Supplemental Information

Oil & Gas Produced Water Retention Ponds as Potential Passive Treatment for Radium Removal and Beneficial Reuse

Bonnie McDevitt[1], Molly C. McLaughlin[2,3], Jens Blotevogel[3], Thomas Borch[3,4,5], Nathaniel R. Warner[1]*

[1] Department of Civil and Environmental Engineering, The Pennsylvania State University, 212 Sackett Building, University Park, PA 16801, USA
[2] Abt Associates Inc., 2755 Canyon Blvd., Boulder, CO, 80301
[3] Department of Civil and Environmental Engineering, Colorado State University, 1170 Campus Delivery, Fort Collins, CO, 80523-1170, USA
[4] Department of Soil and Crop Sciences, Colorado State University, 1170 Campus Delivery, Fort Collins, Colorado 80523, USA
[5] Department of Chemistry, Colorado State University, 1872 Campus Delivery, Fort Collins, Colorado, 80523, USA

*corresponding author, nrw6@psu.edu

2. Materials and Methods

2.1 Site description

The large O&G fields represented in this study are underlain by tertiary and quaternary deposits with soils reported comprising calcium carbonate mineral upwards of 40% and gypsum upwards of 15% [1]. However, McDevitt et al. (2019) previously found only trace amounts of gypsum in grab sediments and while calcium carbonate dominated grab sediments near NPDES discharges, compositions decreased with increasing distance from the discharges[2].

Facility DA-D discharges an average 1.5 million L treated PW/day which flows into a large, shallow, carbonate-terraced PWRP (DA-W1) that is approximately 40,000 m^2. From average DA-D discharge, and assuming the area of DA-W1 with a water depth of ~0.33 m, estimated hydraulic retention time in DA-W1 is approximately 9 days. From the first PWRP,

water flows approximately 0.3 km into a much larger PWRP (DA-W2) that is largely unvegetated apart from some exteriorly established grasses and is approximately 390,000 m^2. Estimated hydraulic retention time based on a depth of ~0.33 m is 86 days. From this second PWRP, treated PW flows approximately 12 additional km prior to discharge into a large perennial river (previously River B). Facility DB-D discharges an average 310,000 L treated PW/day that is combined from 13 wells. Approximately 0.8 km downstream from the discharge is a created PWRP (DB-W1) approximately 350 m^2. Assuming a 0.165 m wide and 0.165 m deep stream for 0.8km, estimated hydraulic retention time in DB-W1 is 0.19 days or ~4.6 hours. Water exits this PWRP from an outfall pipe (DB-DSW1) that feeds a much larger, shallow pond 0.3 km downstream which was dry during this sampling campaign. DB-D is approximately 56 km upstream of the confluence with a perennial river (previously River B). Facility DC-D discharges an average of 4.5 million L treated PW/day. Approximately 1.8 km downstream the water flows into a 450 m^2 created PWRP (DC-W1) after which a dam divides the discharge into two equivalent streams. One of the streams continues 5.2 km where it flows into a second 2500 m^2 created PWRP (DC-W2) and beyond for an additional 25 km prior to emptying into a large perennial river (previously River C). From estimated areas of DC-W1 and DC-W2 and assuming a consistent measured average upstream velocity of 0.036 m^3/s, estimated hydraulic retention time in DC-W1 is approximately 11 hours and in DC-W2 is approximately 4 days. Perennial Rivers B and C ultimately flow into a reservoir which has a municipal drinking water intake and is a crop irrigation source downstream. The second diverted stream flows approximately 2 km into a shallow, playa lake (DC-PLAYA) intended to sustain wildlife habitat.

Table S1: Previous study sample site names from McDevitt et al. 2019, 2020b for discharges represented in the current study.

Current Site ID	Previous Study ID
DA-D	DB-4.0
DB-D	DB-2.0
DB-100m	DB-2.0 100m
DB-DSW1	DB-2.1
DC-D	DC-1
DC-100m	DC-1 100m
DC-W1	downstream of DC-2

2.2 Field Sampling

Grab sediments for Ra analysis were collected in triplicate from the upper 5 cm of sediment and stored in plastic containers. Grab sediments for TOC and TIC analysis were collected in glass bottles. Grab samples were collected along the sides of the stream channels at the water surface level in areas of sediment accumulation.

Plastic core tubes were new and rinsed 3 times with field site water prior to sediment collection. More sediment cores would have been collected if possible but some of the sediments were either too gravelly to allow for pushing a core tube into or too sludgy to obtain enough suction to bring the sediment to the surface. Sediment cores were stored within the plastic tubing until extraction in the laboratory.

2.3 Laboratory Analysis of Samples

Grab sediments were dried in an oven at 60°C, ground to homogeneity with a mortar and pestle, sieved to 1.18 mm diameter in order to exclude rocks and larger material, and then packed into HDPE liquid scintillation vials with a urea gas seal cap. Vials were then wrapped with

parafilm to prevent the escape of ^{226}Ra daughter product, ^{222}Rn. Samples were then incubated for at least 21 days to establish secular equilibrium of ^{226}Ra daughter products, ^{214}Pb and ^{214}Bi. Samples were measured to counting errors < 5% per peak, background counts were subtracted, and a uranium ore tailing standard (UTS-2) from Canadian Certified Reference Material project was utilized as a calibration standard for detector efficiency.

Grab sediments were also analyzed for TC, TIC and TOC at Colorado State University. Frozen grab sediments were thawed, dried within glass storage containers at 105°C in a muffle furnace, and sieved to 2 mm diamater. TC was measured on a LECO TruSpec CN autosampler and reported as percent sediment mass (dry weight). Sidney High soil standard was used to calibrate the instrument. TIC was measured via a calcimeter pressure transducer and voltage meter after adding 2 mL of 6N HCl + 3% ferrous chloride solution to a 1.84 mL vial, capping and shaking for one minute, and allowing to rest for 2 hours. Concentrations were deduced from a 7-point $CaCO_3$ standard curve and reported as percent sediment mass (dry weight).

After each leaching step, sediment core samples were centrifuged for 20 minutes at 10,000 RPM. Leachates were then decanted and filtered through 0.45-µm nylon filters. After steps 2 through 5, residues were rinsed, shaken, and centrifuged three times with 5 mL each of ultra-pure distilled water to ensure adequate removal of target species. Rinse water was filtered and combined with initial leachate. Leachate pH was measured immediately following and preserved with nitric acid to pH < 2 for cation analysis. Sediment residues after each leaching step were freeze dried, packed into 24-mL liquid scintillation vials, and analyzed by gamma spectrometry. Because samples were not incubated for 21 days, ^{226}Ra was analyzed by direct measurement at the 186 keV peak after deconvolution of the ^{235}U 63 keV peak. ^{228}Ra was measured the same way as grab sediment and core samples, via the 911 keV peak.

A subset of sediment samples (n=8) that included the original sediments from the leached DC-W1 and DC-W2 core sections (DC-W1 3-5, DC-W1 13-15, DC-W1 23-25, DC-W2 2-4, DC-W2 10-12, DC-W2 16-18) and two final leaching residues from DC-W1 13-15 cm and DC-W2 10-12 cm were analyzed by X-ray diffraction (PANalytical Empyrean and Jade software) at the Material Characterization Laboratory (MCL) at The Pennsylvania State University. Quantification was only achieved for three samples due to the complexity of the mineralogical matrix. However, minerals are listed in order of relative abundance for all samples in Table S5.

Vegetation was thawed and then rinsed three times in an ultra-pure distilled water bath as a bulk sediment removal step. Vegetation was then soaked in a bath of ultra-pure distilled water and Triton™ X-100 surfactant (~1% solution) to remove recalcitrant clay particles from the vegetation [3]. A final ultra-pure distilled water bath and rinse was employed to remove the surfactant and any remaining sediment. Vegetation was separated into roots, leaves (cattails) or stems (grass and bulrush), and seeds if present. Samples were then freeze dried, ground to homogeneity and a small size fraction utilizing a blender, and then packed densely into 24-mL liquid scintillation vials for gamma analysis after 21 days of incubation similarly to grab and core sediments. Two samples of dried cattail seed heads (CSW and DC-W2) were separately stored upon collection and packed for gamma analysis without any additional preparation.

3. Results and Discussion

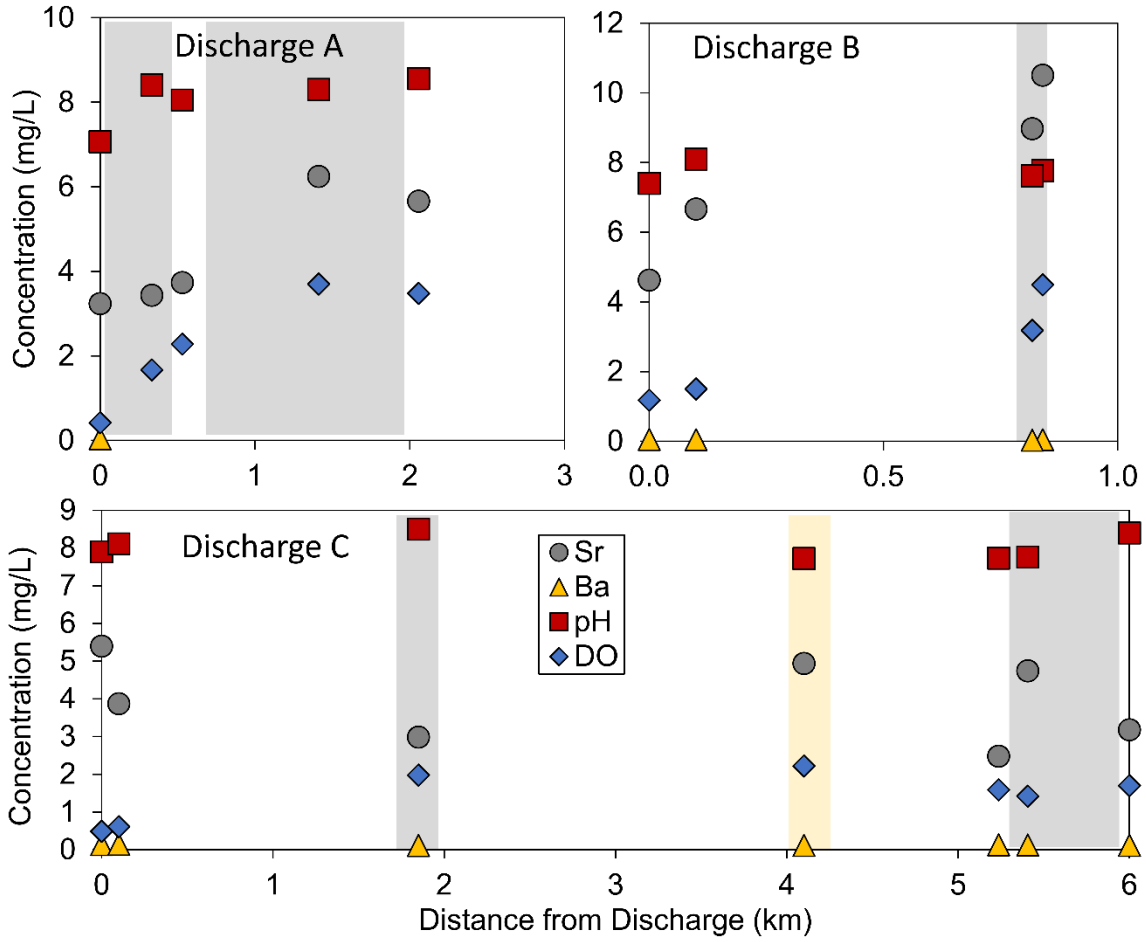

Figure S1. Sr, Ba, pH and dissolved oxygen (DO) concentrations versus distance from respective NPDES discharge A, B and C. Gray bars indicate PWRPs and the yellow bar represents the playa lake on the Discharge C PW stream.

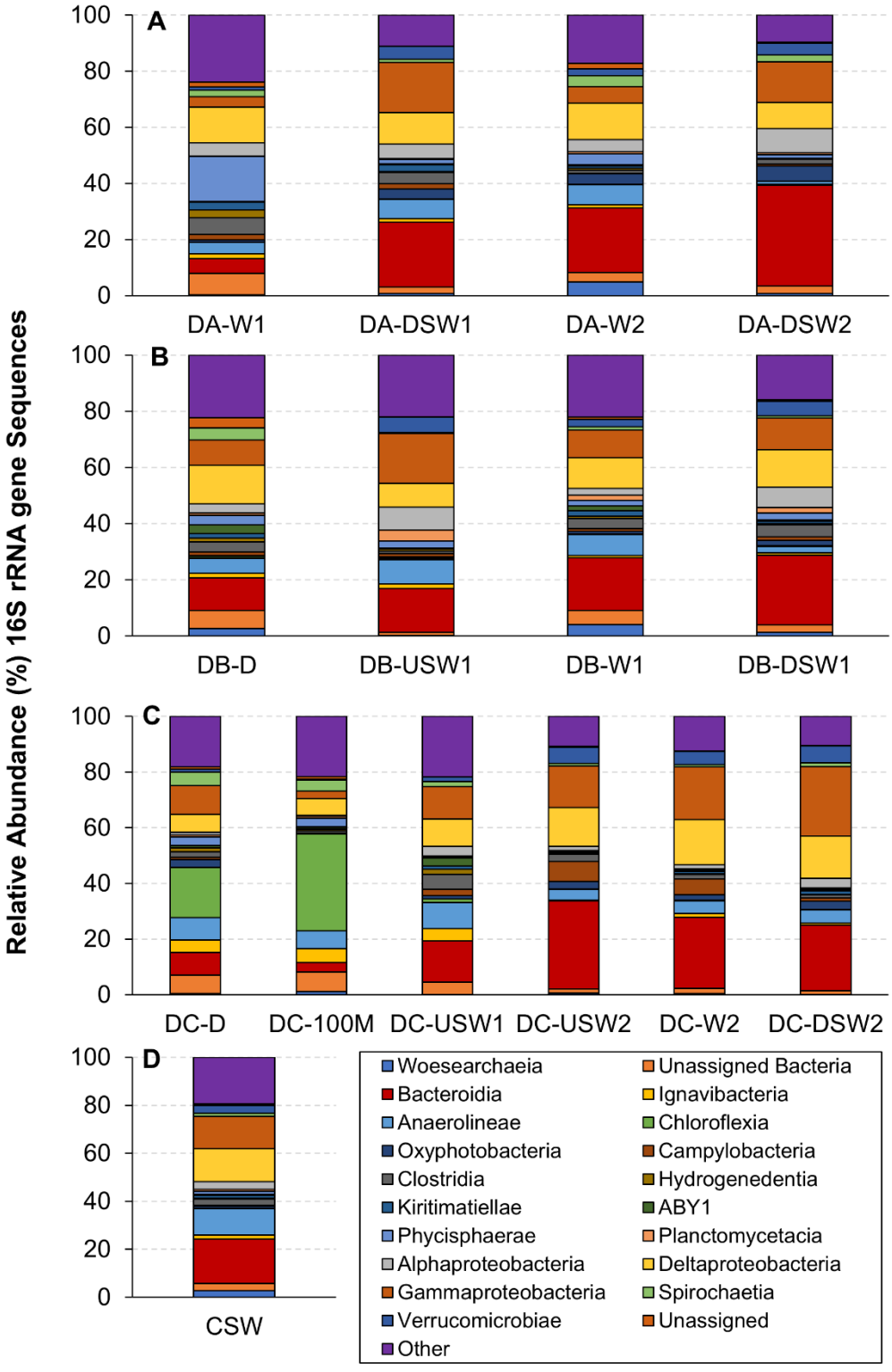

Figure S2. Grab sediment microbial community relative abundance (%) from 16S rRNA gene sequencing. Note the high relative abundance of Chloroflexia at DC-D and DC-100m and increasing relative abundance of Deltaproteobacteria from DC-D to DC-DSW2.

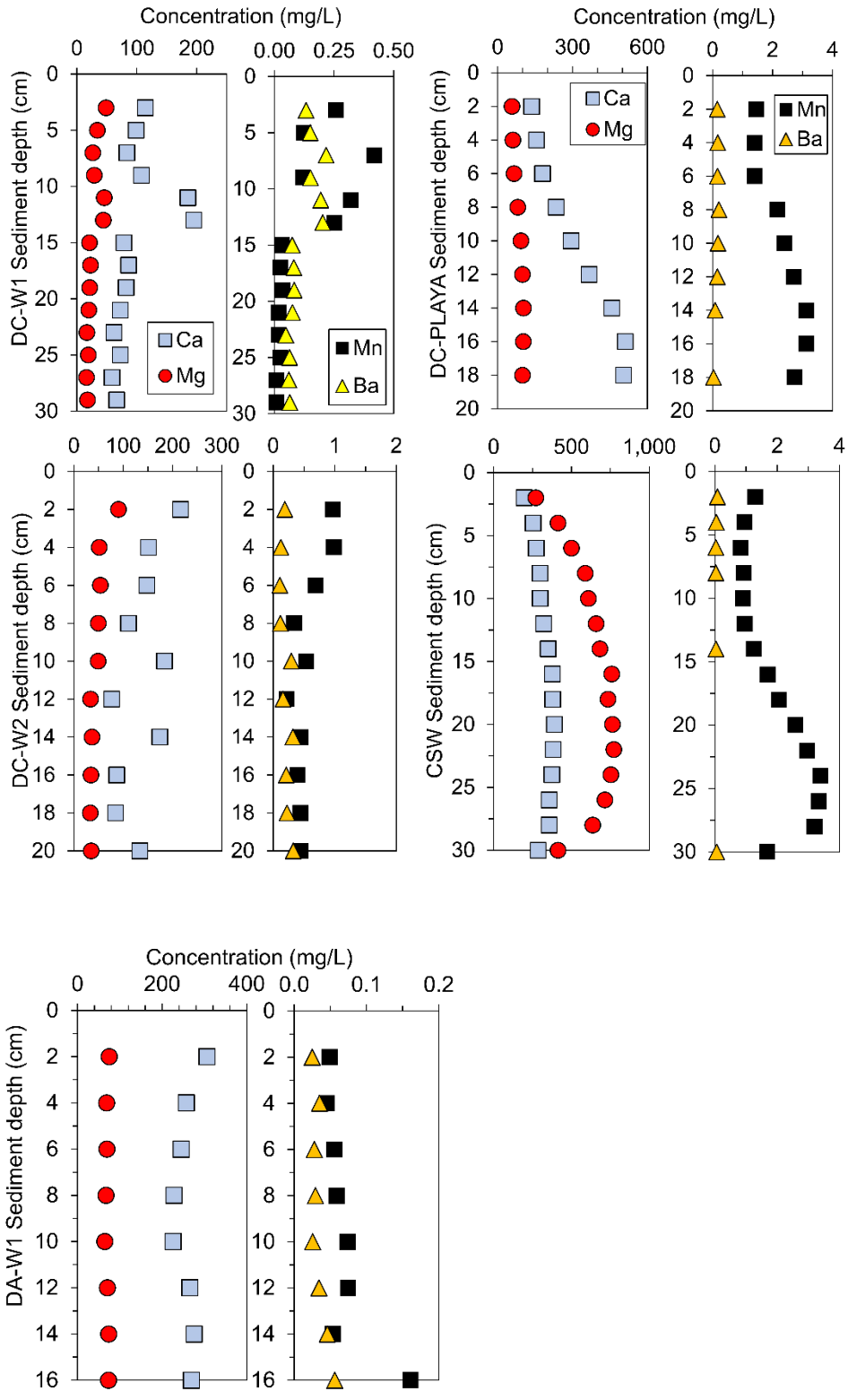

Figure S3. Scaled core porewater profiles for Ca, Mg, Ba and Mn concentrations versus sediment depth.

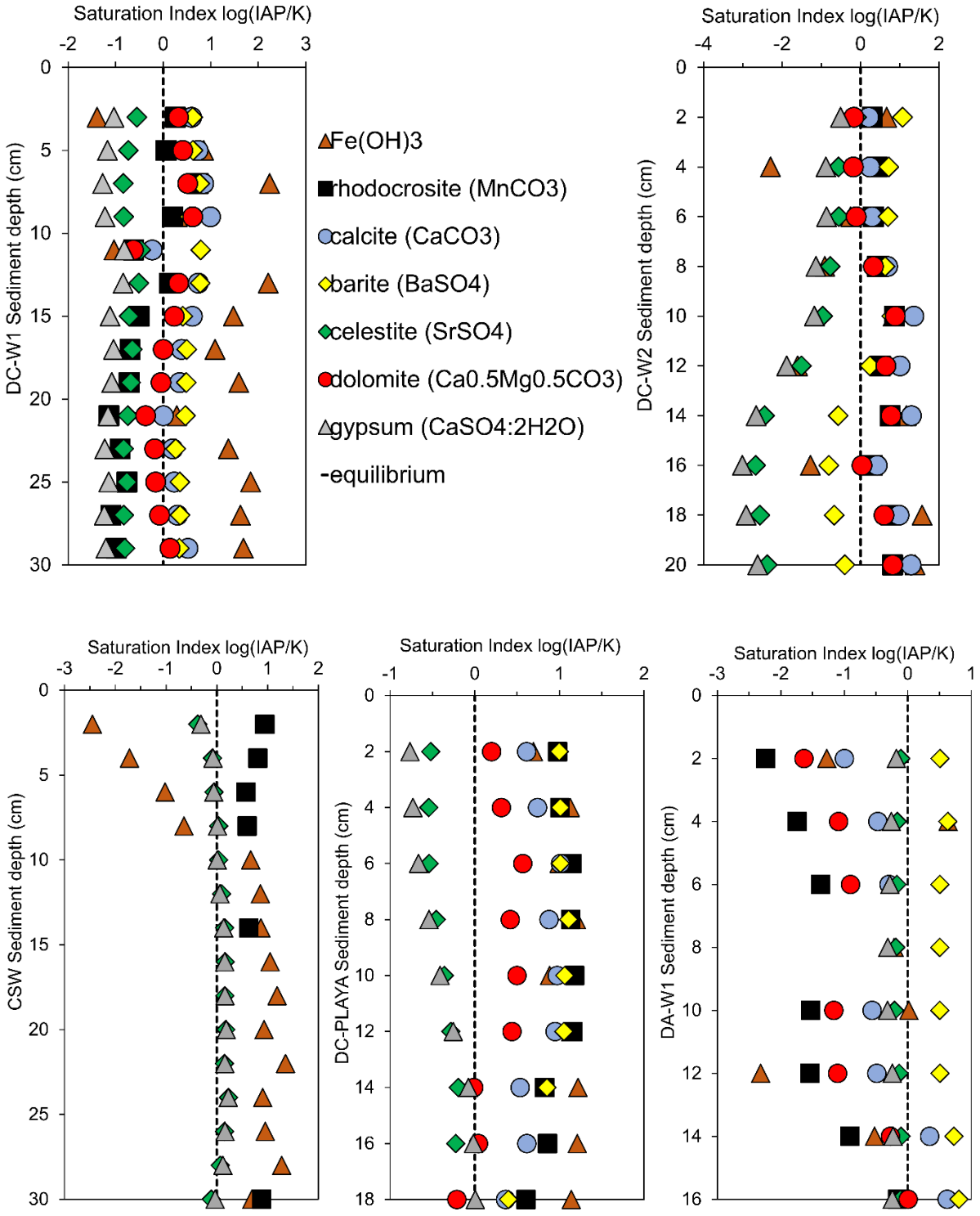

Figure S4. Saturation indices (SI) as log(IAP/K_{eq}) calculated using sediment core porewater chemistry and PHREEQC. The dashed line at an SI value of 0 indicates equilibrium. Minerals with an SI value greater than 0 indicate supersaturation and potential for dissolution whereas SI values less than 0 indicate undersaturation and potential for precipitation.

References

(1) NRCS. NRCS Web Soil Survey https://websoilsurvey.sc.egov.usda.gov/App/HomePage.htm.

(2) McDevitt, B.; McLaughlin, M.; Cravotta, C. A.; Ajemigbitse, M. A.; Van Sice, K. J.; Blotevogel, J.; Borch, T.; Warner, N. R. Emerging Investigator Series: Radium Accumulation in Carbonate River Sediments at Oil and Gas Produced Water Discharges: Implications for Beneficial Use as Disposal Management. *Environ. Sci. Process. Impacts* **2019**, *21* (2), 324–338. https://doi.org/10.1039/C8EM00336J.

(3) Edwards, W. R.; Smith, K. E. Exploratory Experiments on the Stability of Mineral Profiles of Feathers. *J. Wildl. Manage.* **1984**, *48* (3), 853–866. https://doi.org/10.2307/3801432.

Table S2. Field measurements, major anions and cations and grab sediment Ra-226, Ra-228 and Total Ra reported as Bq/kg. All anion and cation concentrations are reported as mg/L.

Site Name	Temperature (°C)	DO (mg/L)	pH	EC (uS/cm)	Cl	SO_4	Br	NO_3	PO_4	Sr	Mn	K	Mg	Ca	Ba	B	Na	Li	^{226}Ra ± error (Bq/kg)	^{228}Ra ± error (Bq/kg)	Total Ra ± error (Bq/kg)
CSW	4.85	1.56	8.80	900	6	318		0.14	0.08	4.3	-	3	41	44	0.02	0.08	81	0.08	44.4 ± 0.6	45.2 ± 2.0	89.6 ± 4.2
DA-D	35.43	0.42	7.07	6404	672	1868	1.0	0.13	0.11	3.2	-	80	71	304	0.04	3.09	780	1.46	1013.3 ± 3.5	739.9 ± 10.0	1753.2 ± 24.4
DA-DSW1	3.62	2.28	8.05	6325	631	1795	1.0	0.15	-	3.7	-	95	90	314	-	4.17	1018	1.68	520.7 ± 5.4	271.8 ± 13.0	792.5 ± 38.7
DA-DSW2	2.37	3.48	8.56	11410	1531	4279	2.3	0.18	0.13	5.7	-	172	168	363	-	7.92	2252	3.02	229.2 ± 1.3	213.3 ± 4.1	442.6 ± 8.8
DA-W1	1.96	1.67	8.41	5788	660	1865	0.9	0.31	-	3.4	-	90	77	275	-	3.33	890	1.52	1518.0 ± 7.2	1202.8 ± 21.2	2720.8 ± 49.6
DA-W2	2.11	3.70	8.30	7829	1038	2711	1.4	0.22	0.12	6.2	-	109	102	286	-	4.63	1303	1.89	233.5 ± 2.4	135.5 ± 6.5	369.1 ± 18.1
DB-100m	6.62	1.50	8.10	4719	391	1749	0.6	0.11	0.07	6.7	-	110	93	315	0.04	2.78	569	1.61	2724.1 ± 9.1	1565.3 ± 24.4	4289.4 ± 68.4
DB-D	10.55	1.18	7.40	5084	437	2068	0.7	0.15	0.12	4.6	-	117	94	340	0.05	2.82	604	1.80	1375.0 ± 9.0	681.7 ± 21.5	2056.7 ± 66.2
DB-DSW1	1.87	4.49	7.77	6431	544	2343	0.9	0.10	0.08	10.5	0.03	153	139	426	0.04	3.56	845	2.12	333.0 ± 2.2	131.6 ± 5.0	464.6 ± 18.0
DB-W1	1.02	3.18	7.62	6420	600	2600	1.0	0.14	0.07	9.0	-	153	137	421	0.03	3.66	852	2.16	724.7 ± 4.9	162.0 ± 8.3	886.7 ± 45.8
DC-100m	41.10	0.61	8.11	2291	176	465	0.2	0.11	-	3.9	-	26	39	86	0.14	0.96	264	0.42	442.0 ± 4.8	237.3 ± 11.1	679.3 ± 32.7
DC-D	40.44	0.49	7.90	2266	174	458	0.2	0.08	-	5.4	-	26	39	90	0.14	0.93	265	0.41	451.2 ± 3.9	197.5 ± 9.9	648.6 ± 32.9
DC-DSW1	25.25	1.85	8.36	2074	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	407.0 ± 2.4	154.7 ± 5.3	561.6 ± 19.4
DC-DSW2	2.42	3.21	7.81	1987	186	593	0.2	3.41	-	3.2	-	28	42	63	0.12	1.04	293	0.48	139.1 ± 0.9	106.3 ± 2.7	245.4 ± 6.4
DC-PLAYA	11.80	2.22	7.72	2090	182	606	0.2	0.01	-	4.9	0.21	27	41	65	0.11	0.99	289	0.45	174.7 ± 1.0	98.7 ± 2.7	273.4 ± 7.6
DC-USW1	29.76	1.70	8.40	2093	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	675.3 ± 4.3	321.0 ± 10.1	996.3 ± 32.1
DC-USW2	16.48	1.59	7.73	1995	172	578	0.2	0.15	-	2.5	-	29	41	61	0.13	1.03	291	0.47	567.1 ± 1.9	240.5 ± 4.3	807.6 ± 14.8
DC-W1	27.14	1.98	8.50	2083	179	472	0.2	0.12	-	3.0	-	30	40	57	0.11	1.28	272	0.51	550.3 ± 3.1	275.1 ± 7.8	825.4 ± 23.8
DC-W2	15.67	1.42	7.76	1908	177	598	0.2	0.22	-	4.7	-	29	42	64	0.12	1.00	280	0.45	780.1 ± 2.2	311.0 ± 5.3	1091.1 ± 18.8

[-] below detection limits

NC indicates water sample not collected for DC-USW1 and DC-DSW1

Table S3. Total carbon, inorganic carbon and organic carbon results (%) for sediment grab samples.

Site Name	TC (%)	TIC (%)	TOC (%)
CSW	5.45	1.96	3.49
Discharge A			
DA-D	1.09	0.05	1.04
DA-W1	12.88	12.17	0.71
DA-DW1	2.44	1.88	0.56
DA-W2	1.78	0.92	0.87
DA-DW2	1.47	0.63	0.84
Discharge B			
DB-D	11.39	8.87	2.52
DB-USW1	4.06	2.31	1.75
DB-W1	7.62	4.93	2.69
DB-DSW1	3.72	3.27	0.45
Discharge C			
DC-D	9.20	8.11	1.09
DC-100m	12.32	11.46	0.86
DC-UPW1	11.16	10.79	0.37
DC-W1	7.01	5.05	1.96
DC-DSW1	0.53	0.11	0.42
DC-USW2	3.97	0.33	3.64
DC-W2	4.22	0.37	3.85
DC-DSW2	1.11	0.92	0.19
DC-PLAYA	2.32	0.57	1.75

Table S4. Porewater probe measurements (DO, pH, conductivity, ORP and temperature) collected in an anaerobic chamber, anion and cation concentrations (all in units of mg/L), and sediment core Ra activities measured via gamma spectrometry (all in units of Bq/kg).

Sample	Depth from surface (cm)	DO (mg/L)	DO (%)	pH	Conductivity (µS/cm)	Eh (mV)	Temperature (°C)	Sr	Mn	K	Mg	Fe	B	Na	Ca	Li	Ba	Cl	SO_4	Estimated HCO_3	^{226}Ra ± error	^{228}Ra ± error	^{228}Ra/^{226}Ra
DC-W1 27-29 cm	29	0.24	3.0	8.12	1661.6	101	26.0	3.05	0.01	23	18	0.14	0.85	250	66	0.31	0.06	180	413	160.5	617.8 ± 4.1	129.6 ± 7.2	0.21
DC-W1 25-27 cm	27	0.14	1.8	8.16	1634.9	93	25.5	2.74	0.01	22	16	0.12	0.67	235	58	0.30	0.06	168	419	98.9	633.0 ± 3.9	123.8 ± 6.8	0.20
DC-W1 23-25 cm	25	0.05	0.6	7.90	1604.1	112	25.1	3.19	0.03	76	19	0.31	0.61	247	73	0.33	0.06	231	451	124.7	590.1 ± 4.1	95.2 ± 7.2	0.16
DC-W1 21-23 cm	23	0.15	1.9	7.95	1568.5	61	24.9	2.80	0.02	24	17	0.35	0.58	218	62	0.31	0.05	160	396	114.3	641.6 ± 3.7	130.9 ± 6.3	0.20
DC-W1 19-21 cm	21	0.26	3.3	7.49	1733.8	112	25.1	3.36	0.02	25	20	0.09	0.59	254	72	0.33	0.08	176	432	186.9	627.4 ± 3.3	125.8 ± 5.8	0.20
DC-W1 17-19 cm	19	0.19	2.4	7.82	1594.4	87	25.5	3.69	0.03	27	21	0.57	0.65	274	82	0.33	0.08	199	482	178.5	605.0 ± 1.7	137.1 ± 3.1	0.23
DC-W1 15-17 cm	17	0.24	3.0	7.95	1055.2	95	24.1	3.91	0.03	29	23	0.07	0.61	294	86	0.34	0.08	237	518	141.7	662.3 ± 4.5	150.5 ± 8.0	0.23
DC-W1 13-15 cm	15	0.62	7.6	8.07	1681.4	117	23.7	3.58	0.03	23	21	0.08	0.63	291	79	0.36	0.07	220	459	195.4	652.1 ± 1.3	149.7 ± 2.4	0.23
DC-W1 11-13 cm	13	0.06	0.8	7.23	2730.7	220	26.5	7.62	0.25	87	44	1.35	1.13	398	195	0.46	0.20	316	501	838.6	623.3 ± 2.5	168.3 ± 4.8	0.27
DC-W1 9-11 cm	11	0.05	0.6	6.34	2811.7	183	24.8	7.61	0.32	86	46	1.03	0.91	389	185	0.47	0.19	315	552	727.5	431.9 ± 2.6	166.9 ± 5.9	0.39
DC-W1 7-9 cm	9	0.15	1.9	7.80	2110.7	60	26.8	4.80	0.12	26	29	0.12	1.13	352	108	0.46	0.15	261	287	638.0	610.2 ± 3.2	222.6 ± 7.2	0.36
DC-W1 5-7 cm	7	0.22	2.8	7.89	1934.6	117	24.7	4.12	0.42	41	27	0.86	1.22	319	84	0.49	0.22	258	309	467.6	708.6 ± 4.2	321.3 ± 9.7	0.45
DC-W1 3-5 cm	5	0.60	7.7	7.64	1719.6	95	26.3	4.93	0.12	31	34	0.30	0.80	333	99	0.44	0.15	237	355	553.0	708.7 ± 3.2	274.1 ± 7.3	0.39
DC-W1 0-3 cm	3	0.32	4.1	7.48	2119.4	7	24.7	6.27	0.26	29	49	0.15	0.76	321	114	0.47	0.13	225	458	528.7	534.8 ± 4.4	261.4 ± 9.9	0.49
DC-W2 18-20 cm	20	0.16	2.0	7.92	2292.8	42	25.6	4.74	0.44	34	35	3.05	1.42	387	133	0.48	0.33	329	9	1091.2	323.5 ± 1.6	114.5 ± 3.3	0.35
DC-W2 16-18 cm	18	0.29	3.7	7.85	2255.3	77	25.5	3.76	0.44	34	33	1.67	1.30	382	84	0.46	0.22	321	6	935.5	267.7 ± 1.1	110.4 ± 2.5	0.41
DC-W2 14-16 cm	16	0.11	1.5	7.23	2181.1	15	25.0	3.91	0.39	50	34	1.69	1.20	398	87	0.48	0.21	311	5	1036.0	377.9 ± 1.8	120.8 ± 3.7	0.32
DC-W2 12-14 cm	14	0.20	2.6	7.74	2241.8	80	25.2	5.77	0.45	33	37	1.48	1.30	386	174	0.46	0.32	273	7	1321.7	368.1 ± 2.6	132.1 ± 5.5	0.36
DC-W2 10-12 cm	12	O.O.R.	O.O.R.	7.94	2155.9	-85	24.4	3.71	0.22	30	34	0.34	1.33	376	77	0.43	0.16	265	78	899.1	385.4 ± 2.1	115.5 ± 4.5	0.30
DC-W2 8-10 cm	10	O.O.R.	O.O.R.	7.87	2346.9	-18	24.0	6.11	0.53	35	49	12.48	1.31	387	184	0.51	0.29	262	216	1170.5	331.3 ± 2.8	134.5 ± 6.1	0.41
DC-W2 6-8 cm	8	0.33	3.9	7.58	2058.0	13	21.7	5.27	0.34	32	49	0.38	1.09	372	111	0.47	0.12	241	354	765.9	355.2 ± 2.2	127.5 ± 4.7	0.36
DC-W2 4-6 cm	6	0.55	6.5	7.18	2476.4	89	21.8	6.33	0.69	27	54	1.31	1.12	353	148	0.45	0.11	238	541	609.9	428.6 ± 1.8	138.5 ± 3.7	0.32
DC-W2 2-4 cm	4	O.O.R.	O.O.R.	7.09	2297.1	-60	21.8	6.57	0.99	39	51	7.27	0.89	322	151	0.41	0.12	214	499	638.5	432.8 ± 3.1	154.1 ± 6.7	0.36
DC-W2 0-2 cm	2	0.99	11.4	7.18	2918.4	126	20.7	11.61	0.97	52	90	2.57	0.92	355	216	0.47	0.19	201	1095	412.2	616.8 ± 4.3	204.2 ± 9.2	0.33
DA-W1 14-16 cm	16	0.57	7.2	8.20	5914.4	59	25.5	9.13	0.16	107	73	0.88	2.64	1044	269	1.31	0.06	743	2108	180.7	304.9 ± 2.5	162.6 ± 5.8	0.53
DA-W1 12-14 cm	14	0.31	4.0	8.08	6057.7	-3	25.8	9.25	0.05	110	74	0.12	2.85	1051	276	1.36	0.05	785	2133	121.4	116.0 ± 1.3	88.8 ± 3.8	0.77
DA-W1 10-12 cm	12	0.17	2.2	7.32	5803.9	-13	25.8	8.64	0.07	105	71	0.52	3.26	1022	265	1.35	0.03	729	2121	103.1	493.7 ± 2.0	159.4 ± 4.1	0.32
DA-W1 8-10 cm	10	0.28	3.5	8.18	5167.0	4	25.8	7.39	0.07	93	65	0.14	2.62	866	226	1.11	0.03	615	1882	14.4	1155.2 ± 6.0	242.6 ± 11.1	0.21
DA-W1 6-8 cm	8	0.45	5.7	7.83	3963.5	44	25.5	7.67	0.06	107	67	0.19	2.80	872	228	1.09	0.03	641	1907	0.0	1218.5 ± 6.2	331.5 ± 11.9	0.27
DA-W1 4-6 cm	6	2.35	28.1	8.08	5075.1	137	22.5	8.04	0.06	92	69	0.00	2.94	885	245	1.07	0.03	635	1949	30.4	1442.7 ± 3.2	613.2 ± 7.4	0.43
DA-W1 2-4 cm	4	0.54	6.9	8.00	5268.4	65	25.7	8.13	0.04	87	69	0.19	2.84	859	257	0.99	0.04	610	1954	23.8	1570.5 ± 6.4	890.1 ± 16.4	0.57
DA-W1 0-2 cm	2	0.08	1.0	7.86	6033.6	-31	25.0	9.12	0.05	162	75	0.26	2.90	888	306	1.10	0.03	716	2116	8.7	1392.1 ± 6.8	911.8 ± 19.1	0.65
CSW 28-30 cm	30	1.19	14.6	7.79	10641.8	84	24.0	5.77	1.67	23	414	0.60	1.74	2576	285	1.79	0.06	37	7691	0.0	55.7 ± 0.5	65.8 ± 1.9	1.18
CSW 26-28 cm	28	0.78	9.6	7.98	16253.5	72	24.6	7.64	3.20	71	637	1.27	2.86	4650	356	3.49	-	87	13811	0.0	57.4 ± 0.9	68.0 ± 3.2	1.18
CSW 24-26 cm	26	0.20	2.5	7.94	19174.8	60	24.8	8.42	3.33	15	715	1.43	2.99	4991	356	3.78	-	24	18211	0.0	54.0 ± 0.5	64.9 ± 1.8	1.20
CSW 22-24 cm	24	0.12	1.5	7.92	18872.5	42	24.6	9.15	3.39	16	753	3.32	3.00	5037	373	3.84	-	26	24025	0.0	57.5 ± 0.7	70.8 ± 2.2	1.23
CSW 20-22 cm	22	0.50	6.2	7.94	19271.4	76	23.8	9.01	2.96	24	772	1.74	2.89	4813	383	3.68	-	30	14890	0.0	58.5 ± 0.6	75.6 ± 2.1	1.29
CSW 18-20 cm	20	0.22	2.7	7.84	18933.6	50	24.5	9.17	2.58	22	763	3.79	2.83	4618	390	3.59	-	26	16445	0.0	59.1 ± 0.6	68.4 ± 2.1	1.16
CSW 16-18 cm	18	0.20	2.5	7.99	17960.9	51	25.3	9.05	2.05	23	734	2.22	2.65	4281	379	3.38	-	27	14751	0.0	66.8 ± 1.5	86.0 ± 5.5	1.29
CSW 14-16 cm	16	0.39	4.8	7.87	17524.5	65	24.3	9.32	1.69	23	759	2.11	2.21	4142	378	3.34	-	26	14455	0.0	54.1 ± 0.9	74.9 ± 3.1	1.39
CSW 12-14 cm	14	0.41	5.0	7.91	16462.7	62	24.3	8.76	1.24	26	683	1.22	2.41	3702	350	3.01	0.03	34	14939	0.0	51.5 ± 0.6	63.2 ± 2.0	1.23
CSW 10-12 cm	12	0.27	3.3	7.93	14551.7	53	25.3	8.26	0.96	23	659	1.34	2.01	3291	320	2.70	-	24	11643	0.0	54.3 ± 0.9	60.1 ± 3.2	1.11
CSW 8-10 cm	10	0.30	3.7	7.92	12994.3	45	23.1	7.66	0.89	22	608	1.15	1.98	2820	299	2.38	-	22	9560	0.0	49.8 ± 0.6	62.9 ± 2.2	1.26
CSW 6-8 cm	8	0.08	1.0	7.89	10589.9	-32	23.2	7.56	0.92	22	588	1.38	1.63	2506	298	2.13	0.03	20	9580	0.0	48.1 ± 0.6	59.8 ± 2.1	1.24
CSW 4-6 cm	6	0.12	1.5	7.96	9618.4	-46	23.4	6.66	0.83	20	500	0.54	1.26	1983	274	1.68	0.03	17	7086	0.0	44.1 ± 0.5	51.0 ± 2.0	1.16
CSW 2-4 cm	4	O.O.R.	O.O.R.	7.87	7926.2	-49	23.7	5.95	0.94	23	413	0.23	1.07	1560	253	1.33	0.05	17	7161	0.0	56.8 ± 0.7	63.8 ± 2.4	1.12
CSW 0-2 cm	2	O.O.R.	O.O.R.	7.59	5437.4	-52	23.8	4.09	1.29	27	271	0.30	0.60	932	197	0.76	0.08	17	3144	456.5	61.0 ± 0.7	65.1 ± 2.5	1.07
DC-PLAYA 16-18 cm	18	0.17	2.1	7.66	4564.4	48	25.5	6.69	2.73	32	100	7.66	2.69	557	504	0.78	0.03	320	2277	128.4	61.8 ± 0.7	76.9 ± 2.39	1.24
DC-PLAYA 14-16 cm	16	0.12	1.5	7.45	4142.7	86	24.4	6.93	3.12	45	103	9.63	2.41	549	511	0.84	-	319	2127	358.0	55.5 ± 0.9	76.5 ± 3.11	1.38
DC-PLAYA 12-14 cm	14	0.18	2.2	7.37	4082.2	91	24.1	7.61	3.12	34	103	13.74	2.22	552	457	0.75	0.09	313	1979	388.9	61.9 ± 0.7	73.3 ± 2.28	1.18
DC-PLAYA 10-12 cm	12	0.05	0.6	7.55	3886.9	68	22.0	7.51	2.71	37	99	9.07	2.18	552	366	0.76	0.16	305	1458	766.4	86.4 ± 1.0	81.8 ± 3.22	0.95
DC-PLAYA 8-10 cm	10	0.01	0.1	7.58	3695.2	51	22.6	7.18	2.39	42	93	8.01	2.25	533	294	0.69	0.18	307	1138	878.5	107.3 ± 1.2	86.5 ± 3.77	0.81
DC-PLAYA 6-8 cm	8	0.00	0.0	7.58	3376.6	76	22.5	6.25	2.15	39	80	6.14	2.20	519	234	0.67	0.21	300	957	829.2	111.2 ± 1.3	82.0 ± 3.77	0.74
DC-PLAYA 4-6 cm	6	0.05	0.6	7.91	3228.8	32	24.7	5.19	1.41	40	65	2.05	1.99	486	179	0.61	0.17	296	839	660.4	141.3 ± 1.3	84.8 ± 3.66	0.60
DC-PLAYA 2-4 cm	4	0.05	0.6	7.73	2965.6	47	22.9	5.18	1.41	35	61	5.04	1.86	460	155	0.61	0.17	272	786	593.4	130.2 ± 2.2	94.9 ± 6.52	0.73
DC-PLAYA 0-2 cm	2	0.07	0.8	7.74	2713.7	14	24.2	5.18	1.46	40	57	5.85	1.88	425	136	0.57	0.16	238	793	482.3	118.8 ± 1.6	98.9 ± 4.39	0.83

[O.O.R.] indicates "out of range"
[-] indicates below detection limits

Table S5. Leachate concentrations (μg/g) from sediment core leaching experiment normalized for both the volume of leachate recovered and mass of sample residue leached. Leachate solution blanks were also measured and reported as mg/L. Detection limits were also included at the bottom of the table for each element. [-] denote below detection limits. [NM] denotes not measured.

Sample Name	Leachate Solution	Al	Ba	Ca	Fe	K	Mg	Mn	Na	P	S	Si	Sr	Ti	pH
DC-W1 3-5 cm	H_2O	-	4.7	736	-	213	154	0.8	394	0.4	1663	29	34.7	-	8.17
	NH_4OAc	-	50.5	5954	-	165	380	16.0	61	1.7	217	23	184.4	-	8.27
	$Na_4P_2O_7.10H_2O$	19.4	11.9	851	445.0	50	273	12.3	115005	57562.5	457	118	246.7	2.8	10.44
	HOAc	1.7	27.1	94653	-	106	2919	39.2	31944	466.5	4199	381	2177.1	-	4.54
	HCl	0.5	4.7	24468	-	33	563	2.6	2362	428.1	843	144	516.9	-	4.32
DC-W1 13-15 cm	H_2O	-	3.6	728	-	113	95	0.7	220	0.3	1037	22	26.8	-	7.98
	NH_4OAc	-	25.0	5342	-	74	186	11.5	35	2.2	232	16	131.8	-	8.30
	$Na_4P_2O_7.10H_2O$	7.2	11.0	690	153.4	32	229	4.1	112384	52266.3	590	54	257.7	1.5	10.63
	HOAc	1.4	21.8	79648	-	68	2336	16.1	31487	457.4	4081	176	1706.1	-	4.46
	HCl	0.6	4.1	23588	-	21	505	1.2	1232	818.6	822	78	489.8	-	4.34
DC-W1 23-25 cm	H_2O	-	3.4	772	-	101	90	0.8	182	0.2	1160	23	26.1	-	8.03
	NH_4OAc	-	21.5	5144	-	61	153	6.1	30	2.3	271	14	109.1	-	8.33
	$Na_4P_2O_7.10H_2O$	9.1	13.0	470	73.8	46	229	2.7	113473	51333.3	1044	48	320.3	1.5	10.83
	HOAc	3.5	111.4	97657	6.1	84	3578	34.8	35334	339.6	5083	135	3093.1	-	4.57
	HCl	2.2	5.4	27993	1.6	18	340	7.6	975	10933.9	835	123	263.1	-	3.78
DC-W2 2-4 cm	H_2O	-	2.5	2029	-	266	368	7.7	386	0.5	2283	54	55.1	-	7.40
	NH_4OAc	-	61.2	2899	-	256	352	18.0	51	2.5	173	35	88.7	-	8.06
	$Na_4P_2O_7.10H_2O$	106.0	15.1	606	1303.7	101	153	29.6	126543	69059.3	211	456	9.5	8.7	9.70
	HOAc	74.6	7.2	2134	96.4	70	1109	11.5	5357	455.3	70	403	5.9	-	3.32
	HCl	473.9	28.2	1583	593.4	116	697	9.7	227	535.0	68	634	6.0	-	1.94
DC-W2 10-12 cm	H_2O	-	3.6	635	-	232	163	1.2	470	0.7	1403	38	28.2	-	7.91
	NH_4OAc	-	68.1	5422	-	293	443	25.2	81	1.7	560	37	145.9	-	8.23
	$Na_4P_2O_7.10H_2O$	77.8	9.6	1317	1913.6	108	115	36.5	133613	69289.7	348	276	13.9	9.6	9.76
	HOAc	61.8	6.7	3975	86.9	87	1598	29.5	4319	373.3	146	326	9.8	-	3.35
	HCl	742.5	37.7	1848	1018.3	185	829	14.7	186	614.3	110	1188	7.2	0.5	1.87
DC-W2 16-18 cm	H_2O	0.3	2.0	354	0.4	204	88	1.0	412	1.0	460	38	14.8	-	8.16
	NH_4OAc	-	63.5	5297	-	306	392	35.4	81	2.4	300	40	132.0	-	8.30
	$Na_4P_2O_7.10H_2O$	65.9	7.1	1255	1040.9	104	119	40.8	131938	70384.5	232	281	10.4	6.3	9.72
	HOAc	50.7	6.4	3917	138.7	88	1591	28.7	3812	202.5	143	367	8.5	-	3.32
	HCl	693.7	32.7	1629	1075.3	169	665	13.1	182	545.0	92	1125	6.3	0.4	1.86
DC-PLAYA 2-4 cm	H_2O	-	2.4	714	-	262	146	2.3	417	1.6	1248	55	20.0	-	7.85
	NH_4OAc	0.6	66.1	6245	0.9	382	393	43.9	80	2.8	206	45	106.5	-	8.25
	$Na_4P_2O_7.10H_2O$	70.8	9.6	1706	1593.0	132	78	89.1	140592	72639.1	125	251	17.4	4.5	9.83
	HOAc	67.1	4.4	2269	115.5	113	733	26.2	4777	322.2	63	558	8.0	-	3.32
	HCl	795.0	21.8	1374	514.3	206	578	15.9	192	503.7	45	1264	6.4	-	1.94
DC-PLAYA 8-10 cm	H_2O	-	2.3	639	-	237	111	0.2	404	1.3	914	52	14.6	-	7.92
	NH_4OAc	-	70.3	6056	0.5	416	382	50.7	78	2.4	170	43	85.3	-	8.25
	$Na_4P_2O_7.10H_2O$	187.3	9.2	1433	1456.7	206	118	84.3	148441	67889.4	97	601	9.1	5.3	9.77
	HOAc	86.3	3.8	1693	73.4	100	559	23.5	3547	396.5	52	489	5.2	-	3.22
	HCl	799.4	18.3	1313	514.4	213	640	17.0	179	461.2	42	1214	5.2	-	1.86
DC-PLAYA 14-16 cm	H_2O	-	1.2	2132	-	232	143	2.5	377	0.6	2073	33	20.3	-	7.71
	NH_4OAc	-	57.2	5522	-	386	230	39.0	60	1.7	150	27	45.7	-	8.21
	$Na_4P_2O_7.10H_2O$	85.7	10.2	921	420.5	133	65	66.3	130137	72946.9	59	191	6.6	2.9	9.83
	HOAc	247.8	2.0	1943	88.9	136	526	30.5	7126	1655.8	53	1073	4.6	-	3.43
	HCl	1235.7	21.6	1769	439.8	328	788	30.6	408	952.5	51	2206	5.9	0.4	1.82
DA-W1 2-4 cm	H_2O	1.3	0.2	399	-	86	48	-	585	1.2	491	24	10.0	-	8.68
	NH_4OAc	-	1.9	5407	1.4	23	53	2.6	84	12.6	223	9	69.8	-	8.34
	$Na_4P_2O_7.10H_2O$	8.3	3.6	210	9.5	53	34	-	97493	35500.8	1307	58	215.8	-	10.96
	HOAc	2.8	36.1	115199	2.6	303	1630	29.2	47687	160.5	10606	44	2883.5	-	4.85
	HCl	0.8	5.1	16157	0.4	23	155	0.9	1165	179.0	882	54	264.9	-	4.50

Table S5. Leachate concentrations (μg/g) from sediment core leaching experiment normalized for both the volume of leachate recovered and mass of sample residue leached. Leachate solution blanks were also measured and reported as mg/L. Detection limits were also included at the bottom of the table for each element. [-] denote below detection limits. [NM] denotes not measured.

Sample Name	Leachate Solution	Al	Ba	Ca	Fe	K	Mg	Mn	Na	P	S	Si	Sr	Ti	pH
DA-W1 8-10 cm	H_2O	0.3	0.9	461	-	121	48	-	488	0.2	608	15	12.0	-	8.43
	NH_4OAc	-	6.4	5568	1.2	40	78	4.1	51	3.7	246	9	79.6	-	8.38
	$Na_4P_2O_7.10H_2O$	13.7	3.2	309	30.6	73	73	0.6	99889	39122.5	1243	45	210.8	0.8	11.01
	HOAc	3.1	23.6	105237	3.3	236	2206	12.4	39672	203.3	9279	75	2248.5	-	4.82
	HCl	0.8	4.0	16166	0.7	24	228	0.8	1176	266.2	903	44	250.3	-	4.61
DA-W1 12-14 cm	H_2O	0.4	1.3	289	-	452	50	0.2	725	0.7	790	39	9.1	-	8.26
	NH_4OAc	-	53.8	4900	-	619	342	12.0	130	1.4	195	36	105.9	-	8.30
	$Na_4P_2O_7.10H_2O$	89.0	9.6	737	202.3	187	81	13.3	133710	64653.9	235	298	40.7	9.6	9.96
	HOAc	528.9	1.2	5056	203.8	167	319	27.5	8987	6434.9	221	1024	32.2	3.5	3.32
	HCl	760.8	15.9	2113	620.4	265	628	22.5	358	813.4	106	1555	16.7	1.0	1.88
CSW 2-4 cm	H_2O	-	1.8	600	-	120	513	2.5	1051	1.2	3649	61	13.1	-	8.22
	NH_4OAc	-	48.9	6311	-	296	1617	68.4	206	2.4	306	61	90.5	-	8.34
	$Na_4P_2O_7.10H_2O$	286.0	7.0	1365	1292.4	213	664	102.5	128374	64764.0	323	1126	59.0	14.6	10.21
	HOAc	440.7	1.0	9028	210.9	145	1396	123.4	15885	11794.4	256	1431	30.9	1.9	3.51
	HCl	1140.1	19.7	5499	555.7	303	1831	78.3	654	2187.0	172	2315	22.8	0.8	1.93
CSW 14-16 cm	H_2O	0.4	0.9	188	0.3	62	132	0.6	1705	2.0	1285	44	4.7	-	8.89
	NH_4OAc	-	59.1	3452	-	287	1170	64.1	336	1.5	117	34	59.3	-	8.21
	$Na_4P_2O_7.10H_2O$	283.7	10.0	1012	788.1	196	270	123.7	131792	67389.3	74	920	7.8	7.3	9.75
	HOAc	115.8	3.2	1407	56.2	92	735	30.0	4183	512.5	41	832	3.9	-	3.15
	HCl	1154.5	9.6	1435	440.8	210	786	23.3	236	898.6	45	1747	5.1	0.5	1.89
CSW 26-28 cm	H_2O	-	0.6	449	-	44	177	1.0	2155	1.2	2090	42	6.3	-	8.71
	NH_4OAc	-	54.0	3697	-	247	818	60.7	348	1.2	105	36	39.8	-	8.17
	$Na_4P_2O_7.10H_2O$	238.8	6.1	813	360.2	141	178	107.1	135771	68626.9	46	672	3.8	4.8	9.88
	HOAc	199.7	2.0	2392	39.7	88	1122	41.3	6366	970.7	63	957	3.5	-	3.20
	HCl	1273.7	8.7	1741	412.4	208	931	32.1	296	1150.3	50	1893	4.5	0.5	1.89
Solution Blanks	H_2O	<0.005	<0.005	<0.01	0.01	<0.01	<0.01	<0.005	<0.01	<0.01	<0.02	0.03	<0.005	<0.005	7.00
	NH_4OAc	<0.005	<0.005	0.01	<0.005	0.02	0.01	0.01	0.05	0.04	<0.02	0.02	<0.005	<0.005	8.00
	$Na_4P_2O_7.10H_2O$	<0.005	<0.005	0.07	0.11	0.15	0.01	<0.005	NM	NM	0.05	0.14	<0.005	<0.005	10.75
	HOAc	<0.005	<0.005	<0.01	<0.005	0.01	<0.01	<0.005	0.02	0.08	<0.02	0.03	<0.005	<0.005	2.67
	HCl	<0.005	<0.005	0.01	0.01	<0.01	<0.01	0.01	0.02	0.02	<0.02	0.01	<0.005	<0.005	1.74
Detection Limits		<0.005	<0.005	<0.01	0.01	<0.01	<0.01	<0.005	<0.01	<0.01	<0.02	0.03	<0.005	<0.005	

Table S6. XRD results for select sediment samples including quantification of minerals for DC-W1 3-5 cm, DC-W2 10-12 cm, and DC-W2 16-18 cm.

Sample	Mineral 1	Mineral 2	Mineral 3	Mineral 4	Mineral 5	Mineral 6	Mineral 7	Mineral 8	Mineral 9	Mineral 10	Mineral 11	Quantified
DC-W1 3-5 cm	calcite (75%)	quartz (11%)	pellyite (6%)	montmorillonite (5%)	halloysite (2%)	dickite (<1%)	celestite (<1%)	anorthite (<1%)	-	-	-	yes
DC-W1 13-15 cm	quartz	calcite	montmorillonite	-	-	-	-	-	-	-	-	no
DC-W1 13-15 cm residue	quartz	calcite	orthoclase	anorthite	montmorillonite	magnetite	-	-	-	-	-	no
DC-W1 23-25 cm	quartz	calcite	montmorillonite	barite (trace)	-	-	-	-	-	-	-	no
DC-W2 2-4 cm	quartz	montmorillonite	halloysite	celestite (trace)	gypsum (trace)	-	-	-	-	-	-	no
DC-W2 10-12 cm	quartz (48%)	albite (15%)	illite (13%)	microcline (9%)	dolomite (5%)	calcite (4%)	kaolinite (3%)	chlorite-serpentine (1%)	montmorillonite (<1%)	barite (<0.5%)	mangano-mangani ungaretite (<0.5%)	yes
DC-W2 10-12 cm residue	quartz	montmorillonite	illite	albite	barite (trace)	-	-	-	-	-	-	no
DC-W2 16-18 cm	quartz (50%)	halloysite (11%)	johannsenite (11%)	albite (7%)	orthoclase (5%)	chlorite-serpentine (5%)	anorthite (4%)	dolomite (4%)	montmorillonite (2%)	barite (<0.5%)	-	yes

Table S7: Vegetation samples (dry weight) analyzed for ^{226}Ra uptake and their corresponding calculated concentration ratio (C_r). Exchangeable soil ^{226}Ra was determined as the percent ^{226}Ra lost during sediment leaching step 2. Exchangeable-normalized ^{226}Ra C_r was calculated by dividing the plant ^{226}Ra activity by the exchangeable ^{226}Ra fraction of the sediment.

Sample	Plant ^{226}Ra (Bq/kg)	± error (Bq/kg)	^{226}Ra C_r*	Exchangeable sediment ^{226}Ra (%)	Exchangeable ^{226}Ra C_r
DA-W1 cattail leaves	79.44	4.64	0.05	11	0.46
DA-W1 cattail roots	53.08	4.97	0.03	11	0.31
DB-D grass leaves	523.08	20.14	0.38	9**	4.29
DB-D grass roots	880.53	30.20	0.64	9**	7.22
DB-100m sedge seeds	473.72	22.64	0.17	-	-
DB-100m sedge stems	293.56	11.98	0.11	-	-
DB-100m sedge roots	75.03	4.37	0.03	-	-
DB-W1 cattail leaves	148.91	7.21	0.21	36**	2.32
DB-W1 cattail roots	32.33	2.68	0.04	36**	0.12
DC-D grass roots + leaves	76.40	5.15	0.17	11**	1.51
DC-100m grass leaves	82.93	3.62	0.19	-	-
DC-100m grass roots	167.11	9.06	0.38	-	-
DC-W1 cattail leaves	40.75	6.34	0.06	22	0.26
DC-W1 cattail roots	57.84	14.26	0.08	22	0.37
DC-W2 cattail seeds	24.60	5.59	0.06	52	0.11
DC-W2 cattail leaves	112.20	6.59	0.26	52	0.50
DC-W2 cattail roots	129.55	3.49	0.30	52	0.58
DC-PLAYA sedge stems	50.59	2.68	0.39	56	0.69
DC-PLAYA sedge roots	66.90	2.43	0.51	56	0.92
CSW cattail seeds	BDL	BDL	-	78	-
CSW cattail leaves	7.62	1.42	0.13	78	0.17
CSW cattail roots	6.96	0.51	0.12	78	0.16

*calculated using 5 cm sediment core section ^{226}Ra activity, DB samples calculated using grab sediment sample

**leaching data taken from McDevitt et al. (2019)

[-] indicates data not collected/calculated

BDL indicates below detection limits