks. Specific mechanisms that could account for the zonally differentiated pore water in the marsh interior were deduced, most of which related to species-specific patterns of plant water uptake, rooting depth, and root zone oxidation by subsurface plant aeration. Tidal flooding, flushing, sediment heterogeneity, and rainfall also influenced the pore water geochemical patterns, but predominately within just a few meters of the creek bank. Although soil texture does influence soil chemistry, vegetation zone-specific plant water uptake was a greater influence in this marsh.

Despite the low hydraulic conductivity of the clay marsh sediments, drainage of the concentrated marsh pore waters appeared to contribute 41% of ebb tide dissolved solutes, on average. The marsh appeared to be a possible sink for Zn but a strong source of Ni (87% of ebb export), possibly to a degree hazardous to aquatic organisms. Future sea level rise might intensify these exports [Wilson and Morris, 2012]. The influences of vegetation on pore water geochemical differentiation may, therefore, not only be significant locally within the marsh but may ultimately influence marsh-estuary solute exchange and ecology. Also, the persistent signal of marsh solute efflux over multiple tidal cycles suggests there are mechanisms for continual renewal of the "supply" of geochemically concentrated pore waters within the marsh, across a broad array of elements, with broad implications for a variety of elemental cycles and budgets in salt marshes.

The correlations identified in this study suggest that the presence of a given vegetation zone uniquely modifies the underlying pore water geochemistry in a manner different than an adjacent zone subject to nearly identical environmental controls. It remains to test in future study whether the resulting root zone

microenvironment may be more favorable to the stable persistence of that vegetation zone than to encroachment by other marsh species, and whether the observed correlations of zones and geochemistry in fact have causal links. If so, geochemical feedbacks would contribute to the self-organized vegetation zonation that can arise in salt marshes despite relatively spatially uniform initial environmental conditions. This mechanism would be analogous and complementary to the plant-sedimentation feedbacks known to support multiple stable biogeomorphic salt marsh states [e.g., Marani *et al.*, 2013]. Spatially variable environmental controls (e.g., heterogeneous groundwater gradients and salinity) have already been shown to be able to control marsh vegetation zonation [Moffett *et al.*, 2012; Wilson *et al.*, 2015]. Positive interspecific interactions among the species composing a zone are also likely an important and related factor in zone stability [Bertness and Hacker, 1994]. Explicitly combining physical, chemical, and biological mechanisms into a broader conceptual framework of marsh organization might be particularly useful for predicting marsh responses to localized patch-scale disturbances (whether natural or anthropogenic) more at the scale of vegetation patches and geochemical gradients, which has so far been less tractable than predicting coarser-scale biogeomorphic responses of whole marsh shorelines to exogenous influences such as changes in sea level and sediment supply [e.g., Mariotti and Fagherazzi, 2010].

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