Uranium delivery and uptake in a montane wetland, north-central Colorado, USA

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Abstract

Comprehensive sampling of peat, underlying lakebed sediments, and coexisting waters of a naturally uraniferous montane wetland are combined with hydrologic measurements to define the important controls on uranium (U) supply and uptake. The major source of U to the wetland is groundwater flowing through locally fractured and faulted granite gneiss of Proterozoic age. Dissolved U concentrations in four springs and one seep ranged from 20 to 83 ppb ($\mu\text{g/l}$). Maximum U concentrations are ~ 300 ppm (mg/kg) in lakebed sediments and >3000 ppm in peat. Uranium in lakebed sediments is primarily stratabound in the more organic-rich layers, but samples of similar organic content display variable U concentrations. Post-depositional modifications include variable additions of U delivered by groundwater. Uranium distribution in peat is heterogeneous and primarily controlled by proximity to groundwater-fed springs and seeps that act as local point sources of U, and by proximity to groundwater directed along the peat/lakebeds contact. Uranium is initially sorbed on various organic components of peat as oxidized U(VI) present in groundwater. Selective extractions indicate that the majority of sorbed U remains as the oxidized species despite reducing conditions that should favor formation of U(IV). Possible explanations are kinetic hindrances related to strong complex formation between uranyl and humic substances, inhibition of anaerobic bacterial activity by low supply of dissolved iron and sulfate, and by cold temperatures.

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1. Introduction

Wetlands function as efficient filters that trap suspended particles, incorporate excess dissolved nutrients, and fix dissolved metals (Cu, Pb, Zn, Cr, Hg, U) and metalloids (As, Se) of environmental concern (Shotyk, 1988; Reddy and DeLaune, 2008). Organic-rich, microbially active wetland soils and sediments provide ideal substrates for metal uptake by a variety of mechanisms including ion exchange, metal-organic complexing, adsorption, and precipitation of metal sulfides and oxides (Walton-Day, 2003). Important controlling variables include pH, Eh, water/sediment chemistry, and the type, diversity, and activity of resident microorganisms. Interplay of the various uptake mechanisms is complex and site

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specific. For example, wetlands that receive sulfate-rich waters can support active bacterially mediated sulfate reduction that is indicated by H₂S generation and precipitation of metal sulfides and reduced oxides (Machemer and Wildeman, 1992; Groudev et al., 1999). In contrast, wetlands that receive sulfate-poor waters may fix metals by one or more mechanisms in which sulfate reduction plays a lesser role. The observed distribution of metals within a wetland is the net result of spatial and temporal variations in these uptake mechanisms and variations in metal supply from source rocks and sediments.

Uranium-bearing wetlands are a relatively well-documented subclass of metal-enriched wetlands. Reported occurrences are from a variety of geologic, hydrologic, and climatic settings, mostly in North America and Europe (Armands, 1967; Halbach et al., 1980; Toens, 1984; Zielinski et al., 1987, 1988; Culbert and Leighton, 1988; Shotyk, 1988; Shotyk et al., 1992; Read et al., 1993; Owen and Otton, 1995; Tixier and Beckie, 2001; Zayre et al., 2006). Many of the reported wetlands are removed from obvious sources of pollution, but some are located near U mines and deposits of U-bearing mining/milling wastes (Akber et al., 1992; Groudev et al., 1999; Schöner et al., 2009). This suggests that in some settings, U-bearing wetlands might be used as indicators of concealed ore deposits, or as pollution controls.

Studies of U-bearing peat and organic-rich lakebeds are of scientific interest because they describe the first stage of U-organic matter associations that, after burial and diagenesis, may form U-bearing coal or shale, respectively. These studies also contribute to broader topics such as the role of metals in soil-forming processes and the environmental controls on contaminant migration. Knowledge gained from natural wetlands can inform the design of engineered wetlands (Hammer, 1989) and improve the design of commercial facilities for the biologically based removal of dissolved U (Lovley, 1995; Kalin et al., 2005). Results also aid land managers who must understand the important controls on U fixation and mobility in order to predict the environmental impacts of wetland disturbances (Owen and Otton, 1995).

Identification of U-rich wetlands is hampered by their generally low radioactivity, because emplacement of uranium is often too recent to permit significant ingrowth of the more radioactive U decay products (Culbert and Leighton, 1988). This means that physical sampling and chemical analyses, rather than ground-based or aerial radioactivity surveys, are needed to locate and characterize these types of deposits. However, uranium remains a preferred element for study because it is amenable to a variety of specialized measurements such as radiography and U decay-series dating that provide unique spatial and temporal information.

This study documents the conditions and processes controlling the efficient uptake of U in a relatively remote, natural wetland that is absent of reported U occurrences, mining impacts, or other obvious sources of pollution. Unlike previous studies of U-rich wetlands, this study is distinguished because it provides an exceptionally detailed three-dimensional view of the distribution of uranium in as much as 3.7 m of Holocene peat, underlain by organic-rich lacustrine silt and clay (gyttja) and organic-poor clay and silt of a precursor post-glacial lake, with a combined thickness of as much as 8.5 m. The hydrologic characteristics of the entire sedimentary package from surface to bedrock were investigated with an extensive array of installed piezometers and water-table monitoring wells. Mechanisms of U uptake were investigated by a variety of techniques that utilize analyses of waters and of core and auger samples collected from depths as great as 11 m. Results of this study provide new insights regarding the spatial association of U with organic matter in peat and, unlike previous studies of metal delivery to wetlands, identify several pathways for the delivery of U to specific areas and horizons in the wetland. The techniques used in this broad-scope investigation can be readily applied to other U-rich or metal-rich wetlands.

2. Physical Setting

The study area is located approximately 13 km north of Rocky Mountain National Park in north-central Colorado, USA, within the upper reaches of the Laramie River valley (Fig. 1). We informally refer to this wetland as the Boston Peak (BP) wetland after the U.S. Geological Survey 7.5' topographic quadrangle in which it is located, and because this name was informally given to the wetland when the occurrence of elevated uranium and trace metal concentrations were first documented there by Sarnecki (1983). The study area lies within the Roosevelt National Forest, in an area that is largely used for recreational activities such as camping, fishing, and hunting. The wetland occupies an area of approximately 3.9 hectares at an altitude of approximately 2700 m above sea level. It is separated from the Laramie River to the west by a low, elongate, north-trending bedrock ridge (Fig. 1). To the east, the wetland is flanked by a steep bedrock slope thinly mantled with glacial deposits, colluvium, and talus. The north (downstream) margin of the wetland is bordered by a large landslide (Fig. 1).

Annual precipitation in the area is typically 50-64 cm, with most falling as snow during the months October-May (Khun et al., 1983). Snowmelt occurs primarily in May-June and is the primary source of water to the area. During the summer months, rain is delivered in the form of occasional afternoon thundershowers (Dougherty et al., 1987). Surface runoff enters the wetland via a small intermittent stream that is sourced by a seep zone at the contact between bedrock and the overlying

landslide deposits, which enters the wetland at its northeastern edge. During spring snowmelt and following heavy rains, surface runoff also enters the wetland via overland flow that runs down the valley side slopes to the east and west of the wetland, through an intermittent surface drainage from the south, and from melting snow on the wetland surface during late spring. Surface water exits the northwestern corner of the wetland via a gap between the bedrock ridge to the west and the landslide to the north (Figs. 1,2).

The primary source of water to the wetland is groundwater from springs and seeps that are sourced in fracture systems in the surrounding crystalline bedrock. The springs and seeps enter the wetland at the edges and from beneath, and form spring pools on the wetland surface (Fig. 2) where the upwelling water is forced through the peat overlying the margins of the lake sediments.

The elevation of the wetland places it in the upper montane life zone, which supports mature forests of spruce/fir, lodgepole pine, and aspen (Willard and Foster, 1990). The dominant vegetation types in the wetland include sedges (*Carex*), spike rush (*Eleocharis rostellata*), mosses (primarily *Sphagnum*), and grasses. Shrubs of willow (*Salix planifolia*) and birch (*Betula glandulosa*) are located primarily at the northern end and margins of the wetland. These hydrologic and vegetative characteristics classify the BP wetland as a minerotrophic fen-carr complex (Mitsch and Gosselink, 1986; Cooper, 1988). The southern half of the wetland (fen) is dominated by dwarf-shrub, sedge, and grass plants. The northern half (carr) primarily hosts grasses, tall shrub, and tree species. Peat comprising the southern (fen) part of the wetland contains very little or no mineral matter. In contrast, peat underlying the northern part is mixed with alluvial sediments derived from the drainage to the northeast, and from slope wash (till, landslide deposits, and weathered crystalline bedrock) from the adjacent valley sides, resulting in deposits that range from a mucky peat to organic-rich mineral soil in composition.

3. Geologic Setting

Bedrock in the study area consists of two-mica granite gneiss, mafic gneiss, and amphibolite, all of Proterozoic age (Fig. 3). Locally these units are overlain by poorly lithified volcanoclastic sediment and tuff of Tertiary age, and by Pleistocene glacial till. A kilometer-wide, north-trending zone of variably sheared and shattered bedrock underlies the study area. This zone represents the trace of the Green Ridge fault, a high-angle reverse fault exhibiting evidence of probable Laramide movement (McCallum et al., 1983). This fault provides some of the structural control of the previously glaciated, north-trending Laramie River valley. Chemical analyses of rock samples and a gamma spectrometric survey of the area indicate that some of the sheared and altered rocks are anomalously uraniferous. Seventeen specimens

of crystalline bedrock collected within a ~0.5 km radius surrounding the wetland contain from 0.4 to 119 ppm U. Higher concentrations of uranium in these samples were associated primarily with iron-oxide stained, granitic and amphibolitic rocks in mineralized shear and fracture zones (F.A. Hills, USGS, written communication, 1992). The source of U is speculative, but highly fractured granite gneiss shows evidence of redistributed U in microfractures and in altered biotite, based on fission track radiography. Three of nine samples of granite gneiss have unusually elevated Th/U ratios (>6.0) compared to average values reported for various granitic rocks of the United States (3.5-6.0; Wedepohl, 1978), suggesting some preferential leaching of U.

The area was glaciated most recently in Pinedale (late Wisconsin) time, and glacial material mantles some of the upland slopes surrounding the wetland. As the valley glacier receded between 18 and 12 ka (Kiver, 1972; Madole, 1980), some glacial till and poorly consolidated volcanoclastic sediments exposed on valley walls slumped downslope onto the valley floor (Griswold, 1980). One such event produced the landslide deposit at the north end of the wetland (Figs. 1, 3). This landslide acted as a dam and a small lake formed behind it. Sediments underlying the Boston Peak wetland record the progressive filling of this lake. The present wetland was established as the lake became shallow enough to permit rooting of aquatic vegetation and accumulation of peat.

4. Methods

Fieldwork and analyses for this study were conducted in 1987-1991. Shortly thereafter, the principal scientists were reassigned to other projects due to an internal reorganization. In 2016, we had the opportunity to prepare this summary and reinterpret the data, incorporating new GPS measurements, 3D modeling, and updated PHREEQC calculations.

Locations of auger and core holes, piezometers, and water sampling locations were initially determined using a measuring tape and compass. Elevations were determined to the nearest 0.1 ft (3 cm) using a tripod-mounted surveying level and stadia rod, relative to an arbitrary datum established at the base of a road sign adjacent to the wetland. In 2014, the two primary authors revisited the field area and more precisely determined the location of the arbitrary surveying benchmark with high-resolution differential GPS, at 105.856309°W, 40.631637°N, elevation 2710.3 m. Locations and elevations of selected features and data collection points in the wetland were also verified with GPS.

Sampling and analyses of sediment and water were conducted during the summers of 1987-1989. Sediments were collected from 48 auger holes and 9 core holes (Fig. 2). Auger sampling utilized a 0.61 m-long stainless steel bit fabricated from welding together two ship's auger bits and adding a

coupling to accept threaded 1 m sections of lightweight aluminum drill rod and a “T”-shaped handle. Sampling to all depths in the peat and lakebed sediments was facilitated by connecting together as many 1 m sections of drill rod as needed. Auger samples were collected in 30 cm increments and placed in cloth or polypropylene sample bags. Recovery of coarse material at the base of the lakebeds was limited because either the substrate was too coarse to allow passage of the auger bit, or because the sand- and greater-sized material did not adhere to the bit. After air drying, the samples were disaggregated and lightly crushed by passing through a mill with ceramic plates set at 2 mm opening.

Coring was performed with a stainless-steel “Livingstone”-type piston corer measuring approximately 1 m long and 5 cm in diameter (Wright et al., 1984). A threaded fitting on top of the corer accommodates the same drill rods as the auger bit. Cores were extruded, described, and sampled in documented intervals. Most core samples were wrapped in plastic wrap and aluminum foil, and placed in core boxes for transport. Core intervals collected for detailed physical and chemical analyses were packaged in field-moist condition in wide-mouth, screw-capped, HDPE bottles, and refrigerated. Splits of core and auger samples submitted for full chemical analysis were dried at 80 °C and ground to pass a sieve of 100 µm (150 mesh) opening.

Organic matter content of samples was estimated by loss-on-ignition (LOI) at 500 °C (Dean, 1974). Uranium concentrations in 680 air-dried samples were determined by delayed neutron activation (Millard and Keaten, 1982). Selected core samples were dissolved in a mixture of acids and analyzed for major and trace elements by inductively coupled plasma-atomic emission spectrometry (ICP-AES) (Lichte et al., 1987). Estimated one-sigma precision for U and most reported elements is better than ±10%. Chemical data for 43 samples of peat and lake sediments (Table A-2 of supplemental information) were evaluated by R-mode factor analysis (Harbaugh and Merriam, 1966) to identify simplifying and correlative relationships between U and other constituents (Table A-3 of supplemental information).

Physical properties (moisture, bulk density, fiber content, ash content, cation exchange capacity, pH, and humification index) of 17 peat samples collected at a variety of depths, and the mineralogy of selected peat and lakebed sediments determined by X-ray diffraction, optical microscope, and scanning electron microscope are available in Tables A-4 and A-5 of supplementary information. The carbohydrate fraction of 18 samples of variably humified peat was determined by ¹³C nuclear magnetic resonance (NMR) spectroscopy using the cross polarization, magic-angle spinning (CPMAS) technique (Hatcher et al., 1983; Orem and Hatcher, 1987) (Figure A-6 of supplementary Information). Eight slices of U-rich peat from various depths near a spring pool were dehydrated, impregnated with low-viscosity

resin (Bailey and Blackston, 1984) and prepared as thin sections for measurements of U distribution by fission-track radiography (Zielinski and Budahn, 1998).

Radiometric ages of discrete samples from cores were determined by Beta Analytic, Inc., and the University of Pittsburgh Radiocarbon Laboratory. The samples were prepared by first dispersing them in hot distilled water and handpicking root material and other possible contaminants from the sample. The samples were treated with a warm dilute acid solution to remove possible carbonate contaminants, then rinsed with distilled water. They were then dried and synthesized to benzene. Radiocarbon ages were adjusted to -25 per mil ^{13}C and calibrated to calendar ages using CALIB 7.1 and the IntCal13 data set (Stuiver and Reimer, 1993; Reimer et al., 2013).

To evaluate the oxidation state of uranium in peat, selective extractions of 15 samples of U-rich peat were performed shortly after collection using a dilute carbonate-bearing solution (0.1 M NaHCO_3 , pH=8.3) which, in the absence of added strong oxidants, is highly specific for dissolving oxidized U(VI) (Zielinski and Meier, 1988, Kohler et al., 2004). One gram of air-dried peat was combined with 100 ml of leaching solution in a sealed flask under nitrogen atmosphere and stirred for 24 hrs. To evaluate the possibility of U oxidation during sample processing and short-term storage prior to laboratory leaching, an extraction of freshly collected peat was performed under nitrogen atmosphere immediately after collection in the field. For all leaching experiments (lab or field-based) the leached peat was recovered by filtration, dried, and analyzed by the delayed neutron method along with a split of the original peat for comparison.

Hydrologic characteristics of the wetland were investigated using a network of 20 piezometer stations (Fig. 2), 14 of which included piezometers installed at multiple depths to facilitate measurements at various depths in peat and in the basal sand beneath the lacustrine sediments. The remaining 6 stations only monitored water table levels of the unconfined aquifer, but all 20 stations included a water table monitor. Piezometers were constructed from PVC pipe with an outside diameter of 4.2 cm. This diameter was slightly larger than the effective diameter of the auger pilot hole, thus a tight fit was obtained when the pipe was manually inserted. A dense rubber stopper was used to close off the bottom end of the pipe during insertion. The installed pipe plus stopper was filled with water to equalize hydrostatic pressure, then a smaller-diameter pipe was used to push the rubber stopper from the end of the piezometer pipe and downward an additional 7.6 cm, creating an open 7.6 cm-long by 4.2 cm-diameter cylindrical sampled interval below the casing. Water-table monitoring wells consisted of PVC pipe with a perforated section (holes drilled in the pipe) extending 0.6-1.0 m below the surface and a non-perforated section extending above ground.

Water levels in the piezometers were measured with a plastic float attached to monofilament line that was lowered until the float made contact with the water surface, then the line was withdrawn and its length measured with a tape measure. Replicate measurements indicated a precision better than ± 0.2 cm. Hydraulic conductivities in peats and basal sands were estimated by withdrawing water from piezometers, monitoring the rate of recovery to equilibrium levels, and applying standard equations (Luthin and Kirkham, 1949; Boelter, 1965).

Water was collected from the bottom of 13 piezometers for chemical analyses (Fig. 2). At least two volumes of water were pumped and constancy of measured temperature and specific conductance were confirmed prior to sampling. Surface waters were collected at four locations; three along the reach of the small stream that enters the wetland at its northeastern arm and one at the outlet of the wetland at its northwestern arm (Fig. 2). Four samples of spring water were collected from open pools on the wetland surface fed by upwelling groundwater. Two seeps were sampled on low slopes near the margin of the wetland. Pore waters from 13 intervals of core C1 were expressed in the laboratory using high-speed centrifugation.

For surface, spring-seep, and piezometer waters, all measurements of temperature, specific conductance, pH, and dissolved oxygen were performed at the time of collection. Water samples (300 ml) were placed in completely filled screw-capped LDPE bottles and refrigerated. Within several hours of collection, water samples were passed through filter membranes of $0.45\ \mu\text{m}$ opening under pressure from nitrogen gas. Filtered samples were split into unacidified and acidified ($\text{pH} < 2$ with ultrapure HNO_3) fractions for analysis. Water samples were promptly submitted for chemical analysis upon return from the field.

Alkalinity (as HCO_3^-) was determined by titration (Brown et al., 1970), dissolved organic carbon (DOC) by oxidation-infrared spectrometry (Wershaw et al., 1983), silica and cations by ICP-AES (Lichte et al., 1987), and anions by ion chromatography (Fishman and Pyen, 1979). Dissolved uranium concentration was measured by inductively coupled plasma-mass spectrometry (ICP-MS) (Date and Gray, 1985). Estimated precision for most reported elements is better than $\pm 10\%$ and for U $\pm 5\%$.

The equilibrium distribution of inorganic aqueous species, including uranium species, and the degree of saturation of the waters with respect to some of the more common uranium minerals was calculated using the computer code PHREEQC (Parkhurst and Appelo, 1999) and the wateq4f.dat database (Ball and Nordstrom, 1991).

EarthVisionTM (<http://www.dgi.com>) modeling software was used to create three-dimensional mathematically defined surfaces of the stratigraphic units from irregularly located x, y, and z data points,

where x and y is a Universal Transverse Mercator (UTM) location in meters, and z is an elevation in meters. In the model, x, y, and z values define the upper surfaces of lithologic units. Volumes of units are defined and shown as the space between two surfaces. EarthVision™ software follows basic geologic rules to define depositional, channel fill, or unconformable contacts and surfaces. These rules can be modified by adding data points to a surface, altering gridding parameters or detail, or using smoothing algorithms in any or all of the x, y, or z dimensions. Modeled surfaces were derived using “minimum surface-tension” gridding algorithms and modified by inserting user-defined data points. Uranium concentration data from auger and core samples were also gridded to create three-dimensional maps of U distribution in the wetland deposits.

5. Results and Discussion

5.1 Stratigraphy of the Boston Peak wetland

Three distinct lithologic units comprise the BP wetland: organic-poor lacustrine clay, gyttja (organic-rich lacustrine clay), and peat. The lacustrine-paludal units are underlain by a presumably thin (<1 m) layer of sandy and gravelly sediment, probably of glacial, glaciofluvial, and/or detrital origin, associated with the withdrawal of glacial ice from the topographic trough that formed the lake, and later, the wetland (Fig. 4). This unit is poorly characterized because often it could not be fully penetrated or retrieved by either the auger bit or the core barrel. Auger and core holes within the trough occupying the southern part of the depression usually encountered gravel or rock, suggesting glacial till, whereas auger and core holes in the northern part of the wetland usually encountered sand to very coarse sand, probably representing fluvial and/or colluvial deposits. In all three of the main wetland stratigraphic units, lithologic variability is more pronounced near wetland margins and in the northern portion due to contributions of mineral detritus from surrounding slopes and alluvial deposition (Fig. 5).

Lacustrine Clay (L) consists of variably greenish-gray to gray, organic-poor, laminated to thinly bedded clay and clayey silt as much as 5.1 m thick. This unit contains diatoms and sponge spicules, and is interpreted as lacustrine (lakebed deposits). This unit is largely confined to the deepest part of the bedrock trough in the central part of the wetland (Fig. 4). Coarser-grained alluvial, colluvial, and landslide sediments intertongue with this unit to the north (Fig. 5).

Gyttja (G) consists of variably olive-gray clay and variably brown clayey silt. This unit ranges in thickness from about 3.9 m in the center of the wetland to absent near the margins of the depression. The light- to dark-olive-gray color is attributed to remains of green algae *Pediastrum* and *Botryococcus* (E.I. Robbins, USGS, written communication, 1992). Browner-colored facies commonly contain abundant

terrestrial plant debris, including wood fragments, twigs, and pine needles. The lowest 0.3-0.6 m of the gyttja contains leaf and stem fragments of *Nymphaea* (water lily) and *Potamogeton* (pondweed), aquatic plants that require shallow water for rooting and germination (less than about 2 m for *Nymphaea*; Sinden-Hempstead and Killingbeck, 1996). Transitional sediments lying directly beneath the leaf- and stem-bearing gyttja layer yielded dates of 16-15 ka (Table 1, Fig. 6). This suggests that the lake was relatively shallow at about 15 ka, but the restricted thickness of the interval containing plant fragments indicates that shallow water conditions suitable for growth of these plants did not persist for very long. Deepening of the lake after about 15 ka may have been caused by the landslide that dammed the northern end of the wetland (Fig. 1).

Peat (*P*) is variably brown and yellow-brown, fibrous sedge (*Carex-Eleocharis*) peat with occasional wood and charcoal fragments. In some intervals, abundant *Sphagnum* and other bryophyte fibers are also present. Charcoal fragments or layers are fairly common. Thickness of the peat deposit ranges to as much as 3.7 m. The peat is thickest at the southern end of the wetland and thins gradually northward. To the north, the peat intertongues with coarse sandy sediment (Fig. 5). LOI generally ranges from 40 to 90% but peat sampled from the central part of the wetland, which contains little or no mineral detritus, is generally 80-90% LOI. Because the samples for U and LOI were integrated over 30 cm (1 ft) increments, some samples that we classified as containing dominantly peat also contained relatively small quantities of interspersed mineral matter by volume, but sufficient by weight to yield LOI values that appear anomalously low. Most of the peat in the Boston Peak wetland is fibric, but near the edges of the wetland, or where peat is in contact with flowing groundwater, such as near seeps or spring pools, some of the peat is hemic or sapric in quality.

A sample of peat collected near the base of the unit yielded an age of 3380 cal BP (Table 1, Fig. 6). This date, together with a date of 4930 cal BP obtained from the uppermost part of the gyttja unit (Fig. 6), suggest that approximately 4000-5000 years ago, the lake became so shallow that wetland vegetation began to take root and accumulate as peat. The shallowing may have occurred gradually or rapidly, probably through a combination of raising the lakebed elevation through steady accumulation of sediment, and lowering of the lake's outlet through erosion of the landslide dam in the northwest corner of the wetland.

5.2 Groundwater Hydrology

The wetland deposits include two aquifers, an unconfined aquifer composed of as much as 3.7 m of peat, and a deeper confined aquifer, probably less than 1 m thick in most places, consisting of till

and glaciofluvial coarse sand and gravel with variable lesser amounts of silt and clay, at the base of the lake and wetland deposits (Fig. 7). The confining layer above this basal aquifer is the lacustrine clay and gyttja, with a combined thickness of less than 1 m toward the margins of the lake to more than 8 m near the center. Both surface water and groundwater are sources for the unconfined peat aquifer, whereas groundwater is the dominant source of water to the basal sand aquifer. Water in the unconfined peat aquifer is influenced by variations in surface runoff, precipitation, and evapotranspiration, whereas water in the deeper confined aquifer is sustained primarily by fracture-controlled flow of groundwater through the surrounding crystalline bedrock.

Measurements of hydraulic conductivity (K) in 8 piezometers accessing the basal sand/gravel unit ranged from 2.0×10^{-6} to 3.0×10^{-3} cm sec⁻¹, with an average of 7.5×10^{-4} cm sec⁻¹, whereas measurements of K in 6 piezometers monitoring water flowing through peat of varying degrees of humification yielded a range of 2.5×10^{-5} to 1.5×10^{-3} cm sec⁻¹, with an average value of 6.3×10^{-4} cm sec⁻¹ (see Table A-7 of supplemental information for individual measurements). The average hydraulic conductivities of the peat and basal sand units are very similar and both units have hydraulic conductivities spanning at least two orders of magnitude. Differences in hydraulic conductivity of the basal sand are probably due in large part to differences in grain size, primarily in the silt and clay content of the glaciofluvial sediments in different parts of the aquifer. Differences in hydraulic conductivity in the peat are controlled by several factors, including degree of compaction, types of vegetation that make up the peat, and degree of humification.

Fault or shear zones appear to be preferential conduits for groundwater delivery to the wetland, in a manner similar to that previously described by Read et al. (1993). A number of north to northwest-trending linear features follow regional structural trends in the area and are associated with faults, fractures, or shears in the bedrock. A strand of the Green Ridge fault passes directly beneath the Boston Peak wetland (Fig. 3). A contour map showing the elevation of the piezometric surface of the confined basal aquifer relative to the elevation of the unconfined water table (Fig. 8) indicates that the basal aquifer is under artesian pressure. The greatest difference in hydraulic head between the two aquifers (more than 0.7 m) occurs near the center of the wetland (Fig. 8), and probably indicates the location of the fracture zone through which groundwater is entering the basal aquifer from the underlying bedrock (shown diagrammatically in Fig. 7).

Because it is under artesian pressure, water entering the basal aquifer from fractures in the underlying bedrock is forced laterally and upward through the basal sand and gravel where it is confined by overlying lacustrine clay and gyttja. As the clay thins toward the edges of the lacustrine deposits, the

aquifer becomes unconfined and groundwater is forced upward through the peat, forming the open spring pools (Fig. 7). The occurrence of the spring pools only near the margins of the wetland (Fig. 2) corresponds with the location of the outer edge of the clay and gyttja deposits in the subsurface (Fig. 4), lending further evidence to this hypothesis for the origin of the spring pools.

5.3 Association of uranium with organics and with other elements

A plot of U concentration versus LOI for subsets of the auger samples (Fig. 9) illustrates the ability of each stratigraphic unit to concentrate uranium. Samples classified as sand (black circles in Fig. 9) generally contain less than 5% organic matter and have uranium concentrations in the 4-30 ppm (mg kg⁻¹) range. Three of the four sand samples with higher uranium concentrations, 50-364 ppm, were collected from near the margins of the wetland where the sand contains as much as 23% intermixed organic matter.

Samples of organic-poor lacustrine clay (blue triangles) generally contain less than ~15% organic matter and have low U concentrations (Table 2; Fig. 9). In the central part of the trough, the organic matter content of the lacustrine clay (estimated by LOI) ranges from 0% near the base of the section to 2% near the top, but the margins of the unit contain as much as 30% organic matter where detrital fine-grained organic matter has washed in. In both the sand and clay units, the relative lack of organic matter accounts for their relatively low U concentrations.

Gyttja samples (green triangles) typically contain less than 40% organic matter. The organic constituents of the gyttja, consisting of plant fragments in the lower 30-60 cm of gyttja, and algae throughout, were exposed to lake water consisting of surface water mixed with uraniferous groundwater. Thus, U concentrations in the gyttja (Table 2) were determined by both the organic matter content of the gyttja relative to inorganic clay, and by the spatial and temporal extent of exposure of individual gyttja layers to U-bearing source waters.

Peat samples contain variable amounts of mineral matter content as determined by LOI (Table 2), but most samples that we classified as containing dominantly peat are composed of more than 75% organic matter. Uranium concentrations in peat are also highly variable (Table 2, Fig. 9), but on average, greater than those in both clay and gyttja. Uranium distribution within peat is influenced by basinward flow of introduced spring water, variations in peat humification and permeability, and ambient geochemical and hydrologic conditions/gradients (Halbach et al., 1980; Hill and Siegel, 1991). The most uraniferous peat is generally confined to within several meters of spring pools, indicating that peat is a highly efficient scavenger of introduced U.

R-mode factor analysis of uranium and other elements in 43 samples of peat, gyttja, and clay from three cores (C1, C2, C3) and one auger hole (A17) indicated that U correlated most strongly with arsenic (As) and molybdenum (Mo) (Table A-3 of Supplemental Information). Co-variation of these three elements was most evident in the 22 samples of peat. Statistically significant correlations of these three elements were observed despite the much lower concentrations of As (4-48 ppm) and Mo (3-51 ppm) in the peats (Table A-2 of Supplemental Information). Both Mo and As occur in dissolved form as oxyanions and can accumulate in peat (Krachler and Shotyk, 2004; Langner et al., 2012).

5.4 Uranium in water

Concentrations of dissolved U in surface water and spring/seeps ranged from <1 to 83 ppb ($\mu\text{g/L}$) and were highest (>20 ppb) in the four sampled spring pools (W3-W6, Fig. 2) and in one seep (W8, Table 3). The concentration of uranium in most spring-seeps that enter and surround the BP wetland is anomalous when compared to a regional average of 3.3 $\mu\text{g/l}$ for surface and groundwater draining crystalline rocks of the Colorado Front Range (Bolivar et al., 1978). The spring pools are fed by freshly discharging oxygenated groundwater, based on their relatively high DO, low Fe concentrations, and low DOC concentrations compared to piezometer waters (Table 4). Chemical gradients within the spring pools are indicated by floating films of iron oxides produced by iron-oxidizing bacteria (*Leptothrix discophora*) and submerged filamentous sulfur-oxidizing bacteria (*Thiothrix* sp. and *Lamprocystis* sp.) located ~10 cm below the pool surface (Robbins et al., 1990). Some spring pools have bottom turbulence from inflowing water indicated by gently roiling suspensions of humified peat fibers. Spring pool waters also contain high concentrations of dissolved ^{222}Rn , in the 10^2 - 10^3 Bq L^{-1} range (D.E. Owen, USGS, written communication, 1991). High concentrations of dissolved U in the spring pools imply sufficiently oxidizing conditions to maintain U as the relatively soluble U(VI) species.

Uranium speciation calculations with PHREEQC (Table A-9 of Supplemental information) indicated that Ca-uranyl-carbonate complexes are the dominant dissolved U species in 12 of 13 well waters (assumed Eh=0.1 volts) and in 5 of 6 spring/seeps (assumed Eh=0.4 volts). Uranium speciation in 3 of 4 surface waters (assumed Eh=0.4 volts) indicated major contributions from uranyl phosphate complexes that are favored by greater abundances of dissolved phosphate compared to the other waters (Tables 3, 4). All waters were also calculated by PHREEQC to be undersaturated with respect to uranium(VI) and uranium(IV) minerals at the assumed values of Eh. Speciation calculations did not consider uranium-organic complexing which could be important in piezometer waters containing relatively high concentrations of dissolved organic carbon (Table 4; Campbell et al., 2016).

Away from spring pools, the supply of uranium to peat is diminished because waters derived from other sources (stream, snowmelt, rain, runoff) contain lower concentrations of dissolved U than spring water (groundwater). Dilution of spring water with water from these other sources is non-uniform within the wetland, episodic when related to storms, and variable with the seasons. Persistence of the springs throughout the summer indicates that there is considerable recharge and storage capacity in the system of bedrock fractures that feeds the basal sand/gravel aquifer (also see Fig. A-8 of supplemental information).

5.5 Spatial distribution of uranium

The distribution of uranium within each of the three major stratigraphic units is heterogeneous. In lacustrine clay and gyttja, the highest U concentrations are generally stratabound in the more organic-rich layers, but exhibit variations within strata of very similar organic matter content. Peats of very similar composition and organic matter content exhibit a wide range (>1000X) of U concentrations (Table 2). Cross-section “slices” through a three-dimensional model of the wetland (Fig. 10) reveal areas with elevated U concentrations and facilitate correlation of U with stratigraphy and proximity to topographic and hydrologic features. Uranium in peat is concentrated in three main areas: (1) along the central axis of the southern part of the wetland (Fig. 10, slices 1-4); (2) related to the groundwater seep zone along the western margin of the wetland (slices 5-12); and (3) in the vicinity of spring pools on the east and west sides of the wetland (slices 6-9, 12-14).

U concentrations at the base of spring pools in the western part of the wetland (slices 7-10) are greatest just above the peat-gyttja contact. In this area, it appears that uraniferous groundwater was forced upward through relatively thin clay and gyttja layers overlying a subsurface bedrock high. In contrast, U concentrations in peat and gyttja along the eastern margin of the wetland, seen in slices 5-12, are highest in proximity to the till-mantled and fractured bedrock valley wall. This area is notable for numerous groundwater seeps issuing from the relatively steep valley side slopes above and at the wetland margin. The yellow-shaded zones of higher U concentrations extend westward into the peat layer in zones underlying spring pools (slices 4-7, 12-14). These postulated shallow subsurface and groundwater flowpaths are illustrated in Fig. 7. Water from springs and seeps at the south end of the wetland flows directly into the peat, then flow becomes concentrated along a shallow trough in the upper surface of the gyttja layer along the central axis of the wetland, indicated by the higher U concentrations shown in Fig. 10, slices 1-3.

5.6 Uranium uptake in lakebed sediments

When a shallow lake occupied the site of the BP wetland, dissolved U in the water column was relatively well mixed and average concentrations of dissolved U responded quickly to changes in the supply and volume of water. Average concentrations of U were likely highest during drier periods when spring water (i.e., groundwater) contributions were proportionally greater. Dissolved U in the lake was subject to additional variability caused by the abundance of suspended algae with probable high affinity for U. Algae have been shown to concentrate U by as much as 1000-10,000X compared to coexisting water (Degens et al., 1979; Mann and Fyfe, 1984; Dienemann et al., 2002). Algal remains settled to the lake bottom and were incorporated into silt and clay lake sediments, forming gyttja. The amount of U removed by settling algae was probably greatest during drier *and* warmer periods, the latter condition fostering high algal productivity. Variables influencing the efficiency of U uptake by algae include pH and temperature of the water, and the proportion of algal remains relative to other types of organic matter that are less efficient concentrators of U. In BP lakebed sediments, other organic matter includes minor amounts of woody tissue, conifer needles, fragments of pondweeds, and sedge leaves.

Studies of uranium distribution in shallow cores of organic-rich marine sediments suggest several processes that could redistribute U immediately after deposition of BP lakebed sediments. These processes include (1) diffusive transfer of U across the sediment/water interface; (2) movement of particle-bound or dissolved U during initial burial and compaction; and (3) fixation of U by reduction (Cochran et al., 1986; Anderson et al., 1989a, b). In another study, Duff et al. (1997) used X-ray near-edge absorption measurements (XANES) to identify oxidation states of U in anoxic algal pond sediments. They noted that the pond sediments contained a mixture of both U(IV) and U(VI) species, with a gradual increase of U(IV) species with depth.

An additional longer-term influence on U concentration and distribution in BP lakebed sediments is post-depositional delivery of U from flowing groundwater. Such additional delivery of U is most likely to occur near the contacts of lakebed sediments with more transmissive units of underlying glaciofluvial sands or overlying peat. Similar enrichments of U in wetland sediments located near contacts with more transmissive units were also noted by Shotyk et al. (1990, 1992). Concentration of U near such contacts in the BP wetland is indicated by U distributions shown in Fig. 10.

5.7 Uranium uptake in peat

As the lake filled with sediment and became shallow enough to support rooted vegetation, areas of formerly open water were occupied by stationary and floating mats of plants and plant remains. Circulation and mixing of water became more restricted, governed by rates of exchange of pore waters

stored in sponge-like peaty sediments. Transport and mixing of water near spring pools was affected to some degree, but the dispersion of dissolved U was very highly restricted because of its strong affinity for peat. At slightly acidic pH, peat can concentrate U by as much as 10,000X compared to coexisting water (Szalay, 1964; Armands, 1967). The uranium concentrations in four samples of peat from core C1 were enriched by a factor of $12,900 \pm 2800$ compared to coexisting pore water. This compares favorably with enrichment factors of approximately 10,000 reported from similar studies of uranium accumulation in peat in the natural environment (Szalay, 1964; Armands, 1967; Halbach et al., 1980; Idiz et al., 1986). Enrichment factors of 10,000 or more imply strong preferential uptake of U by peat despite competing formation of stable dissolved complexes of U (organic or inorganic).

The mode of occurrence of U in peat was directly observed using fission-track radiography of eight samples of variably uraniferous peats from core C5 near a spring pool (Fig. 2). Results indicate that charcoal fragments and finer-grained organic matter showed highest concentrations of U, perhaps related to their relatively large surface areas (Fig. 11). Larger structures of twig, bark, and conifer needle fragments showed U enrichment on external surfaces and on interior surfaces that comprised the original cell wall framework (Fig. 11a-d). Uranium was not concentrated in the immediate vicinity of occasional framboidal pyrites (Fig. 11e-f). However, in adjacent core C8, the most uraniferous peat horizons, containing 1000-3280 ppm (mg kg^{-1}) U, did contain trace amounts of framboidal pyrite.

Humification alters the physical, chemical, and hydrologic characteristics of peat, any or all of which could potentially affect uranium availability and uptake. In core C8, peat humification increased with depth based on scattered, but generally diminishing, carbohydrate fractions of 55 to 38% determined by ^{13}C -NMR (Figure A-5 of Supplemental Information). A similar range of carbohydrate (55 to 44%) was measured in U-poor core C9 taken from the center of the wetland. In U-bearing intervals of core C8, there is a poor correlation between U content and depth or degree of humification. These limited results suggest that peat humification is not markedly greater near a spring pool and that uptake of U is not strongly dependent upon the degree of humification.

Fifteen samples of uraniferous peat were collected from various depths within the 1.5 m core C8 (Fig. 2) during initial sampling of the wetland. The samples were extracted with 0.1 M NaHCO_3 to estimate the fraction of loosely bound or specifically adsorbed U(VI). Uranyl forms strong complexes with dissolved carbonate species (Langmuir, 1978) and is readily leached from peat by sufficiently concentrated bicarbonate solutions (Zielinski and Meier, 1988). Leached U ranged from 80 to 87% and was independent of sample depth or original U concentration. To address the possibility that U could have been oxidized during sample preparation, a sample of freshly collected peat of 1500 ppm U was

leached in the field under nitrogen atmosphere. The leached fraction of U for this sample was 78%, suggesting that oxidation of U in laboratory-leached peats was, at most, minor.

These observations of U distribution and leachability are most consistent with initial uptake of dissolved U(VI) by specific adsorption onto ion-exchange sites of organic matter. Such uptake involves ion exchange and complexation of uranyl ion (UO_2^{+2}) with carboxylic and other acidic functional groups of humic substances (Szalay, 1964; Borovec et al., 1979; Halbach et al., 1980; Disnar, 1981; Idiz et al., 1986). Field and laboratory experiments have documented the efficient uptake of U(VI) onto peat (Szalay, 1964) and controls on the leachability and reversible uptake of U(VI) (Zielinski and Meier, 1988). Recent studies of U-rich peats, organic soils, and sediments using synchrotron-based X-ray absorption spectroscopy have confirmed the dominance of oxidized U(VI) species (Duff et al., 1997; Bolanos, 2006; Regenspurg et al., 2010). Similar spectroscopic measurements of naturally reduced sediments near a former U mill at Rifle, Colorado, indicate incomplete reduction of U(VI) delivered by local groundwater (Campbell et al., 2012; Qafoku et al., 2014; Janot et al., 2016).

Readily leachable U(VI) is present at all depths in peats of core C8. Water table fluctuations at this site are minimized by the presence of a nearby spring, thus redox gradients with depth are relatively stable. Observed framboidal pyrite in some of the deeper peats from cores C5 and C8 indicates at least locally reducing conditions of $E_h < 0.0$ V. Mineral/solution equilibria calculations performed on waters of this study indicate that U(IV) oxide minerals such as uraninite and coffinite are thermodynamically stable at $E_h < 0.1$ V. Sulfate-reducing bacteria that mediate formation of framboidal pyrite are also capable of reducing dissolved U (Lovley and Phillips, 1992; Lovley et al., 1993). Factors that lessen the amount of uranium reduction in core C8 might include sluggish kinetics and strong complexing of U(VI) with peat or dissolved humic substances that renders U less bioavailable (Suzuki and Banfield, 1999). Potentially limiting factors for bacterially mediated processes at the BP wetland include low concentrations of dissolved sulfate and iron in source waters (Table 3) and sub-freezing temperatures throughout much of the year.

6. Long-term fate of uranium in the wetland system

Peat accumulation and uranium uptake from groundwater are ongoing processes in the BP wetland. Environmental changes, in the relatively short or long term, could potentially alter the fate of uranium in the BP wetland. Because it is located within a national forest, anthropogenic disturbances such as mining or other land development are unlikely. More likely scenarios include climate change or, in the long term, burial of peats with attendant organic-matter diagenesis and coalification.

If a warming climate translates to lower water tables, it is reasonable to expect more drying, oxidation, and decomposition of near-surface peats. During peat decomposition, U that is presently bound to organic matter may become available for leaching. Repeated raising and lowering of the water table may then cause some of this U to be flushed from the wetland, whereupon it could enter downstream surface-water systems. At the time of sampling for this study, the outlet water at the BP wetland contained 10 ppb ($\mu\text{g L}^{-1}$) U (W1, Table 3). A useful benchmark for comparison is the drinking water standard established by the U.S. Environmental Protection Agency of 30 ppb U (US EPA, 2001).

Decomposition of peat during burial will proceed at a much slower rate. Rate-determining variables include the viability of anaerobic bacteria and the abundance of easily metabolized organic matter. In this scenario, slow release of organically bound U(VI) may facilitate reduction to U(IV) by resident bacteria, or further enable abiotic reduction mechanisms that are thermodynamically favored within pore waters of persistently low Eh. Formation of reduced U(IV) species may provide some additional resistance to remobilization of U.

Loss of U during organic matter diagenesis is strongly indicated because large ($\sim 10\times$) volume reductions during coalification of peat would otherwise greatly increase U concentrations. For example, if U is conserved during coalification then a peat containing only 10 ppm (mg kg^{-1}) U on a wet basis would yield a coal containing ~ 100 ppm U. Most coals in the United States contain < 4 ppm U and coals of > 20 ppm are rare (Zielinski and Finkelman, 1997). Lignites represent an intermediate stage in the coalification of peat and can contain as much as 200 ppm U (Denson and Gill, 1956). A study of highly uraniferous lignite within a sandstone U deposit in Texas indicated that the vast majority (60-80%) of U was dispersed in organic matter (Mohan et al., 1991). The authors used selective extraction results to propose a uranyl-humate species. Future studies of U speciation in lignites and peats will benefit from more direct measurements of U oxidation state and bonding environment using synchrotron-based X-ray absorption spectroscopy.

7. Conclusions

Uranium in the BP wetland and its underlying lakebed sediments is associated with organic matter, but the distribution of U is heterogeneous and the relationship between U and organic matter is complex. Samples of similar organic content have highly variable U concentrations because the U concentration in sediments is strongly influenced by proximity to U-rich groundwater that emerges from local fractured and faulted granite gneiss. U-rich groundwater enters the wetland as artesian springs, seeps, and as shallow and deeper subsurface flows directed along the upper and lower contacts of

relatively impermeable lakebed sediments. Extensive sampling of sediments and water, and determination of hydrologic sources and inputs at the BP wetland reveal these multiple pathways for delivery of U-bearing groundwater and thus expand upon more limited observations of controls on U distribution and concentration in other wetlands.

Fission-track radiography of U-rich peat provides an extremely detailed view of the intimate association of U with structured and variably humified organic constituents, and also indicates an absence of U enrichment near framboidal pyrite. Selective extraction of freshly collected peat with a sodium bicarbonate solution that is specific for U(VI) indicated that the majority (~80%) of U is oxidized, despite reducing conditions. Possible explanations include slow kinetics of reduction, low bioavailability of U resulting from strong bonding of adsorbed uranyl with humic substances, and low activity of anaerobic bacteria capable of reducing U.

The unusually broad scope of sampling and investigations at the BP wetland provides a resource base of information that could be exploited further to investigate aspects of montane wetland function, environmental sensitivity, hydrology, and U uptake. Generally speaking, greater consideration of the third dimension during sampling, and greater appreciation of the broader context of wetland geologic history that includes pre-peatland sediments could enhance future studies of wetlands.

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812

FIGURE CAPTIONS

Figure 1. Location and physiographic setting of the Boston Peak wetland, Larimer County, Colorado, USA. Simulated oblique aerial view looking northward along the Laramie River valley. Aerial photo from U.S. Department of Agriculture, National Agriculture Imagery Program (NAIP), image date 2013, draped over USGS National Elevation Dataset 0.3 arc-second DEM data. Scale of image is variable due to perspective tilt.

Figure 2. Maps showing locations of stratigraphic core/auger holes, piezometers and numbered spring pools, and water samples in the Boston Peak wetland. All elevations in the study were measured relative to a local datum determined by high-resolution differential GPS.

Figure 3. Geologic map of the Boston Peak wetland and immediate surrounding area. Explanation of map symbols: Qls, landslide (Quaternary); Qal, alluvium; Qg, glacial till (may include colluvium and alluvium); Twt, rhyolite tuff, probably part of White River Formation (Tertiary); Twr, White River Formation; Ta, andesite; Xg, granite gneiss (Proterozoic); Xm, mafic gneiss. Cross-hatched features indicate fracture or shear zones, queried where inferred. Dotted lines indicate inferred traces of concealed faults. GRFZ – Green Ridge fault zone. Geology mapped by F.A. Hills, USGS (1992, written comm.), based in part on unpublished mapping of W.A. Braddock, USGS and University of Colorado (1989, written comm.).

Figure 4. Oblique view of Boston Peak wetland showing the pre-lake topography of the basin and extent of basin fill of successive lacustrine clay, gyttja, and peat layers. Contour interval = 1 meter; selected contours labeled in meters above sea level.

Figure 5. North-south cross section through the BP wetland. Vertical exaggeration approx. 10:1. Elevation of “datum” is 2710.3 m ASL. Core and auger holes used to construct the cross-section are shown at top of figure; refer to Figure 2 for locations. P, peat; G, gyttja; L, lacustrine (lake) clay; S, sand. Stippled areas represent sandy and gravelly deposits that intertongue and interlayer with peat, gyttja, and clay deposits.

Figure 6. Calibrated radiocarbon ages of sediments of the BP wetland, in calendar years before 1950. See Table 1 for details. Core diagrams are placed in order from west to east. Elevation of “datum” is

840 2710.3 m ASL. P, peat; G, gyttja; T, transitional sediments consisting of interlayered clay and gyttja; L,
841 lacustrine clay.

842 Figure 7. Schematic cross-section illustrating the stratigraphic units comprising aquifers (peat and sand)
843 and aquitards (clay and gyttja), and showing postulated groundwater sources and flowpaths, in the
844 Boston Peak wetland. Not drawn to scale.

845 Figure 8. Contour map showing the difference in elevation between the piezometric surfaces of the
846 confined basal aquifer and the unconfined peat aquifer, measured 20 July 1989. Units in meters. Positive
847 values indicate that the potentiometric surface of the basal aquifer is higher than that of the unconfined
848 peat aquifer, i.e., that the basal aquifer is under artesian pressure and has potential for upwelling of
849 groundwater.

850 Figure 9. Plot of U concentration versus organic content (LOI) for auger samples of peat and sediments
851 of the BP wetland. Refer to Table 2 for ranges of values.

852 Figure 10. Three-dimensional model showing uranium distribution in peat and sediments of the Boston
853 Peak wetland. Cross-section “slices” through the model, numbered 1-17, reveal relationships among
854 uranium concentrations, stratigraphy, and topography. Locations of individual slices shown in index
855 map, lower right. Thin, sub-horizontal black lines indicate contacts between peat, gyttja, and lacustrine
856 clay layers in each slice.

857 Figure 11. Fission-track images of the uranium distribution in thin sections of peat. Dark areas
858 correspond to high concentrations of uranium. All samples are from core C5. (A) Low-magnification view
859 showing the variety of uranium enrichment in constituent organic matter. Areas of low fission-track
860 density (low U) are generally associated with the epoxy resin that fills pore spaces originally occupied by
861 water. Sample depth=1.4 m, long dimension of view=15 mm. (B, C, D) Uranium distribution in sedge
862 leaves, twig fragment and conifer needles, respectively. Sample depths=0.9-1.2 m, long dimension of
863 views=2.5 mm. (E) Photomicrograph and (F) corresponding fission-track image showing the absence of
864 anomalous uranium enrichment in the immediate vicinity of framboidal pyrite that occupies the interior
865 of the rootlet (darkest zone in E). Sample depth=1.4 m, long dimension of view=1.0 mm.

866

TABLE 1. Radiocarbon ages for sediment and wood fragments from the Boston Peak wetland (shown graphically in figure 6). The core is identified by the C-designation in the sample number, e.g., C4 is core 4. Radiocarbon ages are shown $\pm 1\sigma$ (68%) standard deviation. Cal. age is the median calibrated age reported by CALIB 7.1 (Stuiver and Reimer, 1993; Reimer et al., 2013), in calendar years before 1950.

Sample number	Depth (cm)	Lab number	Material	^{14}C Age (yr)	Cal. Age (yr)
BP-C4,157-164	493	PITT-0432	Gyttja	10,380 $\pm$ 145	12,220
BP-C6,188-195	650	PITT-0433	Gyttja/transitional	13,250 $\pm$ 160	15,920
BP-C7-6	388	Beta-44842	Gyttja	4330 $\pm$ 70	4930
BP-C7-11	520	Beta-44843	Gyttja	8460 $\pm$ 100	9460
BP-C7-13	598	Beta-44844	Gyttja	10,120 $\pm$ 80	11,730
BP-C7-18	742	Beta-44845	Gyttja/transitional	12,720 $\pm$ 90	15,140
BP-C7-21	820	Beta-44846	Gyttja/transitional	12,820 $\pm$ 180	15,280
BP-C1, 82-96	226	Beta-22463	Peat	3165 $\pm$ 105	3380
BP-C1, 231	587	Beta-22465	Wood fragment	11,880 $\pm$ 150	13,720

TABLE 2. Summary of uranium and organic matter concentration data for the primary sediment types in the Boston Peak wetland. Units of LOI are percent loss by weight. n, number of samples analyzed.

	PEAT		GYTTJA		CLAY		SAND	
	U (ppm)	LOI (%)	U (ppm)	LOI (%)	U (ppm)	LOI (%)	U (ppm)	LOI (%)
MIN	2	7	24	7	4	1	4	2
MAX	3280	91	318	43	107	30	364	23
MEDIAN	39	81	73	31	18	7	10	4
MEAN	119	74	78	29	26	8	24	5
n	290	290	209	209	128	128	46	46

Table 3. Chemistry of surface water and spring-seeps at the Boston Peak wetland.

Sample	surface				spring seep					
	W1	W2	W23	W24	W3	W4	W5	W6	W7	W8
Major chemical species (mg L ⁻¹)										
SiO ₂	26.1	31.0	30.2	40.5	19.4	13.3	18.2	12.9	12.1	19.0
Na	12.4	10.5	10.0	12.3	24.1	9.1	25.9	5.5	3.7	8.8
K	1	2	2	<1	3	3.	3	2	1	2
Mg	5.22	4.63	4.07	4.54	9.65	8.28	8.23	8.77	2.51	7.66
Ca	22.4	19.4	17.3	20.9	20.3	21.7	19.3	22.9	8.0	15.3
Fe (total)	0.43	0.05	0.05	0.09	0.02	0.02	0.04	0.02	2.09	0.20
HCO ₃	132	115	94.5	112	183	135	181	132	50	110
SO ₄	<0.5	3.4	3.4	0.4	6.3	6.5	4.6	6.3	<0.5	4.0
PO ₄	0.06	0.04	0.04	0.08	0.006	0.003	0.002	0.002	<0.1	<0.1
Cl	0.5	0.7	0.7	0.3	2.1	4.9	2.8	0.7	0.6	0.6
F	0.14	0.11	0.27	0.29	0.32	0.16	0.33	0.13	<0.1	0.13
NO ₃	<0.1	<0.1	0.06	0.02	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Dissolved oxygen	2.0	3.8	2.4	---	2.3	3.3	4.0	4.0	0.5	2.0
Diss. org. carbon	18	6.3	8.0	16	1.4	1.6	3.8	2.0	7.2	4.3
Total diss. solids (calc)	133.8	128.5	114.6	135.5	175.7	133.4	171.5	124.2	55.4	112
T (°C)	15	5	3.5	14	5.5	5.0	9.0	8.5	8.5	8.5
Spec. cond. (µS/cm)	220	230	140	160	240	250	330	250	95	215
pH	7.45	7.30	7.15	7.35	7.65	7.60	7.55	7.65	6.20	6.95
Minor and trace chemical species (µg L ⁻¹)										
Al	<100	<100	200	<100	<100	<100	<100	<100	200	<100
Ba	40	56	53	49	45	10	25	9	52	27
Cu	<10	<10	20	<10	<10	<10	20	<10	20	<10
Li	10	10	9	13	15	11	18	10	<4	7
Mn	44	6	4	15	5	2	5	4	291	12
Sr	256	223	182	270	377	337	323	351	72.2	301
Zn	4	4	7	7	<3	3	24	7	14	6
U	10	5.4	0.8	0.2	56	32	83	36	<0.1	20

---, not determined. Other elements sought but below the detection limit (µg L⁻¹)= Ag (2), Be (1), Bi (10), Cd (1), Co (3), Cr (1), Ga (5), Mo (10), Ni (5), Pb (10), Sn (6), Ti (1), V (6), Zr (1). Sample W14 was not collected.

Table 4. Chemistry of water sampled from piezometers at the Boston Peak wetland. Sample W14 was not collected.

Sample	W9	W10	W11	W12	W13	W15	W16	W17	W18	W19	W20	W21	W22
Total depth (m)	6.1	3.7	2.7	5.8	2.4	7.6	7.0	2.4	5.8	9.1	2.1	4.9	5.2
Lithology at depth	sand	peat	peat	sand	peat	sand	sand	peat	sand	sand	peat	sand	sand
Major chemical species (mg L ⁻¹)													
SiO ₂	28.3	25.9	21.1	40.9	40.7	28.7	32.1	17.5	46.3	44.6	48.4	47.6	23.4
Na	16.7	19.3	8.0	37.9	30.0	20.3	50.5	10.3	60.6	17.6	22.0	29.9	11.7
K	3	4	3	4	4	3	1	2	3	4	2	4	3
Mg	6.17	8.48	8.59	10.40	9.75	10.70	2.17	9.04	6.87	10.0	14.3	7.21	6.75
Ca	14.3	18.2	24.6	28.3	24.4	29.0	6.1	19.7	20.4	28.1	39.0	23.4	18.1
Fe (total)	0.21	0.07	1.17	7.56	3.20	0.09	1.50	0.16	6.86	3.53	5.80	1.58	2.83
HCO ₃	131	157	150	266	215	200	162	136	277	214	278	189	120
SO ₄	0.5	4.7	0.5	0.5	2.6	4.4	0.4	2.4	1.0	1.3	0.5	5.7	7.4
PO ₄	0.005	0.009	0.003	0.001	0.003	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cl	1.5	2.4	0.7	6.5	3.1	2.1	3.3	1.0	4.2	1.6	0.4	5.0	0.7
F	0.22	0.25	0.18	0.39	0.36	0.55	1.35	0.48	1.26	0.66	0.47	0.91	0.46
NO ₃	<0.1	0.1	<0.1	<0.1	<0.1	0.94	0.18	0.03	0.04	0.03	0.04	0.04	0.05
Dissolved oxygen	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Diss. org. carbon	10	22	6.0	3.5	9.0	11	5.5	2.9	9.3	11	13	1.8	1.5
Total diss. solids (calc)	135.4	160.6	141.7	267.3	223.9	198.1	178.3	129.5	286.7	216.6	269.6	218.3	133.4
T (°C)	9.5	10.5	10.0	10.5	10.0	7.0	8.5	8.0	7.5	6.5	10.0	4.0	5.0
Spec. cond. (µS/cm)	190	240	250	450	350	305	310	200	420	325	400	300	195
pH	7.05	6.25	6.15	6.85	6.35	6.95	6.85	7.00	6.65	7.05	6.30	7.20	7.15
Minor and trace chemical species (µg L ⁻¹)													
Al	<100	<100	<100	<100	<100	100	100	<100	<100	100	<100	100	200
Ba	85	57	37	139	91	105	40	30	99	127	149	51	85
Cu	10	<10	<10	<10	10	30	20	10	20	140	20	30	20
Li	12	13	11	12	32	16	9	5	16	14	10	7	7
Mn	115	28	48	351	120	186	75	17	262	527	296	417	240
Sr	246	336	352	368	355	409	70	327	255	363	558	262	236
Zn	18	14	11	12	32	16	9	5	16	14	10	7	7
U	1.9	0.9	<0.1	1.4	1.5	1.1	1.5	0.9	3.7	3.5	1.2	0.3	0.9

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10