

Paleochannel Deposits of Natural Uranium at a Former Air Force Landfill

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ABSTRACT

The US Air Force has sought to understand the provenance of radionuclides that were detected in monitor wells surrounding a closed solid-waste landfill at the former Lowry Air Force Base in Denver, Colorado. Groundwater concentrations of gross alpha, gross beta, and total uranium were thought to exceed regulatory standards. Downgradient concentrations of these parameters exceeded upgradient concentrations, suggesting that the landfill is leaching uranium to groundwater.

Alternate hypotheses for the occurrence of the uranium included that either equipment containing refined uranium had been discarded or that uranium ore may have been disposed in the landfill, or that the uranium is naturally-occurring. Our study has concluded that the elevated radionuclide concentrations stem from naturally-occurring uranium in the regional watershed which has been preferentially deposited in paleochannel sediments beneath the site.

This study shows that a simple comparison of upgradient versus downgradient groundwater samples can be an inadequate method for determining whether heterogeneous geosystems have been contaminated. It is important to understand the geologic depositional system, plus local geochemistry and how these factors impact contaminant transport.

INTRODUCTION

Lowry Air Force Base (Lowry) was closed in 1994 as part of the Base Realignment and Closure (BRAC) program. As part of the due diligence activities conducted by the Air Force, a series of remedial investigations were conducted across the base. A closed waste landfill, designated Operable Unit 2 (OU2), was initially assessed in a series of three investigations. The three studies indicated that Gross Alpha, Gross Beta, and Uranium concentrations were consistently above regulatory standards and that there were detections of low concentrations of other radionuclides. Gross alpha and gross beta concentrations in groundwater were consistently thought to exceed regulatory limits. The highest gross alpha activity was 11.7 Becquerels per liter (Bq/L) (315 picocuries per liter (pCi/L)) and the highest gross beta activity was 23.3 Bq/L (630 pCi/L). The Air Force and regulators agreed to use a one-year monitoring and sampling program to seek to explain the provenance of the radionuclides that were detected and to

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determine if radionuclides potentially buried within OU2 are leaching into groundwater or surface water.

OU2 is a 30 hectare parcel located in the south-central portion of Lowry, as shown in Figure 1. OU2 was used from 1948 until 1986 for disposal of general base-related waste. The volume of waste has been estimated to be approximately 1.4 million cubic meters. Some drums were disposed in the 1950s and 1960s. Waste thickness averages 4.5 meters [1]. Historical records indicate that a drum containing electron tubes was disposed at the site sometime during the 1950s [2].

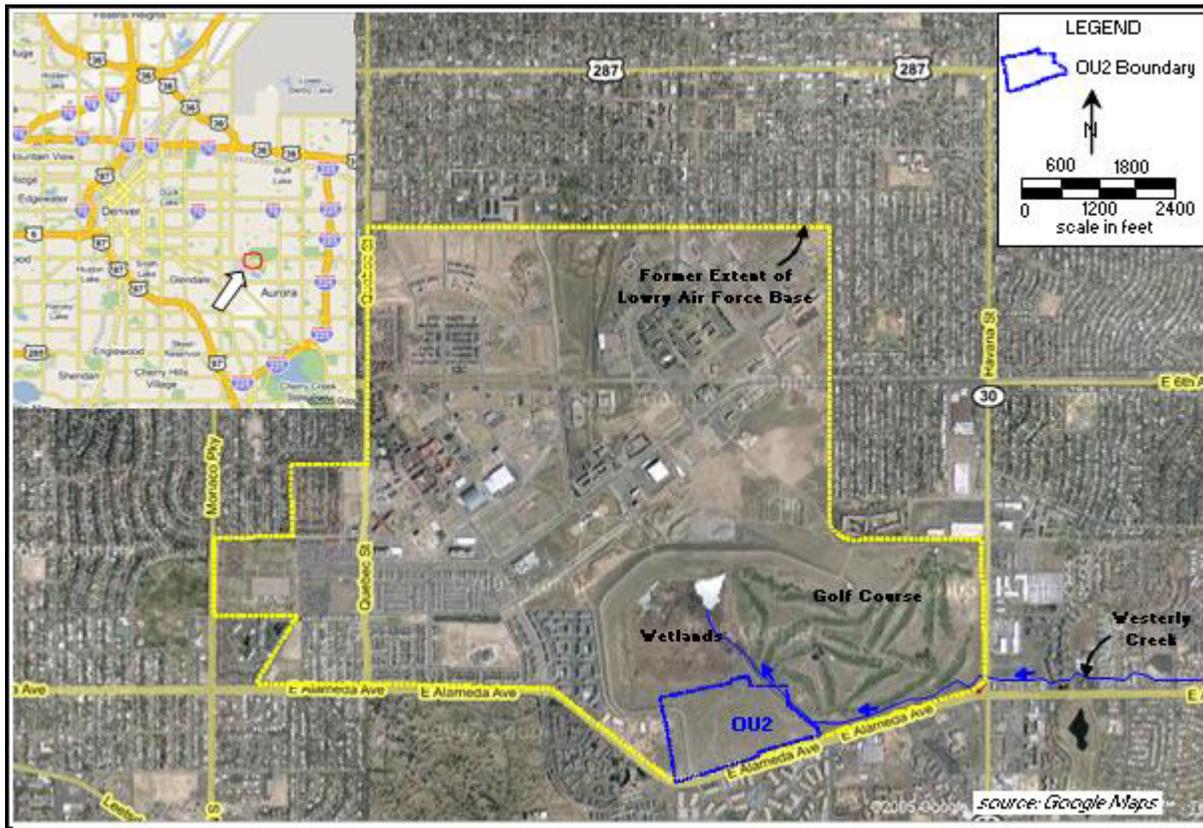


Figure 1. Location of Operable Unit 2, Lowry Air Force Base, Colorado

OU2 is bounded by residential areas to the south and west, a flood-control dam and wetlands to the north, and a creek and golf course to the east. The flood-control dam and wetlands are new features dating from 1992. The creek is a perennial stream that was re-routed from an original run farther to the east. OU2 is located within the Westerly Creek drainage basin, which comprises a 50 km² drainage area. Regional stream flow directions are from the southeast to the northwest.

The native soils consist of Holocene-age alluvium and loess deposits. These deposits range in thickness from 10 m in the south of the site to 3 m at the northern edge [3]. The alluvium

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unconformably overlies a weathered bedrock surface developed on the claystones, siltstones, and sandstones of the Cretaceous and Tertiary Denver Formation. A paleochannel, which is incised into the bedrock surface, trends diagonally across the site from southeast to northwest [1].

A 1998 study included sampling for the anthropogenic radionuclides Cesium-137 (Cs-137), Cobalt-60 (Co-60), Nickel-63 (Ni-63), Plutonium-238 (Pu-238), Plutonium-238/240 (Pu-238/240), and tritium (H-3, which also occurs naturally) [4]. The activities of all these radionuclides were at background concentrations and up-gradient versus down-gradient comparisons were not statistically separable [5].

EVALUATION OF RADIONUCLIDES OF POTENTIAL CONCERN

Electron tubes are a potential source of low-level radioactivity. Beginning around 1940, radionuclides were added to a number of tubes designed for military applications including radar transmit-receive tubes, trigger tubes and tubes for voltage regulation and surge protection. The earliest tubes used Ra-226 and Cs-137. Later versions of these tubes used Co-60, H-3, and Kr-85. Up to a microcurie of radioactive Cs-137, Co-60, Kr-85 or Ni-63 was used to insure high-speed operation with minimal voltage variation in the absence of light. Other radioactive materials used in tubes included C-14, Pm-147 and U-238. Individually, electron tubes contain little or no hazard. However, large collections of buried electron tubes may represent a substantial risk of radioactive contamination, specifically if the metal or glass envelopes containing the radioisotopes are broken or compromised.

Radioisotopes with self-luminous properties were widely used in aircraft gauges, sights, switches telescopes, and other devices. Primary ROPCs from these sources include Radium, Thorium, H-3, Sr-90, Pm-147, and Kr-85.

Various types of components were identified that contained uranium. Components containing U-238 and U-235 include various types of weights (e.g., ballast and counter-weights), and in electron tubes. Depleted uranium (DU) was also used as weights (e.g., balance weight, horizontal stabilizer, counter-balance) as well as munitions projectiles, cartridges, and special weapons mockups used for training purposes. Mockups of this kind were used at Lowry. Natural uranium (U-nat) was also used in counter weights.

Historical information and previous investigations for Lowry were reviewed to identify radionuclides of potential concern (ROPCs). The Air Force Aircraft Radioactive Material Database was searched for radionuclides that could have been used at Lowry [6]. The search identified 6623 components containing radioactive material. All 6623 records were reviewed to identify the equipment that may have been used at Lowry, the radioactive material used, and the amount of radioactivity that the equipment contained. A list of 23 Radionuclides of Potential Concern (ROPCs) was selected and is shown in Table I. The list includes parameters that were previously detected at OU2 as well as ROPCs identified in the database search.

STUDY METHODS

Groundwater samples were collected from 17 perimeter wells and surface water and sediments were collected from upstream and downstream locations from an adjacent creek over four consecutive quarters. Samples were analyzed for concentrations of the 23 ROPCs. In addition, concentrations of major ions and total metals were analyzed. Ten geochemical parameters were measured onsite, including pH, specific conductance, temperature, turbidity, dissolved oxygen, redox potential, water elevation, and concentrations of sulfide, ferrous iron and nitrite ions. The 'low-flow' sampling protocol was used and filtered and nonfiltered sample splits were compared to evaluate the potential impact of sample turbidity and colloidal transport. The National Uranium Resource Evaluation (NURE) database was searched for the presence of naturally-occurring radionuclides in the area.

A review U.S. Geological Survey databases was conducted to determine the background levels of uranium in the Denver region. The USGS maintains a database of natural uranium concentrations in sediments and groundwater samples that were collected from across the United States as part of the National Uranium Resource Evaluation (NURE Database) [7].

Table I. Radionuclides of Potential Concern and Applicable Groundwater Standards

ROPC	Regulatory Standard [pCi/L]	ROPC	Regulatory Standard [pCi/L]
Gross Alpha	15 ^a	Am-241	0.15 ^{e,f}
Gross Beta	50 ^b	H-3	20,000 ^g
Ra-226	5 ^c	C-14	2,000 ^h
Ra-228	5 ^c	Ni-63	50 ⁹
U _{total}	30 µg/L ^d	Sr-90	8 ^g
U-234	≈ 9.5 ^d	Pm-147	600 ⁱ
U-235	≈ 1.1 ^e	Re-187	9,000 ^h
U-238	≈ 9.5 ^d	K-40	Not listed
Pu-238	15 ^a	Co-60	100 ^h
Pu-239/240	0.15 ^e	Cs-137	200 ^h
Th-232	15 ^a		
Th-228	15 ^a		
Th-230	15 ^a		

- Stated in 40 Code of Federal Regulations (CFR) 141.66(c), the MCL for gross alpha particle activity, including radium-226 but excluding radon and uranium
- Stated in 40 CFR 141.26. For community water systems designated as vulnerable, the screening level for gross beta particle activity minus the naturally-occurring K-40 beta is 50 pCi/L. For community water systems contaminated by effluents from nuclear facilities, the screening level for gross beta particle activity minus the naturally-occurring K-40 beta is 15 pCi/L
- Stated in 40 CFR 141.66(b), the MCL for combined Radium-226 and 228 is 5 pCi/L.
- Stated in 40 CFR 141.66(e). Apportioning the 30 micrograms per liter [µg/L] limit to the uranium isotopes assumed 0.67 pCi/µg and uranium activity fractions of 0.473 for U-234 and U-238 and 0.054 for U-235.

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- e. Stated in Regulation 41, Section 41.5.C.2, Basic Standards for Groundwater, CDPHE, Water Quality Control Commission.
- f. Non-descript americium standard in CDPHE Regulation 41.5.C.2 applied to Am-241.
- g. Stated in 40 CFR 141.66, Table A
- h. Stated in 40 CFR 141.66(d). 4 mrem/yr limit as specified in Table IV-2A and IV-2B of EPA76 and in Table 3-3 of EPA00
- i. Stated in 40 CFR 141.66(d) calculated from NBS Handbook 69.

Samples were collected over four consecutive quarters. Groundwater samples were collected from the existing monitoring wells located around the perimeter of OU2. No wells remained within the interior of the former landfill. Low-flow sampling protocols were used to ensure sample consistency and representativeness [8]. Surface water samples and sediment were collected from creek locations both upstream and downstream of OU2.

Groundwater, surface water and sediment samples were analyzed for ROPCs, 23 metals and 14 geochemical parameters. Laboratory analytical data was reduced utilizing guidance from the Interim Guidance on Statistical Analysis of Groundwater Monitoring Data at RCRA Facilities [9] as well as the Addendum to the Interim Final Guidance [10]. This guidance outlines analysis techniques and protocols for evaluating contaminant levels in groundwater wells against hydraulically up-gradient (or background) wells and/or fixed standards or limits (e.g., MCLs). The data was analyzed using standard descriptive statistics as well as parametric and non-parametric analysis of variance statistical techniques in order to ascertain the likelihood of the presence of ROPCs in wells near OU2.

RESULTS

A search of the NURE database found 29 groundwater analysis results and 28 sediment analysis results from samples collected from an approximate 10-mile radius of former Lowry AFB [11]. As shown in Figure 2, a dissolved-phase plume of natural uranium trends from southeast to northwest, directly up-gradient of OU2. Dissolved uranium concentrations in the water-table aquifer are as high as 147 $\mu\text{g/L}$ up-gradient of OU2.

Groundwater flow direction and flux are very similar during each of the four quarterly monitoring events. The groundwater flow direction was to the west-northwest and the gradient was 2%. Over the course of the investigation, groundwater pH averaged 7.3, specific conductance averaged 1078 microsiemens per centimeter ($\mu\text{S/cm}$), turbidity averaged 0.8 nephelometric turbidity units (NTU), dissolved oxygen averaged 1.1 milligrams per liter (mg/L), and oxidation-reduction potential averaged 21 millivolts (mV).

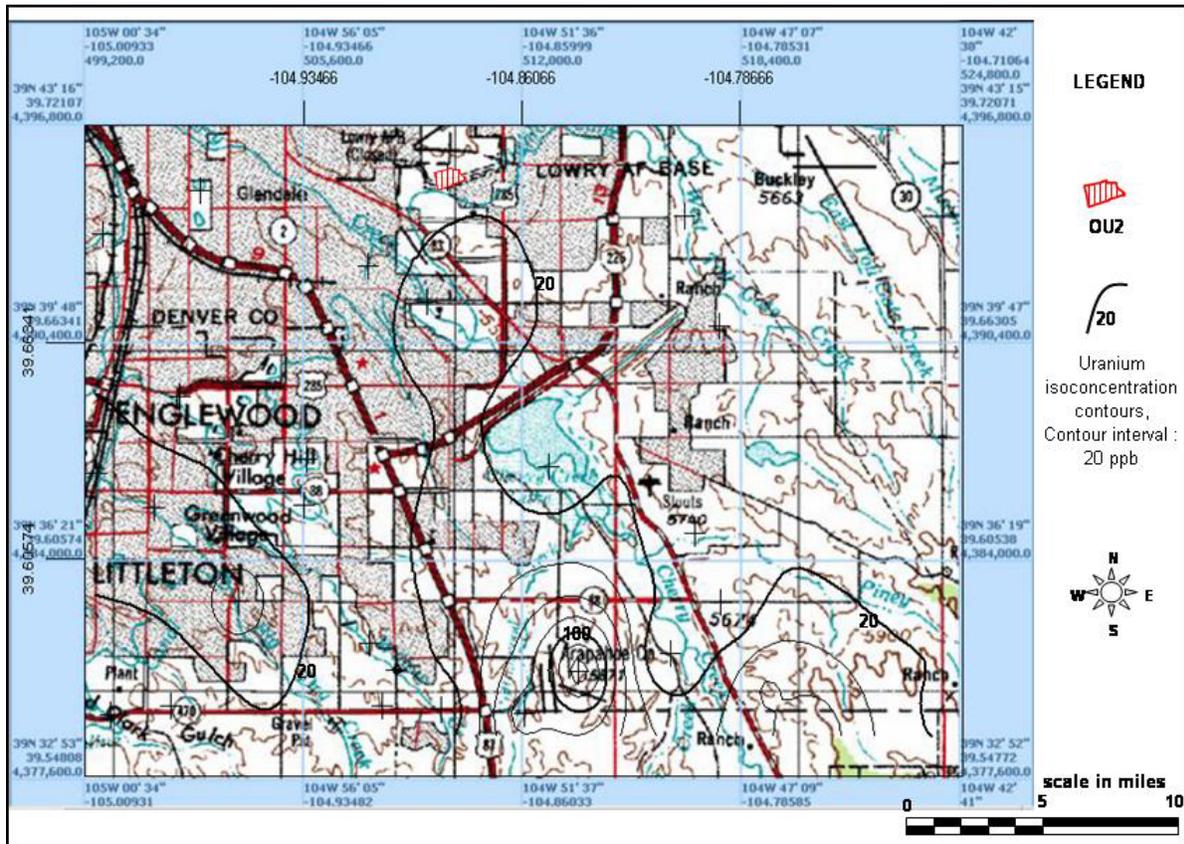


Figure 2. Dissolved Uranium in the Regional Watershed

Analysis of major anion concentrations indicated that OU2 groundwater has consistently had high alkalinity. Total hardness averages 300, which classifies the water as ‘very hard’. Total dissolved solids in all wells were above the USEPA secondary drinking water standard of 500 mg/L [12]. Chlorides and sulfate concentrations are elevated, which in the western United States is reflective of recharge by irrigation [13]. Nitrate and orthophosphate concentrations were not elevated. The averaged ion charge balance for the 17 wells was 0.2%, which indicates that the charges balance [13]. Among cations, calcium and sodium dominate (potassium amounts to less than 10% of summed potassium + sodium concentrations). Among anions, bicarbonate is the dominant ion, except that sulfate is the dominant anion in wells located near the golf course east of the site. The groundwater at OU2 is a sodium-calcium-bicarbonate water type.

Gross Alpha results are presented as raw and ‘corrected’ that exclude contributions from uranium in accordance with the guidance in the Safe Drinking Water Act [14]. After correction, all Gross Alpha results were less than the 15 pCi/L MCL. Gross Beta results were also elevated throughout OU2 but none exceeded the 50 pCi/L MCL. Gross Alpha, Gross Beta concentrations in groundwater are shown in Figure 3.

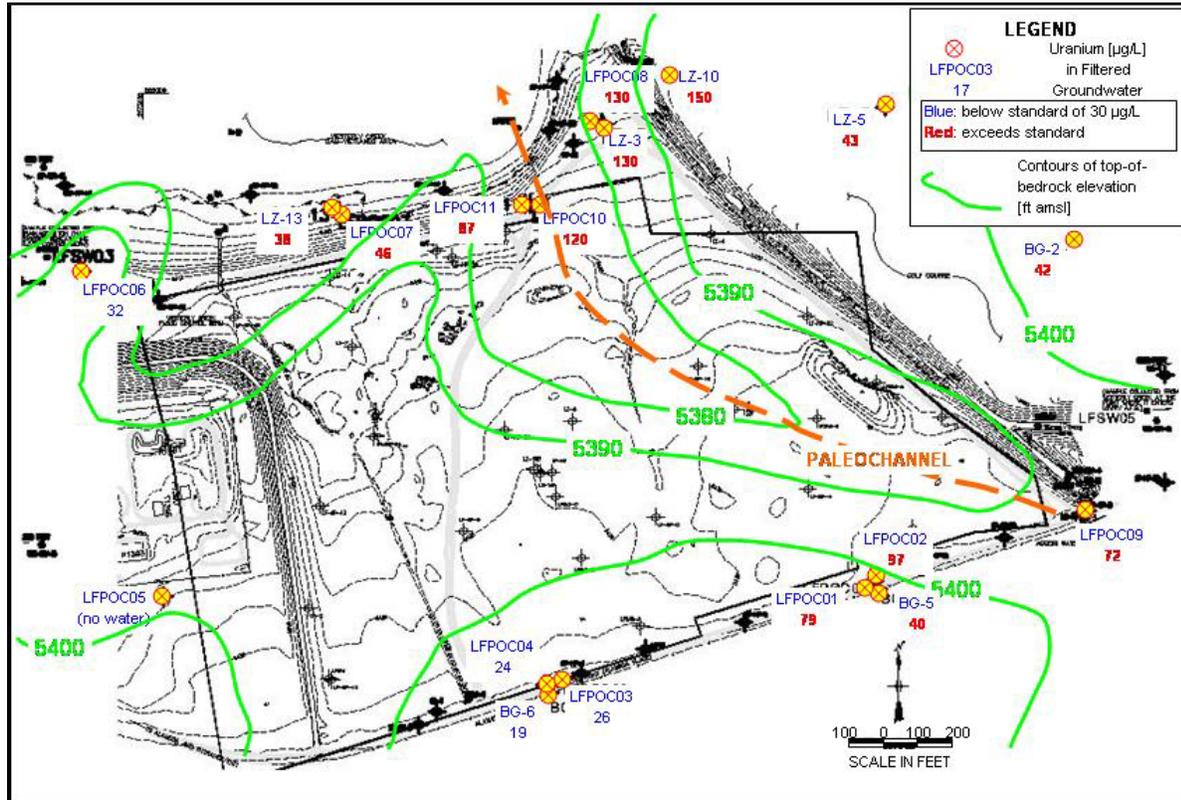


Figure 3. Dissolved Uranium in OU2 Groundwater

Uranium concentrations exceeded the standard of 30 $\mu\text{g/L}$ in 6 of 9 up-gradient wells with a maximum of 97 $\mu\text{g/L}$ at LFPOC02, and in all eight down-gradient wells with a maximum of 150 $\mu\text{g/L}$ at LZ10. The location and magnitude of each maximum value were near identical the values observed during previous quarters. Uranium concentrations in groundwater are shown in Figure 4.

Uranium concentrations showed similar up-gradient and down-gradient relationships in all four quarterly sampling events. The analysis of the means of the up-gradient wells versus down-gradient wells by the non-parametric Wilcoxon-Rank Sum statistical test resulted in 8 ROPCs rejecting the Null Hypothesis of equal means. Results of the statistical tests on groundwater samples collected during each quarter indicate that Gross Beta, uranium (both isotopic and total), H-3, Ni-63, and Pm-147 were found to be statistically distinguishable in down-gradient well locations versus up-gradient locations.

CONCLUSIONS

Comparison of up-gradient versus down-gradient concentrations of uranium in groundwater shows that the averaged uranium concentration is higher in the down-gradient wells compared to the up-gradient wells. However, we do not conclude that this is the result of disposal of

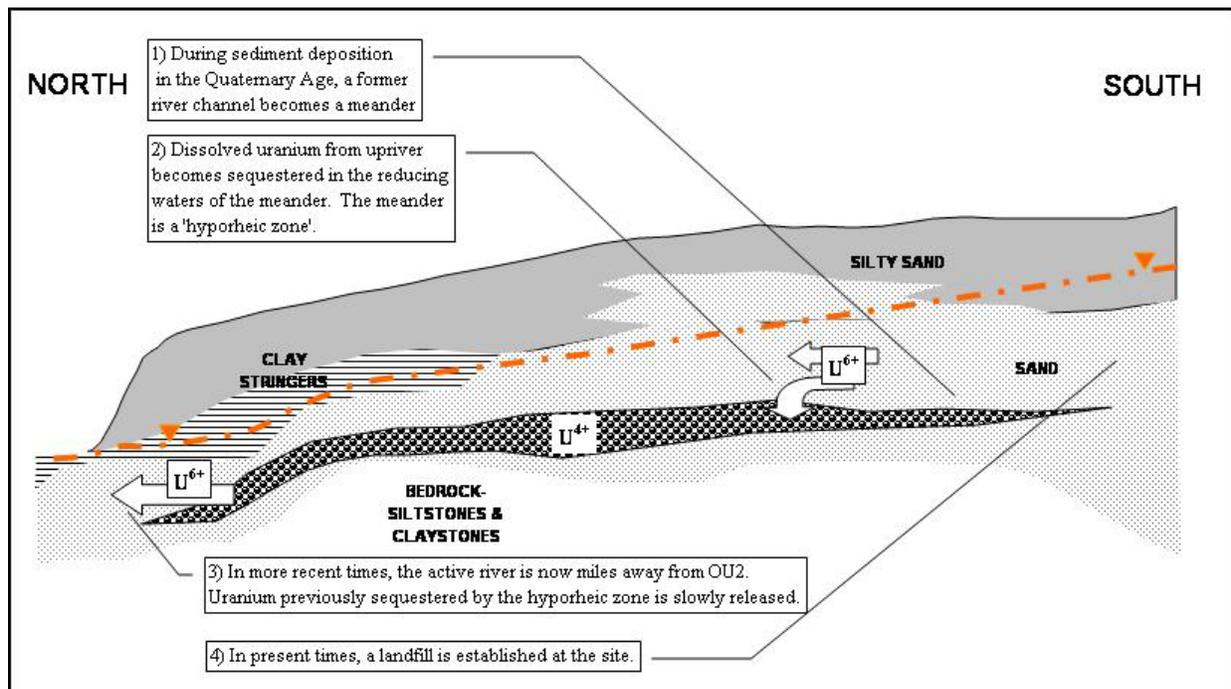


Figure 4. Conceptual Diagram of Uranium Sequestration Beneath OU2

uranium-bearing materials. Several lines of evidence indicate that natural uranium occurring in streams has been preferentially deposited beneath OU2 in the geologic past.

In essentially all geologic environments, the most important oxidation states of uranium are (4+) and (6+). Uranium will exist in the (6+) oxidation state in oxidizing environments and in the (4+) state in more reducing environments. U^{6+} species dominate in oxidizing environments. U^{6+} retention by soils and rocks in alkaline conditions is poor because of the predominance of neutral or negatively charged species. U^{4+} species dominate in reducing environments. U^{4+} tends to form low-solubility precipitates that commonly control U^{4+} concentrations in groundwater [15].

There are several environmental parameters that affect uranium migration. These parameters include redox condition, pH, ligand concentrations (carbonate, fluoride, sulfate, phosphate, and dissolved carbon), iron-oxide concentrations, and uranium concentrations. The most important factor controlling uranium solubility in a geosystem is thought to be redox potential [16, 17]. Redox potential is important because it most directly relates to the valence state of the uranium ion; whether it exists in the more mobile U^{6+} state or the less mobile U^{4+} state.

Uranium ions have weak charge bonds, so in water with high ionic content, other ions gain bonding sites at the expense of uranium, causing its mobilization [18, 15]. Uranyl acetates, carbonates and sulfates are among the most soluble complexes [17]. Uranyl nitrates are moderately soluble, while uranyl oxides, hydrides and carbides are relatively insoluble. Complexes with sulfate, fluoride, and chloride are potentially important uranyl species where concentrations of these anions are high. However, their stability is considerably less than the carbonate and phosphate complexes [15]. In the presence of sedimentary organic or humic

substances, uranium precipitation is believed to be the result of uranium reduction to form insoluble precipitates, such as uraninite. USEPA reports that UO_2^{2+} -phosphate complexes can be important in aqueous systems with a pH between 6 and 9 where the total concentration ratio of PO_4 (total) / CO_3 (total) is greater than 0.1 [15].

Alternate Hypothesis: Disposal of Refined Uranium

One alternate hypothesis is that aircraft components or weapons-training mock-ups containing depleted uranium components were disposed at OU2. Isotopic ratio analysis supports the conclusion that the uranium measured at OU2 is naturally-occurring. As shown in Table II, the U-238:U-235 ratio is consistent with standard published ratios for water-transported uranium [19, 20]. The results of the pCi:µg ratio calculations using USEPA conversion factors indicate that OU2 samples fall within a range of 0.68 to 0.71 pCi/µg. This is within the recognized range of 0.67 to 1.5 pCi/µg for natural uranium sources as presumed by USEPA [14]. Furthermore, the calculated values are consistent from all sample locations (wells, surface water, and sediment).

Table II. Uranium Activity Ratios in OU2 Media

	Natural Uranium	Depleted Uranium	Commercial Reactor Uranium	Weapons-grade Uranium	OU2 Ground-water	OU2 Surface water	OU2 Sediment
Activity percentages							
U-234	48.8%	28%	79%	97.4%	52.7%	52.6%	51.8%
U-235	2.3%	1%	4%	2.6%	2.4%	2.7%	2.3%
U-238	48.8%	71%	17%	0.01%	44.9%	44.7%	45.9%
Activity ratios							
U234: U238	1	0.4	4.6	9740	1.17 ± 0.03	1.18 ± 0.09	1.13 ± 0.11
U238: U235	21.1	71	4.3	0.004	19.6 ± 6.4	16.8 ± 8.8	19.6 ± 23.4

Alternate Hypothesis: Disposal of Uranium Ore

An alternate hypothesis is that the uranium found at OU2 resulted from the disposal of ore samples. Evidence supporting this theory includes the fact that some bags of ore samples were found in a building at Lowry. But several lines of evidence discount this theory. Virtually all commercial uranium ore deposits are Eocene-Age or older (more than 30 Ma) and thus always contain ingrown radium and thorium. Most of the radioactivity in uranium ore, some 85%, comes from the in-grown radium and thorium. But almost no radium or thorium is found at OU2 because the soils of the water-table aquifer and the natural uranium deposited in it are Holocene Age (less than 0.01 Ma) and thus too young for radium in-growth. Also, there is no evidence of the other ions that would be expected where uranium ore is disposed, such as excess sulfate, aluminum, manganese, nitrate, iron, or selenium.

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Radium and thorium account for 85% of the radioactivity in uranium ore and radium would be soluble in reducing groundwater water, but there appears to be almost no radium present at OU2. This is because the uranium minerals in the paleochannel are Holocene-Age and too young for radium in-growth, and the uranium has been transported in the oxidized watershed while the radium, which is mobile under reducing conditions, could not have been.

Accessory minerals that would co-occur with ore are not present. Ions of radium, arsenic, nickel, aluminum, manganese and iron co-occur in U tailings sites and are not seen at OU2.

Alternate Hypothesis: Paleochannel Sequestration of Natural Uranium

We believe that the most plausible explanation for the observed downgradient increases in uranium in site groundwater is the presence of a paleochannel beneath the eastern edge of OU2 that had sequestered uranium from the upgradient drainage system in the past. The transition to oxidizing conditions in portions of the channel allows the formerly precipitated uranium species to re-mobilize, hence increases the uranium concentrations downgradient.

Paleochannels may exhibit geochemical behavior identical to that of modern stream hyporheic zones, and in fact may be their historical analogues. Hyporheic zones are locally-reducing environments found in modern braided stream systems [21]. Hyporheic zones are known to be responsible for both the sequestration and mobilization of metal species [22, 23]. In hyporheic zones, oxygenated water from streams is carried below and away from the stream sediments and into the underlying aquifer where oxygen is depleted. The mechanism for concentration of the natural uranium is that streams carry dissolved hexavalent uranium through the oxidized water table aquifer until it encounters reducing conditions of a hyporheic zone, where the uranium was precipitated and deposited as less-soluble U^{4+} minerals [24].

Because of the soil texture difference in paleochannels compared to surrounding sediments, hydraulic conductivity (and thereby, groundwater flux) can increase by orders of magnitude. The much higher flow through these zones can redirect surface water into the subsurface, where chemical conditions are markedly different. Evidence that natural uranium was sequestered and released in the paleochannel includes: a) redox conditions and uranium concentrations correlate with the axis of the paleochannel, b) redox conditions are seldom at equilibrium in natural environments and range from oxidizing to reducing beneath OU2, and c) hyporheic zones both sequester and release heavy metals depending on changes in local geochemistry.

High levels of dissolved uranium were observed both upstream and downstream of OU2. High dissolved uranium correlates with the axis of the paleochannel rather than the location of the landfill. Excess uranium is naturally occurring and being released from paleochannel sediments.

Based on the results of this study, it is thus concluded that the elevated uranium levels measured in OU2 groundwater, surface water, and sediment are due to the high dissolved uranium content in the regional watershed and not due to anthropogenic causes at OU2.

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