Hydrogeologic Modeling and the Development of Preliminary Remediation Goals for the Guterl Specialty Steel Site, New York - 15431

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ABSTRACT

The Former Guterl Specialty Steel Corporation Site (Guterl Site) is located 32 kilometers (20 miles) northeast of Buffalo, New York, in Lockport, Niagara County, New York. Between 1948 and 1952, up to 15,875 metric tons (35 million pounds) of natural uranium metal (U) were processed at the Guterl Site. The resulting milling dust, shavings, thermal scale, and associated on-site land disposal contaminated both the facility and soils. These combined manufacturing and disposal processes promoted the development of a large-scale uranium plume in the groundwater underlying the site.

Site soils are anthropogenic fill and re-worked, glacially-derived sediments that vary between 0.6 meters (m) and 3 meters (2.0 to 10.0 feet [ft]) in thickness. These soils blanket a highly permeable weathered and fractured zone in the upper Lockport Dolostone bedrock. Aerial recharge through the thin soils flushes soluble constituents to groundwater, as exemplified by variations in specific conductivity and groundwater levels in the bedrock during precipitation events. Seasonally higher groundwater levels also promote contact with uranium contamination in deeper soil, thus complicating the transport characteristics of the site (e.g., oxidizing metallic uranium in the soil that increases leach rates).

The Seasonal Soil (SESOIL) compartment model was used to simulate a series of site soil conditions and predict the leaching of uranium to groundwater. The SESOIL input included physical soil characteristics, uranium profiles common in five soil areas, climate data, and an assumed oxidation-state (or chemical valence) for metallic uranium in the soil. The iterative modeling process evaluated site-specific variables to achieve a final deterministic set of input that was applied to the five soil areas. The leaching simulations estimated time-dependent influx rates for uranium, which were input to a numerical groundwater flow (MODFLOW) and transport (MT3DMS) model.

The combined modeling effort produced spatially and temporally variable transport conditions that were simulated over a 1,000-year period. The leaching models predicted a preliminary remediation goal (PRG) for uranium of 11 milligrams per kilogram (mg/kg) that is protective of groundwater. However, this low uranium PRG has field implementation concerns, so a modified direct-exposure PRG of 70 mg/kg was developed and evaluated in the flow and transport model. The primary difference in the PRGs is the longevity and location of the uranium plume, which will affect the remedial costs and potential stakeholder acceptance.

INTRODUCTION

From 1948 to 1956, the former Guterl Specialty Steel Corporation processed up to 15,875 metric tons (35 million pounds) of uranium (U) ingots in Lockport, NY, which is located approximately 40 kilometers (25 miles) northeast of Buffalo, NY. Soil contamination from on-site land disposal of metallic dust, shavings, oxide scale, and pickling fluids derived from billet heating and milling led to contaminated soil areas and groundwater conditions in the underlying
carbonate bedrock aquifer (Figure 1). These operational sources for contamination appear
evident in historical aerial photographs of the site and are integral to remedial alternative
evaluations and remedy selection.

Figure 1. Uranium Contamination in Soil and Groundwater
METHOD

Site hydrogeology

The site is underlain by an anthropogenic sandy silt fill, re-worked native soil, and a native clayey silt soil that together overlie the Lockport Dolostone. The fill material and native soil are prevalent throughout the operational areas of the site and ranges from 0.06 m to 2.8 m (0.2 to 9.25 ft) in thickness. Hydraulic conductivity (K) of the native soil varies around $1 \times 10^{-5}$ centimeters per second (cm/s) and has documented poor yields [1]. The fill and reworked soils are not hydraulically characterized due to thinness, yet appear more porous and permeable in boring samples due to coarser-grained fractions and disturbance [1]. This thin and relatively permeable soil layer provides a uranium source for the observed groundwater plume.

The upper 3 m to 4.5 m (10 ft to 15 ft) of the Lockport Dolostone is a highly weathered and fractured preferential flow zone exhibiting a horizontal K ranging from $7.1 \times 10^{-5}$ cm/s to $0.089$ cm/s; K arithmetically and geometrically averages $1.1 \times 10^{-3}$ cm/s and $4.9 \times 10^{-3}$ cm/s, respectively. This zone has an estimated effective porosity of 0.09 [2]. Groundwater levels in the shallow bedrock are within 0.6 m to 2.4 m (2 ft to 8 ft) of grade and fluctuate up to 1.2 m (4 ft) seasonally, which can result in groundwater contacting the uranium impacted soils and fill, as illustrated in Figure 1. Groundwater in the bedrock flows mainly southeasterly under a 0.06 m/m (ft/ft) gradient towards the Erie Canal.

A deeper groundwater bearing zone occurs from 9.1 m to 12 m (30 ft to 40 ft) below grade and exhibits a K range from $3.0 \times 10^{-7}$ cm/s to $1.0 \times 10^{-2}$ cm/s, which indicates a wider variability in fracturing. Groundwater levels in the deep bedrock are within 0.9 m to 10.7 m (3 ft to 35 ft) of grade and produce a more variable flow field that also has a southeasterly gradient of 0.09 m/m (ft/ft) [3].

Below these zones, the Lockport Dolostone becomes argillaceous and transitions conformably into the Rochester Shale, which acts as a lower aquitard that lies approximately 13.7 m (45 ft) to 20.4 m (67 ft) below ground surface. Observations of this transitional facies (e.g., rock quality designation [RQD] and bedding) do not indicate significant water bearing zones at depth.

Groundwater predominantly flows to the southeast across the site toward the Erie Barge Canal, but has a localized westerly vector due to pumping stresses from a bedrock quarry just west of the site. Groundwater in the upper zone is generally higher than the lower zone, indicating that vertical flow barriers (e.g., competent bedding layers) are common. This vertical variability is demonstrated by uranium (U) distributions, where the upper zone has three-times greater concentrations than the deeper zone. Generally, the deeper zone is not impacted with U until nearer the Erie Canal. Figures 1 and 2 together depict the shallow and deep groundwater uranium plumes exceeding the United States Environmental Protection Agency (USEPA) maximum contaminant level (MCL) of 30 micrograms per liter (µg/L) superimposed on the shallow groundwater potentiometric surface map, groundwater flow paths, and uranium-impacted soils.

Site contamination

Uranium contamination at the site is typically present as uranium dioxide (UO₂) or triuranium octoxide (U₃O₈) in soils; UO₂ slowly converts to U₃O₈ at ambient air temperatures [4]. Uranium in UO₂ is present in the reduced, tetravalent (U⁴⁺) form, which has exceedingly low solubility in water (approximately $1 \times 10^{-26}$ µg/L at pH 7) [5]. Uranium in the U₃O₈ state is present as both
U$^{4+}$ and oxidized U$^{6+}$ valence states, as stochastically noted: $(2\text{U}^{6+})\text{U}^{4+}\text{O}_8$. U$_3$O$_8$ also is known for low solubility in water, yet can vary with redox-sensitive species (e.g., iron and manganese), pH, and the presence of calcium, carbonates, and humic substances. Aqueous chemistry variations can increase the U$^{6+}$ solubility more readily than U$^{4+}$ and produce U concentrations that exceed the groundwater screening level of 30 µg/L. Carbonate ions, in particular, form complexes with uranium and increase its solubility and mobility [6]. Since the Lockport Dolostone is a CaMgCO$_3$ based Silurian bedrock, oxidized uranium will speciate into mobile uranyl-carbonate in the aquifer [2].

The on-site pH ranges from 6.6 to 11.1 and reduction-oxidation (redox) conditions vary from -285 millivolts (mV) to 192 mV, which is a range that can lessen uranium mobility. However, the presence of high sulfate in the bedrock groundwater mitigates the lower redox conditions and thus hexavalent uranium still is promoted in the aquifer [7]. A maximum uranium solubility was geochemically modeled at 100 milligrams per liter (mg/L) [2].

Routine groundwater sampling conducted since 2009 produced results consistent with previous Remedial Investigation (RI) sampling (2005-2006), which shows natural isotopic signatures in a mostly dissolved state (i.e., filtered and unfiltered samples yield the similar results). The uranium concentrations in most wells show steady-state conditions, with only two wells showing upward trends.

**Groundwater exposure**

Site groundwater is not used as a drinking water or industrial source, yet other exposure pathways exist and land-use uncertainties in developing this potable water supply raises concerns due to the large-scale, above-MCL (30 µg/L) contamination in site groundwater (Figure 1). The Erie Canal is 91 m (300 ft) southeast of the site and excavated into bedrock along the reach adjacent to the site. The potential influx of contaminated site groundwater to the canal is a concern because the emergency drinking water intake for the City of Lockport is located across the canal from the Guterl site [8, 9]. The hydrogeologic conceptualization of the flow system shown on Figure 2 provides a basis to estimate the risk of exposure from the ingestion of uranium released from the Guterl Steel Site.

The Feasibility Study (FS) for the site includes the modeling of soil-based uranium leaching to groundwater and the transport of the leachate to the Erie Canal. A potability (geochemical) analysis of the site groundwater indicates it qualifies as a USEPA Class IIB groundwater (or an unexploited [potential] drinking water source). Consequently, the remedial alternatives for the Guterl site soils and groundwater must account for the long-term potential that site groundwater can be a drinking-water resource. This two-phased analysis coupled an uranium leaching model with a groundwater flow and contaminant-transport model of the site to assess optimal source and plume remediation alternatives. The period of protection is 1,000 years, so a short-to medium-term solution is desired.
RESULTS

Soil-source characteristics

The metallic U processing and associated plant operations indicate the soil was predominantly impacted with uranium in the $U^{4+}$ valence state, which is solubility limiting and tends to precipitate under reducing conditions in groundwater. The nearly 60-year aerial exposure of the impacted soils has promoted oxidizing conditions that increased the mobility of uranium by converting $U^{4+}$ to mobile $U^{6+}$ species. This is observed through the persistent groundwater plume of dissolved U. Once in the carbonate aquifer, reduction-oxidation (redox) conditions (i.e., iron redox couples, nitrate and bicarbonate concentrations) are favorable for uranium migration in groundwater. Where nitrate exceeds 0.5 milligrams per liter (mg/L), uranium exceeds the groundwater screening level, or MCL of 30 µg/L [3].

Soil-source modeling input

The site-wide soil impacts were modeled using SESOIL by first dividing the site into five areas of similar hydrogeology and soil impacts (Figure 1). Soil properties were derived directly or indirectly from Guterl Site data and SESOIL guidance [2, 3, 10]. Since the half-lives for uranium are long, only advection, dispersion, and adsorption were modeled for metallic uranium (i.e., first-order decay was ignored).

Site-specific and calibration-derived SESOIL input parameters for the contaminated fill include the following variables:

- Hydraulic conductivity of the five impacted soil zones ranged between $2.8 \times 10^{-3}$ cm/s and $5.7 \times 10^{-3}$ cm/s to reflect reworked/disturbed sandy silt textures.
- A soil disconnectedness index of 4 that is indicative of mixed soils (silty sand).
• An effective porosity of 0.25 was input to account for the mixed soil texture [3].
• A soil moisture of 15% is indicative gravity drained soil zone.
• A bulk density of 1.30 grams per cubic centimeter (g/cc) (81.2 pounds per cubic foot [lbs/ft³]) reflects the disturbed sandy silt soils [3].
• Uranium solubility was estimated using site geochemical profiles at 100 mg/L.
• Monthly climatic data for Lockport includes:
  o Temperature
  o Cloud cover
  o Relative humidity
  o Short wave albedo
  o Evapotranspiration
  o Precipitation
  o Storm length
  o Number of storms
  o Length of rainy season
• An average groundwater recharge of 0.38 meter per year (m/yr) (15 inches per year) was calculated via calibration and matches a rate of 0.39 m/yr determined by a previous radiologic exposure model [2].

Uranium adsorption (or soil distribution) coefficients (Kd) are a highly sensitive parameter in the SESOIL model and were estimated using two methods. First, a twenty-four hour batch test was performed using uranium spiked groundwater of differing concentrations (380 ug/L to 25,665 ug/L) shaken with site soils having a narrow range of uranium concentrations [3]. Secondly, eight undisturbed contaminated soil samples were exposed to simulated precipitation in the form of synthetic rain water using the Synthetic Precipitation Leach Procedure (USEPA Method 1312) to define actual leaching concentrations from on-site soils.

The tests produced the following Kd results:

• Native reworked uncontaminated soil: 1247 and 1452 milliliters per gram (mL/g), with a three point average of 1356 mL/g
• Contaminated soil/fill: 5 to 97 mL/g, with a nine point average 39 mL/g.
• Lockport Dolostone (gravel-sized grains): 0.22 mL/g.
• Undisturbed native soils: 1,052 to 95,667 mL/g, with a 24-point average of 17,699 mL/g (USEPA Method 312 dataset).

The Kd values for U-impacted fill or reworked native soils are lower than the undisturbed native soils due to the coarser texture of the fill and reworked/disturbed soils (i.e., boring logs indicate a more sandy texture than native soils). The Kd values are also lower for the high-concentration solute tests due filled adsorption capacity, whereas the native soil exposed to lower solute concentrations produced higher Kd values due to continued availability of exchange sites throughout the test. This condition is important when evaluating the potential for leaching from high U-concentration soils, where interstitial pore-water concentrations would be high and leaching rates that may advance faster than ambient soil data would predict.

The concentrations of uranium in soil profiles of the five soil-source areas were calculated using the following process:
• Uranium-concentration data from each soil area was vertically divided into up to 15-cm (6-in) thick, vertical compartments.
Statistical profiles were generated to describe each the vertical compartment and thus a detailed profile of each soil area.

The average vadose zone thickness for each soil area was calculated from site data (Table 1).

Each soil area was represented in SESOIL as three to four main layers subdivided into 15-cm (6-in) sublayers to match the soil-concentration dataset.

The main layers varied between 15 cm (6 in) to 60 cm (24 in) in thickness based upon soil texture, concentration ranges, and proximity to groundwater (outflow boundary).

Table I provides an example summary of the soil-concentration input for Soil Area 2; similar input tables were created for all five soil areas.

Table I. Area 2 Contaminated Soil Profile for SESOIL

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<th>Bottom inches</th>
<th>Sample Count</th>
<th>Average-U238 pCi/g</th>
<th>Min_U238 pCi/g</th>
<th>Max_U238 pCi/g</th>
<th>Total U mg/kg</th>
<th>SESOIL Layer Number</th>
<th>Sublayer Number</th>
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<th>Sublayer Bottom inches</th>
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Notes:
1. Use sample depth to water of 5.79 ft (69.5 inches) as SESOIL model bottom depth
2. Soil U activity exceeds PRG soil to be excavated
3. No excavation

Rules:
1. If the average soil U activity exceeds PRG-GW OF 3.66 pCi/g (11 mg/kg Total U), excavate and replace with backfill.
2. Backfill U = background 0.74 pCi/g U (2.2 mg/kg Total U)
3. All soils overlying the lowest depth at which average soil 238U activity exceeds the PRG will be excavated.

Once all the input to SESOIL was complete, output results (leachate concentrations and flow rates over time) were input to the numerical groundwater flow and contaminant-transport model to assess the fit with the observed system. This initial SESOIL modeling used a Kd range that reflected the native undisturbed soil conditions, or values from 1,052 to 95,667 mL/g, with an average of 17,699 mL/g. The results did not produce appreciable uranium leachate flow to groundwater, so the conditions that created the observed plume were not simulated.

An iterative process ensued using the whole range of site-specific Kd values to best estimate leachate concentrations that would produce the observed plume. To bracket the target concentration, the dilution factor for the bedrock aquifer was estimated using the numerical flow and transport model and the USEPA Soil Source Leaching (USEPA SSL) equations [11]. The dilution factor varied between 2.7 and 4.0; a value of 4.0 was used to estimate the required leachate concentration of ~1,000 µg/L. The Kd value that produced this leachate for most soil areas is 91 mL/g, which falls into the low range of measured site-specific Kd values. This indicates that 1) the impacted fill and reworked fill/native soil mixtures liberate U more than native undisturbed soils and 2) the thin vadose zone (< 1.5 m or <5 ft) has little capacity to redistribute uranium while it transports through the soil profile.
Soil-source modeling results

The final SESOIL models for each soil area and the site-wide saturated-zone models were then used to assess the following three remedial alternatives for groundwater:

1. **No Action** - the soil impacts are not remediated and the site continues to release to the environment.
2. **Soil Excavation to a Soil Preliminary Remediation Goal (PRG) Protective of Groundwater (SOIL PRG-GW Scenario)**
3. **Soil Excavation to a Soil PRG Protective of Construction Workers with Groundwater Ingestion (SOIL PRG-CW Scenario)**

The SESOIL output (or unit area flow of uranium-impacted leachate to the groundwater) was then assessed to determine the degree of contamination (mass) and the length of delivery time (longevity) to the aquifer. These variables govern the amount of uranium mass that will be entered into the numerical flow and contaminant-transport model to assess aquifer response and potential remedial alternatives.

The Alternative 1 (No Action) simulation allows the current site conditions to persist, so the groundwater continues to receive uranium leachate for over 700 years, with the following peak concentrations and times:

- **Soil Area 1:** 28,370 μg/L at 265 ± 5 years
- **Soil Area 2:** 7,682 μg/L at 255 ± 5 years
- **Soil Area 3:** 5,298 μg/L at 235 ± 5 years
- **Soil Area 4:** 35,280 μg/L at 15 ± 5 years
- **Soil Area 5:** 281 μg/L at 235 ± 5 years

The resulting plumes persist above the U MCL for 700 years in the shallow groundwater zone and over 1,000 years in the deep zone due to the large mass of uranium that is transferred from the soil. Consequently, Alternative 1, No Action, is not protective of human health and the environment.

The Alternative 2 (SOIL PRG-GW Scenario) simulation was designed to predict a leachate concentration that would promote the natural attenuation of the existing plumes and achieve the MCL. This objective required the iterative SESOIL modeling of soil remediation goals that would limit uranium leachate concentrations to values that can be diluted to 30 μg/L in the aquifer. The USEPA SSL equations were used to estimate a starting soil goal for SESOIL to test; the results ranged between 9 mg/kg and 19 mg/kg total uranium for the five soil areas. The soil sampling statistics for each 6-in vertical compartment in SESOIL were then recalculated to reflect the removal of soil concentrations (from highest values downward) to achieve an average soil concentration of 9 mg/kg or 19 mg/kg per 6-in compartment. This range was iteratively narrowed using SESOIL to obtain a single uranium PRG of 11 mg/kg total U for all soil areas (i.e., one remedial goal for all five soil areas). This averaging process indicates that some isolated U concentrations would remain in the soil above 11 mg/kg in each compartment.

The SESOIL simulations resulted in the following leachate conditions for total uranium, which are then diluted to below the MCL in the aquifer:

- **Soil Area 1:** 77 μg/L at 25 ± 5 years
• Soil Area 2:  95 µg/L at 15 ± 5 years
• Soil Area 3:  36 µg/L at 140 ± 5 years
• Soil Area 4:  37 µg/L at 25 ± 5 years
• Soil Area 5:  37 µg/L at 25 ± 5 years

SESOIL predicts the leachate will persist above the MCL for 200 years, although the existing plumes will attenuate to below the MCL in 45 years in the shallow groundwater zone and 115 years in the deep zone. Consequently, Alternative 2, SOIL PRG-GW Scenario, is protective of human health and the environment after a period of 115 years of land use controls.

Alternative 3, the Soil PRG-CW Scenario, was developed using the Residual Radiation Model (RESRAD) [12] that estimated a construction-worker exposure PRG of 69 mg/kg for total uranium (as converted from uranium-238). This PRG is based on summed dose-to-source ratios from exposure to each site contaminant (isotopic radium, thorium, and uranium). The worker is assumed to incidentally ingest 0.2 liters per day (6.8 fluid ounces per day) of contaminated groundwater at a RESRAD-calculated peak concentration of 895 µg/L. The total pathway exposure to the isotope-specific PRGs, including groundwater, equates to the 25 mrem/yr exposure limit [2].

The isotopic PRGs were then used in a sum-of-ratios (SOR) analysis to designate soil samples as “in need of remediation” in the database. Consequently, new uranium statistics in the 6-in soil compartments for SESOIL did not directly reflect 69 mg/kg, but the influence of radium and thorium in the removal of samples from the soil dataset (i.e., some uranium values below 69 mg/kg were removed to account for collocated impacts). It was assumed that all soils overlying the deepest sample greater than 69 mg/kg in each boring would be excavated, so that after excavation the average of each 6-inch interval was not to exceed the Soil PRG-CW. All the soil values that were removed from the dataset were then replaced with background-level U values of 2.2 mg/kg to reflect the backfilling of remedial excavations with native materials.

The SESOIL models of Alternative 3 produced uranium leachate concentrations that will exceed the MCL for over 490 years. The leachate concentrations are expected to peak in the future at the following concentrations and timeframes:

• Soil Area 1:  71 µg/L at 15 ± 5 years
• Soil Area 2:  306 µg/L at 25 ± 5 years
• Soil Area 3:  607 µg/L at 85 ± 5 years
• Soil Area 4:  36 µg/L at 135 ± 5 years
• Soil Area 5:  279 µg/L at 235 ± 5 years

The contaminant-transport modeling of this SESOIL output indicates the site groundwater will remain impacted above the MCL for 435 years in the shallow groundwater zone and over 655 years in the deep zone. This attenuation timeframe is not uniform for all soil areas; leachate from soil areas 1, 2, and 4 will reduce to concentrations that will achieve the MCL within 100 years. Consequently, Alternative 3, SOIL PRG-CW Scenario, is protective of human health via radiation exposure and will require a period of 655 years of land use controls.

For guiding soil excavation during field implementation of these PRGs, the 11 mg/kg or 69 mg/kg PRGs will be treated as an exposure concentration or never-to-exceed concentration, and thus soil areas exceeding the PRG would be excavated. This conservative approach would lower the actual uranium averages for the 6-inch vertical soil compartments, which indicates
these SESOIL-based leachate values represent an upper bound of the period of performance (i.e., estimated number of years) to achieve the MCL. To exemplify the SESOIL outputs, the results for soil area 2 are presented in Figure 3; the leachate-concentration versus time curves indicate the effects of the two PRGs with respect to the No Action Alternative.

CONCLUSION

The groundwater contamination at the Guterl site is above the drinking water standard of 30 μg/L for uranium and discharges to the nearby Erie Canal at concentrations near and above this standard. Soil and groundwater contaminant assessments indicate that the uranium in soils provide a source for an existing uranium plume in groundwater. The selection of a remedial alternative for soil will affect the remedial strategy for groundwater.

The soils alternative that is most protective of groundwater also presents implementation challenges; the uranium PRG of 11 mg/kg is about five-times background and not a value readily observed using field-survey instruments (e.g., sodium-iodide detectors). The alternative that is protective of construction-worker exposure to site soils (69 mg/kg of total U) is more
implementable (field detectable), but predicted to prolong the existing groundwater contamination for hundreds of years. Figure 4 presents the waning plume condition for Alternative 2 and maximum plume condition Alternative 3; this comparative figure indicates the temporal and spatial considerations that will need to be addressed for either alternative (i.e., as the soil areas differentially leach uranium, the plumes and monitoring arrays will change too).

An interim remedial strategy may include the remediation of soils to the construction worker PRG, followed by a groundwater monitoring period to assess whether model predictions were too conservative and the natural attenuation of the uranium is more viable than estimated (i.e., will not take hundreds of years to meet MCLs). This approach will require an optimized monitoring well array in areas where uranium residuals are greater than 11 mg/kg, as well as a property boundary array to monitor the potential for additional off-site migration.

Figure 4. Predicted Uranium Plumes
REFERENCES


