EFFECTS OF EXTREME EVENTS ON ARSENIC

CYCLING IN SALT MARSHES

by

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A thesis submitted to the Faculty of the University of Delaware in partial fulfillment of the requirements for the degree of Master of Science in Plant and Soil Sciences

Fall 2017

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ACKNOWLEDGMENTS

I wish to thank my committee members, Dr. Angelia Seyfferth, Dr. Donald Sparks and Dr. Bruce Vasilas, for their encouragement and guidance during this experience. I would also like to thank Dr. Rodrigo Vargas for providing expertise and Maggie Capooci for directly contributing to this research. This work would not be possible without the sampling assistance from Frances Bothfeld, Julia Guimond, Erica Loudermilk, Gretchen Dykes, Patrick Wise, Fred Teasley, Heather Eby, and Alesia Hunter as well as the moral support from the entire Seyfferth lab.

Caroline Golt and the UD Soil Testing Laboratory were also instrumental for their analytical assistance with the numerous soil and water samples. Thank you to Kari St. Laurent and the other staff members of the St. Jones Estuarine Research Reserve for the use of their facilities.

A special acknowledgement goes out to my family members and significant other for their outstanding support through this journey. Despite the late nights and weekends in the lab or office, you have been so understanding and encouraging along the way.

This work was funded by the National Science Foundation Grant No. 1350580 and Grant No. 1652594. Data from the figures and tables can be obtained on the figures and tables and in the Appendix. Precipitation data was obtained from the St. Jones meteorological station and can be accessed at http://cdmo.baruch.sc.edu/get/export.cfm.

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ABSTRACT

Extreme events such as storm surges, intense precipitation, and supermoons cause anomalous and large fluctuations in water level in tidal salt marshes, which impacts the sediment biogeochemistry that dictates arsenic (As) cycling. In addition to changes in water level, which impacts soil redox potential, these extreme events may also change salinity due to freshwater inputs from precipitation or saltwater inputs due to surge. It is currently unknown how As mobility in tidal salt marshes will be impacted by extreme events, as fluctuations in salinity and redox potential may act synergistically to mobilize As. To investigate impacts of extreme events on As cycling in tidal salt marshes, we conducted a combined laboratory and field investigation. We monitored pore water and soil samples before, during, and after two extreme events: a supermoon lunar eclipse followed by a storm surge and precipitation induced by Hurricane Joaquin in Fall 2015 at the St. Jones Reserve in Dover, Delaware, a representative tidal salt marsh in the Mid-Atlantic United States. We also conducted soil incubations of marsh sediments in batch and in flow-through experiments in which redox potential and/or salinity were manipulated. Field investigations showed that pore water As was inversely proportional to redox potential. During the extreme events, a distinct pulse of As was observed in the pore water with maximum salinity. Combined field and laboratory investigations revealed that this As pulse is likely due to rapid changes in salinity. These results have implications for As mobility in the face of extreme weather variability.

Chapter 1

INTRODUCTION

Intense storms and the lunar orbit cause extreme fluctuations in coastal hydrology. These extreme events disproportionately disrupt tidal salt marshes where fresh and brackish water intersect (Capobianco et al., 1999). Water level in these salt marshes reaches radical heights at high and low tide during perigee, which is an event that occurs once a year when the moon is at its absolute closest to the earth (Morrow, 2015). At perigee, the moon appears approximately 14% larger, and hence is often referred to as a supermoon event. Aside from the effects of the lunar cycle, salt marsh water level is also influenced by large storm events. Hurricanes are well known for the flooding caused by heavy precipitation and in coastal zones, flooding is amplified by the tides. At high tides during an extreme storm as well as during a supermoon, water reaches abnormally high levels, covering land that does not usually experience inundation or rapid changes in salinity (Baldwin & Mendelssohn, 1998). Pore water salinity in the salt marsh may be increased by ocean water influx or decreased by precipitation. Pore water redox potential may be decreased by water inundation, or increased if the rising water is well oxygenated (Gong et al., 2007). Fluctuating salinity paired with the dynamic of changing redox potential trigger shifts in the biogeochemical cycling of redox-sensitive elements, such as arsenic (As).

Arsenic is a ubiquitous toxin that occurs naturally or anthropogenically in the environment and its cycling is largely driven by redox state. Under oxic conditions, As is usually strongly attenuated in soil because of its high affinity for iron (oxyhydr)oxides, manganese and aluminum oxides, and 2:1 phyllosilicate clays, depending on the As speciation (Al-Sid-Cheikh et al., 2015; Gu et al., 1994; Wu et al., 2015). Under suboxic conditions, the iron(oxyhydr)oxides and manganese oxides are reductively dissolved, releasing the previously sorbed As to interstitial pore water (Olivie-Lauquet et al., 2001). If strongly reducing conditions persist, arsenic may instead be sequestered by iron-sulfide bearing minerals (Wilkin & Ford, 2006). While the influence of redox potential on As partitioning is well known, it is still uncertain how As behaves upon shifting salinity. Arsenic can be mobilized via ligand exchange with sulfide on amorphous iron (oxyhydr)oxides (Kocar et al., 2010) or by exchange with other salt water oxyanions from organo-As complexes (Sinha & Bhattacharyya, 2011). However, high sulfate in the pore water may impede dissimilatory iron reduction, leading to more As retention with iron sulfides (Burton et al., 2013). Elucidating the impact of increasing salinity on As availability is critical for understanding potential As-induced toxicity to estuarine biota.

Salt marsh organisms could encounter geogenic As, which is naturally occurring in soil minerals, as well as As derived from anthropogenic activities. For example, As leaches from copper chromated arsenate (CCA)-treated wood, which is a common material for commercial boardwalks often present in salt marshes in the Mid-Atlantic (Vasilas et al., 2011; Weis & Weis, 1996, 2002). Soil As concentrations above the biological threshold of 41.6 mg As/kg soil (Jones et al., 1997) can be toxic to grazing organisms like fiddler crabs, which consume detritus by sifting through salt marsh sediments (Weis & Weis, 1992). Given the potential risk of As toxicity to the salt marsh biota, understanding how As release may occur in salt marsh sediments under normal and extreme conditions is warranted.

Here, we report on a combined laboratory study and field sampling campaign at the St. Jones Reserve aimed to elucidate how sediment biogeochemistry including As cycling is affected by redox and salinity changes that occur during extreme events. We monitored As concentrations in soil and in pore water before, during and after two extreme events that occurred in September 2015, and we continued to monitor pore water As concentrations for one year after the events. The extreme events were a supermoon lunar eclipse, followed by the intense rainfall and storm surge from Hurricane Joaquin. The supermoon caused extreme oscillations in the tidal pattern of the channel and inundation of the nearby marsh banks that are not usually flooded, which consequently influenced the redox potential, salinity, and availability of As in the pore water. Just after the supermoon event, Hurricane Joaquin entered the Mid-Atlantic and while it did not make landfall, it caused intense rainfall at the St. Jones Reserve, which decreased the pore water salinity and amplified the storm surge into the marsh. Together, the fluctuations in redox potential and salinity created a natural experiment where we could examine the impact of extreme events on As cycling in the salt marsh. The field experiment was combined with laboratory incubations to further clarify the behavior of As under increasing salinity and shifting redox potential. The major objective of this study is to address the mechanistic reason(s) for the observed As pulse, which has implications for As behavior over the long-term transition of sea level rise in tidal salt marshes and the expected increase in extreme weather variability.

Chapter 2

MATERIALS AND METHODS

2.1 Field Setting

The field sampling campaign was conducted at the St. Jones Reserve, which is part of the National Estuarine Research Reserve System (NERRS) located southeast of Dover, Delaware, USA (Figure 1). The field site is a representative Mid-Atlantic brackish salt marsh, with daily tidal influence from the St. Jones River, which connects to the Atlantic Ocean via the Delaware Bay. Within the marsh, two subsites have been identified based on differences in hydrology and biogeochemistry: the Near Channel site and the Interior Marsh site (Bothfeld, 2016). The Near Channel site is located near a tributary channel of the St. Jones River and is also situated near a portion of the 20-year old visitor boardwalk that runs through the marsh; CCA-treated wood was used to create the boardwalk with consequently high soil As concentrations of 50-150 mg/kg in sediments nearest the boardwalk and < 25 mg/kg closer to the pore water sampling devices (Figure 1). In contrast, the Interior Marsh site is farther away from both the main tributary and boardwalk path with soil As concentrations of ~ 10 mg/kg (Figure 1). In addition to different sediment As concentrations, these two subsites have distinct geochemical signatures: the Near Channel site is slightly higher in elevation, the sediments to a depth of 25 cm are typically not saturated, and its hydrology is mainly influenced by spring-neap cycles whereas the Interior site is consistently saturated to a depth of 5 cm or shallower with daily tidal fluctuations that exchange more brackish water (Bothfeld, 2016). Due to the differences in hydrology,

the redox potentials of the interstitial pore water of the subsites' soils are vastly different. The Interior Site is strongly reducing, with evidence of sulfides in the pore water, whereas the Near Channel site is moderately reducing, with evidence of ferrous iron in the pore water (Bothfeld, 2016). Preliminary monitoring efforts revealed that As was quite low and unchanging in pore water at the Interior site (Figure A1 in the Appendix), so we focused our field efforts on the Near Channel site, which is closer to the source of As contamination (i.e., the boardwalk) (Figure 1).



Figure 1: Panel a: Aerial map of St. Jones Reserve boardwalk and spatial distribution of pore water and surface soil sampling points (0-10 cm depth relative to soil surface). Letters marking soil samples indicate elevation in the salt marsh. L=low-marsh, M=mid-marsh, H=high-marsh, and B = replicate samples taken from 0, 25, and 50cm lateral distances from boardwalk. Panel b: Arsenic concentration in sediments at various locations in the marsh (0-10 cm depth relative to soil surface). Bar height indicates average \pm 95% CI concentration for entire core. Letters indicate significantly different means using Tukey-Kramer HSD at alpha = 0.05.

2.2 Pore Water Collection and Analysis

Pore water samples were collected at the Near Channel site (Figure 1) and in the main channel of the salt marsh using passive and instantaneous methods of water collection during the extreme events induced by the supermoon and storm surge following Hurricane Joaquin in Fall 2015. In June 2014, prior to the extreme events, three passive pore water (peeper) samplers were installed at the Near Channel site with nine 13 mL capacity peeper cells in each PVC housing at discrete depths determined by soil profiling (Bothfeld, 2016). The depths of sample collection were +3, 0, -8, -21, -36, -46, -61, -71, and -86 cm relative to the soil surface. Each peeper cell was fitted with a 0.2 µm nylon membrane and filled with deoxygenated ultrapure water (Seyfferth et al., 2013). Interstitial pore water was collected by osmosis and equilibrium was reached in ca. 10 days (LaForce et al., 2000). Peeper cells were monitored bimonthly from 2014 to 2016 whereby peeper cells were sampled and replaced with fresh cells (Bothfeld, 2016), but only data from September 2015 -November 2016 are reported in the present study because As was only monitored during that time. Five days before the supermoon lunar eclipse, a set of fresh peeper cells were installed on September 22, 2015 and allowed to equilibrate until October 6, 2015, four days after the storm surge from Hurricane Joaquin. During the extreme events, instantaneous pore water samplers (sippers) were also used to collect interstitial water at discrete depths of 0, 25, 35, 45, 55, 65, 75, and 90 cm relative to the soil surface. Sipper samples were collected from the Near Channel site at high tide during the supermoon lunar eclipse, high tide on the following day, and four days after the storm surge, simultaneously with the peeper collection on October 6. To establish seasonal trends in As cycling and pore water geochemistry, peeper pore water sampling and analysis continued from April to November 2016.

For both passive and instantaneous water samples collected from September 2015 to November 2016, pore water chemistry was evaluated. Water samples were aliquoted into polypropylene tubes and analyzed immediately for redox potential (Orion 9179E Triode), pH (Orion Ross Ultra pH/ATC Triode), and electrical conductivity (Orion DuraProbe 4-Electrode Conductivity Cells) with a portable meter (Orion STAR A322). The electrical conductivity values were converted to salinity with the following equation: Salinity (ppt) = 0.4665*Conductivity (mS/cm)1.0878 (Williams, 1986). As outlined by Bothfeld (2016), samples were analyzed colorimetrically for ferrous iron (Stookey, 1970) and sulfide (Cline, 1969) using a Thermo Scientific Evolution 60S UV-Visible Spectrophotometer. Water samples were acidified and analyzed for total As with an Agilent 7500cx Series inductively coupled plasma-mass spectrometry system (ICP-MS) operated in helium collision mode with 1500 RF power at the University of Delaware. Quality assurance and quality control were maintained with standard checks and blanks included in each sample set.

In the main channel, water quality was monitored following NERRS protocol (NERRS, 2017b). Dissolved oxygen, temperature, water level, turbidity, and salinity were measured at 15 minute intervals (YSI 6600) during the course of the extreme events (Figure 2). Precipitation data was retrieved from the meteorological monitoring station at the St. Jones Reserve (NERRS, 2017a) (Figure 2). In addition to collecting the channel measurements, we utilized well level data obtained from a monitoring well installed at the Near Channel site (Bothfeld, 2016)(Figure 2).



Figure 2: Precipitation, water level, and salinity data at the St. Jones Reserve during the extreme events of a supermoon lunar eclipse (September 27, 2015) and a hurricane-induced surge and storm event that caused anomalously high water inundation at the site. Panel a: Water level and salinity data in the main channel where vertical lines indicate sipper sampling events. Panel b: Water level and salinity data in the monitoring well at the Near Channel site where red star symbols indicate sipper sample salinity at 25cm depth.

2.3 Soil Incubations at Varying Salinity and Redox Potential

2.3.1 Batch Soil Incubation

Soils were incubated at different salinities under oxic and anoxic conditions to isolate the mechanistic relationship between pore water As and salinity at varying redox potentials. St. Jones River water was collected in polyethylene bottles and stored in a cooler until transported to a refrigerator at the University of Delaware. The river water was filtered through a 0.45 µm filter, diluted to prepare two levels of salinity (5 and 15 ppt), and then each saline solution was buffered with HEPES to control the soil pH to 7. Surface soil (0-10 cm depth) was collected from the Near Channel site, placed into gas-impermeable bags with oxygen scrubbers, and transported to the University of Delaware on ice. The soil was stored in an anoxic glove bag (95% $N_2/5\%$ H₂) until dry. Six grams of air-dried soil were weighed into 15 mL vials and 9 mL of buffered saline water was added. Three replicates of each salinity were set-up for both an oxic treatment, for which the vials were kept on the benchtop, as well as an anoxic treatment, for which the vials were stored in an anaerobic glove bag. Nine total sets of vials for the redox and salinity treatments were prepared to allow for the complete destruction of one set at each designated sampling timepoint. Sample vials were lightly agitated on a reciprocating table in an incubation chamber set to a constant temperature of 25°C. At 0.5, 1, 6, 12, 24, 48, 72, 144, and 168 hours, one set of samples (three replicates for each treatment) was removed from the shaker table, centrifuged, decanted, and filtered through a 0.2 µm nylon syringe filter. Redox potential, pH, salinity, and As concentration were evaluated in the filtrate as described in section 2.2.

2.3.2 Flow-Through Incubation

In addition to the batch incubation, a flow-through experiment was designed to more accurately emulate field conditions of the tidal salt marsh. Six soil cores, approximately 20 cm in diameter and 9 cm deep, were collected within seven meters from the boardwalk at the Near Channel site. Soil cores were collected within a PVC collar and sealed to a plastic board to prevent water and soil loss during transport to the University of Delaware. In the laboratory, the collar and board junction was waterproofed with silicone, outfitted with an input and output tube, and equipped with a Rhizon sampler (Eikjelkamp, The Netherlands), inserted at a 45° angle, for pore water collection. River water collected for the batch incubation was also used for the continuous flow-through experiment. A peristaltic pump controlled the input of river water to each core while another pump controlled the output of water to maintain flooded conditions in the cores.

Three replicate cores were designated as controls, for which the input salinity was maintained at 17 ppt and, the 2016 average high tide salinity in the main channel of the St. Jones Reserve (NERRS, 2016), for the duration of the experiment. The three treatment cores were subjected to changing salinities based on channel data collected during the Hurricane Joaquin storm surge (Figure 2a) (NERRS, 2016). The impacts of salinity treatment were evaluated over four phases: Phase One (P1), where the salinity in both control and treatment cores was maintained at 17 ppt; Phase Two (P2), where the salinity in the treatment cores decreased from 17 to 12 ppt; Phase Three (P3), where the salinity in the treatment cores increased from 12 to 17 ppt. During Phase One and Phase Three, pore water was collected every other day, while during Phase Two and Phase Four, pore water was collected every day. Vials for pore water

collection were purged and crimp sealed in the glove bag (95% $N_2/5\%H_2$) to maintain anoxic conditions (Seyfferth et al., 2016). Redox potential, pH, salinity, sulfide, ferrous iron, and As concentration were evaluated as described in section 2.2.

2.4 Statistical Analyses

This study involved five total data sets: three sets of pore water data from field collections and two sets of pore water data from laboratory incubations. Field data sets included pore water chemical data from peeper samples collected before and after the 2015 extreme events, sipper samples from before, during, and after the 2015 extreme events, and peeper samples from baseline conditions throughout 2016. Laboratory data sets included pore water chemical data from samples collected during the batch incubation and the flow-through incubation. For each data set, normality and homogeneity of variance tests were performed. In the case of all three field data sets and the batch incubation, As was not normally distributed and so it was log transformed to achieve normality.

The difference in pore water As, pH, redox potential, and salinity for each depth between the pre- and post-storm surge peepers was evaluated with a one-way ANOVA difference of means analysis. Partial correlation with As were computed for each pore water parameter controlling for other covariates (redox, salinity, pH, ferrous iron, and depth) to address the unique contribution of redox and salinity on As mobility. The pore water data from the sipper collections were also used to compute partial correlation coefficients between As and its covariates (redox, salinity, pH, and depth).

For the peeper samples collected throughout 2016, partial correlation coefficients were calculated between As and salinity, redox potential, ferrous iron, and

sulfide. One-way ANOVA was also used to compare average pore water parameters at each depth. Tukey-Kramer groupings at $\alpha = 0.05$ were reported, with common letters indicating no significant difference.

For the batch soil incubation, a one-way ANOVA was used to compare the average pore water As between the salinity treatments and the redox treatments for each time of collection. In the case of unequal variances, Welch ANOVA was used to evaluate significant differences. Partial correlation coefficients were calculated for the relationships between As and redox, salinity, and time. From the flow-through incubation, the difference in the average As concentration between the treatment and control cores was also analyzed by means of one-way ANOVA for each phase of the experiment and each individual date. Partial correlation coefficients were computed between As and sulfide, ferrous iron, redox potential, pH, and salinity. All statistical analyses were performed with JMP Pro 13 statistical software.

Chapter 3

RESULTS

3.1 Pore Water Chemistry During 2015 Extreme Events

The supermoon lunar eclipse and hurricane-induced precipitation and surge caused anomalously high water levels in the main channel and at the Near Channel site (Figure 2). Prior to the extreme events, the water level in the main channel ranged -0.08 to +1.22 m during low and high tide (average = $+0.67 \pm 0.01$ m, N = 2880) but became more inundated during the extreme events. Low and high tide shifted to +0.10 to +1.34 m during the supermoon (average = $+0.82 \pm 0.02$ m, N = 384) and to +0.63 to +1.77 m during the storm and surge (average = $+1.28 \pm 0.02$ m, N = 480) (Figure 2a). The effects of the extreme events were more pronounced in the monitoring well at the Near Channel site. Prior to the extreme events, water level in the monitoring well at the Near Channel site ranged from -0.15 to +0.33 m during low and high tide (average = -0.03 ± 0.001 m, N = 2880) but became more inundated during the extreme events. Low and high tide shifted to -0.04 to +0.30 m during the supermoon (average = $+0.10 \pm 0.005$ m, N = 384) and to +0.03 to +0.64 m during the storm and storm surge (average = $+0.24 \pm 0.004$ m, N = 480) (Figure 2b). The change in water level and salinity during the extreme events altered the pore water chemistry.

Arsenic concentrations in pore water increased near the 21 cm depth during the extreme events (Figure 3 and Table A6 in the Appendix). This As bulge at the 21 cm depth was observed for pore water samples collected with both peepers (Figure 3a) and sippers (Figure 3e) but was more pronounced with sippers. At the 21 cm depth,

the post-storm surge peepers (collected October 6, 2015) had significantly higher As concentrations compared to the pre-storm surge peepers (collected September 22, 2015) (p = 0.0344, F = 9.9536). While there was no significant difference in pore water pH in peepers between the two sampling dates until the 86 cm depth (p = 0.0453, F = 8.2581, Figure 3b), the redox potential was significantly higher for the post-storm samples for the entire depth profile (p < 0.0001, F = 209.0239, Figure 3c). The salinity was significantly higher in both the 36 and 71 cm depths for the post-surge peeper samples (p = 0.0421, F = 8.6890 and p = 0.0314, F = 10.5478, respectively, Figure 3d). Overall, the natural logarithm of As was strongly positively correlated with ferrous iron (r_{InAs} , Fe partialed for Eh, pH, logSalinity, Depth = 0.73, p < 0.0001, F = 32.8173), and also positively correlated with redox potential (r_{InAs} , Eh partialed for pH, Fe, logSalinity, Depth = 0.44, p = 0.0158, F = 6.6366), although this relationship was not as strong.



Figure 3: Arsenic, pH, redox potential, and salinity data for pore water samples.
Ferrous iron data is illustrated alongside salinity data for peepers in Panel d. Panels a-d illustrate peeper pore water samples from before and after extreme events. Symbols for peeper parameters indicate average value ± 95% CI for the three peepers at the Near Channel site. Panels e-h show sipper pore water samples from high tides during the extreme events.

In contrast to peeper collection (Figure 3a-d), which averages over ca. 14 days, the sipper method of collecting instantaneous pore water allowed the examination of rapid changes to pore water chemistry during the extreme events (Figure 3e-h and Table A7 in the Appendix). Sipper collection revealed that As concentrations during the supermoon increased from < 0.3 up to 0.75 μ M at the 21 to 45 cm depth and returned to < 0.3 μ M by four days after the surge and storm events (Figure 3e). The sipper sample collection at midnight on September 28 occurred at maximum high tide during the supermoon lunar eclipse whereas the two sipper collections on September 29 occurred just before and after the following high tide. The collection on October 6 coincided with the peeper pore water collection, four days after the intense rain and

storm surge. Sipper samples collected September 28-29 had similar pH and redox potential down to the 45 cm depth, but on October 6, pH increased by ~0.25 units and redox potential increased by ~200 mV (Figure 3g). The salinity profile also changed during the extreme events whereby samples collected at the peak high tide during the supermoon were ~17 ppt at 21-45 cm depth and returned to ~13 ppt by October 6 (Figure 3h).

Overall for the four sampling points, the natural logarithm of As was strongly positively correlated with salinity ($r_{InAs, Salinity partialed for pH, Eh, Depth = 0.57, p = 0.0056, F$ = 9.6541) and depth ($r_{InAs, Depth partialed for Salinity, pH, Eh = 0.36, p = 0.0955, F = 3.0618$). The strength of the partial correlation coefficient was less than the Pearson correlation coefficient for the relationship between redox potential and As, indicating that the covariates partially explained the As response to changes in redox potential (Table A1 in the Appendix).

3.2 Pore Water Chemistry During 2016 Baseline Conditions

Typical baseline As concentrations in the Near Channel site during the growing season varied with depth along with ferrous iron, sulfide, pH and salinity (Figure 4, and Figure A2 and Table A8 in the Appendix). Average As concentrations in pore water at the Near Channel site during the growing season from late April to early November 2016 ranged from 0.013 (\pm 0.005) to 0.234 (\pm 0.048) µM with the highest concentrations at the 21 cm depth and during the summer months (Figure 4 and Figure A2). Pore water As was significantly higher at the 8, 21, and 36 cm depths compared to the +3, 0, and >61 cm depths (p = <0.0001, F = 19.4721; Tukey-Kramer Grouping found in Table 1). On average for the growing season, the pore water As and salinity followed a similar pattern in the upper 36 cm, while the As exhibited an

inverse relationship with the redox potential at these depths. However, at a depth of 46 cm and deeper, the pore water As concentration and the salinity decreased while the redox potential did not change (Figure 4). Although salinity was not significantly different at depths below the soil surface, the redox potential was significantly lower at depths 36 cm and deeper compared to the pore water in the upper 21 cm (p < 0.0001, F = 7.4848). Partial correlation coefficients between the natural logarithm of As and the other covariates were calculated for above and below the 40 cm depth as this was the most obvious transition zone in the geochemical signature (Figure 4). At the depths corresponding to maximum pore water As (8 to 46 cm), the natural logarithm of As was strongly positively correlated with ferrous iron (r_{InAs, Fe} partialed for Eh, pH, Sulfide, Salinity = 0.55, p < 0.0001, F = 36.9427) and negatively correlated with the redox potential (r_{InAs, Eh partialed for pH, Fe, Sulfide, Salinity} = -0.22, p = 0.0395, F = 4.3715).



Figure 4: Average (\pm 95% CI, N = 30 per depth) pore water As and salinity (a), redox potential (b), ferrous iron (c), and sulfide (d) for Near Channel peepers obtained during the growing season from May to September 2016. Correlations between As and other parameters are partialed to other covariates and represent unique strength of relationship for above and below 40 cm depth.

Depth Relative	Average As	Tukey	Average	Tukey
to Surface	(µM)	Grouping for	Redox	Grouping for
(cm)		As	Potential (mV)	Redox
+3	0.015 ± 0.005	D	252.46 ± 78.93	А
0	0.063 ± 0.059	CD	215.48 ± 70.31	AB
-8	0.167 ± 0.058	AB	139.98 ± 52.18	BC
-21	0.234 ± 0.048	А	117.95 ± 81.64	С
-36	0.216 ± 0.059	AB	98.76 ± 25.86	С
-46	0.139 ± 0.036	BC	100.54 ± 25.52	С
-61	0.071 ± 0.019	CD	107.29 ± 24.02	С
-71	0.044 ± 0.010	D	106.51 ± 24.32	С
-86	0.031 ± 0.006	D	111.66 ± 25.88	С

Table 1:Average (± 95% CI) Pore Water As and Redox Potential With Depth and
Tukey-Kramer Grouping for Baseline Peeper Samples

3.3 Soil Incubations at Varying Salinity and Redox Potential

3.3.1 Batch Soil Incubation

The batch soil incubation was carried out to elucidate the separate effects of redox potential and salinity on As availability. Over the course of the entire incubation (Figure 5 and Table A9 in the Appendix), average As concentrations were significantly higher in the anoxic treatment than the oxic treatment (p = 0.0101, F = 7.0685). A difference of means test was performed for each sampling time to compare the salinity treatments. Within the first 48 hours of the experiment, the 5 ppt treatment resulted in significantly higher pore water As than the 15 ppt treatment (Table 2, Figure 5). From 72 hours on, the pore water As for the two salinity treatments were

not significantly different at $\alpha = 0.05$. In contrast to the salinity results, the As concentrations between redox treatments only became significantly different at 72 hours and after, with the anoxic samples having higher As than the oxic samples (Table 2). These results indicate salinity control on As release early in the experiment and redox control on As release later in the experiment.



Figure 5: Progression of pore water [As] (a), redox potential (b), pH (c), and salinity (d) during soil batch incubation. Symbols indicate average (± 95% CI) of three replicates of each treatment. Horizontal lines represent target salinity values of 5 and 15 ppt.

Time	Mean 5 ppt	Mean 15 ppt	One-way	Mean	Mean Oxic	One-way
(hours	Treatment	Treatment	ANOVA	Anoxic	Treatment	ANOVA for
)	As (µM)	As (µM)	for Salinity	Treatment	As (µM)	Redox
				As (µM)		
0.5	0.233 ±	$0.182 \pm$	p = 0.0002	$0.202 \pm$	0.212 ±	p = 0.6080*
	0.018	0.013	F =	0.021	0.041	F = 0.2869
			34.7771			
1	0.273 ±	0.136 ±	p = 0.0212	0.177 ±	0.232 ±	p = 0.4510
	0.126	0.065	F = 9.5839	0.141	0.165	F = 0.6498
6	0.199 ±	0.135 ±	p = 0.0005	0.157 ±	0.178 ±	p = 0.4004
	0.024	0.023	F =	0.047	0.037	F = 0.7714
			25.3767			
12	$0.212 \pm$	$0.128 \pm$	p < 0.0001	0.169 ±	$0.171 \pm$	p = 0.9468*
	0.017	0.029	F =	0.036	0.068	F = 0.0048
			40.8275			
24	0.201 ±	0.147 ±	p =	0.183 ±	0.166 ±	p = 0.4106*
	0.243	0.008	0.0017*	0.046	0.017	F = 0.7760
			$\mathbf{F} =$			
			29.1824			
48	0.371 +	0.226 +	p = 0.0020	0.337 +	0.261 +	p = 0.1764
	0.067	0.061	F =	0.104	0.086	F = 2.1166
	0.007	0.001	17 0680	0.101	0.000	1 2.1100
72	0.830 +	0.437 +	n =	0.987 +	0 280 +	p = 0.0049*
. –	0.556	0.286	0 1475*	0.392	0.096	F = 20.2523
	0.000	0.200	F = 2.6101	0.072	0.070	1 20.2020
144	5 21 + 3 85	5.14 + 3.73	p = 0.9750	8 37 + 1 29	1.99 ± 0.447	p < 0.0001
	0.21 - 0.00	0.11 = 0.70	F = 0.0010	0.07 - 1.27		F =
			1 = 0.0010			1/1 6/30
168	3.00 ± 1.28	4.04 ± 2.14	n –	103 ± 268	2.20 ± 0.522	n = 0.0464*
100	5.07 ± 1.30	4.04 ± 3.14	P^{-}	4.73 ± 2.00	2.20 ± 0.333	$P = 0.0404^{\circ}$
			$0.49/1^{\circ}$			$\Gamma = 0.0201$
			F = 0.5140			

Table 2:Average (± 95% CI) Pore Water As for Salinity and Redox Treatments in
Batch Incubation. *Indicates Welch ANOVA for unequal variance

Partial correlation coefficients were calculated for the relationships between As and salinity, redox potential, and time, controlling for the covariates. Arsenic data was transformed to the natural logarithm to achieve normality. Over the course of the entire incubation, the natural logarithm of As was strongly correlated with time (r_{lnAs} , Time partialed for Salinity, Eh = 0.87, p < 0.0001, F = 297.1699) and redox potential (r_{lnAs} , Eh partialed for Salinity, Time = -0.50, p < 0.0001, F = 33.2419). The correlation between As and salinity (r_{lnAs} , Salinity partialed for Eh, Time = -0.15, p = 0.1265, F = 2.3753) was partially explained by the covariates.

3.3.2 Flow-Through Incubation

To further examine the potential role of changes in salinity on As release, a flow-through incubation was conducted to simulate field conditions experienced during extreme events. Saline water was pumped through six soil cores until the pore water salinity reached increased from the native 13 ppt to the 17 ppt of the input water, signaling the start of Phase 1. During Phase 1, when salinity between treatment and control cores were similar, pore water As was strongly correlated with salinity (r_{As}, Salinity partialed for Sulfide, Fe, Eh, pH = 0.76, p = 0.0271, F = 8.4558), sulfide (r_{As}, Sulfide partialed for Fe, Eh, pH = 0.80, p = 0.0162, F = 10.9579), ferrous iron (r_{As}, Fe partialed for Sulfide, Eh, pH, Salinity = 0.89, p = 0.0027, F = 24.0638), and pH (r_{As}, pH partialed for Sulfide, Fe, Eh, Salinity = - 0.63, p = 0.0925, F = 3.9991) (Figure 6 and Table A10 in the Appendix). Although the redox potential decreased during this time, the relationship between redox and As was not significant at the α = 0.05 level. Additionally, there was not a significant difference between the pore water As in the treatment and control cores in this phase.

Salinity was manipulated in treatment cores during Phases 2-4, resulting in changes between treatment and control cores. During Phase 2, the salinity of the treatment cores decreased to 12 ppt while the control cores were kept at 17 ppt. During Phase 2, average As concentrations were higher for the treatment cores compared to the control cores and were 0.139 (\pm 0.035) μ M and 0.093 (\pm 0.040) μ M, respectively,

but only significantly different at the $\alpha = 0.1$ level (p = 0.0808, F = 3.5425). During Phase 3, when the 12 and 17 ppt salinities were maintained, the treatment cores still had slightly higher pore water As compared to the control, significant at the $\alpha = 0.1$ level (p = 0.0955, F = 3.1390). Point-by-point comparison revealed that As concentrations were significantly higher in treatments than controls for the last sampling point of Phase 3 (p = 0.022, F = 13.2202), which may reflect a lag response to salinity treatments. During Phase 4, when salinity in treatment cores was brought up to 17 ppt, As concentrations were significantly higher in the treatment cores at the $\alpha =$ 0.1 level (p = 0.0614, F = 4.4376), averaging 0.262 (± 0.042) µM for treatments and 0.206 (± 0.042) µM for the controls.



Figure 6: Progression of pore water [As] (a), redox potential (b), ferrous iron (c), and salinity (d) during continuous flow-through incubation. Symbols indicate average (± 95% CI) of three replicates. Shaded backgrounds indicate phase of the experiment.

Chapter 4

DISCUSSION

Our combined laboratory and field observations indicate that both redox potential and salinity interact to contribute to As release in tidal salt marshes, and this interplay is affected by extreme events. Extreme events such as supermoons and intense storms have the potential to shift biogeochemical cycling of unwanted environmental contaminants, such as As (Thomson & Rose, 2011), but few studies have been published about the direct effects of extreme phenomena on water quality (Haddout et al., 2016). Both the supermoon and hurricane-induced storm and surge extreme events we examined increased water inundation at our field site, but affected pore water chemistry in different ways. These rapid changes in soil biogeochemical conditions due to extreme events have a direct impact on the mobility of As. While the pore water As concentration increased up to four-fold due to extreme events, it returned to normal baseline conditions within one week of the events. Therefore, our data illustrate that rapid changes in salinity and redox potential due to extreme events result in an As pulse to the aquatic environment, but we suggest the impact of released As on the aquatic system is relatively small and short-lived.

4.1 Arsenic Cycling During Extreme Events

Our field data used both passive sampling devices (peepers) and instantaneous sampling devices (sippers) to examine both baseline conditions and impacts of extreme events on As cycling in the tidal salt marsh. It was evident that the 14d equilibration time of the peepers smoothed out the short-term chemical signature of the supermoon and storm surge observed with the sipper data (Figure 3a and 3e).
Sipper pore water As reached roughly four times the magnitude of the As concentration in the peeper samples. Yet within a week of the extreme events, the sipper pore water As concentrations returned to baseline levels, indicating sequestration or a flux out of the marsh. Despite significant differences between the pre- and post-storm surge peeper samples, the values for all measured pore water parameters with peepers (As, pH, redox, and salinity) fell within the range of the baseline conditions monitored throughout 2016 (Figure 4). If only peepers were utilized, impacts of the extreme events on rapid changes in pore water chemistry would have been missed.

Only with the instantaneous pore water samples were we able to observe the effect of changes in salinity due to extreme events on As release. In contrast to the trends observed with peepers, the sipper samples illustrated a significant and positive relationship between the natural logarithm of As and salinity ($r_{InAs, Salinity partialed for pH, Eh$, $D_{pepth} = 0.57$, p = 0.0056, F = 9.6541). This was the unique contribution of salinity on As cycling when accounting for the covariance in the other parameters measured, including redox and pH. At the 25 cm depth, the typically dominant variables in As cycling (redox potential and pH) were approximately the same for the first three sampling events during the extreme events (Figure 3e). This suggests that the pulse of As that occurred on September 29 is likely attributed to a rapid change in salinity (Figure 3h).

4.2 Mechanisms of Arsenic Release During Extreme Events

The short-term mechanism for As release due to salinity from the salt marsh sediments could be electrostatic competition with anions in the salt water (Chapman & Wang, 2001). These include bicarbonate and carbonate (Appelo et al., 2002; Ardau et

al., 2013; Kim et al., 2000), sulfate (Borda & Sparks, 2007), sulfide (Kocar et al., 2010), nitrate (Xu et al., 2009), and phosphate or molybdate (Barrow, 1974; Manning & Goldberg, 1996; Melamed et al., 1995; Roy et al., 1986; Smith et al., 2002; Woolson et al., 1973). As the salinity increases, both cations and anions are introduced, increasing ionic strength of the solution. While a solution's ionic strength affects As sorption on laterite iron concretions (Partey et al., 2008), goethite (Antelo et al., 2005), and variable charge soils (Xu et al., 2009), the effect is contingent on the As speciation and how the As anions are sorbed to the soil surface (Hayes et al., 1988). In batch experiments, higher ionic strength (up to 0.1 M) resulted in less arsenite sorption but more arsenate sorption to laterite iron concretions (Partey et al., 2008). Antelo et al. (2005) similarly demonstrated that arsenate sorption on goethite increased in the solution with a higher ionic strength (up to 0.1M KNO₃), but arsenite behavior was not investigated in that study. In this present study, an increase in As desorption was observed with an increase in salinity/ionic strength. Arsenic concentration was lowest in peeper samples, which also had the lowest ionic strength of $0.04 (\pm 0.04)$ M. In contrast, As desorption was higher from our other samples which had nearly an order of magnitude higher ionic strength in sippers (average = 0.33 ± 0.02 M), the batch incubation (average = 0.27 ± 0.11 M), and flow-through incubation samples (average $= 0.33 \pm 0.04$ M). While As speciation was not evaluated, we suggest that arsenite was the dominant As phase that was desorbed in our experiments.

Electrostatic competition has the potential to displace easily exchangeable outer-sphere As, but As can also be associated with the solid phase in lower degrees of lability. Arsenic fractionation of soils from the St. Jones Reserve was assessed using sequential extraction procedure (Wenzel et al., 2001) (Figure A3 and Table A11 in the Appendix). Only about 2% of total As was nonspecifically sorbed to soil surfaces, representing the easily exchangeable fraction of As in outer-sphere complexes. Given the high lability of this fraction, As release through the mechanism of ligand exchange was expected to be rapid. Sipper samples collected during the first 36 hours of the supermoon showed a quick pulse of As, which was likely due to this mechanism of ionic competition. After the pulse, the pore water As concentration decreased to baseline conditions within a week likely because the easily exchangeable pool was depleted.

The majority of As (>40%) in the St. Jones soil was not as easily exchanged because it was associated in inner-sphere complexes with amorphous and poorly crystalline hydrous oxides of iron (Figure A3), potentially as As on ferrihydrite. Additionally, close to 20% of the total As was complexed with well-crystalized iron oxides. In these two pools, As was dominantly retained in an inner-sphere complex with iron oxides, which could be freed under suboxic conditions. Arsenic release could occur if arsenate is chemically reduced to arsenite or if ferric iron in the iron oxides is reduced to ferrous iron (Jones et al., 2000; Langner & Inskeep, 2000). Although dissimilatory arsenate reduction could have a large role in As availability (Fendorf & Kocar, 2009), arsenite could still repartition with other iron oxides such as hematite, goethite, and lepidocrocite (Ona-Nguema et al., 2005). Prolonged suboxic conditions would be expected to free the As in these complexes, which is supported by the long-term pore water collection throughout 2016. Despite low baseline As concentrations, the continuous peak in As around the 21 cm depth was positively correlated to ferrous iron (r_{lnAs} , Fe partialed for Eh, pH, Sulfide, Salinity = 0.55, p < 0.0001, F = 36.9427) and negatively correlated with the redox potential (r_{lnAs, Eh partialed for pH, Fe},

sulfide, salinity = -0.22, p = 0.0395, F = 4.3715). Therefore, the long-term mechanism for As release in the St. Jones Reserve was likely driven by suboxic redox potential, leading to more ferrous iron and As in the pore water.

Batch incubations with field sediments under oxic and anoxic conditions and at low and high salinity revealed that while redox potential had a dominant control on As release, salinity also played a role within the first 72 hours (Figure 5). Like the sequential extraction suggested, the quick release in the first 72 hours could have represented desorption of the easily extractable fraction due to ligand exchange. Then at 72 hours and after, redox potential had more of an effect on As release, likely indicating the destabilization of inner-sphere-associated As on iron oxides (Root et al., 2007; Tufano & Fendorf, 2008). Although the samples from the high salinity treatment resulted in lower pore water As than the low salinity treatment during this time, the high salinity treatment samples also had a slightly higher redox potential throughout the experiment (Figure 5). Consequently, the redox covariate could have skewed the effect of salinity. Here, the partial correlations were beneficial to disentangle the unique contributions of each covariate on As release. The partial correlation coefficient for the relationship between salinity and As was still negative after accounting for covariates, but it was not statistically significant at the $\alpha = 0.05$ level. The high salinity treatment, which added more anions like sulfate to each sample, could have inhibited the reductive dissolution of iron (oxyhydr)oxides, resulting in lower As mobility and more As sequestration with iron sulfides (Burton et al., 2013). Additionally, high ionic strength in the high salinity treatment samples could have impeded the microbially-mediated iron reduction (LeMonte et al., 2017). A steady increase in salinity occurred in all of the treatment groups over time, which can

be attributed to the style of experiment (i.e., batch) and the high soluble salt content of the salt marsh soils. Because this was a batch study, there was no flow to remove excess salts from building up in the solution. Column experiments have demonstrated the importance of advective flow in observing secondary mineral formation in the effluent end of the column compared to the influent end (Benner et al., 2002). The lack of flow in our batch experiment may have influenced the solid phase mineral formation so that it was no longer representative of field conditions.

To more closely represent field conditions, we conducted a continuous flowthrough incubation in order to overcome some of the limitations of batch experiments and to examine impacts of salinity changes on As release from intact marsh sediments. Pore water salinity in sediments was initially ~2 ppt (Figure 4) and was increased in treatments and controls during Phase 1 to 17 ppt, which was the measured salinity in sippers during extreme events (Figure 3). While as expected there was no significant difference in As concentrations between treatments and controls for Phase 1, there was a significant and positive relationship between As release and salinity after accounting for the covariance in other parameters ($r_{As, Salinity partialed for Sulfide, Fe, Eh, pH = 0.76, p =$ 0.0271, F = 8.4558). This relationship likely illustrated a lag effect of the rising salinity in the beginning of the experiment and desorption of As from the easily exchangeable fraction. Throughout the incubation when salinity in treatment cores decreased to 12 ppt (Phase 2), remained at 12 ppt (Phase 3) and increased again to 17 ppt (Phase 4), the pore water As was consistently higher in the treatment cores compared to the control cores (Figure 6). The increased As mobility in the treatment cores could be attributed to rapid changes in salinity, similar to the data obtained via sippers during the extreme events (Figure 3e-h).

4.3 Long-term Implications

Given the strong affinity As has for iron (oxyhydr)oxides (Al-Sid-Cheikh et al., 2015; Gu et al., 1994), long-term As cycling in a salt marsh is likely dominated by the redox potential. As the redox potential decreases, usually due to increasing water level, the iron (oxyhydr)oxides will be reductively dissolved (Olivie-Lauquet et al., 2001). Therefore, when the soil is inundated, lower redox potential will lead to more ferrous iron in the pore water and consequently As release. If reducing conditions persist, sulfate reduction could occur and any available As might be repartitioned with iron sulfide phases such as greigite and mackinawite, which are more thermodynamically favorable (Kocar et al., 2010). Over the long-term, we could expect As to be less bioavailable when it is sequestered in these iron sulfides (Wilkin & Ford, 2006). Arsenic stabilization is therefore reliant on the iron and sulfur minerals present. Both iron and sulfur cycling are closely entwined in a salt marsh environment and strongly depend on tidal movement and salinity (Luther & Church, 1988).

Arsenic may be the most available to pore water within the zone of intermediate redox potential, where both iron reduction and sulfide oxidation could be present. This relationship was demonstrated by the 2016 peeper samples, where the bulge in As was inversely proportional to the redox potential down to about 40 cm (Figure 4) and strongly correlated with the reduced iron in the pore water ($r_{InAs, Fe}$ partialed for Eh, pH, Sulfide, Salinity = 0.55, p = <0.0001, F = 36.9427). The major source of this As and ferrous iron could be pyrite oxidation, which is promoted by the tidal cycle and occurs quickly (Luther & Church, 1988). The rate of pyrite oxidation in tidal salt marshes has been found to be less than 6 days (Luther & Church, 1988), which also aligns with the time frame between the supermoon As pulse and post-storm surge return to baseline conditions.

The depths of the As bulge also correspond to the approximate rooting depths of the dominant salt marsh grasses at the reserve. These grasses are well adapted to periods of saturation with adventitious roots and cavities called aerenchyma, which allow gas exchange with the atmosphere (Armstrong, 1967; Armstrong et al., 1991). Gases diffuse to the roots, where some oxygen may be leaked due to differences in root development (Kotula et al., 2009). This creates a gradient of oxidation from the rhizosphere outwards, which would influence the stability of the soil minerals, and in turn, the attenuation of As within this depth range (Seyfferth et al., 2010).

Unlike the well-established trend between redox potential and As availability, the definitive relationship between As and salinity remains elusive. Salinity is thought to be the controlling factor for contaminant partitioning in estuaries, whereas pH is usually the dominant variable in freshwater environments (Chapman & Wang, 2001). Increased salinity in a salt marsh may cause competition between seawater anions and As oxyanions for soil sorption sites, but it is also possible that increased salinity may encourage flocculation of iron oxides and precipitation of As (Chapman & Wang, 2001). Higher salinity may have a detrimental impact on the microbial activity that would otherwise be responsible for dissimilatory iron reduction, ultimately inhibiting the major long-term mechanism for As availability in this environment (LeMonte et al., 2017). Results from the batch incubation demonstrated that the direction of change in salinity may not be as critical as the absolute rate of change in pore water salinity. However, the limitations of batch experiments may prevent the accurate emulation of field conditions. In comparison, the field results from this study were consistent with the flow-through incubation, which both suggested that increasing salinity led to more short-term As release in the pore water.

Chapter 5

CONCLUSIONS

Extreme shifts in tidal patterns impact the biogeochemical cycling of redoxsensitive elements, such as As. We observed 4-fold increase in porewater As due to extreme events that returned to baseline conditions within one week. The environmental conditions that change during extreme events drive the soil and water biogeochemistry, namely through the potentially additive effects of lower redox potential and higher salinity. If extreme storms continue to occur more frequently due to climate variability, the As pulses released to pore water in salt marshes could approach acute toxicity thresholds for salt marsh organisms. This work is applicable to how other salt marsh environments behave in the immediate response time of an extreme event and to the effects of more gradual climate variability. In general, increased salinity may result in the short-term release of contaminants, like As, from the solid phase to the interstitial pore water. Long-term effects of salinity may instead act to stabilize the iron-sulfide minerals, leading to more As sequestration and consequently less available for biotic uptake.

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Appendix

Supporting Information

The following supporting information includes supplementary data and statistical analyses for pore water samples collected at the St. Jones Reserve. Pore water was collected as outlined in the main text. Pore water chemical data was analyzed in JMP statistical software for Pearson and partial correlation coefficients. Meteorological data can be accessed at http://cdmo.baruch.sc.edu/get/export.cfm.



Figure A1: Pore water arsenic for Near Channel and Interior Marsh peepers. Data points represent average (± 95% CI) of 3 replicate peepers at each site on October 15th, 2014.



Figure A2: Progression of pore water depth profiles of [As] (a, d, g, and j), salinity (b, e, h, and k), and redox potential (c, f, i, and l) during the growing season of 2016. Symbols represent the average (± 95% CI) of three peepers at each depth at the Near Channel site. Sampling occurred on May 20th (Spring), June 8th (Early Summer), August 31st (Late Summer), and September 21st (Fall).



Figure A3: Arsenic concentration within sequential extracts of soil. Fractions of soil As range from most labile (Fraction 1) to the most recalcitrant (residual, Fraction 5). Bar height indicates average As (\pm 95% CI) for that fraction (n= 2).



Figure A4: Aerial map of St. Jones Reserve with sampling points for soil and vegetation labeled with transect numbers 1-5.



Figure A5: Arsenic and iron concentrations in root dithionite citrate bicarbonate (DCB)-extracts for five vegetation sampling sites at the St. Jones Reserve. Transects 1-5 correspond to locations in Figure A4.



Figure A6: Average (±95% CI) arsenic concentration in shoot (positive axis, striped bar) and root (negative axis, solid bar) tissue digests for 5 transect sampling sites at the St. Jones Reserve.

Table A1:Pearson and partial correlation coefficients for relationships between pore
water covariates for sipper samples collected during the supermoon and
Hurricane Joaquin extreme events.

Relationship Pearson Correlation		Partial Correlation	p-value for Partial	
	Coefficient	Coefficient	Correlation	
ln[As] and Salinity	0.57	0.57	0.0056	
ln[As] and pH	0.03	-0.02	0.9124	
ln[As] and Eh	0.38	0.10	0.6614	
ln[As] and Depth	0.54	0.36	0.0955	
Salinity and pH	0.17	0.16	0.4856	
Salinity and Eh	0.19	-0.09	0.6743	
Salinity and Depth	0.15	-0.08	0.7331	
pH and Eh	0.55	0.76	< 0.0001	
pH and Depth	-0.22	-0.61	0.0028	
Eh and Depth	0.47	0.63	0.0015	

Table A2:Pearson and partial correlation coefficients for relationships between
pore water covariates for peeper samples collected during the supermoon
and Hurricane Joaquin extreme events.

Relationship	Pearson Correlation	Partial Correlation	p-value for Partial
-	Coefficient	Coefficient	Correlation
ln[As] and Eh	0.23	0.44	0.0158
ln[As] and pH	0.51	0.18	0.3564
ln[As] and Ferrous	0.70	0.73	< 0.0001
Iron			
ln[As] and	0.52	-0.11	0.5741
ln(Salinity)			
ln[As] and Depth	0.84	0.74	< 0.0001
Eh and pH	0.23	-0.19	0.3234
Eh and Ferrous Iron	-0.14	-0.52	0.0042
Eh and ln(Salinity)	0.46	0.47	0.0107
Eh and Depth	0.15	-0.30	0.1175
pH and Ferrous	0.18	-0.27	0.1623
iron			
pH and ln(Salinity)	0.66	0.53	0.0029
pH and Depth	0.58	0.13	0.4993
Ferrous Iron and	0.28	0.25	0.1864
ln(Salinity)			
Ferrous Iron and	0.47	-0.35	0.0660
Depth			
ln(Salinity) and	0.52	0.15	0.4285
Depth			

Table A3:Pearson and partial correlation coefficients for relationships between
pore water covariates for peeper samples collected during baseline
conditions at depths corresponding to maximum pore water As.

Relationship	Pearson Correlation	Partial Correlation	p-value for Partial
	Coefficient	Coefficient	Correlation
ln[As] and Eh	-0.21	-0.22	0.0395
ln[As] and pH	-0.12	-0.11	0.2945
ln[As] and Ferrous	0.55	0.55	< 0.0001
Iron			
ln[As] and Sulfide	0.05	0.13	0.2289
ln[As] and Salinity	-0.28	-0.16	0.1259
Eh and pH	0.06	0.03	0.7574
Eh and Ferrous Iron	-0.06	0.08	0.4844
Eh and Sulfide	0.02	0.04	0.7184
Eh and Salinity	0.00	-0.05	0.6574
pH and Ferrous	0.03	0.07	0.5159
Iron			
pH and Sulfide	-0.17	-0.13	0.2258
pH and Salinity	0.09	0.03	0.7628
Ferrous Iron and	-0.20	-0.28	0.0072
Sulfide			
Ferrous Iron and	-0.20	-0.13	0.2412
Salinity			
Sulfide and Salinity	-0.23	-0.25	0.0191

Table A4:Pearson and partial correlation coefficients for relationships between pore
water covariates for batch incubation samples.

Relationship	Pearson Correlation	Partial Correlation	p-value for Partial
	Coefficient	Coefficient	Correlation
ln[As] and Time	0.91	0.87	< 0.0001
ln[As] and Eh	-0.71	-0.50	< 0.0001
ln[As] and Salinity	0.22	-0.15	0.1265
Salinity and Eh	-0.20	-0.10	0.2995
Salinity and Time	0.29	0.24	0.0157
Eh and Time	-0.60	0.18	0.0660

Relationship	Pearson Correlation	Partial Correlation	p-value for Partial
	Coefficient	Coefficient	Correlation
[As] and Salinity	0.03	0.76	0.0271
[As] and Sulfide	-0.10	0.80	0.0162
[As] and Ferrous	0.68	0.89	0.0027
Iron			
[As] and pH	-0.30	-0.63	0.0925
[As] and Eh	-0.38	0.28	0.5003
Sulfide and Ferrous	-0.55	-0.85	0.0083
Iron			
Sulfide and Eh	0.11	-0.34	0.4153
Sulfide and pH	0.56	0.70	0.0541
Sulfide and Salinity	0.21	-0.64	0.0891
Ferrous Iron and Eh	-0.64	-0.54	0.1672
Ferrous Iron and	-0.28	0.50	0.2019
pH			
Ferrous Iron and	-0.46	-0.77	0.0255
Salinity			
Eh and pH	-0.15	-0.06	0.8931
Eh and Salinity	0.32	-0.20	0.6348
pH and Salinity	0.13	0.51	0.2014

Table A5:Pearson and partial correlation coefficients for relationships between pore
water covariates for Phase One of the flow through incubation samples.

			Electrical Conductivity	Redox Potential		Ferrous	Arsenic	
Location	Date	Depth (cm)	(mS/cm)	(mV)	pН	iron (dM)	(uM)	Salinity
TG1A	9/22/2015	3		-28.2	6.61		0.003	
TG1B	9/22/2015	0	0.07	-21.7	5.66		0.003	0.03
TG1C	9/22/2015	-8	0.33	-2.9	6.74		0.012	0.14
TG1D	9/22/2015	-21	1.13	2	7.14		0.039	0.53
TG1E	9/22/2015	-36	2.43	17.8	6.7	15.06	0.101	1.23
TG1F	9/22/2015	-46	2.46	12.3	7.01	11.20	0.043	1.24
TG1G	9/22/2015	-61	1.56	12.4	6.73	10.88	0.027	0.76
TG1H	9/22/2015	-71	1.62	8.9	6.9	6.14	0.011	0.79
TG1I	9/22/2015	-86	0.97	22.3	6.5	4.96	0.011	0.45
TG2A	9/22/2015	3		-0.7	6.6		0.001	
TG2B	9/22/2015	0	0.00	-6.8	6.41		0.000	0.00
TG2C	9/22/2015	-8		7.1	6.6		0.000	
TG2D	9/22/2015	-21	0.97	32.6	6.91		0.074	0.45
TG2E	9/22/2015	-36	1.61	38.8	6.9	25.36	0.199	0.78
TG2F	9/22/2015	-46	2.67	35.8	7.02	8.29	0.104	1.36
TG2G	9/22/2015	-61	2.37	22.4	7.07	19.26	0.087	1.19
TG2H	9/22/2015	-71	1.15	33	6.53	20.60	0.027	0.54
TG2I	9/22/2015	-86	1.01	30.5	6.62	13.23	0.022	0.47
TG3A	9/22/2015	3	0.01	16.3	7.07		0.002	0.00
TG3B	9/22/2015	0		24.5	6.99			
TG3C	9/22/2015	-8	1.19	44.8	7.29		0.034	0.56
TG3D	9/22/2015	-21	2.14	54.6	6.9		0.052	1.07
TG3E	9/22/2015	-36	2.94	41	7.08	18.13	0.183	1.51
TG3F	9/22/2015	-46	3.51	51.9	7.02	22.05	0.066	1.83
TG3G	9/22/2015	-61	1.28	47.4	6.8	10.12	0.033	0.61
TG3H	9/22/2015	-71	1.06	-3.1	6.7	2.11	0.014	0.50
TG3I	9/22/2015	-86	1.33	42.6	6.56	10.74	0.014	0.64
TG2A	10/6/2015	3	2.33	218.1	7.19		0.002	1.17
TG2B	10/6/2015	0	2.67	226.6	7.21		0.015	1.35
TG2C	10/6/2015	-8	0.98	238.7	6.87		0.010	0.46
TG2D	10/6/2015	-21	1.21	201.6	6.77	5.32	0.104	0.58
TG2E	10/6/2015	-36	5.33	144.5	6.92	15.97	0.118	2.88

Table A6:Pore water data for peeper samples collected during supermoon and
Hurricane Joaquin in Fall 2015

TG2F	10/6/2015	-46	8.21	146.1	7.01	6.55	0.074	4.61
TG2G	10/6/2015	-61	5.28	135.4	6.94	11.47	0.045	2.85
TG2H	10/6/2015	-71	4.44	153.3	6.79	15.89	0.040	2.36
TG2I	10/6/2015	-86	0.94	173.5	6.68	10.99	0.074	0.44
TG1B	10/6/2015	0	2.76	115	6.98		0.007	1.41
TG1C	10/6/2015	-8	4.97	152	6.73		0.007	2.67
TG1D	10/6/2015	-21	4.79	137.3	7.03	18.43	0.147	2.57
TG1E	10/6/2015	-36	13.61	123.4	6.94	13.41	0.125	7.98
TG1F	10/6/2015	-46	7.88	111.5	7.05	16.90	0.130	4.40
TG1G	10/6/2015	-61	4.35	122.8	6.94	3.78	0.019	2.31
TG1H	10/6/2015	-71	4.13	131.4	6.9	9.15	0.031	2.18
TG1I	10/6/2015	-86	3.22	139.8	6.74	7.80	0.018	1.67
TG3B	10/6/2015	0	8.60	218.4	6.87		0.009	4.85
TG3C	10/6/2015	-8	8.81	230.9	7.11		0.019	4.97
TG3D	10/6/2015	-21	7.09	123.2	7.03	18.70	0.203	3.93
TG3E	10/6/2015	-36	10.29	113.8	7	18.92	0.168	5.89
TG3F	10/6/2015	-46	3.69	113.9	6.9	16.44	0.113	1.93
TG3G	10/6/2015	-61	2.09	100.7	7.01	1.65	0.016	1.04
TG3H	10/6/2015	-71	2.25	129.3	7	2.90	0.019	1.13
TG3I	10/6/2015	-86	2.39	160.1	6.9	2.69	0.013	1.20

Sampling	Location	Data	Donth (cm)	Electrical Conductivity	Redox Potential		Arsenic	Water	Salinity
nine	Creak	Dale	Depth (cm)	(ms/cm)	(1117)	рп	(uivi)	Level (III)	(ppr)
0:00	Сгеек	9/28/2015	0		8.4	7.46	0.013	1.23	
0:00	IGY	9/28/2015	-25	24.75	26.1	6.56	0.090	1.23	15.30
0:00	TG B	9/28/2015	-35	22.55	28.2	6.8	0.203	1.23	13.83
0:00	TG R	9/28/2015	-45	29.34	8.7	7.03	0.198	1.23	18.41
0:00	TG DG	9/28/2015	-55	26.47	-46.7	7.04	0.107	1.23	16.46
0:00	TG DY	9/28/2015	-65	24.37	-83.2	6.97	0.034	1.23	15.05
0:00	TG DB	9/28/2015	-75	22.84	-90.6	6.9	0.040	1.23	14.02
0:00	TG DR	9/28/2015	-90	25.13	-118.6	7.06	0.050	1.23	15.56
9:47	Creek	9/29/2015	0		-9.1	7.2	0.016	1.13	
9:47	TG Y	9/29/2015	-25	28.19	2.2	6.97	0.141	1.13	17.63
9:47	TG B	9/29/2015	-35	27.52	-1.4	6.94	0.167	1.13	17.18
9:47	TG R	9/29/2015	-45	25.80	-13.5	6.97	0.607	1.13	16.01
9:47	TG DG	9/29/2015	-55	24.27	-85.8	7.1	0.107	1.13	14.98
9:47	TG DB	9/29/2015	-75	24.18	-122.5	6.9	0.022	1.13	14.92
9:47	TG DR	9/29/2015	-90	24.94	-119.8	6.98	0.034	1.13	15.43
12:03	Creek	9/29/2015	0		-5.7	7.38	0.016	1.22	
12:03	TG Y	9/29/2015	-25	27.24	9.8	6.94	0.751	1.22	16.98
12:03	TG R	9/29/2015	-45	27.43	1.1	7.06	0.218	1.22	17.11
12:03	TG DG	9/29/2015	-55	25.04	-40.8	7.16	0.057	1.22	15.50
12:03	TG DY	9/29/2015	-65	25.42	-80.6	6.95	0.056	1.22	15.75
12:03	TG DB	9/29/2015	-75	25.13	-94.7	7.04	0.100	1.22	15.56
12:03	TG DR	9/29/2015	-90	25.52	-54.1	7.25	0.062	1.22	15.82
11:40	TG Y	10/6/2015	-25	22.84	222.2	7.25	0.050	1.21	14.02
11.40	TG B	10/6/2015	-35	23.89	214	7 36	0.082	1 21	14 73
11:40	тар	10/6/2015		23.03	175 5	7.50	0.002	1 21	17.21
11.40		10/6/2015	-+ EF	27.71	1/ 3.5	7.2	0.230	1.21	17.31 16 of
11.40		10/0/2015	-55	27.04	40.1	7.02	0.119	1.21	17.40
11:40		10/6/2015	-65	27.52	212.8	/.34	0.216	1.21	17.18
11:40	TG DB	10/6/2015	-75	27.14	186.7	7.33	0.140	1.21	16.92

Table A7:Pore water data for sipper samples collected during supermoon and
Hurricane Joaquin in Fall 2015

Location	Date	Depth (cm)	Arsenic (uM)	Electrical Conductivity (mS/cm)	Redox Potential (mV)	рН	Ferrous Iron (ppm)	Sulfide (ppm)	Salinity (ppt)
TG1A	4/29/2016	3		0.25	626.5	7.2	0.40		0.11
TG1B	4/29/2016	0		0.03	597.1	7.07			0.01
TG1C	4/29/2016	-8	0.010	0.03	613.9	7	0.44		0.01
TG1D	4/29/2016	-21	0.309	0.05	221.6	7.01	28.91		0.02
TG1E	4/29/2016	-36	0.197	0.04	156.4	6.96	23.01		0.01
TG1F	4/29/2016	-46	0.107	0.04	140.9	7.01	20.50		0.02
TG1G	4/29/2016	-61	0.027	0.04	134.4	6.82	3.69	1.81	0.01
TG1H	4/29/2016	-71	0.044	0.05	129.2	6.78	12.28		0.02
TG1I	4/29/2016	-86	0.039	0.04	142.9	6.79	11.05		0.01
TG2A	4/29/2016	3	0.015	0.04	301.1	6.69			0.01
TG2B	4/29/2016	0		0.04	315.8	6.52			0.01
TG2C	4/29/2016	-8	0.024	0.04	322.2	7.4			0.01
TG2D	4/29/2016	-21	0.193	0.05	280.9	7.11	1.63		0.02
TG2E	4/29/2016	-36	0.345	0.04	152.2	7.06	29.36		0.01
TG2F	4/29/2016	-46	0.164	0.03	136.1	6.91	20.40		0.01
TG2G	4/29/2016	-61	0.226	0.04	151.2	6.85	22.56		0.01
TG2H	4/29/2016	-71	0.022	0.04	148	6.77	25.25		0.01
TG2I	4/29/2016	-86	0.028	0.03	147	6.61	6.20	1.33	0.01
TG3A	4/29/2016	3	0.007	0.03	281.6	6.85			0.01
TG3B	4/29/2016	0	0.010	0.03	295.4	7.29			0.01
TG3C	4/29/2016	-8	0.099	0.03	305.1	7.06			0.01
TG3D	4/29/2016	-21	0.277	0.17	113.8	7.06	21.72		0.07
TG3E	4/29/2016	-36	0.087	0.06	110.2	7.07	12.65		0.02
TG3F	4/29/2016	-46	0.135	0.05	105.4	7.01	20.50		0.02
TG3G	4/29/2016	-61	0.101	0.05	123.2	6.87	16.10		0.02
TG3H	4/29/2016	-71	0.091	0.02	126.7	6.75	25.13		0.01
TG3I	4/29/2016	-86	0.049	0.04	162.7	6.45	19.62		0.02
TG1A	5/20/2016	3	0.012	0.72	617.8	7.37		2.81	0.33
TG1B	5/20/2016	0	0.154	4.69	603.1	7.81	1.62	2.72	2.51
TG1C	5/20/2016	-8	0.222	3.37	154.4	7.61	21.22	2.74	1.75
TG1D	5/20/2016	-21	0.317	2.15	100.5	8.11	27.88	2.98	1.07
TG1E	5/20/2016	-36	0.069	5.32	104.1	7.82	12.24	3.03	2.87
TG1F	5/20/2016	-46	0.073	2.81	105.9	7.78	13.65	2.86	1.43
TG1G	5/20/2016	-61	0.059	1.05	120.1	7.61	20.22	2.91	0.49
TG1H	5/20/2016	-71	0.041	4.11	123.5	7.55	20.20	2.79	2.17

Table A8:Pore water data for peeper samples collected during baseline conditions
from April to November 2016.

TG1I	5/20/2016	-86	0.032	1.93	150.7	7.67	14.58	2.74	0.96
TG2A	5/20/2016	3	0.010	1.06	256.1	7.61		2.58	0.50
TG2B	5/20/2016	0	0.033	2.51	303.3	7.94		2.56	1.27
TG2C	5/20/2016	-8	0.148	1.33	121.7	7.77	15.77	2.84	0.63
TG2D	5/20/2016	-21	0.385	5.22	81.8	7.62	35.41	2.79	2.82
TG2E	5/20/2016	-36	0.128	3.33	132.1	7.61	12.48	2.63	1.72
TG2F	5/20/2016	-46	0.053	1.97	129.3	7.75	11.58	3.03	0.98
TG2G	5/20/2016	-61	0.017	2.17	127.1	7.59	8.15	3.80	1.08
TG2H	5/20/2016	-71	0.092	4.11	136.2	7.85	10.44	2.67	2.17
TG2I	5/20/2016	-86	0.023	4.48	132.4	8.05	3.19	8.30	2.38
TG3A	5/20/2016	3	0.012	0.85	302.7	7.69		2.53	0.39
TG3B	5/20/2016	0	0.056	7.83	291.2	7.84	0.58	2.53	4.37
TG3C	5/20/2016	-8	0.145	0.01	140.7	7.65	17.91	3.24	0.00
TG3D	5/20/2016	-21	0.111	3.70	115.5	7.45	19.38	3.19	1.94
TG3E	5/20/2016	-36	0.172	1.87	92.6	7.69	19.59	2.96	0.92
TG3F	5/20/2016	-46	0.201	5.00	82.3	7.76	21.10	2.98	2.68
TG3G	5/20/2016	-61	0.071	4.80	98	7.67	17.79	2.88	2.57
TG3H	5/20/2016	-71	0.086	5.69	104.5	7.57	21.51	2.67	3.09
TG3I	5/20/2016	-86	0.054	3.37	127.9	7.6	18.01	2.77	1.75
TG1A	6/8/2016	3	0.017	0.02	586.2	3.59	0.76	2.48	0.01
TG1B	6/8/2016	0	0.023	1.90	537.4	6.31	0.52	2.46	0.94
TG1C	6/8/2016	-8	0.244	2.93	126.6	6.75	13.22	2.60	1.50
TG1D	6/8/2016	-21	0.103	2.23	87.7	6.83	19.40	3.36	1.12
TG1E	6/8/2016	-36	0.113	1.94	90.6	6.86	16.77	2.67	0.96
TG1F	6/8/2016	-46	0.071	3.44	94	6.81	11.52	2.62	1.79
TG1G	6/8/2016	-61	0.086	1.12	112.9	6.74	13.32	2.50	0.53
TG1H	6/8/2016	-71	0.018	1.54	104	6.83	6.84	4.50	0.74
TG1I	6/8/2016	-86	0.044	1.58	131.7	6.8	9.05	2.50	0.77
TG2A	6/8/2016	3	0.009	0.00	471.6	3.34	0.70	2.36	0.00
TG2B	6/8/2016	0	0.021	0.00	245.4	7.03		2.41	0.00
TG2C	6/8/2016	-8	0.452	3.63	67.2	7.04	34.41	2.69	1.90
TG2D	6/8/2016	-21	0.545	1.02	78.4	6.84	50.78	2.80	0.48
TG2E	6/8/2016	-36	0.400	4.63	65.4	6.95	33.60	2.62	2.47
TG2F	6/8/2016	-46	0.211	1.33	93	6.83	19.77	2.46	0.63
TG2G	6/8/2016	-61	0.058	1.08	122.5	6.75	12.03	2.55	0.50
TG2H	6/8/2016	-71	0.044	1.06	117.2	6.64	9.27	2.64	0.49
TG2I	6/8/2016	-86	0.058	1.46	126.4	6.7	13.24	2.48	0.70
TG3A	6/8/2016	3	0.057	0.00	-16.6	6.85		2.43	0.00
TG3B	6/8/2016	0	0.755	0.61	8.6	6.9	22.06	2.80	0.27

TG3C	6/8/2016	-8	0.393	4.03	-7.9	6.88	25.34	2.55	2.12
TG3D	6/8/2016	-21	0.261	5.55	-2	6.96	14.64	3.04	3.01
TG3E	6/8/2016	-36	0.462	2.78	4.7	6.86	24.81	2.60	1.42
TG3F	6/8/2016	-46	0.240	0.80	-8.7	6.86	17.09	2.53	0.36
TG3G	6/8/2016	-61	0.160	0.91	3.6	6.74	17.48	2.48	0.42
TG3H	6/8/2016	-71	0.066	2.65	-7.2	6.67	18.02	2.78	1.35
TG3I	6/8/2016	-86	0.039	1.99	-11.5	6.66	9.17	3.22	0.99
TG1A	6/27/2016	3		0.24	458.8	3.51		4.19	0.10
TG1B	6/27/2016	0	0.015	2.51	146.2	5.73		3.20	1.27
TG1C	6/27/2016	-8	0.030	4.28	113.8	6.83		3.34	2.27
TG1D	6/27/2016	-21	0.302	7.05	76.2	6.95	16.10	4.50	3.90
TG1E	6/27/2016	-36	0.118	6.10	58.3	6.94	12.20	6.15	3.34
TG1F	6/27/2016	-46	0.046	4.93	89.1	6.93	11.24	0.19	2.65
TG1G	6/27/2016	-61	0.192	6.58	69.1	6.8	29.15	4.26	3.62
TG1H	6/27/2016	-71	0.072	3.45	73.4	6.85	21.21	3.70	1.79
TG1I	6/27/2016	-86	0.020	4.46	101.2	6.76	12.20	4.71	2.37
TG2A	6/27/2016	3		0.00	511	3.67		3.06	
TG2B	6/27/2016	0	0.014	0.05	385.8	5.6		3.03	0.02
TG2C	6/27/2016	-8	0.011	0.19		6.09		4.08	0.08
TG2D	6/27/2016	-21	0.136	2.96	189.5	6.98	4.27	2.96	1.52
TG2E	6/27/2016	-36	0.110	13.99	127.3	7.03	24.53	4.12	8.23
TG2F	6/27/2016	-46	0.214	4.52	103.8	6.85	24.21	0.19	2.41
TG2G	6/27/2016	-61	0.044	1.04	132.7	7.12	6.24	3.80	0.49
TG2H	6/27/2016	-71	0.015	0.22	132.3	6.87	7.78	6.22	0.09
TG2I	6/27/2016	-86	0.052	11.18	134.4	6.9	8.95	3.87	6.45
TG3A	6/27/2016	3	0.018	0.37	87.1	3.86		2.89	0.16
TG3B	6/27/2016	0	0.058	13.06	8.1	7.1	1.13	3.13	7.63
TG3C	6/27/2016	-8		19.08	79.3	6.77		2.92	11.53
TG3D	6/27/2016	-21	0.175	3.09	39.4	6.89	11.10	4.05	1.59
TG3E	6/27/2016	-36	0.264	2.63	16	7.01	13.23	6.18	1.34
TG3F	6/27/2016	-46	0.137	2.42	42.7	6.93	8.69	0.16	1.22
TG3G	6/27/2016	-61	0.079	7.59	40.1	6.78	17.69	5.55	4.23
TG3H	6/27/2016	-71	0.073	1.70	7.6	6.78	18.37	4.64	0.83
TG3I	6/27/2016	-86	0.048	1.39	67.1	6.77	14.67	4.89	0.67
TG1A	7/20/2016	3	0.016	0.16	228.6	3.72		1.44	0.06
TG1B	7/20/2016	0	0.009	0.00	229.3	5.13	1.11	1.70	0.00
TG1C	7/20/2016	-8	0.057	1.56	227.6	7		2.90	0.76
TG1D	7/20/2016	-21	0.269	0.00	225.6	6.97	14.29	3.16	0.00
TG1E	7/20/2016	-36	0.209	0.00	228	7.13	17.03	2.65	0.00

TG1F	7/20/2016	-46	0.061	1.32	226.9	6.91	9.08	5.82	0.63
TG1G	7/20/2016	-61	0.051	0.02	227.2	6.82	14.63	1.57	0.01
TG1H	7/20/2016	-71	0.029	0.01	226.5	6.75	14.23	3.73	0.00
TG1I	7/20/2016	-86	0.026	1.70	224.8	6.64	15.74	5.88	0.83
TG2A	7/20/2016	3		0.15	249.2	3.65		2.27	0.06
TG2B	7/20/2016	0		0.02	248.2	3.52		1.32	0.01
TG2C	7/20/2016	-8	0.071	0.00	236.8	7.57		0.87	0.00
TG2D	7/20/2016	-21	0.105	1.15	238.1	6.86		1.00	0.54
TG2E	7/20/2016	-36	0.188	0.00	238.3	6.84	17.14	4.36	0.00
TG2F	7/20/2016	-46	0.166	0.00	241.1	6.82	22.47	3.03	0.00
TG2G	7/20/2016	-61	0.057	0.01	230.3	6.73	19.84	5.12	0.00
TG2H	7/20/2016	-71	0.032	0.00	227.4	6.75	14.94	4.99	0.00
TG2I	7/20/2016	-86	0.018	0.01	220.5	6.52	5.17	4.61	0.00
TG3A	7/20/2016	3	0.007	0.00	220.8	3.71		0.94	0.00
TG3B	7/20/2016	0	0.039	0.01	219.5	7.18		1.19	0.00
TG3C	7/20/2016	-8	0.217	2.70	221	7.46		0.81	1.37
TG3D	7/20/2016	-21	0.040	0.01	221	6.91		1.38	0.00
TG3E	7/20/2016	-36	0.595	0.01	218.9	7.01	26.05	3.54	0.00
TG3F	7/20/2016	-46	0.120	0.01	214.9	6.86	8.83	7.46	0.00
TG3G	7/20/2016	-61	0.069	1.13	214.6	6.77	18.75	4.36	0.53
TG3H	7/20/2016	-71	0.083	0.01	215.1	6.78	26.32	3.28	0.00
TG3I	7/20/2016	-86	0.040	2.44	231	6.45	19.46	2.40	1.23
TG1A	8/8/2016	3	0.016	0.24	-15.4	4.39		7.47	0.10
TG1B	8/8/2016	0	0.011	7.80	-13.8	6.55		8.26	4.36
TG1C	8/8/2016	-8	0.290	0.96	-11.7	7.16	6.58	3.67	0.45
TG1D	8/8/2016	-21	0.413	2.75	-15.7	7.12	15.28	3.45	1.40
TG1E	8/8/2016	-36	0.084	5.91	-12.8	7.07	13.46	3.62	3.22
TG1F	8/8/2016	-46	0.057	13.90	-8.5	6.93	16.50	2.86	8.17
TG1G	8/8/2016	-61	0.109	2.67	-18.1	6.95	20.08	4.76	1.35
TG1H	8/8/2016	-71	0.022	5.82	-14.9	6.82	9.10	4.73	3.17
TG1I	8/8/2016	-86	0.017	3.64	-32	6.69	9.49	3.70	1.90
TG2A	8/8/2016	3	0.018	2.51	264.1	6.54	3.44	3.76	1.27
TG2B	8/8/2016	0	0.033	16.32	258.4	7.02		3.31	9.73
TG2C	8/8/2016	-8	0.196	5.39	130.3	6.94	8.94	10.38	2.91
TG2D	8/8/2016	-21	0.284	0.53	147.6	6.8	14.27	10.63	0.23
TG2E	8/8/2016	-36	0.378	0.00	110.5	6.95	18.40	9.15	0.00
TG2F	8/8/2016	-46	0.395	1.90	90.2	6.81	32.84	10.46	0.94
TG2G	8/8/2016	-61	0.056	2.01	116.4	6.89	15.85	8.00	0.99
TG2H	8/8/2016	-71	0.037	3.66	100.9	6.76	16.11	8.09	1.91

TG2I	8/8/2016	-86	0.024	2.13	125.7	6.65	13.35	4.23	1.06
TG3A	8/8/2016	3	0.013	0.55	236.9	6.71	0.80	7.72	0.24
TG3B	8/8/2016	0	0.136	5.91	231.2	7.01		4.71	3.22
TG3C	8/8/2016	-8	0.427	2.02	97.1	7	14.38	6.72	1.00
TG3D	8/8/2016	-21	0.373	2.85	105.7	6.78	18.45	4.93	1.46
TG3E	8/8/2016	-36	0.442	0.60	75.3	7.01	18.92	6.49	0.27
TG3F	8/8/2016	-46	0.260	15.26	74.3	6.97	19.55	3.39	9.04
TG3G	8/8/2016	-61	0.041	11.64	103.2	6.61	17.46	13.03	6.74
TG3H	8/8/2016	-71	0.069	3.12	101.2	6.72	19.04	6.19	1.61
TG3I	8/8/2016	-86	0.050	3.76	100.6	6.71	16.79	4.09	1.97
TG1A	8/31/2016	3	0.013	0.00	67	3.52	3.39	4.79	0.00
TG1B	8/31/2016	0	0.010	0.01	82.5	3.79		4.82	0.00
TG1C	8/31/2016	-8	0.127	0.86	66.9	6.17	1.34	4.86	0.40
TG1D	8/31/2016	-21	0.410	1.96	73.3	6.77	20.44	4.93	0.97
TG1E	8/31/2016	-36	0.169	0.62	63.2	6.86	24.63	5.46	0.28
TG1F	8/31/2016	-46	0.060	3.30	63.3	6.79	19.88	5.88	1.71
TG1G	8/31/2016	-61	0.021	2.25	70.7	6.68	12.97	6.45	1.12
TG1H	8/31/2016	-71	0.014	2.09	75.4	6.8	16.82	5.35	1.04
TG1I	8/31/2016	-86	0.009	1.01	79.9	6.51	14.42	6.27	0.47
TG2A	8/31/2016	3	0.010	0.00	24	4.35		5.00	
TG2B	8/31/2016	0	0.007	0.01	12.5	6.95		4.68	0.00
TG2C	8/31/2016	-8	0.028	1.39	-4	7.06		4.61	0.67
TG2D	8/31/2016	-21	0.131	2.47	11.4	6.87		4.72	1.25
TG2E	8/31/2016	-36	0.267	0.64	23.9	6.99	11.17	5.74	0.29
TG2F	8/31/2016	-46	0.129	2.92	5.3	6.9	17.85	6.16	1.50
TG2G	8/31/2016	-61	0.102	3.03	58.6	6.79	30.03	5.42	1.56
TG2H	8/31/2016	-71	0.045	1.57	62.9	6.74	20.07	5.35	0.76
TG2I	8/31/2016	-86	0.025	1.28	13.1	6.36	14.50	5.81	0.61
TG3A	8/31/2016	3	0.007	0.00	-1.4	4		4.79	
TG3B	8/31/2016	0	0.024	0.00	-9.4	6.87		4.72	0.00
TG3C	8/31/2016	-8	0.040	3.82	-13.4	7.23		4.72	2.00
TG3D	8/31/2016	-21	0.323	2.44	-9.6	6.97	1.76	4.75	1.23
TG3E	8/31/2016	-36	0.309	3.42	-3.4	6.81	14.88	5.95	1.78
TG3F	8/31/2016	-46	0.324	4.97	-7.7	6.92	15.73	6.13	2.67
TG3G	8/31/2016	-61	0.044	3.96	-6.4	6.78	12.37	6.91	2.08
TG3H	8/31/2016	-71	0.024	4.21	-7.9	6.63	10.19	8.64	2.23
TG3I	8/31/2016	-86	0.016	1.12	-19	6.6	11.57	7.47	0.53
TG1A	9/21/2016	3	0.016	0.04	133.2	6.11		4.17	0.02
TG1B	9/21/2016	0	0.044	6.21	130.6	7.13		4.27	3.40

TG1C	9/21/2016	-8	0.156	2.05	178.6	6.73	7.16	4.90	1.02
TG1D	9/21/2016	-21	0.259	5.93	164.8	6.74	23.54	6.70	3.23
TG1E	9/21/2016	-36	0.066	1.79	179.9	7	10.00	8.31	0.88
TG1F	9/21/2016	-46	0.055	6.17	185.3	6.84	9.10	9.96	3.38
TG1G	9/21/2016	-61	0.038	7.62	157.5	6.66	14.63	6.46	4.25
TG1H	9/21/2016	-71	0.023	0.83	150.7	6.7	16.38	5.83	0.38
TG1I	9/21/2016	-86	0.036	4.82	147.9	6.67	20.25	5.92	2.58
TG2A	9/21/2016	3	0.010	1.07	85.9	4.4		4.37	0.50
TG2B	9/21/2016	0	0.017	1.68	93.2	4.67		4.42	0.82
TG2C	9/21/2016	-8	0.062	9.19	77.4	7.22	1.90	8.84	5.21
TG2D	9/21/2016	-21	0.125	5.78	77.4	7.03	10.75	9.28	3.15
TG2E	9/21/2016	-36	0.172	6.02	78.1	7.24	21.83	5.97	3.29
TG2F	9/21/2016	-46	0.171	4.31	77.8	7.01	23.54	7.19	2.28
TG2G	9/21/2016	-61	0.077	4.94	84.6	6.84	31.71	5.58	2.65
TG2H	9/21/2016	-71	0.042	3.95	87.7	6.83	24.32	5.78	2.08
TG2I	9/21/2016	-86	0.014	3.89	77.7	6.72	13.80	6.22	2.04
TG3A	9/21/2016	3	0.014	8.65	19.1	4.14		4.42	4.87
TG3B	9/21/2016	0	0.071	7.36	3.2	7.23		4.47	4.09
TG3C	9/21/2016	-8	0.096	6.79	13.5	7.51		4.42	3.74
TG3D	9/21/2016	-21	0.159	6.19	89.4	7.22	5.34	8.11	3.39
TG3E	9/21/2016	-36	0.121	4.54	71	7.05	5.53	10.98	2.42
TG3F	9/21/2016	-46	0.088	4.61	85.3	7.01	13.09	8.31	2.46
TG3G	9/21/2016	-61	0.035	4.08	91.5	6.83	15.42	5.58	2.15
TG3H	9/21/2016	-71	0.049	3.90	96.6	6.5	11.94	5.78	2.05
TG3I	9/21/2016	-86	0.029	3.06	94.9	6.64	13.55	6.02	1.58
TG1A	10/12/2016	3	0.010	2.84	169.5	7.05		0.38	1.45
TG1B	10/12/2016	0	0.024	2.98	145.9	7.06		0.37	1.53
TG1C	10/12/2016	-8	0.413	3.95	79.2	7.11	41.93	0.70	2.08
TG1D	10/12/2016	-21	0.121	5.75	113.2	6.99	12.70	1.21	3.13
TG1E	10/12/2016	-36	0.044	4.38	90.5	7.09	7.75	1.45	2.32
TG1F	10/12/2016	-46	0.057	1.63	113.3	7.04	11.25	1.39	0.80
TG1G	10/12/2016	-61	0.018	5.10	97.6	6.63	12.34	1.00	2.74
TG1H	10/12/2016	-71	0.017	3.42	117.9	6.85	9.97	0.99	1.78
TG1I	10/12/2016	-86	0.050	3.56	109.6	6.77	21.05	0.65	1.86
TG2A	10/12/2016	3	0.042	0.00	147.1	4.54		0.36	
TG2B	10/12/2016	0	0.009	4.40	102.9	6.42		0.32	2.33
TG2C	10/12/2016	-8	0.217	7.23	85.8	7.14	15.32	0.42	4.01
TG2D	10/12/2016	-21	0.250	10.55	89.1	7.05	20.58	0.66	6.05
TG2E	10/12/2016	-36	0.110	6.83	92.6	7.08	16.58	0.87	3.77

TG2F	10/12/2016	-46	0.079	3.18	91.8	7.05	16.85	0.97	1.64
TG2G	10/12/2016	-61	0.049	8.96	96.7	6.9	28.05	0.57	5.06
TG2H	10/12/2016	-71	0.022	6.79	100.6	6.94	22.77	0.76	3.75
TG2I	10/12/2016	-86	0.017	2.72	101.7	6.7	13.37	1.03	1.38
TG3A	10/12/2016	3	0.007	0.00	16.7	6.66		0.32	0.00
TG3B	10/12/2016	0	0.047	0.01	15.9	7.23		0.30	0.00
TG3C	10/12/2016	-8	0.130	12.93	27.3	7.19	5.74	0.73	7.55
TG3D	10/12/2016	-21	0.259	13.37	32.3	7.15	10.69	0.77	7.83
TG3E	10/12/2016	-36	0.207	7.55	36.8	7.12	17.40	0.70	4.20
TG3F	10/12/2016	-46	0.129	8.47	47.7	7	21.82	0.66	4.77
TG3G	10/12/2016	-61	0.036	4.29	50.3	6.61	18.91	0.71	2.27
TG3H	10/12/2016	-71	0.026	6.99	59.3	6.94	16.10	1.05	3.86
TG3I	10/12/2016	-86	0.015	3.20	52.1	6.77	11.27	1.03	1.65
TG1A	11/9/2016	3	0.009	0.20	398.3	3.39		0.16	0.08
TG1B	11/9/2016	0	0.009	0.66	277.8	5.78		0.08	0.30
TG1C	11/9/2016	-8	0.466	0.12	132	6.74	38.09	0.14	0.05
TG1D	11/9/2016	-21	0.173	3.26	133	6.99	12.27	0.44	1.69
TG1E	11/9/2016	-36	0.073	3.59	128.8	7.02	11.58	1.04	1.87
TG1F	11/9/2016	-46	0.081	4.65	125.3	6.87	19.08	0.53	2.48
TG1G	11/9/2016	-61	0.063	18.28	154.6	6.91	21.22	6.03	11.01
TG1H	11/9/2016	-71	0.039	5.20	130.4	6.77	15.59	0.61	2.80
TG1I	11/9/2016	-86	0.017	1.87	140.4	6.67	15.15	0.44	0.92
TG2A	11/9/2016	3		0.02	481	3.34		0.27	0.01
TG2B	11/9/2016	0		0.02	439.1	3.81		0.09	0.01
TG2C	11/9/2016	-8	0.029	10.25	251.4	7.79		0.10	5.87
TG2D	11/9/2016	-21	0.086	29.53	256	7.3	0.72	0.34	18.54
TG2E	11/9/2016	-36	0.098	9.30	121.6	6.95	18.39	0.60	5.28
TG2F	11/9/2016	-46	0.037	20.13	151.2	7	8.56	0.72	12.22
TG2G	11/9/2016	-61	0.051	2.54	119.7	6.78	30.63	0.22	1.28
TG2H	11/9/2016	-71	0.042	0.44	125.6	6.76	26.14	0.29	0.19
TG2I	11/9/2016	-86	0.023	1.85	134.3	6.62	18.39	0.65	0.91
TG3A	11/9/2016	3		0.02	365.3	3.64		0.01	0.01
TG3B	11/9/2016	0	0.011	0.43	260.1	7.52			0.18
TG3C	11/9/2016	-8	0.032	6.97	226.7	7.71		0.12	3.86
TG3D	11/9/2016	-21	0.115	4.76	202.6	7.27		0.01	2.54
TG3E	11/9/2016	-36	0.494	5.79	111.8	7.04	31.14	0.23	3.15
TG3F	11/9/2016	-46	0.236	3.72	125	6.93	21.92	0.34	1.95
TG3G	11/9/2016	-61	0.084	3.48	134.9	6.78	28.30	0.43	1.81
TG3H	11/9/2016	-71	0.027	3.54	144.4	6.55	18.18	0.80	1.85

TG3I 11/9/2016 -86 0.026 3.31 133.8 6.58 21.83 0.44 1.7	TG3I	11/9/2016	-86	0.026	3.31	133.8	6.58	21.83	0.44	1.72
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Table A9:	Pore water da	ta for sample	es collected	during bate	ch incubation
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Sample ID	Salinity Treatment	Redox Treatment	Vial Number	Time (bours)	Arsenic (uM)	Redox Potential (mV)	рН	Salinity (ppt)
15ppt Oxic 1	15 ppt	Oxic	1	0.5	0.165	355.4	6.96	18.19
15ppt Oxic 2	15 ppt	Oxic	2	0.5	0.176	353.4	6.93	20.57
15ppt Oxic 3	15 ppt	Oxic	3	0.5	0.190	355.3	6.88	22.03
15ppt Oxic 4	15 ppt	Oxic	4	1	0.175	372.9	7.12	6.97
15ppt Oxic 5	15 ppt	Oxic	5	1	0.167	379.9	7.08	17.99
15ppt Oxic 7	15 ppt	Oxic	7	6	0.155	369.1	6.88	18.45
15ppt Oxic 8	15 ppt	Oxic	8	6	0.130	370.8	6.94	18.39
15ppt Oxic 9	15 ppt	Oxic	9	6	0.156	370.8	6.87	21.16
15ppt Oxic 10	15 ppt	Oxic	10	12	0.123	375.5	7.16	0.34
15ppt Oxic 11	15 ppt	Oxic	11	12	0.078	374.8	7.19	14.87
15ppt Oxic 12	15 ppt	Oxic	12	12	0.146	377	7.19	19.90
15ppt Oxic 13	15 ppt	Oxic	13	24	0.154	358.1	7.17	14.54
15ppt Oxic 14	15 ppt	Oxic	14	24	0.148	357.6	7.07	15.58
15ppt Oxic 15	15 ppt	Oxic	15	24	0.154	357.2	7.08	15.64
15ppt Oxic 16	15 ppt	Oxic	16	48	0.192	325.7	6.84	16.94
15ppt Oxic 17	15 ppt	Oxic	17	48	0.188	324.1	6.87	17.73
15ppt Oxic 18	15 ppt	Oxic	18	48	0.180	322.3	6.97	17.01
15ppt Oxic 19	15 ppt	Oxic	19	72	0.185	275.5	6.8	16.68
15ppt Oxic 20	15 ppt	Oxic	20	72	0.196	270.2	6.82	17.80
15ppt Oxic 21	15 ppt	Oxic	21	72	0.210	265.1	7.23	17.01
15ppt Oxic 22	15 ppt	Oxic	22	144	1.896	247.4	6.7	19.90
15ppt Oxic 23	15 ppt	Oxic	23	144	2.115	246	6.74	20.37
15ppt Oxic 24	15 ppt	Oxic	24	144	1.724	245.3	6.76	20.57
15ppt Oxic 25	15 ppt	Oxic	25	168	2.449	205.7	6.73	20.17
15ppt Oxic 26	15 ppt	Oxic	26	168	1.893	205.8	6.77	22.03
15ppt Oxic 27	15 ppt	Oxic	27	168	1.872	206	6.78	22.10
15ppt Anoxic 1	15 ppt	Anoxic	1	0.5	0.200	173.1	7.05	16.29
15ppt Anoxic 2	15 ppt	Anoxic	2	0.5	0.175	165.4	7.07	16.03
15ppt Anoxic 3	15 ppt	Anoxic	3	0.5	0.184	152.2	7.08	
15ppt Anoxic 4	15 ppt	Anoxic	4	1	0.104	169.6	7.15	15.58
15ppt Anoxic 5	15 ppt	Anoxic	5	1	0.098	175.5	7.03	15.84
15ppt Anoxic 7	15 ppt	Anoxic	7	6	0.110	217.2	7.02	14.87

15ppt Anoxic 8	15 ppt	Anoxic	8	6	0.110	217.1	7.07	15.64
15ppt Anoxic 9	15 ppt	Anoxic	9	6	0.149	231.5	7	15.90
15ppt Anoxic 10	15 ppt	Anoxic	10	12	0.159	276.2	7.09	14.93
15ppt Anoxic 11	15 ppt	Anoxic	11	12	0.123	272.1	7.13	15.77
15ppt Anoxic 12	15 ppt	Anoxic	12	12	0.138	269.5	7.15	15.90
15ppt Anoxic 13	15 ppt	Anoxic	13	24	0.151	259.4	7.17	15.19
15ppt Anoxic 14	15 ppt	Anoxic	14	24	0.135	255.1	7.22	15.77
15ppt Anoxic 15	15 ppt	Anoxic	15	24	0.144	211.5	7.08	15.71
15ppt Anoxic 16	15 ppt	Anoxic	16	48	0.294	112.4	6.82	17.14
15ppt Anoxic 17	15 ppt	Anoxic	17	48	0.198	206.8	6.87	18.25
15ppt Anoxic 18	15 ppt	Anoxic	18	48	0.307	128.4	6.87	18.12
15ppt Anoxic 19	15 ppt	Anoxic	19	72	0.547	85.5	6.79	17.34
15ppt Anoxic 20	15 ppt	Anoxic	20	72	0.752	74.2	6.86	17.93
15ppt Anoxic 21	15 ppt	Anoxic	21	72	0.733	69.8	6.9	18.25
15ppt Anoxic 22	15 ppt	Anoxic	22	144	8.695	10	7.07	21.70
15ppt Anoxic 23	15 ppt	Anoxic	23	144	7.832	8.1	7.07	21.63
15ppt Anoxic 24	15 ppt	Anoxic	24	144	8.608	7	7.1	21.56
15ppt Anoxic 25	15 ppt	Anoxic	25	168	8.987	-1	7	22.50
15ppt Anoxic 26	15 ppt	Anoxic	26	168	2.528	-3.1	7.08	22.23
15ppt Anoxic 27	15 ppt	Anoxic	27	168	6.528	5.7	7	22.83
5ppt Oxic 1	5 ppt	Oxic	1	0.5	0.249	346.2	6.93	9.87
5ppt Oxic 2	5 ppt	Oxic	2	0.5	0.236	349.7	6.88	14.48
5ppt Oxic 3	5 ppt	Oxic	3	0.5	0.256	343.3	6.96	11.54
5ppt Oxic 4	5 ppt	Oxic	4	1	0.386	350.5	6.94	1.16
5ppt Oxic 6	5 ppt	Oxic	6	1	0.201	366.4	7.01	7.07
5ppt Oxic 7	5 ppt	Oxic	7	6	0.216	361.3	6.91	7.58
5ppt Oxic 8	5 ppt	Oxic	8	6	0.200	362.9	6.89	10.55
5ppt Oxic 9	5 ppt	Oxic	9	6	0.207	362.8	6.94	12.62
5ppt Oxic 10	5 ppt	Oxic	10	12	0.230	376.9	6.97	10.37
5ppt Oxic 11	5 ppt	Oxic	11	12	0.219	375.5	7.07	13.07
5ppt Oxic 12	5 ppt	Oxic	12	12	0.231	371.2	7.09	10.72
5ppt Oxic 13	5 ppt	Oxic	13	24	0.176	350.3	7.25	4.84
5ppt Oxic 14	5 ppt	Oxic	14	24	0.181	348.2	7.24	5.31
5ppt Oxic 15	5 ppt	Oxic	15	24	0.183	348.2	7.19	5.45
5ppt Oxic 16	5 ppt	Oxic	16	48	0.345	317.9	7	6.80
5ppt Oxic 17	5 ppt	Oxic	17	48	0.322	315.3	7.06	6.89
5ppt Oxic 18	5 ppt	Oxic	18	48	0.337	315.4	7.16	7.33
5ppt Oxic 19	5 ppt	Oxic	19	72	0.364	273.4	7.63	6.83
5ppt Oxic 20	5 ppt	Oxic	20	72	0.364	278.2	7.07	6.49

5ppt Oxic 21	5 ppt	Oxic	21	72	0.362	285.4	6.95	7.21			
5ppt Oxic 22	5 ppt	Oxic	22	144	1.974	244.2	7.01	7.83			
5ppt Oxic 23	5 ppt	Oxic	23	144	2.734	247.1	7.32	10.44			
5ppt Oxic 24	5 ppt	Oxic	24	144	1.481	254.3	6.92	11.27			
5ppt Oxic 25	5 ppt	Oxic	25	168	2.021	221.8	6.82	10.74			
5ppt Oxic 26	5 ppt	Oxic	26	168	3.125	213.7	6.88	12.36			
5ppt Oxic 27	5 ppt	Oxic	27	168	1.829	201.8	7.47	12.56			
5ppt Anoxic 1	5 ppt	Anoxic	1	0.5	0.214	232	6.9	16.42			
5ppt Anoxic 2	5 ppt	Anoxic	2	0.5	0.226	203.6	6.96	17.40			
5ppt Anoxic 3	5 ppt	Anoxic	3	0.5	0.215	191.7	6.92	21.23			
5ppt Anoxic 4	5 ppt	Anoxic	4	1	0.252	168.3	7.16	5.66			
5ppt Anoxic 6	5 ppt	Anoxic	6	1	0.255	166.7	7.12	5.98			
5ppt Anoxic 7	5 ppt	Anoxic	7	6	0.180	225.8	7	5.70			
5ppt Anoxic 8	5 ppt	Anoxic	8	6	0.166	215	7.09	5.74			
5ppt Anoxic 9	5 ppt	Anoxic	9	6	0.227	211.6	7.07	5.84			
5ppt Anoxic 10	5 ppt	Anoxic	10	12	0.196	283.8	7.08	0.07			
5ppt Anoxic 11	5 ppt	Anoxic	11	12	0.194	291.8	7.17	5.65			
5ppt Anoxic 12	5 ppt	Anoxic	12	12	0.205	287.9	7.18	5.81			
5ppt Anoxic 13	5 ppt	Anoxic	13	24	0.223	222.6	7.1	4.71			
5ppt Anoxic 14	5 ppt	Anoxic	14	24	0.218	211.3	7.15	5.69			
5ppt Anoxic 15	5 ppt	Anoxic	15	24	0.225	213.1	7.15	5.56			
5ppt Anoxic 16	5 ppt	Anoxic	16	48	0.488	105.4	7.03	6.94			
5ppt Anoxic 17	5 ppt	Anoxic	17	48	0.401	104.1	7.08	7.15			
5ppt Anoxic 18	5 ppt	Anoxic	18	48	0.334	112.3	7.08	6.49			
5ppt Anoxic 19	5 ppt	Anoxic	19	72	1.160	43.2	6.96	8.12			
5ppt Anoxic 20	5 ppt	Anoxic	20	72	1.550	31.8	7.12	9.68			
5ppt Anoxic 21	5 ppt	Anoxic	21	72	1.182	36.6	7.1	10.40			
5ppt Anoxic 22	5 ppt	Anoxic	22	144	9.298	29.1	7.15	11.29			
5ppt Anoxic 23	5 ppt	Anoxic	23	144	6.194	36.1	7.11	9.84			
5ppt Anoxic 24	5 ppt	Anoxic	24	144	9.592	8.6	7.2	12.30			
5ppt Anoxic 25	5 ppt	Anoxic	25	168	4.845	-3.8	7.02	13.26			
5ppt Anoxic 26	5 ppt	Anoxic	26	168	2.178	56.5	6.95	13.64			
5ppt Anoxic 27	5 ppt	Anoxic	27	168	4.516	19.7	7.1	12.56			
Date	Collar Treatment	Collar Number	Abbreviation	Electrical Conductivity (mS/cm)	Salinity (ppt)	Arsenic (uM)	Sulfide (ppm)	Ferrous Iron (ppm)	Redox Potential (mV)	Ηα	Phase of Experiment
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2/6/2017	Control	1	C1	28.78	17.78	0.034	0.21	2.60	195.8	6.83	1
2/6/2017	Control	2	C2	28.62	17.66	0.028	0.24	0.00	231.6	6.59	1
2/6/2017	Control	3	C3	28.32	17.46	0.088	0.25	3.80	174.4	6.69	1
2/6/2017	Treatment	1	T1	29.03	17.94	0.101	0.22	4.07	180.1	6.45	1
2/6/2017	Treatment	2	Т2	28.01	17.25	0.100	0.23	7.64	168.4	6.54	1
2/6/2017	Treatment	3	Т3	28.51	17.59	0.055	0.22	3.97	164.8	6.52	1
2/8/2017	Control	1	C1	29.05	17.94	0.064	0.28	0.43	165.6	6.95	1
2/8/2017	Control	2	C2	29.04	17.99	0.088	0.25	0.00	243.5	6.61	1
2/8/2017	Control	3	C3	28.66	17.67	0.114	0.23	6.90	152.3	6.71	1
2/8/2017	Treatment	1	T1	29.40	18.17	0.115	0.22	5.18	183.2	6.58	1
2/8/2017	Treatment	2	Т2	28.47	17.54	0.119	0.22	9.95	175.2	6.63	1
2/8/2017	Treatment	3	Т3	28.69	17.69	0.077	0.23	5.88	162.4	6.67	1
2/14/2017	Control	1	C1	29.45	18.24	0.122	0.20	8.42	116.8	7.05	2
2/14/2017	Control	2	C2	28.34	17.48	0.041	0.19	2.26	153.5	6.93	2
2/14/2017	Control	3	C3	28.27	17.43	0.102	0.20	3.88	132.4	6.83	2
2/14/2017	Treatment	1	T1	22.45	13.55	0.080	0.20	4.81	124.5	6.8	2
2/14/2017	Treatment	2	Т2	24.81	15.11	0.150	0.22	24.91	137	6.88	2
2/14/2017	Treatment	3	Т3	28.15	17.35	0.251	0.23	12.32	88.6	6.83	2
2/15/2017	Control	1	C1	29.07	17.99	0.121	0.38	6.82	157.5	7.08	2
2/15/2017	Treatment	1	T1	23.53	14.26	0.104	0.40	3.36	151.4	6.79	2
2/15/2017	Treatment	2	Т2	24.12	14.66	0.115	0.42	8.35	134.6	6.78	2
2/15/2017	Treatment	3	Т3	23.34	14.14	0.159	0.44	7.60	120.9	6.83	2
2/16/2017	Control	1	C1	27.81	17.12	0.110	0.23	8.68	169.1	6.74	2
2/16/2017	Control	2	C2	28.35	17.49	0.023	0.22	0.67	212	6.79	2
2/16/2017	Control	3	С3	27.90	17.18	0.130	0.06	5.77	194.1	6.81	2
2/16/2017	Treatment	1	T1	21.43	12.88	0.082	0.25	4.98	200.6	6.58	2
2/16/2017	Treatment	2	Т2	21.34	12.81	0.139	0.25	32.10	178.4	6.68	2
2/16/2017	Treatment	3	Т3	21.09	12.66	0.172	0.27	10.07	157.5	6.7	2
2/18/2017	Control	1	C1	27.99	17.24	0.133	0.23	8.80	99.6	6.73	3
2/18/2017	Control	2	C2			0.045	0.22	0.86	111.3	6.77	3
2/18/2017	Control	3	С3	27.81	17.11	0.153	0.27	5.85	107.7	6.71	3
2/18/2017	Treatment	1	T1	21.35	12.81	0.158	0.23	7.07	100.6	6.65	3
2/18/2017	Treatment	2	Т2	18.66	11.07	0.077	0.25	6.45	117.5	6.92	3

Table A10: Pore water data for samples collected during flow through incubation

2/18/2017	Treatment	3	Т3	19.99	11.92	0.193	0.26	6.37	94.5	6.94	3
2/22/2017	Control	1	C1	27.83	17.12	0.153	0.24	12.56	144.6	6.76	3
2/22/2017	Control	2	C2	26.84	16.45	0.071	0.21	3.24	188.1	6.71	3
2/22/2017	Control	3	C3	27.93	17.20	0.103	0.20	5.00	221.7	6.91	3
2/22/2017	Treatment	1	T1	21.15	12.69	0.219	0.21	9.36	203.7	6.82	3
2/22/2017	Treatment	2	Т2	20.88	12.52	0.110	0.21	7.87	138.1	6.85	3
2/22/2017	Treatment	3	Т3	19.17	11.40	0.125	0.24	5.54	135	6.93	3
2/28/2017	Control	1	C1	27.54	16.94	0.210	0.25	16.02	163.1	6.7	3
2/28/2017	Control	2	C2	28.01	17.26	0.151	0.22	4.42	146.5	6.73	3
2/28/2017	Control	3	С3	27.91	17.19	0.199	0.22	7.94	153.1	6.76	3
2/28/2017	Treatment	1	T1	20.43	12.22	0.259	0.22	7.46	106.5	7	3
2/28/2017	Treatment	2	Т2	20.73	12.42	0.280	0.24	18.58	163.2	6.83	3
2/28/2017	Treatment	3	Т3	19.87	11.86	0.335	0.22	5.36	110.7	7.09	3
3/4/2017	Control	1	C1	28.01	17.25	0.254	0.27	15.40	159.1	6.87	4
3/4/2017	Control	2	C2	27.25	16.75	0.228	0.22	10.33	135.3	6.93	4
3/4/2017	Control	3	С3	28.33	17.48	0.192	0.22	5.97	140.4	6.85	4
3/4/2017	Treatment	1	T1	21.76	13.10	0.291	0.23	15.48	173.5	7.02	4
3/4/2017	Treatment	2	Т2	20.86	12.31	0.243	0.22	20.71	168.3	6.82	4
3/4/2017	Treatment	3	Т3	21.05	12.63	0.306	0.22	13.89	149.2	6.9	4
3/6/2017	Control	1	C1	28.82	17.79	0.186	0.26	13.49	154.2	6.95	4
3/6/2017	Control	2	C2	27.95	17.20	0.173	0.25	11.69	130.8	6.9	4
3/6/2017	Control	3	С3	29.29	18.11	0.204	0.25	12.71	152.9	6.69	4
3/6/2017	Treatment	1	T1	24.31	14.77	0.176	0.25	13.14	155	6.95	4
3/6/2017	Treatment	2	Т2	22.74	13.73	0.332	0.26	23.57	160.8	6.88	4
3/6/2017	Treatment	3	Т3	23.83	14.46	0.225	0.24	14.86	155.8	6.92	4

Table A11:Soil data for samples collected adjacent to the boardwalk, in the high-
elevation marsh, mid-elevation marsh, and low-elevation marsh of the St.
Jones Reserve. Data includes Sample ID, depth of collection, marsh
elevation zone, total As, and sequential extraction fractions of As
(abbreviated SEP)

		Marsh Elevation	Arsenic (mg/kg	SEP F1 As	SEP F2 As	SEP F3 As	SEP F4 As	SEP F5 As
Sample ID	Depth (cm)	Zone	soil)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)
Tr1	0	Mid	17.16					
Tr1	5	Mid	18.32					
Tr1	10	Mid	18.83					
Tr1	15	Mid	18.28					
Tr2	0	Low	9.48					
Tr2	5	Low	9.37					
Tr2	10	Low	10.21					
Tr2	15	Low	11.68					
Tr3	0	High	6.66					
Tr3	5	High	7.32	0.108	1.128	1.169	1.125	2.069
Tr3	10	High	6.03					
Tr3	15	High	5.91	0.227	0.759	1.033	1.111	2.003
Tr4	0	Mid	16.63					
Tr4	5	Mid	16.67	0.374	1.963	6.960	2.801	4.225
Tr4	10	Mid	15.54					
Tr5	0	Mid	16.84					
Tr5	5	Mid	18.03					
Tr5	10	Mid	16.55					
Tr5	15	Mid	14.9					
Tr5	20	Mid	15.57					
NC	0	Mid	14.9					
NC	5	Mid	18.37					
Path 1	0	High	4.49					
Path 2	0	High	5.59					
Path 3	0	High	7.55					
Ag 1	0	Low	14.37					
Ag 2	0	Low	12.23					
Ag 3	0	Low	12.46					
Ag 4	0	Low	8.81	0.085	2.878	3.793	1.755	2.791
Ag 5	0	Low	7.96					

Ag 6	0	Low	10.98					
SG1 C 50	0	NB 50	31.25					
SG2 C 50	0	NB 50	38.13	0.436	8.789	19.743	6.041	5.779
SG1 P 50	0	NB 50	50.13					
SG2 P 50	0	NB 50	56.89	0.490	11.829	22.110	8.199	13.484
TG3 C 50	0	NB 50	66.69	0.118	11.021	46.338	7.375	6.361
TG3 P 50	0	NB 50	31.21	0.062	5.291	18.549	4.869	3.153
TG1 C 50	0	NB 50	55.91					
TG1 P 50	0	NB 50	17.81					
SG1 C 25	0	NB 25	42.99					
SG2 C 25	0	NB 25	68.56					
SG1 P 25	0	NB 25	128.6					
SG2 P 25	0	NB 25	57.65					
TG3 C 25	0	NB 25	89.5					
TG3 P 25	0	NB 25	58.34					
TG1 C 25	0	NB 25	77.96					
TG1 P 25	0	NB 25	52.91					
SG1 C 0	0	NB 0	169.5					
SG2 P 0	0	NB 0	95.92					
TG3 C 0	0	NB 0	171.5	0.896	35.929	117.589	22.754	9.811
TG1 C 0	0	NB 0	209.1					
TG1 P 0	0	NB 0	164.3					
TG3 P 0	0	NB 0	79.4	0.328	15.288	53.700	15.151	7.262
SG2 C 0	0	NB 0	140.5	0.510	28.729	75.222	28.887	20.931
SG1 P 0	0	NB 0	232.6	1.418	51.228	151.673	32.819	13.585
TG 0-27	0	Mid	11.89					
TG 27-73	27	Mid	18.23					
TG 73+	73	Mid	23					