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Supplementary material

Effect of biogeochemical redox processes on the fate and transport of As and U at an abandoned uranium mine site: an X-ray absorption spectroscopy study

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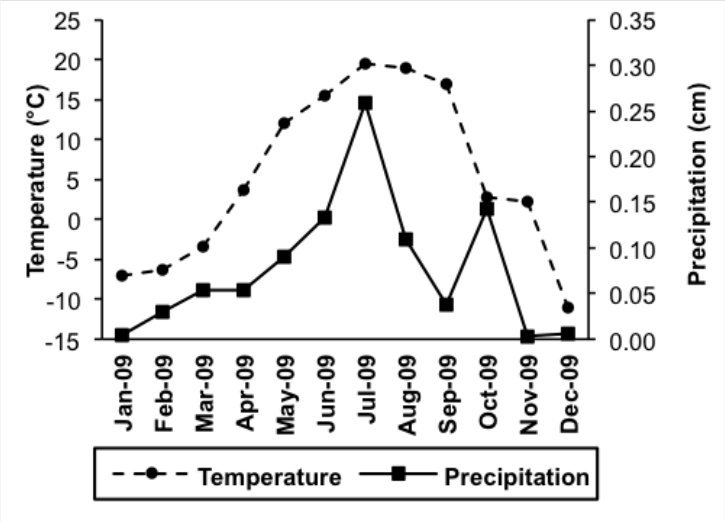

Fig. S1. Average monthly climate data for the year of 2009 collected at Antelope Range from the South Dakota Climate and Weather's Automatic Weather Data Network. The left axis corresponds to temperature (dashed line) and the right axis corresponds to precipitation (solid line).

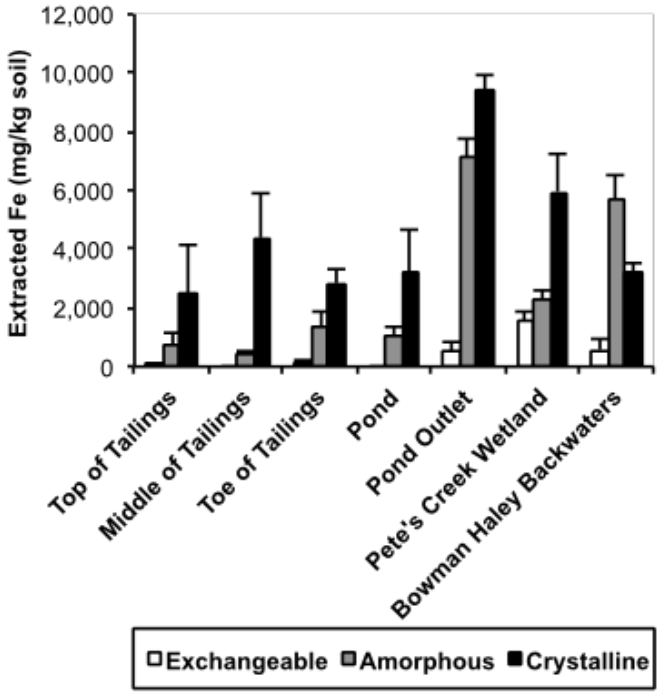

Fig. S2. Sequential extractions of sediment-associated iron. Bars indicate percentage exchangeable (white), amorphous (grey) and crystalline (black) iron of the total extracted iron. Error bars indicate one standard deviation from the mean of triplicate extractions.

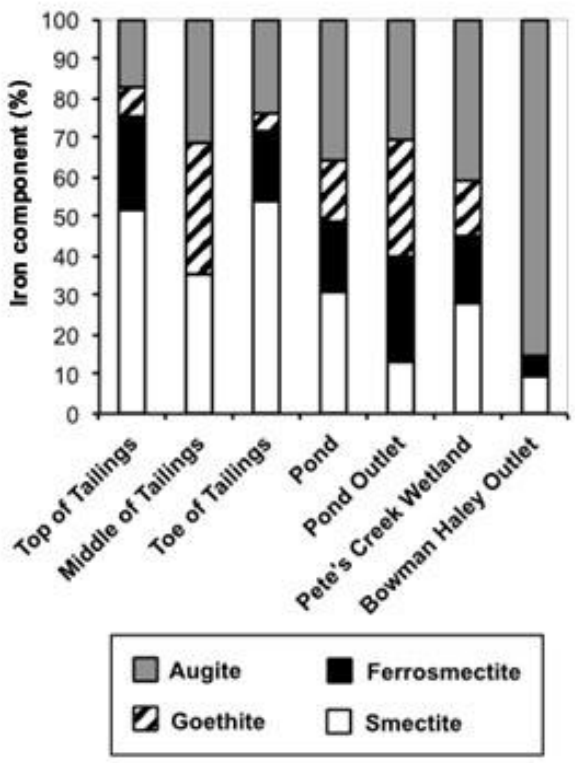

Fig. S3. Distribution of Fe minerals in sediments as determined by linear-combination fitting of k^3 Fe extended X-ray absorption fine structure (EXAFS). Stacked bars represent reference compounds including augite (grey), goethite (dashed), ferrosmectite (black), and smectite (white). Components are plotted as percentage of the total, with ~5 % error.

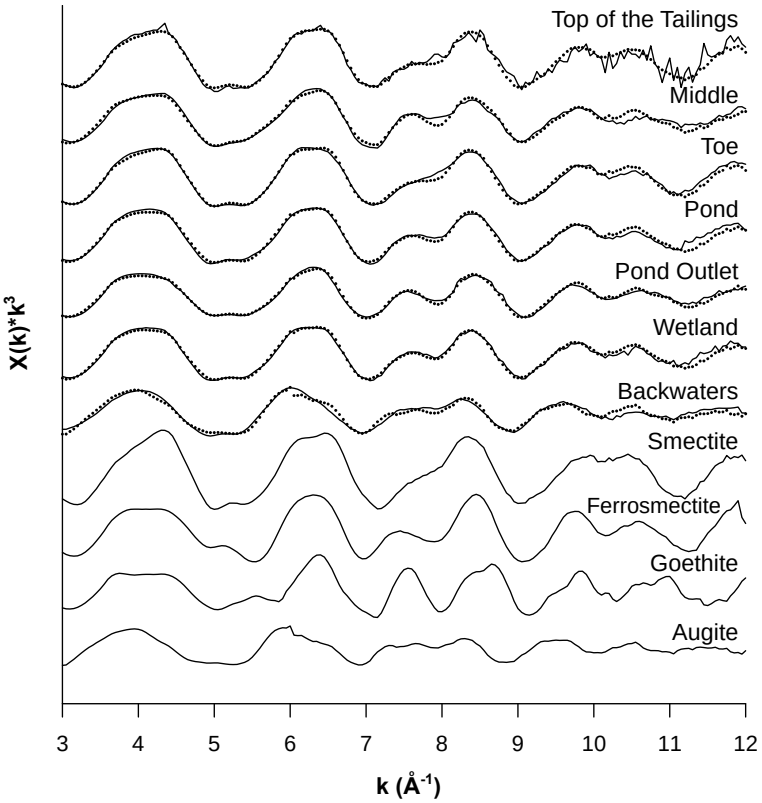

Fig. S4. Linear-combination fits (dots) of Fe extended X-ray absorption fine structure (EXAFS) spectra (solid lines) from all of the sampling locations. The reference compounds used in fitting are plotted at the bottom for comparison.

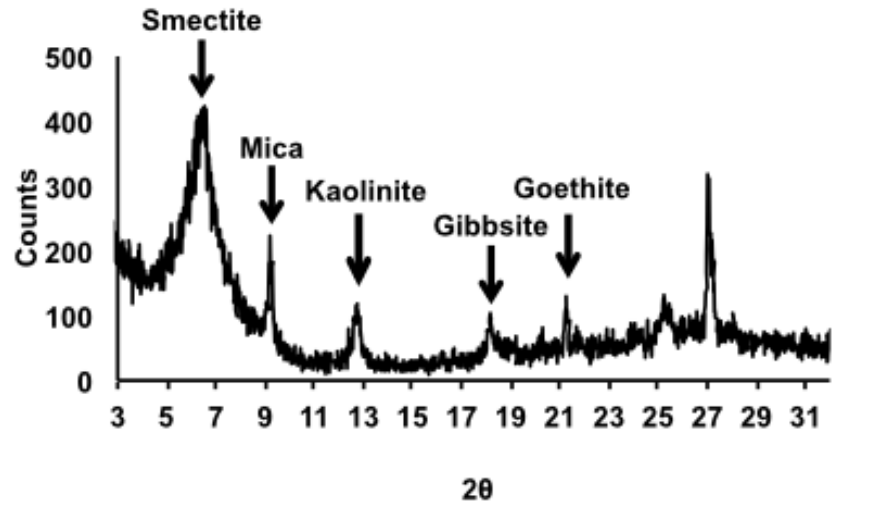

Fig. S5. X-Ray diffraction pattern of clay sediment fraction from the sedimentation pond sampling site. Diffraction peaks are labelled with their corresponding clays and minerals.

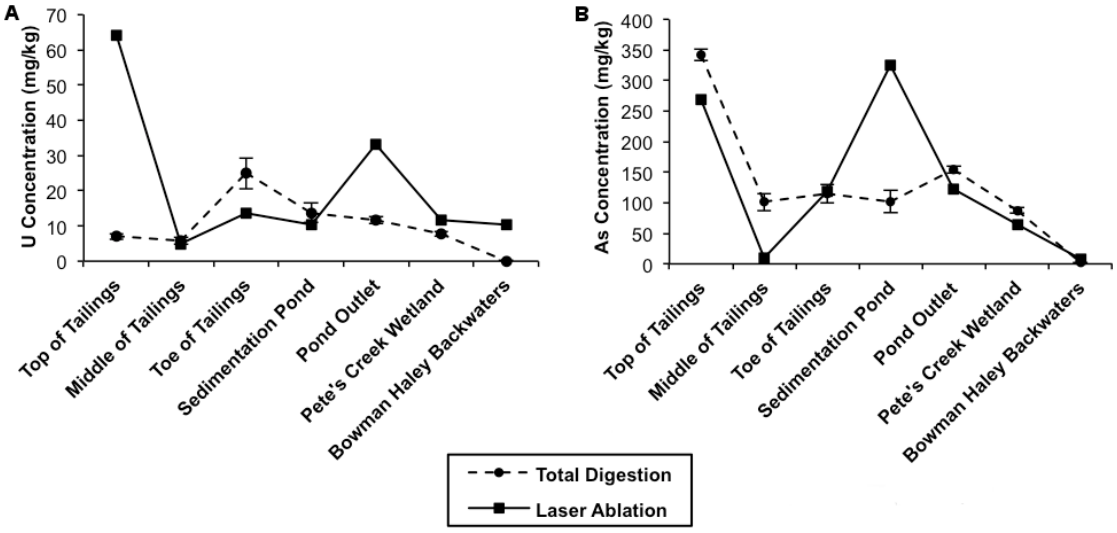

Fig. S6. Total concentrations of solid phase uranium (a) and arsenic (b) in the top 0–5 cm of sediment cores as determined by total digestions analysed by ICP-MS (dotted lines) and by laser ablation ICP-MS (solid lines). Error bars on the total digest values indicate one standard deviation from the average of triplicate digests.

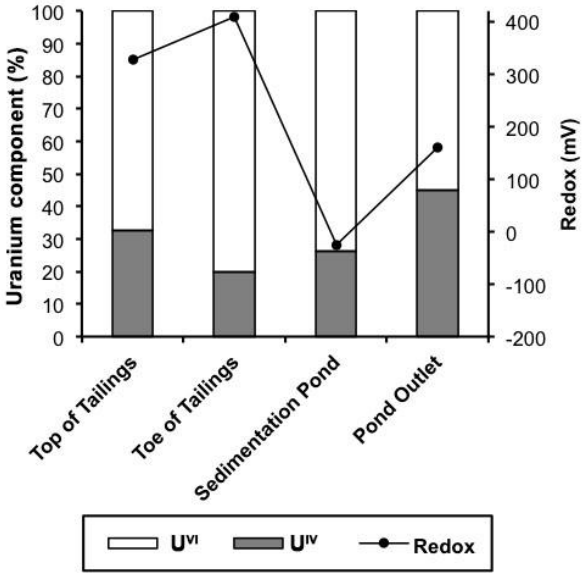

Fig. S7. Uranium speciation as determined by linear-combination fitting of uranium X-ray absorption near-edge fine structure (XANES) spectra of sediments collected 0–5 cm from the surface. Spectra were fit with the following two reference compounds – uranyl acetate (white) and uraninite (grey). Components are plotted as percentage of the total, with ~5 % error. The line indicates field ORP measurements taken at a depth of 0–5 cm from the surface during field sampling.

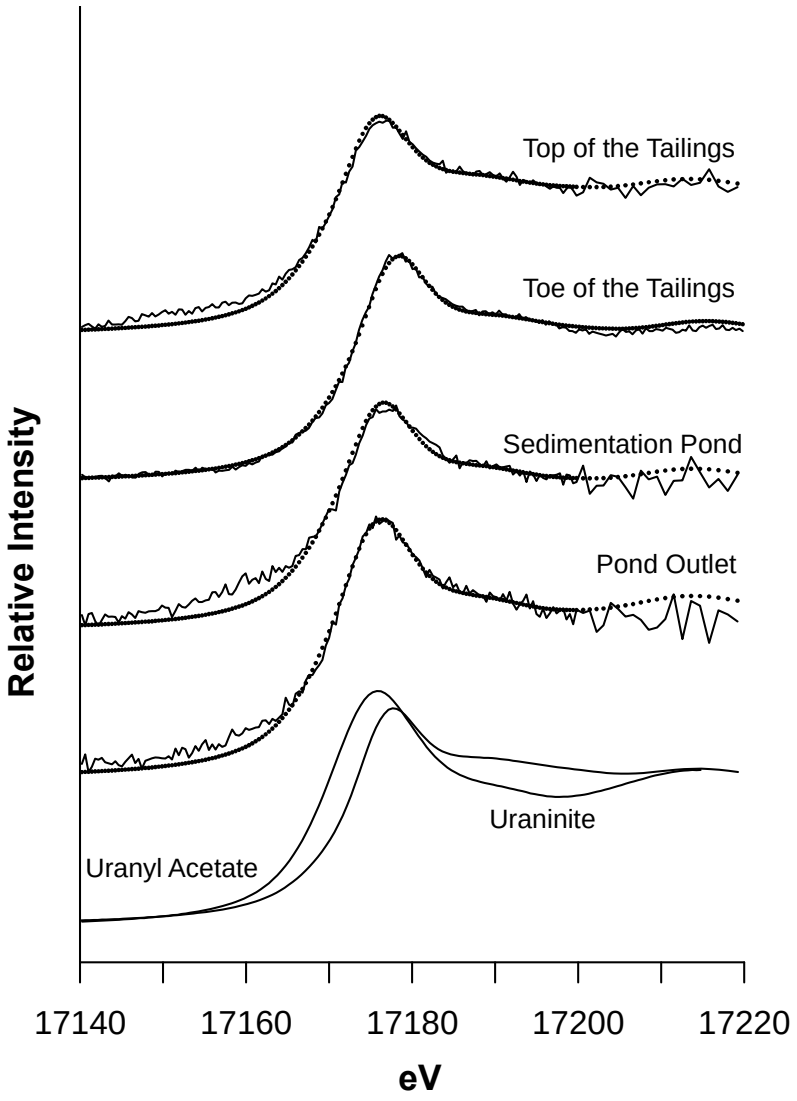

Fig. S8. Linear-combination fits (dots) of U X-ray absorption near-edge fine structure (XANES) spectra (solid lines) from the top of the tailings pile, toe of the tailings, sedimentation pond, and the pond outlet. The reference compounds used in fitting are plotted at the bottom for comparison.

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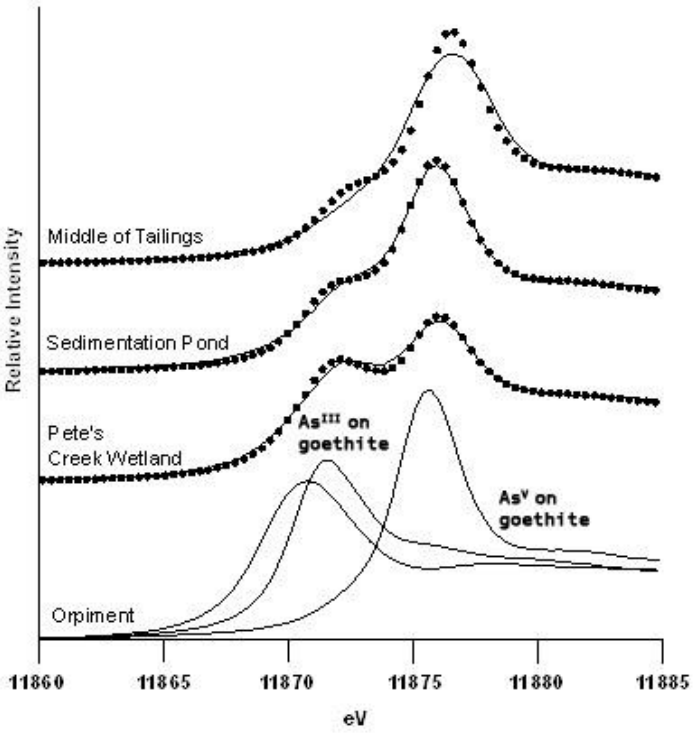

Fig. S9. Linear-combination fits (dots) of arsenic X-ray absorption near-edge fine structure (XANES) spectra (solid lines) from the middle of the tailings pile, sedimentation pond, and Pete's Creek wetland. The reference compounds used in fitting are plotted at the bottom for comparison.

Table S1. Summary of Fe extended X-ray absorption fine structure (EXAFS) linear-combination fitting

Location name	Percentage smectite	Percentage ferrosmectite	Percentage goethite	Percentage augite	R factor
Top of the tailings	51.6	23.9	7.1	17.4	0.068693
Middle of the tailings	35.3	0	33.1	31.5	0.041328
Toe of the tailings	54.3	17.4	4.6	23.7	0.020924
Sedimentation pond	31.0	17.9	15.5	35.6	0.029843
Pond outlet	13.0	27.0	29.5	30.5	0.018432
Pete's Creek wetland	27.9	17.0	14.4	40.7	0.023450
Bowman-Haley backwaters	9.7	4.8	0	85.5	0.065118

Table S2. Summary of U X-ray absorption near-edge fine structure (XANES) linear-combination fitting

Location name	Percentage U^{IV}	Percentage U^{VI}	R factor
Top of the tailings	32.5	67.5	0.003771
Middle of the tailings	–	–	–
Toe of the tailings	19.7	80.3	0.001874
Sedimentation pond	26.4	73.6	0.001293
Pond outlet	45.1	54.9	0.000481
Pete's Creek wetland	–	–	–
Bowman-Haley backwaters	–	–	–

Table S3. Summary of As X-ray absorption near-edge fine structure (XANES) linear-combination fitting

Orpiment was used as a reference compound for the As XANES LC-fitting in order to represent an oxidation state rather than an exact chemical structure

Location name	Percentage Orpiment	Percentage As^{III}	Percentage As^{V}	R factor
Top of the tailings	30.1	0	69.9	0.014789
Middle of the tailings	0	11.3	88.7	0.003969
Toe of the tailings	0	13.8	86.2	0.002586
Sedimentation pond	0	33.0	67.0	0.001037
Pond outlet	0	43.8	56.3	0.001209
Pete's Creek wetland	17.6	40.8	41.6	0.000903
Bowman-Haley backwaters	25.0	38.3	36.8	0.006110