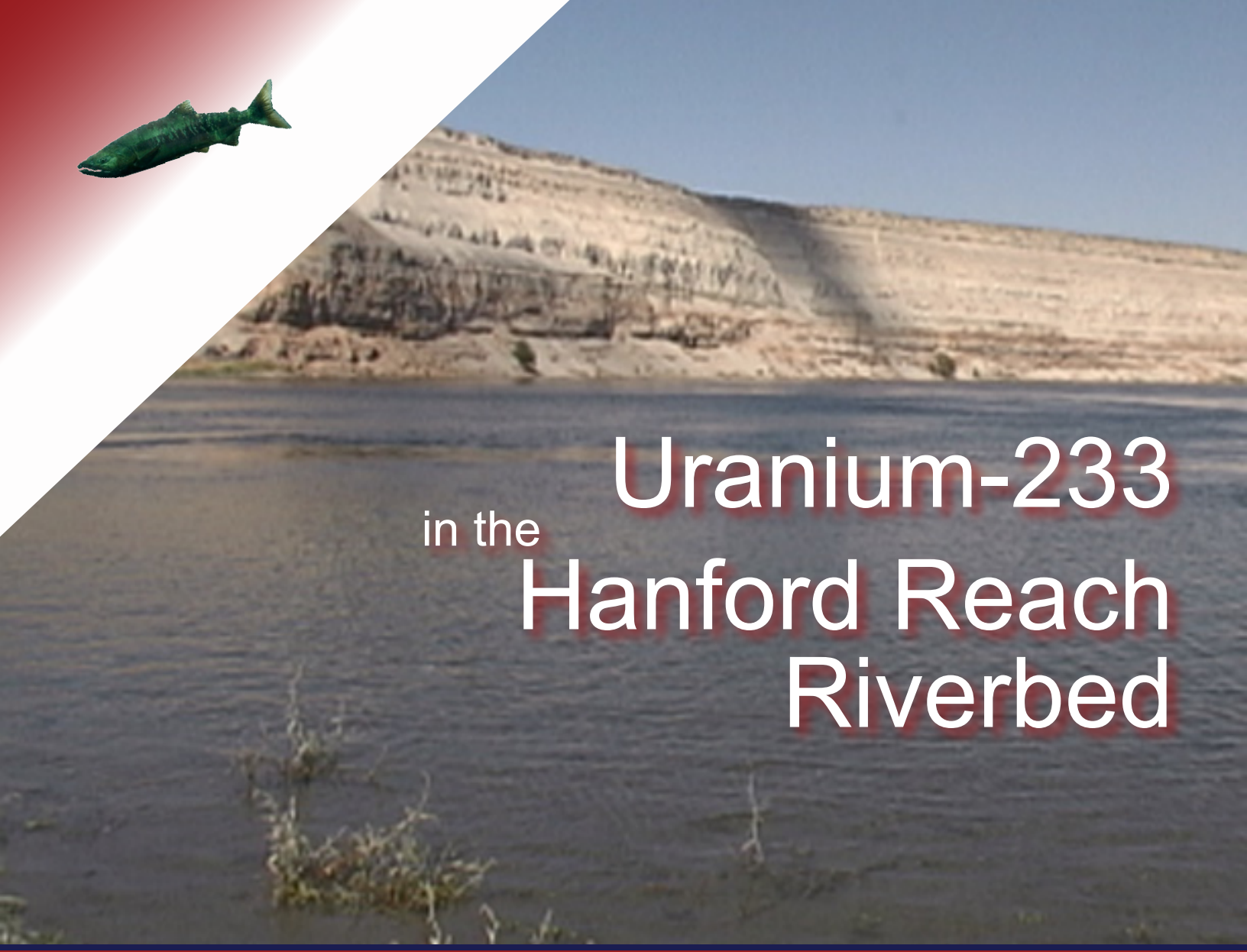

Uranium-233 in the Hanford Reach Riverbed

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☢ Contents

☢ Summary

The 50-mile Hanford Reach of the Columbia River in Eastern Washington is recognized as

- **a scenic recreation area and national monument.**
- **crucial spawning habitat for wild chinook salmon.**
- **part of the Columbia Basin irrigation project and the mid-Columbia hydro-electric power generation system.**
- **collecting radioactive and toxic wastes from old nuclear weapons production at the Hanford Site.**

The present study confirms that some of Hanford's worst radioactive waste is sticking to Columbia riverbed sediments.

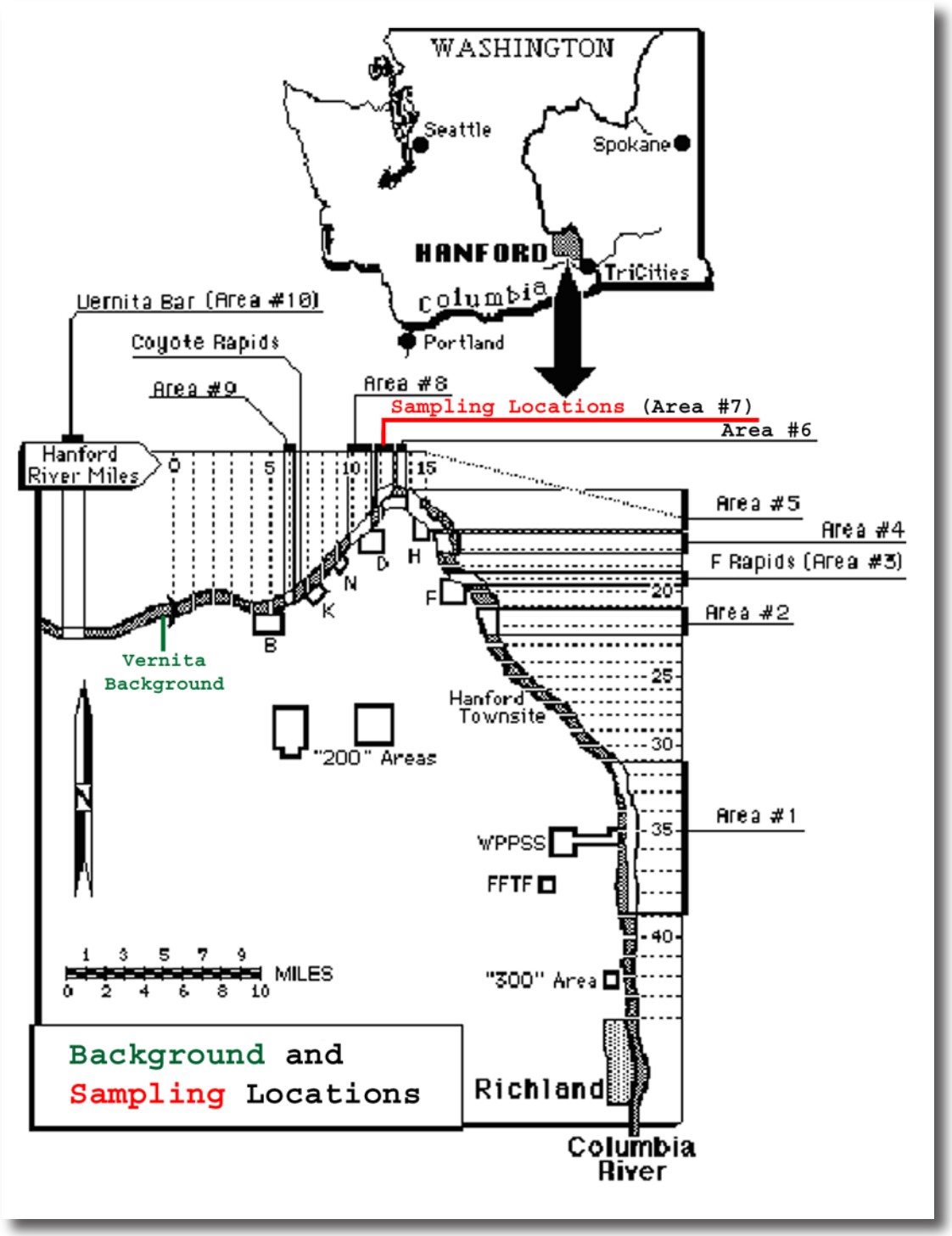
Hanford reactors produced uranium-233 (U-233) for tactical nuclear weapons during the Cold War. TRAC has surveyed Hanford's radioactivity in the riverbed since 1999. Based on the evidence in TRAC's studies, the Washington State Ecology Department jointly sampled riverbed sediments with TRAC on 23 September 2004. Ecology then analyzed *colloids* from the sediment samples to test for U-233. The objective was to answer the question: Is U-233 of Hanford origin present or absent in Hanford Reach sediment?

TRAC and Ecology agreed on sample locations. One background sample was collected at Vernita, upstream of Hanford and so not influenced by Hanford discharges into the river. Three samples were collected from the riverbed, near the upstream end of D-Island. Those three D-Island samples provided the test data for this study.

Unfortunately, the detection level to identify U-233 was not clearly specified for Ecology's analyses. Ecology's ICP-MS analyses had a detection level a hundred times above the level required for detection of U-233 and so provided only nil results. Ecology's alpha spectrometry did not report U-233 distinct from naturally occurring U-234.

Ecology provided its alpha spectrometry data files to TRAC. TRAC applied an ordinary analytical procedure to separate a U-233 peak from the U–234 peaks in the spectra. The three test samples from D-Island yielded U-233 in the range of **0.010 to 0.024 pCi/g**. The background (Vernita BKG), blank, and spike results were each 0.000 pCi/g. **These results confirm the presence of U-233 of Hanford origin, on sediments in the riverbed of the Hanford Reach.** The level of confidence in this confirmation depends on previously held beliefs.

The U-233 waste is present in *colloids* that stick on sediments under the Hanford Reach. The colloids interact chemically with water in the riverbed, where salmon hatchlings live.



Reactor areas are identified by letter. Major salmon spawning areas are identified by numbered black bars (Areas #1 - #10). The Vernita background sample was collected a half mile upstream of Hanford River Mile (HRM) zero at Vernita Bridge. The three test samples were collected from the upstream end of D-Island, about HRM 10.5.

☢ Introduction

Wild fall chinook salmon spawn in ten areas that underlie the 50-mile Hanford Reach of the Columbia River in Eastern Washington; see Locations on Page 2. These areas are crucial habitat for the wild salmon hatchlings (*alevin*) that are believed to maintain the strength of the salmon stock for a billion-dollar-a-year commerce.

Beginning in World War II, Hanford's nuclear reactors, built on the shore of the Columbia, used river water to cool their cores. Radioactive and toxic wastes from the reactors were discharged back into the river.

D-Reactor is one of Hanford's 9 nuclear reactors that produced U-233 for nuclear weapons during the Cold War.

Hanford's best known nuclear product was plutonium-239 (Pu-239). Pu-239 powered the Trinity Test at Alamogordo, New Mexico, on 16 July 1945 and the bomb dropped on Nagasaki, Japan on 9 August 1945, ending World War II. Plutonium from Hanford's reactors provided much of the strength of the U.S. nuclear deterrent during the Cold War.

In addition to Pu-239, Hanford produced a wide variety of special nuclear materials, ranging from test specimens to full production. During the Cold War, all Hanford's 9 reactors produced uranium-233 (U-233) for tactical nuclear weapons.

In 2000, the U.S. Department of Energy, owner of the Hanford Site, began to declassify information about its still semi-secret production of U-233 in Hanford's reactors. Following a request under the Freedom of Information Act, TRAC looked over tens of thousands of pages of documents relating to U-233 production, processing, and waste disposal at Hanford. See References 4 and 5, for a summary of TRAC's discoveries.

Since 1999, TRAC has surveyed artificial radioactivity in the sediments under the Hanford Reach of the Columbia River. See References 3, 4, and 5. Based on TRAC's accumulating evidence of U-233 waste in the riverbed, the Washington State Ecology Department jointly sampled riverbed sediments with TRAC on 23 September 2004. The samples were split three ways for quality control and assurance.

TRAC extracted the *colloidal* and fine fraction from one set of the sample splits and returned that fraction to Ecology. Ecology then analyzed those four sample fractions, as *Stakeholder Columbia River Samples*, to test for the presence or absence of U-233. The objective was to answer the question: **Is U-233 of Hanford origin present in Hanford Reach sediment?** —This was a confirmation or refutation test, not an indication of the importance of any U-233 that might be present.

Representatives of Ecology, of the state Department of Health (Division of Radiation Protection), of the Pacific Northwest National Laboratory, and of TRAC participated in sample collection on 23 September 2004. These participants collaborated to select the exact sampling locations. This assured that sample location was unobjectionable from any known standpoint. One sample was collected near Vernita, upstream of Hanford operations and so was not influenced by Hanford discharges into the river. That sample represented background (BKG) for this study. Three samples were collected from the riverbed, near the upstream end of D-Island. Those three D-Island samples provided the test data for this study.

The required detection level was discussed, but evidently not agreed. Ecology understood that their contract laboratories would "run the samples at the lowest detection limit for which they are quality assured/quality controlled [Reference 1]." TRAC understood that detection level to be 1 part/trillion (=0.1 µg/kg) by mass or 0.01 pCi/g activity, the level required to detect U-233. Ecology's actual detection level was 100 parts/trillion (=10 µg/kg), by ICP-MS. Ecology's ICP-MS analyses had a detection level a hundred times above the required level and so could not provide any meaningful results.

Ecology's alpha spectrometry did not report U-233 separately from U-234 in their isotopic uranium analyses.

The alpha spectra of the samples and reference materials, for isotopic uranium, contain alpha energy peaks for naturally occurring U-234, U-235, and U-238, as well as peaks for artificial U-233 if it is present in the analyzed material. The U-233 alpha peaks are within the general region of the U-234 peaks. Therefore, Ecology's contract laboratory did not report U-233 and U-234 separately, but instead reported the sum of those two isotopic activities as "U-233/U-234".

Nonetheless, the main alpha emission from U-233 has distinctively higher energy than the highest alpha energy from U-234 decay. Therefore, the activity of U-233 can be analytically separated from the activity of U-234, even though Ecology's contract laboratory did not make this distinction in their report.

TRAC has performed this ordinary, post-analysis separation of U-233 from U-234. This is the report of that analytical separation of the three test samples, the background sample, a tracer, and a blank.

☢ **Procedure**: Chronology

One background (BKG) sample was collected a half mile upstream of Vernita, which is upstream of Hanford Site operations. This location (on WGS 84 datum) is a usual reference background for Hanford Site impact studies:

Vernita BKG:	North Latitude:	46.63435°	West Longitude:	119.75458°

The Vernita sample provided a check for a non-Hanford source of U-233, such as worldwide fallout from atmospheric testing of nuclear weapons or a false positive analytical result.

Three sediment samples were collected from the riverbed, on the upstream end of D-Island.

The locations of these D-Island samples were as follows:

D-Island "1":	North Latitude:	46.70151°	West Longitude:	119.54201°
D-Island "2":	"	46.70153°	"	119.54167°
D-Island "3":	"	46.70175°	"	119.54136°

Those three samples were collected upstream of the D-/DR-Reactor discharge pipes to eliminate the possibility of sampling U-233 debris that might have been discharged into the river from failed target rods in D- or DR-Reactor. The D-Reactor discharge pipe was marked by riser vents located between:

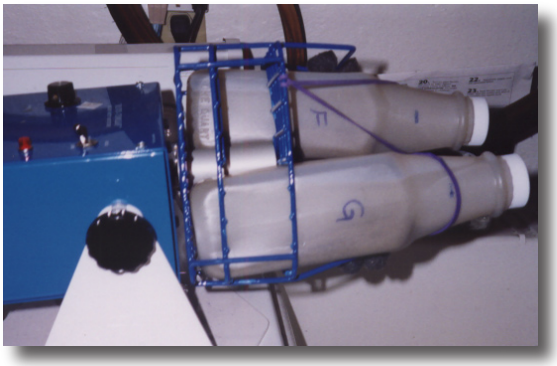
north riser:	North Latitude:	46.70245°	West Longitude:	119.54135°
south riser:	"	46.70224°	"	119.54117°

The DR-Reactor discharge pipe was farther downstream.

At each sampling location, large gravels and cobbles were removed by hand and shovel from the sediments.

Finer sediments were shoveled into a 2-mm standard stainless steel sieve, over a collecting pan (see photo sidebar on next page). The contents of the collecting pan were poured into a large sample bag. The participants repeated the process until the sample bag was filled for three-way splitting: to Ecology, to the state Health Department's Division of Radiation Protection, and to TRAC. TRAC prepared and re-submitted the samples to Ecology.

TRAC extracted colloids from the surfaces of the sediment samples, as follows: About 750g of sediment were placed into rounded square plastic bottles (Cole-Parmer Cat. No. A-62270-00) which were then filled with distilled water. The bottles were rotated horizontally at 6 rpm for at least 5 hours (Cole-Parmer Roto-Torque Rotator, Cat. No. A07637-00 with a test tube basket modified to rotate two, quart bottles at a time).



Rotating two bottles with sediment samples and distilled water to extract colloids from the sediment surfaces.

The rotated samples were then flushed with distilled water through a standard 0.475 mm stainless steel sieve. The extracted colloids and the very fine sediments that passed through the sieve were oven dried at <90°C and weighed. On 7 October 2004, TRAC shipped each sample (one of 3 ounces and three of 4 ounces) to Ecology for analysis, along with a Chain of Custody letter.

On 28 October 2004, Ecology shipped splits of the four prepared samples, along with a Chain of Custody and an Analytical Request to Paragon Analytics. Ecology's analytical request included uranium isotopes ("ISO UR") with a *remark* for uranium-233 ("UR 233").

Paragon Analytics used their standard Procedure PA SOP14R9 for isotopic uranium analysis: "In this report, any uranium in this region of interest [in the alpha energy spectrum] will be reported as U-233/234 [Paragon Laboratories Case Narrative (7 December 2004)]." The sample analyses were completed on 2 December. The results included a duplicate ("Dup.") of the D-Island "1" sample analysis, a Method Blank analysis, and a Method Tracer analysis.

Although Procedure PA SOP14R9 does not separate U-233 from U-234, Paragon's alpha analysis is *sharp* enough to allow complete separation of the 4825 keV alpha emission of U-233 decay from the 4776 keV emission of U-234 decay. In order to separate U-233 from U-234 in Ecology's alpha spectra, TRAC requested the spectral files from Ecology on 30 January 2005. Ecology provided those data files by 23 March.

Each alpha spectrum consists of a list of counts in each of 508 energy *channels*. Each spectrum is documented with the sample description; spectrum acquisition information; and calibration information including (by channel) energy, efficiency, and sharpness (FWHM). This information allows a reviewer to replicate any of the computer-generated analytical reports of radioactivity.

Uranium-233 has a distinct alpha emission peak at 4825 keV, with an intensity of 84.4% [D.R. Lide, ed., "CRC Handbook of Chemistry and Physics," 84th ed., CRC Press, Boca Raton (2003) **11**-184]. The nearest natural uranium peak is at 4776 keV, arising from U-234 decay. TRAC selected a 50 keV wide, spectral region of interest, just above 4800 keV. TRAC picked the particular channels based on the energy calibration information for each alpha spectrum. Then TRAC read the number of counts in each of those

Joint Sampling Process

Step 1: Select a sample site and remove large cobbles and gravel. This is one of the three sites where the samples were collected upstream of D-Island.

Step 2: Shovel finer sediments into a 2-mm stainless steel sieve (on right).

Step 3: Empty sieved sediments from collection pan into sample bag. The sample is then divided among the sampling participants.

channels. TRAC used the same procedure as the laboratory used for U-233/U-234, U-235, and U-238 analysis, with the intensity for the 4825 keV peak of U-233 to separate U-233 from U-234.

TRAC provided to Ecology the final break-out of U-233 on 25 March 2005, for the blank, tracer, and the Hanford samples, including the background sample from Vernita and a duplicate analysis, providing quality assurance. See the Analytical Results on the next page.

☣ Analytical Results — U-233 separated analytically from "U-233/234" in alpha spectra

in Paragon Analytics "Isotopic Uranium By Alpha Spectroscopy Raw Data Report" (7 December 2004).

Symbol:				N	m	E	Y	A	2s
Sample Name	File	Start Energy [keV]	Channels	Counts	Aliquot Mass [g]	Base Efficiency	Chemical Yield	Activity [pCi/g]	Uncertainty [pCi/g]
Method Tracer	URB225L	4804.	181-185	0	2.	0.3163	0.754	0.000	--
Method Blank	UB225B	4803.	179-183	0	2.	0.3163	0.754	0.000	--
Vernita (BKG)	UA2541	4802.	180-184	0	2.02	0.2839	0.847	0.000	--
D-Island "1"	UA2542	4802.	180-184	3	2.	0.3141	0.815	**0.010**	±0.012
D-Island "1" Dup.	UA2542D	4802.	179-183	7	2.01	0.3015	0.845	**0.024**	±0.018
D-Island "2"	UA2543	4806.	181-185	5	2.01	0.2976	0.766	**0.019**	±0.017
D-Island "3"	UA2544	4807.	180-184	3	2.01	0.3107	0.804	**0.011**	±0.012

For each sample:

"Start Energy" is the lowest energy in the first channel in the sample spectrum File, entirely above 4800 keV. Spectral channels above this energy are insensitive to the highest energy emission of U-234 at 4776 keV. This distinguishes the 4825 keV alpha emission of U-233 decay from the closest emission from U-234 decay.

"Channels" is the counting region that comprises the first five channels beginning above 4800 keV in each sample spectrum File. Each channel is about 10 keV wide. The 4825 keV alpha emission of U-233 is thus centered close to the middle of the 5 channels of region counted for U-233.

N = Number of counts in "Channels" counting region in the sample spectrum File.

Activity (A) of each sample is calculated as follows:

A = (27 pCi-seconds/counts) x N / T / m / I / E / Y

where: T = Live Count Time = 18,000 seconds

and: I = Intensity of 4825 keV alpha emission of U-233 = 0.844

Uncertainty (2s) of the activity (A) of each sample is calculated as follows (nominally, 95% confidence interval):

2s = Two Standard Deviations of N counts within the sample counting region (with nil blank and background) = $2A / N^{1/2}$

To convert µg/kg of U-233 to pCi/g: multiply by 9.63

☢ Confidence

The isotopic separation procedure for the Analytical Results of this report is an ordinary analytical practice. However, this analysis is not part of the analytical laboratory's standard Procedure PA SOP14R9 for isotopic uranium analysis.

There is some concern that the few alpha counts in the U-233 region of spectral interest might be non-random, false positives. Besides U-233, the other radionuclide having intense alpha emissions in the 4800 to 4850 keV energy region-of-interest reported in the Analytical Results is

Thorium-229: 4814 keV (9% intensity), and 4845 keV (56% intensity)

Thorium-229 (Th-229) is the first radionuclide product of U-233 decay chain. Ecology's contract laboratory for alpha spectrometry uses Th-229 as a quantitative spike in its isotopic thorium analyses. Therefore, Th-229 is a plausible laboratory contaminant in sample preparation or other processes in the laboratory. There are scenarios in which the positive sample counts in the Analytical Results might be attributed to Th-229 instead of U-233. These scenarios are precluded by the zero counts in the region-of-interest in the background sample, tracer, and blank spectra.

The reliability of the confirmation of U-233 in Hanford riverbed colloids reported here depends primarily on zero (N) counts in each of

tracer
blank
background (Vernita BKG sample)

These zeros weigh heavily against the counts for the three samples and the duplicate being false. These three zeros provide cumulative confidence of the validity of the positive sample counts in the three test samples, ranging from 3 to 7 counts.

The Ecology Department does not presently accept *any* conclusion regarding the presence or absence of U-233 in its alpha results, because its contract laboratory's uranium isotopic analysis is not certified for U-233. Ecology has asked its contract alpha laboratory to undertake a special study in 2006 to "show the 'peaks' for U-233 in a spiked sample and compare that to the peaks in the U-234, U-235, and U-238, so there will be a clear understanding of how the laboratory is able *to determine that U-233 is not present* [emphasis added, Reference 1]."

The Ecology Department's effort to determine that U-233 is not present in the Hanford Reach riverbed calls attention to the strength of different beliefs in regard to U–233. Many believe that U-233 of Hanford origin certainly does not contaminate riverbed colloids; it is a matter of identifying whatever errors have led to false claims for the presence of U-233. For them, several more independent studies will be required to convince them to change their belief.

TRAC had proposed and tested a theory that U-233 of Hanford origin contaminates riverbed sediments. TRAC estimated the likelihood of U-233 in the riverbed at 80 to 95%, before the present study. For TRAC, the present study was anticipated to be an independent, *skeptical* confirmation of an already established theory with its supporting evidence.

As a confirmation study, the 18 counts (sum of N column in the Analytical Results) in the three test samples plus duplicate are **positive 4.2 standard deviations**, in comparison to a total of 0 counts in the background plus tracer plus blank. As a skeptical confirmation study, these results are **positive beyond reasonable statistical doubt**.

With this independent, skeptical confirmation, beyond statistical doubt, it would be just as difficult to convince TRAC that U-233 is not present in the riverbed as it would be to convince Ecology that U-233 is present.

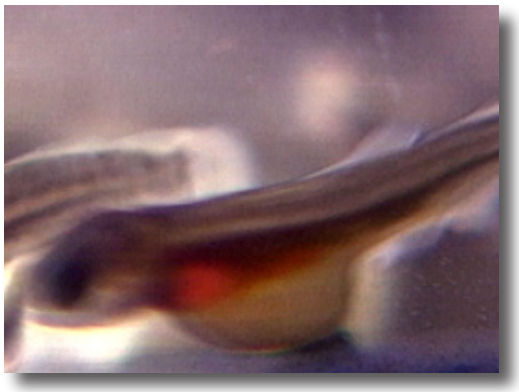
This great difference in opinion has arisen from correspondingly great differences in pre-existing beliefs about the nature of contamination of the Hanford Site and of the Hanford Reach of the Columbia River. To appreciate the present difference of opinion, one must recognize how strongly present interpretations of data depend on past experiences and firmly held beliefs.

The issues of what constitutes a sufficient determination of scientific fact in the context of pre-existing beliefs is the subject of Bayesian statistics. See Reference 7 for an informal introduction to *inference* and the influence of evidence on firmly held, but opposing beliefs.

The difference between the previously held, strongly confirming and strongly refuting beliefs was incorporated into the design of the reported study. Advocates of the two different beliefs participated in all phases of this study, under the auspices of the state Ecology Department. Each participant agreed with the procedure. There was an informal agreement to accept the outcome of this study. Sample splits were retained by the Ecology and the Health Departments and TRAC for their own, independent checks, at their option.

☢ Implications

The reader might already anticipate from the above discussion of Confidence, that there is much at stake over the answer to the technical question of whether U-233 of Hanford origin is present in sediments underlying the Hanford Reach. Hundreds of thousands of dollars of labor and laboratory costs have been spent on this question since 1999.



Salmon hatchling (*alevin*) - Photo: Army Engineer Corps.

The presence of U-233 in riverbed colloids raises a specific concern for the *future* health of biota in the riverbed of the Hanford Reach. The poster children of this concern are the hatchlings (*alevin*) of wild-spawning fall chinook salmon that spend their first months among the cobbles in the Hanford Reach riverbed.

This specific concern focuses on radium-225 (Ra-225), which is the second decay product of U-233. Radium-225 mimics calcium. Calcium is an element essential for life. Salmon hatchlings are believed to absorb calcium —and hence Ra-225— from riverbed water in their habitat under the Hanford Reach.

Radium-225 has a half-life of only 15 days. Thus, any harm Ra-225 might do the salmon would occur only during the alevin stage. The Ra-225 might reduce the strength of the wild salmon stock, which would affect a billion-dollar-a-year commerce.

This study raises general concern for the degree that public and regulatory oversight of the Hanford Site is weighted by pre-existing beliefs. Reference 2 reports unmonitored radium entering the river in seepages from Hanford's 300-Area. Thus, the present study justifies review of the degree to which pre-existing beliefs influence the management strategy and clean-up of the site.

An important result of this study is the wide range of its interpretations: between positive confirmation of U-233 in the riverbed at one extreme to mere analytical data requiring further procedural

evaluation in the laboratory at the other extreme. This wide range reveals how little is yet known about Hanford Site's present and future impacts on the Columbia River.

A comprehensive, independent, environmental assessment of the Hanford Reach river corridor is needed to support decision-making in order that clean-up of Hanford Site will protect the present and the future of the Columbia River.

☢ References (hyperlinked within www.radioactivist.org)

1• **Hanford State of the Site Meeting Comments, Uranium-233**
—letter by Jay Manning, Ecology Director (December 2005)

2• **Radioactive Bioaccumulation in Clams along the Hanford Reach**
(March 2005)

3• **Uranium-233 Detected in Hanford Reach Sediments** (November 2003)

4• **Trouble in the Columbia Riverbed**
—increasing radioactivity under the Hanford Reach (October 2003)

5• **Hanford Radioactivity in Salmon Spawning Grounds**
—quality, extent and some implications (August 2002)

6• **Questions and Answers on Uranium-233 at Hanford** (August 2002)

7• **Why Reasonable Minds Differ**
—learning modeled on changing confidence in beliefs (April 1997)

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