Final Report

An Investigation of Contaminants and Contaminant Migration at the Hanford Site

November 2003

Hanford Action of Oregon

funded by
Resolve's Citizens' Monitoring and Technical Assessment Fund Round 3

Introduction

The vastness in both size of the U.S. Department of Energy's nuclear weapons manufacturing site at Hanford, and the extent and diversity of its contamination, combined with a lengthy history of poor waste management practices and annual cleanup budget shortfalls, has created an ongoing need for external citizen sampling of contaminants of Hanford origin. History has consistently shown that modeling and existing USDOE monitoring alone have been less than adequate at both comprehensively identifying contaminated areas around the Hanford site and at predicting mobility of contaminants through soils. Yet rapid migrations of dangerous chemicals and radionuclides place groundwater and the Columbia River at risk, and greatly encumber cleanup. There is a need to conduct science in the public interest that considers gaps in monitoring and preferential pathways of contamination, and samples these locations for suspect species.

This report summarizes the findings of an investigation undertaken by Hanford Action of Oregon whereby soil and vegetation sampling was conducted at select site locations in search of suspected pathways of radiological contamination.

Background

Uranium 233 Production Wastes

Uranium 233 was generated in large quantities at Hanford as part of the U.S. government's mini-nuke or tactical nuclear weapons program. The wastes from that production activity were discharged to the environment from pipes emanating from the reactor to the river or possibly dumped from barges. However, the extent of the U233 contamination to the river has not been clear. U233 and its daughter products of decay are of concern.

U233 decays to Radium 225. Both elements are of concern to the welfare of the regional environment and salmon stocks. U233 is insoluble and long-lived. So any U233 in the riverbed, if not retrieved, will remain there decaying for a long time. All the while, the U233 is generating Ra225 which is soluble, mimicking calcium (like strontium 90) and taken up readily by aquatic animals, collecting in bone and shell. If found to be accumulating in the Columbia River's Hanford Reach, then the health of the Reach and its aquatic life may be in jeopardy.

Previous investigation by the Government Accountability Project revealed evidence of U233 production contamination in the riverbed with the finding of a widespread presence of Europium 152, a 'fingerprint' indicator of U233 production. GAP tested riverbed water and found that the U233 was in colloidal form adhering to sediments and indeed decaying to soluble Ra225. Direct measurements of colloidal U233 in Hanford Reach sediment had yet to be made. Such measurements are one of the subjects of this investigation and report.

300-Area

Another subject focuses on Hanford's 300-area which harbors immense quantities of buried radioactive waste in massive dumpsites and trenches, many without any barrier between wastes and soils. More nuclear wastes, including some imported from off-site locations, continue to be disposed of at Hanford's 300-area. Currently, the U.S. DOE plans to limit cleanup of the 300-area to industrial use standards despite public concern that the buried wastes are migrating through the soils, creating potential health hazards.

K-Basins Area

Ten years ago old corroded spent fuel rods buried in Hanford's K-Basins storage pools along the Columbia River were found to be leaking highly radioactive material, including plutonium, into the storage pool water. Because of the risk of criticality, USDOE made safe removal of the spent fuel a top safety priority for the nation. However, recent evidence of leakage of the contaminated water from the basins has prompted concern as to the extent of underground contamination and whether or not it has found a pathway to the river. The K-Basins area near KW and KE Reactors is off-limits to the public so there has not been previous public interest science conducted there.

Purpose of Investigation

The purpose of this investigation was to collect and analyze soil and vegetation samples from specified areas at Hanford considered likely to be contaminated with radioactivity, to identify possible contaminant pathways and accumulations that could pose a threat to the health of the local environment and the adjacent Columbia River.

A significant portion of the investigation was a search for radiological anomalies in Hanford's F-slough of the Columbia River in order to obtain the first direct measurements of artificial uranium-233 (U-233) in the riverbed of the Hanford Reach National Monument where eighty percent of the Columbia River's salmon spawn.

In addition, the investigation sought to flag a possible groundwater pathway from the KE-Basin to the Columbia River by mapping Strontium 90 in deep-rooted vegetation, and to check vegetation uptake of buried radioactive wastes in the 300-area.

Description of Investigation

An investigation was conducted of possible radioactive contamination at key sites near and along the Columbia River, in particular around Hanford's F-Slough, 300-Area and KE and KW-Reactors. The locations were chosen because of their importance as a public concern.

300-Area and K-Basins Area

Sampling of deep-rooted vegetation, namely gray rabbit brush and tumbleweed, was conducted in November 2002. Such vegetation was chosen for test because these

plants take up water through their root systems from deep underground in an arid climate to nourish the plant at the surface. In this way deeply buried contaminants may be carried to the surface where they can then be detected by collection of plant material without excavation.

This sampling effort took place on the site in a restricted area that required USDOE permission for access. That permission had been granted in accordance with a Site Access Agreement between the Government Accountability Project and USDOE that allowed scientist Norm Buske to conduct this work. The agreement was signed in March 2001. However, following the collection and preliminary reporting of the data and results below and in Attachment 2, site access was effectively revoked by USDOE. USDOE cited lack of staff resources available to provide the required official escort for Mr. Buske and to analyze sample splits.

F-Slough Anomalies

Access to the F-Slough area is unrestricted. So after the effective loss of site access in the 300 and K-basins areas, the investigation turned to sampling the F-slough area down river from F-reactor. Sediment was examined to see if U233 could be detected directly.

Approach and Methods

Part I: Sampling in the 300-Area and K-Reactors area.

Nine samples of rooted vegetation were collected altogether, namely tumbleweed and gray rabbit brush. Five rooted samples of tumbleweed were collected from the 300-Area, as well as one sample of moss with soil. One of the tumbleweed samples was used to establish background for the other tumbleweed samples. No sample was taken to establish background for the moss with soil sample. Three samples of gray rabbit brush were collected from unfenced areas northeast of KE-Reactor for analysis. One of the rabbit brush samples was used to establish background for the other gray rabbit brush collected.

The samples were collected from areas showing relatively high radioactivity using a Geiger counter. It was found that excessive time was required to achieve the sensitivity necessary for Sr-90 mapping near K-basins.

All samples were split USDOE/RL.

Following collection, the samples were dried and counted for twenty-three hours. Results reported are the root product of the absolute and above-background measurements. More detail on Methods is reported in Attachment 2.

Part II: Sampling in the F-slough.

Sampling locations near F-Slough were selected on the basis of previous work, reported in Hanford Radioactivity in Salmon Spawning Grounds and Trouble in the Columbia Riverbed , both by Norm Buske.

Five samples of fine sediments were collected from the near shore riverbed of the Hanford Reach near F-Slough and gently tumbled to remove and collect "colloidal" contaminants sticking to the sediment grains. In addition, mulberry leaves were collected from trees on shore near two of the sediment sampling locations and tested. One sediment sample, collected from Hanford Slough served as a "background" for the sediment analyses. Radiological results for the other four sediment samples and the two mulberry leaf samples are reported here.

Sediment samples were analyzed one at a time (due to limitations in equipment). For consistent analyses over time, the samples of wet sediment were sealed and refrigerated for three weeks. Sediment sample preparation consisted of first weighing and splitting each, and then screening them to extract only the sediments' finest grain and surface material including contamination. The extracted material was then evaporated, reduced to a paste, and oven-dried and weighed.

Data was collected by counting photon emissions from each sample over twenty-three hours. This was repeated twice for each sample every four days.

In reporting and analyzing the data, it was necessary to distinguish interfering emissions, including one particularly strong and potentially conflicting one originating from Pb-210 generated by U-238, and then subtract the unwanted contributions to the measurements accordingly. More method details are described in Attachment 1.

Results

Part I: 300-Area and K-Basins area samples

Each of the six samples tested positive for at least one artificial radionuclide, indicating uptake of radioactivity by deep-rooted plants. Am241, Cs137, Sr90, ThNat and UNat were found in plants growing outside of fenced areas.

Part II: F-Slough samples.

Three samples (372008, 372010, and 372012) tested positive for U-233 in the range of one picocurie/gram. The U233/Eu152 ratio of these three samples was 0.6.

That is to say, random uncertainties as evidenced by "counting uncertainty" are far less than the known but uncontrolled, systematic errors. Please note the total uncertainty is stated at four times the "±" counting error tabulated in the Results, below.

Data

Part I: 300-area data.

Sample	**Location**	**Coordinates**	**Results** pCi/Kg	**Notes**
1 gray rabbit brush	N of KE Reactor	N46deg 39.169min W119deg 35.866min	none	Used as background for gray rabbit brush
2 gray rabbit brush	N of KE Reactor	N46deg 39.248min W119deg 35.891min	**Am241** 100	
3 gray rabbit brush	NE of KE Reactor	N46deg 39.273min W119deg 35.850min	**ThNat** 100 **Cs137** 50	
4 tumbleweed	NE of 309 Bldg.	N46deg 22.068min N119deg 16.564min	**UNat** 1800 **Cs137** 60	
5 tumbleweed	NE of 324 Bldg.	N46deg 22.139min W119deg 16.439min	**Am241** 100 **Cs137** 30	
6 tumbleweed	NW of 3707F Bldg.	N46deg 22.180min N119deg 16.589min	none	Used as background for tumbleweeds
7 tumbleweed	S of 305 Bldg.	N46deg 22.326min W119deg 16.879min	**ThNat** 60 **Cs137** 30 **Unidentified beta emitter**	
8 moss & soil	E of 313 Bldg.	N46deg 22.306min W119deg 16.788min	**UNat** 4300 **ThNat** 1900 **Cs137** 90 **Sr90** trace	Tentative results. Sample collector between concrete
9 tumbleweed	E of 321 Bldg.	N46deg 22.196min W119deg 16.878min	**UNat** 220 **Sr90** 3200	

Part II: F-Slough data

Sample	**Location**	**Coordinates**	**wet/ dry**	**Results** pCi/g (dry)	**Notes**
372011 sediment	Hanford Slough HRM 25.3 west side	N46deg 35.470min W119deg 23.135min	129	none	Used as background for colloids, sediments
372014 sediment	F-Slough NW HRM 19.6 west side	N46deg 39.352min W119deg 26.140min	104	**U233** –0.13 ±0.04 **Eu152** 0.07 **Cs137** 0.29 ±0.03	
372008 sediment	F-Slough Mid HRM 20.6 west bank	N46deg 38.780min W119deg 25.182min	55	**U233** 0.56 ±0.03 **Eu152** 0.91 **Cs137** 0.18 ±0.02	
372010 sediment	F-Slough, off HRM 22.0 west side of river	N46deg 37.967min N119deg 24.655min	85	**U233** 2.22 ±0.04 **Eu152** 3.68 **Cs137** 0.95 ±0.03	
372012 sediment	F-Slough, downstream HRM23.0	N46deg 37.077min W119deg 24.617min	87	**U233** 2.29 ±0.04 **Eu152** 3.69 **Cs137** 1.12 ±0.03	
372008m leaves	F-Slough NW HRM 19.6	N46deg 38.767min N119deg 25.190min	2.9	**U233** 0.076 ±0.017 **Eu152** not detected **Cs137** not detected	
372012m leaves	F-Slough, off HRM 22.0	N46deg 37.080min W119deg 24.627min	3.8	**U233** not detected **Eu152** not detected **Cs137** not detected	

Conclusion

Detecting Sr90 in gray rabbit brush, the prevalent deep-rooted biota in the unfenced area around the K-Basins, in an effort to reveal a contaminant flow to the river was considered an unlikely find because rabbit brush does not readily uptake Sr90. However, while the Sr90 detection attempt was inconclusive, other man-made radionuclides were found in the same rabbit brush samples that warrant further investigation. This project verified underground long-lived radiological contamination and revealed mobility of the contaminants to the surface via deep-rooted vegetation. This is important to note because such 'leaks' to the environment where the radioactivity will readily spread necessitate one of two measures for prevention of spread: intensive institutional control whereby the responsible agency ensures all vegetation and surface matter is permanently purged, or extraction and cleanup of the underground contamination flows. Hanford Action of Oregon recommends the latter because it does not rely on impractical institutional control over a long time. The only real way to ensure environmental protection is to remediate the underground contamination.

All of these results point to the need for USDOE to obtain more data and launch an investigation into the likely contamination pathways at Hanford flagged by this work, and ultimately establish sufficient remediation and cleanup of both the migrating contaminants and their respective sources.

References

Attachment 1. U233 Detected in Hanford Reach Sediments, by Norm Buske November 2003

Attachment 2. November 2002 Report On-Site Sampling, by Norm Buske November 2002

Access Agreement between the Government Accountability Project and United States Department of Energy Richland Operations Office, DOE/RL document signed March 2001

Trouble in the Columbia Riverbed , by Norm Buske <radioactivist.org> 2003

Hanford Radioactivity in Salmon Spawning Grounds, by Norm Buske <radioactivist.org> 2001

Sample collection, analyses and reporting for this investigation were conducted by scientist Norm Buske, of TRAC and the Radioactivist Campaign, for Hanford Action of Oregon. This report to the Citizens' Monitoring and Technical Assessment Fund was assembled by Robin Klein for Hanford Action of Oregon and summarizes the work undertaken by Norm Buske for Hanford Action of Oregon using a Round Three grant award from the fund.

Attachment 1.

Data Report:

URANIUM-233 DETECTED IN HANFORD REACH SEDIMENTS

November 04, 2003

By: Norm Buske <search@igc.org> The RadioActivist Campaign <radioactivist.org>

The RadioActivist Campaign (TRAC) conducted this study for Hanford Action of Oregon, supported by a grant from the Citizens' Monitoring and Technical Assessment Fund.

SUMMARY:

Sediment samples were collected from the F-Slough area to explore radiological anomalies. The background anomaly was europium-152 (Eu-152) reported in HANFORD RADIOACTIVITY IN SALMON SPAWNING GROUNDS, dated August 2002.

The sediment samples were gently tumbled to remove and collect "colloidal" contaminants sticking to sediment grain surfaces, thus leaving behind irrelevant analytical "noise" of natural radioactivity within sediment particles. The main purpose of this work was to obtain the first direct measurements of artificial uranium-233 (U-233) in the riverbed of the Hanford Reach.

Five samples of fine sediments were collected from the near shore riverbed of the Hanford Reach, near F-Slough, on July 20, 2003. Mulberry leaves were collected from trees on shore, near two of the sediment sampling locations. One sediment sample, collected from Hanford Slough served as a "background" for the sediment analyses. Radiological results for the other four sediment samples and the two mulberry leaf samples are reported here.

Three samples (372008, 372010, and 372012) tested positive for U-233 in the range of one picocurie/gram. The U233/Eu152 ratio of these three samples was 0.6.

METHOD:

General sampling locations near F-Slough were selected on the basis of previous work, reported in HANFORD RADIOACTIVITY IN SALMON SPAWNING GROUNDS and in TROUBLE IN THE COLUMBIA RIVERBED. One sampling location (372012) was selected as an exact replicate. Others were selected to test and extend previous results. One sediment sample was to be collected from Hanford Slough, to serve as study background, while also checking Hanford Slough into this analytical schema.

Sediment samples were passed through a 2 mm screen, discarding large fractions irrelevant both to radiochemical processes in the riverbed and to Hanford wastes in the riverbed.

Experience has shown that the Hanford Reach riverbed is “radiologically dynamic.” That is, *dis*equilibria of radionuclides in decay chains must be addressed. In consideration of TRAC’s single in-house spectrometer, the sediment samples collected on July 17th had to be analyzed sequentially, one after the other. Therefore, the wet sediment samples were refrigerated in sealed bags for three weeks, so that subsequent analyses of the samples, one after another over one week, would not differentiate the samples appreciably.

Each wet sediment sample, of about 1500 g, was then weighed (“wet” weight tabulated, below) and split into one of two quart, square rounded bottles, filled with distilled water. The bottles were rotated horizontally at 6 rpm for 5 hours. The samples were then screened through an 0.475 mm sieve with distilled water flushing. The very fine sediments and the material thus extracted from the sediment surfaces was then evaporated quiescently to a paste and then oven dried <90 C, and weighed. That weight of dried “colloids” is the “dry” weight tabulated in the Results, below.

Each sample was counted for 23 hours in a highly stabilized sodium-iodide, photon detector, with an energy window of 5-3000KeV. Each of these initial spectra were checked to assure conformity for the work-in-progress. Then each sample was recounted for 23 hours, twice, at four day intervals. This allowed quality assurance checks that potential interferences from naturally occurring lead-210 (Pb-210), in the uranium-238 decay chain, were within bounds.

The primary interference is the strong photon peak from Pb-210 (lead-210) in the natural U-238 decay chain. That interference peak is at 46.5 KeV energy. I have to distinguish a weaker 42.4 KeV peak for U-233 analysis. The subtraction of Pb-210 involves counting the strength of Ra-226 (in the U-238 decay chain, but with most photon activity before Pb-210 removed). Then the Ra-226 spectrum (and hence Pb-210) is subtracted accordingly. The check on this is that the residual U-233 peak does appear at lower energy than the eliminated Pb-210 peak.

Radioactive decay of Pb-210 emits intense (Intensity ~85%) gamma photons at 46.5 KeV energy. The present study seeks to clearly distinguish emissions from U-233decay (at merely 13% Intensity) at 42.4 KeV energy (from relaxation of daughter, thorium-229, from its born-excited state). This distinction was made by reference to an appropriate “background” reference that included Pb-210 and then by one adjustment for the difference in Pb-210 activity between the sample and the background.

This distinction is crucial because the energy difference between detection of Pb210 decay and U-233 decay is only 4.1 KeV, to be differentiated in a spectrometer having a photopeak width defined of 9 KeV (Full Width at Half Max = 3 channels).

The acquired spectra were transformed to constant photopeak width and three acquisitions added. The stated background and other reference spectra (including potassium-40, Eu-152, and Cs-137) were then subtracted, yielding final spectra that could be counted for U-233.

The U-233 counts in this report are based on the gamma intensities listed in C.M. Lederer and V.S. Shirley's TABLE OF THE ISOTOPES, 7th ed. (1978), p. 1411, in preference to more recent reports like Brookhaven's NUDAT.

Even with the most careful technique, considerations such as these increase the uncertainty of results. In the results reported here, uncontrollable, deterministic errors likely dominate. That is to say, random uncertainties as evidenced by "counting uncertainty" are far less than the known but uncontrolled, systematic errors. Please note,

the total uncertainty is stated at four times the "±" counting error tabulated in the Results, below.

RESULTS:

U-233 = uranium-233, a transuranic, fissile product, used to nuclear weapons and other purposes. Halflife: 159,200 years.

Eu-152 = europium-152, a fingerprinting byproduct of U-233 production, resulting from neutron activation of a natural impurity in feed stocks (Eu-151). See HANFORD RADIOACTIVITY IN SALMON SPAWNING GROUNDS, pp. 9-11. Halflife: 13.5 years.

Cs-137 = cesium-137, a product of nuclear fission, either in nuclear reactors like those at Hanford or from fallout from atmospheric testing of nuclear weapons in the 1950s and 60s. Halflife: 30.2 years.

Material tumbled and rinsed off sediment grain surfaces.

Sample Number	Coordinates North 46°	West 119°	Location
372011	35.470'	23.135'	Hanford Slu, HRM 25.3 west side

wet/dry = 129 No results reported: Sample used as background for colloids from sediments.

372014	39.352'	26.140' NW F-Slu, HRM 19.6 west side

wet/dry = 104 U-233 = –0.13 ±0.04 pCi/g(dry) Eu-152 = 0.07 " Cs-137 = 0.29 ±0.03 "

372008	38.780'	25.182' Mid F-Slu, HRM 20.6 west bank

wet/dry = 55 U-233 = 0.56 ±0.03 pCi/g(dry) Eu-152 = 0.91 " Cs-137 = 0.18 ±0.02 "

372010 37.967’	24.655’ Off F-Slu, HRM 22.0, west side river
wet/dry = 85	U-233 = 2.22 ±0.04 pCi/g(dry) Eu-152 = 3.68 “ Cs-137 = 0.95 ±0.03 “

372012 37.077’	24.617’ Downstream of F-Slu, HRM 23.0
wet/dry = 87	U-233 = 2.29 ±0.04 pCi/g(dry) Eu-152 = 3.69 “ Cs-137 = 1.12 ±0.03 “

Mulberry leaves, collected near two sediment sampling locations, above.

372008m 38.767’	25.190’ NW F-Slu, HRM 19.6, by 372008
wet/dry = 2.9	U-233 = 0.076 ±0.017 pCi/g(dry) Eu-152 = nd Cs-137 = nd

372012m 37.080’	24.627’ Off F-Slu, HRM 22.0, by 372012
wet/dry = 3.8	U-233 = nd Eu-152 = nd Cs-137 = nd

“±” one standard deviation counting error. **The uncertainty & minimum detection level for this study is 4 times the listed “±” value.**

nd not detected.

Attachment 2.

Report of Hanford Site Access of 11/12/02 (80.3 KB) by Norm Buske

TRAC's data from on-site sampling at Hanford shows cleanup of 300 Area has failed.

http://www.radioactivist.org/hanfordreports.html