# Trouble in the Columbia Riverbed:

increasing radioactivity under the Hanford Reach



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TRAC's Moon Callison, Just Before Finding D/DR Discharge Pipeline Vents

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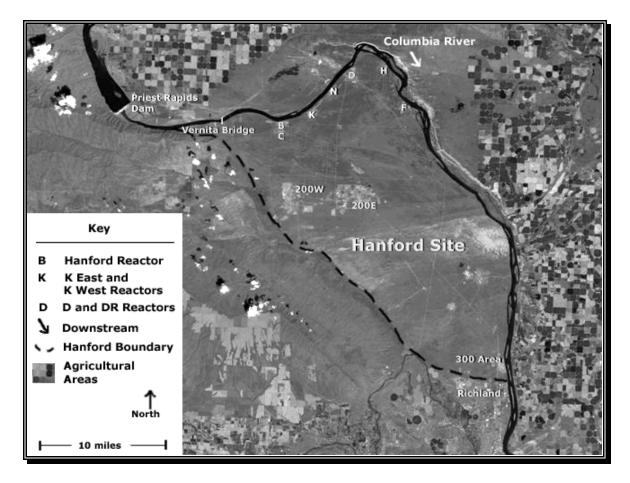
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# **Trouble in the Columbia Riverbed:**

## increasing radioactivity under the Hanford Reach



The Columbia River runs through Hanford Site, in eastern Washington State.

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## Summary

The public has expressed concern that radioactivity from Hanford's wartime nuclear weapons production might contaminate salmon spawning grounds in the Columbia River. Salmon hatchlings (*alevin*) grow among the gravels in the riverbed, where they are susceptible to environmental stresses that include toxic chemicals and radioactivity. Some people worry that radioactivity from Hanford will damage the alevin genetically and slowly reduce the strength of the salmon stock.

In 2001, The RadioActivist Campaign (TRAC) found a radioactive fingerprint (europium-152) of old Hanford nuclear waste in 60% of sediments lying under the Hanford Reach and in 7 of 10 major salmon spawning areas. Those results prompted this laboratory study, to examine the water in which the salmon alevin live. The question was, What radioactivity dissolves from contaminated Hanford Reach riverbed sediments into the riverbed water?

In 2002, the Government Accountability Project (GAP) contracted TRAC to evaluate radioactivity in riverbed water along the 51 mile stretch of the Columbia River that is the Hanford Reach. The main result of this study is the discovery that artificial radium-225 (Ra-225) is dissolving into the riverbed water from Hanford Reach sediments.

Radium is a radioactive element that mimics calcium, which is essential for control of body functions at the cellular level. Radium is readily taken into animal cells as a calcium substitute. When radium decays, *alpha* particles are ejected. Alpha particles are heavy and energetic. Like atomic cannon balls, alpha particles do massive damage on the molecular scale. This is the basis for concern that developing salmon alevin might take up Ra-225, as a substitute for natural calcium, and might incorporate the Ra-225 into cells susceptible to alpha particle damage.

Alpha particle damage, caused by Ra-225 decay, might account for chromosomal anomalies reported in salmon spawning in the Hanford Reach, by University of Idaho researchers in 2001 [Ref **5**. References and Notes are identified by a number in brackets, and they are located at the end of this report.]

Radium-225 is an artificial isotope of the element radium. Ra-225 is created by radioactive decay from artificial uranium-233 (U-233). During the Cold War, Hanford produced U-233 for tactical nuclear weapons, like the *Davy Crockett* rocket, launched from a bazooka. Based on the results of this study, the amount of U-233 in the Hanford Reach riverbed might be about ten kilograms, stuck to the surfaces of particles of sediment.

Uranium is nearly insoluble in water. Some uranium compounds are *polar*, making them sticky or gummy. Finely dispersed uranium wastes in the riverbed adhere to sediment particles and remain there, relatively harmlessly, rather than dissolving and being washed away.

Ra-225 is the first radionuclide in the U-233 decay chain that is very soluble in water. When U-233 decays to Ra-225, this radioactive radium is dissolved into the riverbed water where aquatic organisms live.

TRAC has identified three separate plumes of Ra-225 in riverbed water, downstream of three barge-and-tug terminals that had supported wartime operations at Hanford. TRAC compared those locations and the finely dispersed form of the radioactive waste in the riverbed with documentation of Hanford's operations. TRAC concluded that the U-233 waste disposed in the river probably came from processes that separated Hanford's U-233 product from byproducts.

Soon after a little of Hanford's newly produced U-233 first entered the riverbed, the radionuclides along the U-233 radioactive decay chain might have posed little hazard to biota. U-233 decays into thorium-229 (Th-229) which has a fairly long radioactive halflife of 7,300 years. That means that over the next 7,300 years, Th-229 will "grow in" at a nearly constant rate of 2 - 4% annually, until Th-229 decays match U-233 decays. Ra-225 activity in the riverbed will increase at the same rate Th-229 increases.

Unless remedial action is taken, Ra-225 activity in the Hanford Reach riverbed water will probably increase more than a hundred fold, over the next several thousand years. Unfortunately, there are already early warning signs of biological trouble in the riverbed.

These results remind us that the Columbia River runs through Hanford Site, the most contaminated place in North America. As more of Hanford's historical impacts on the Columbia River are discovered, Hanford Site becomes ever more clearly an unwise choice for a waste disposal site, for the present and for our future.



Warning sign on the Columbia River shore at N-Springs (Hanford River Mile 9).

## **Questions and Answers**

#### What's the problem?

- Preliminary evidence associates radioactivity in the Columbia Riverbed with chromosomal anomalies in salmon spawning in the Hanford Reach.
- Radioactivity of Hanford-origin in riverbed water will multiply a hundred times where salmon hatchlings live, unless the problem is fixed.

#### Why should I care?

- —So the problem gets addressed and is fixed, so salmon stocks remain strong. The Department of Energy *is* responsive to pressure from the public and will fix it if the public demands it.
- —So DOE stops making Hanford's waste problems worse by importing more radioactive and toxic wastes.
- "Saving the salmon" has become a cliché. But it's still better to save them than to wreck their genetics. If we do save the salmon, we'll have begun to save the Columbia River too.—Great endeavors always begin with a first little step: "Remember the alevin!"

#### How strong is the evidence?

- —Fairly.
- This first-of-its-kind study provides strong evidence, but not a proof. The seriousness of the evidenced problem demands follow-ups, for confirmation and then for problem solving.

#### Why is radioactivity in the riverbed increasing, rather than decaying away?

• The decay chain of artificial uranium-233 is releasing radium-225 at a rate that increases 2 — 4% per year (linearly, not compounding). When the decays of Ra-225 match the decays of U-233, the activity of Ra-225 in the riverbed will be more than a hundred times its present activity.

#### Why does Hanford radioactivity threaten salmon spawning?

- Radioactive waste dumped into the Columbia River decades ago sank to the bottom of the river and has stuck to sediments in the riverbed, where it remains in the salmon spawning grounds. The U-233 radioactivity has begun to break through a thorium-229 block, releasing increasing doses of radium-225 into riverbed waters where salmon hatchlings live. See Fig. 1, on the next page.
- Radioactive radium-225 mimics biologically essential calcium in animals. This report presents the first evidence that Ra-225 is already at levels of concern in the riverbed, and increasing.

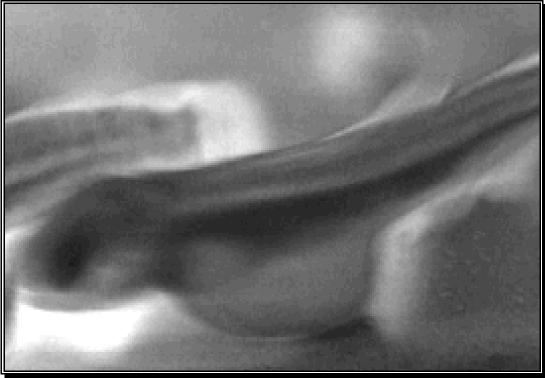


Fig. 1. Salmon hatchling (alevin) [Army Engineer Corps photo].

#### What should I do?

- Tell your family, friends, and co-workers. Speak up! Get the word out.
- Contact the Department of Energy, tell them you want the river corridor cleaned up, and not to bring any more waste to Hanford.

Department of Energy contact points

- Public Affairs: "Andrea Powell" <Andrea\_S\_Powell@rl.gov>.
- Switchboard: (509) 376-7411. --Ask for the right person for you to talk to regarding your stated concerns.
- Snail mail: U.S. Department of Energy

Office of River Protection P.O. Box 450 Richland, WA 99352

## **Problem Statement**

TRAC undertook an experimental evaluation of artificial radioactivity in riverbed water along the 51 mile Hanford Reach. See Fig. 2, below.

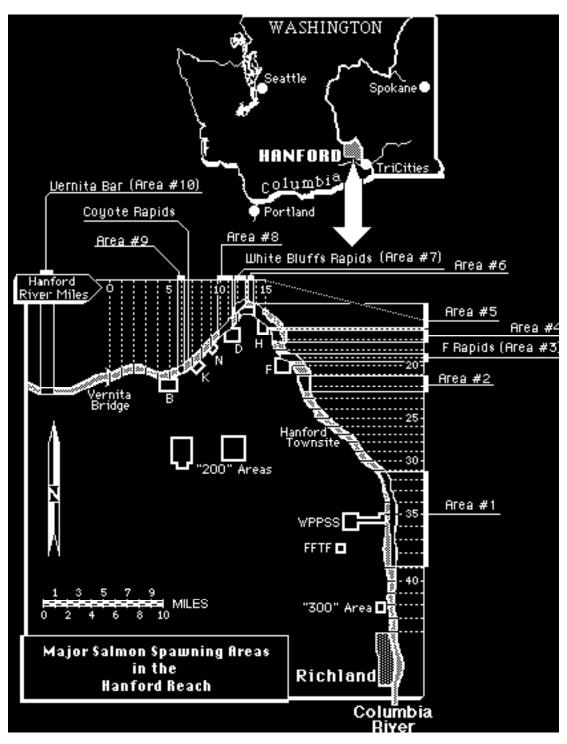


Fig. 2. Reactor Areas and Other Features Along the Hanford Reach.

## **Historical Perspective**

The United States government scouted the remote Hanford, Washington area in January 1943, looking for a site to produce plutonium for an atomic bomb. Hanford satisfied all the requirements, and land acquisition began a month later. The Army Engineer Corps had responsibility for management, contacting with the DuPont corporation to build and run the plant. The challenge was enormous; Hanford was the largest scale-up from a pilot plant, in human history.

Hanford's B-Reactor provided the plutonium explosive for the world's first nuclear bomb. "Trinity" detonated at Alamogordo, New Mexico, on 16 July 1945, with explosive power equivalent to 23,000 tons of TNT [1].

The world's second atomic bomb was built from uranium, highly enriched in isotope number 235 at a plant at Oak Ridge, Tennessee. "Little Boy" fell on Hiroshima, Japan, on 6 August 1945, with two-thirds the explosive power of "Trinity." The third atomic bomb was "Fat Man," another Hanford product of B-Reactor. "Fat Man" destroyed Nagasaki, Japan, three days later. Thus ended the Second World War with powerful new images of war won in a flash.

During the Cold War years from 1950 to the end of the 1980s, Hanford produced most of the material for the U.S. side of the nuclear standoff with Russia. We convinced the Russians that we were willing to use our overwhelming nuclear power, thus avoiding the need to actually use our lethal arsenal. That finesse involved scaring the Russians while reassuring ourselves.

#### Throughout the history of the Hanford Site, managers of the complex consistently attempted to reassure the public by proclaiming that the installation was safe with minimal threat to human health [2].

Nuclear weaponry played a key role in the Cold War and, ultimately, in that U.S. victory also. The Cold nuclear War was won, almost without a flash. The workers and managers of Hanford and other nuclear plants deserve credit for their contributions to both those American victories.

But the Cold War was also a war of images and of nerves. In some ways, the images were harsher at Hanford than anywhere else. Hanford's now-silent, monolithic reactors along the Columbia River, and its huge processing plants on the central plateau, draw the eye across miles of arid land, with a hint of sage on the hot breeze.

Hanford's larger-than-life architecture is a development of the utilitarian International/Modernist Style, called "Brutalism." The name derives from the French term for rough, untreated concrete: *breton brut* [1]. The style is the statement: *A structure is beautiful to the degree it is functional* [3]. The huge scale of the rough-poured, raw structures with their exposed pipes and open waste disposal trenches is exaggerated beyond any functionality. Hanford is an *awesome* image. Hanford served our nation well in two wars, the Second World War and the Cold War. Looking back over those decades of war, the mind's eye is drawn to values that came out of Hanford's hot dawning of the nuclear age: ...power... ...expediency... ...functionality... ...service... ...secrecy... ...imagery...

Hanford's Cold War values are carrying forward into another era, in which they are transmuting into new meanings.

Hanford's new, primary mission is restoring the site and releasing most —about 500 square miles to be cleaned up— back to public uses by 2012 [4]. To accomplish this mission, DOE's first agenda item is **restoring the Columbia River corridor**, maybe one of the largest challenges in human history.

This report explores and discusses Hanford's radioactivity in the riverbed of the Columbia River corridor.

## Methods

This study is a laboratory experiment based on sediments collected from the Hanford Reach riverbed in 2001, with a few follow-up sediments collected in 2002. An alternative to this experiment would have involved pumping water directly from the riverbed and analyzing it for radioactivity. TRAC chose the laboratory method to obtain better definitional control, to improve reproducibility, and to gain an understanding of some of the complicated processes that occur in the natural riverbed environment. The focus was on the interactions between the sediments and the riverbed water.

As TRAC learned of that interaction, the original assumptions of the study were adjusted and the experimental procedure was modified accordingly. Two assumptions misguided the *beginnings* of this laboratory study. Those incorrect assumptions, along with their corrections, are highlighted here for clarity, and further discussed in the description of the procedural steps, to show their relationship to the experiment and its outcome.

**First** <u>incorrect assumption</u>: Radioactivity in riverbed water is more or less in balance with radioactivity in the riverbed sediments. <u>Correction</u>: The preparation of water samples revealed the presence of an unexpectedly large amount (0.4 to 1.2%) of material that adheres weakly to the sediment grains in the reference sediment samples. Further work suggested that this adhered material contains most of the radioactivity that dissolves into the riverbed water. What had previously been conceptualized as a sediment and water interaction was thus re-conceptualized as primarily a *colloid* (the adhering

material) and water interaction, with riverbed sediment acting as the framework for that interaction. This correction was applied in Step #2 of the procedure; on the next page.

Hanford's radioactivity in the riverbed is *sticking to* sediment particles.

**Second** <u>incorrect assumption</u>: Uranium, thorium, plutonium, and other very heavy (*actinide*) element wastes from Hanford are dissolved into riverbed water. <u>Correction</u>: TRAC's evaluation of the gamma and alpha data from this study revealed that the relatively insoluble, radioactive actinides adhere to sediment particles in the riverbed until they decay to radium. Radium, which mimics calcium, is soluble in the riverbed water. TRAC accordingly changed the interpretation of alpha and gamma results to radium, in Step #5 of the procedure. Likewise, TRAC failed to schedule alpha and gamma analyses for best detection levels, because TRAC had anticipated that long-lived actinide elements would dominate the radiological results, rather than short-lived radium isotopes. That scheduling lowered most data quality from "proof of" to "evidence of".

The procedural steps of this experiment are listed chronologically, beginning on the next page.

#### Step #1: collection of sediment samples from the Hanford Reach.

TRAC collected sediments from the Hanford Reach of the Columbia River, in 2001, with a few follow-up samples collected in 2002. At the time of their collection, the sediment were passed through a sieve (Tyler Standard Test Sieve No. 10, 2 mm), and then the very fine, suspendable material was poured off [6].

#### Step #2: laboratory preparation of samples.

TRAC added 1.6 liters of filtered Columbia River water to 220 grams of the reference sediment [7]. This mixture was divided equally into a pair of square, quart

bottles (Cole-Parmer A-62270-00) and loaded onto a rotator (Cole-Parmer Model 7637); see Fig. 3.

The rotator turned the samples around the long (horizontal) axes of the bottles, at 6 revolutions/minute for 24 hours. The liquid contents were then poured through two, 8 micron filters (Whatman Grade 2V, paper); see Fig. 4. The suspended fraction was greater than anticipated, which affected

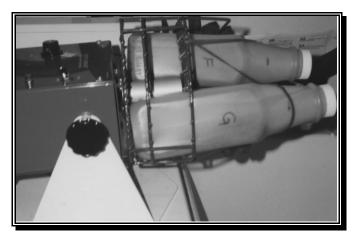


Fig. 3. Rotator with two sample bottles loaded.

TRAC's interpretation of the results. See the Discussion on Page 27.

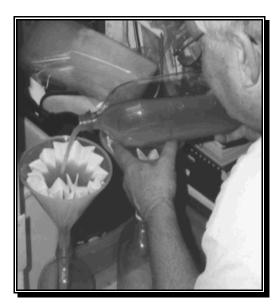


Fig. 4. Pouring liquid filled with suspended material through paper filters.

The filtered water was allowed to stand for at least three days so that any radon-220 gas that might have remained from the reference sediments would have decayed away before radiological analysis.

TRAC bottled 130 mL of filtered water to send to a contract laboratory for alpha analysis. TRAC put the remaining 1.3 liters of filtered water into a microwave oven and steamed the water down to a residue on a plastic film. The residue on plastic film was bottled immediately to keep any short-lived radon gases, that were generated in the prepared samples, from escaping.

#### Step #3: analysis of water samples for gamma and alpha radioactivities.

TRAC submitted an initial batch of ten of the 130 mL filtered water samples to a

contract laboratory (Environmental Inc.) for alpha analysis of 100 mL by EPA Method 900.0, allowing 30 mL spare [8].

TRAC analyzed each of its sixty-two water sample residues for gamma radioactivity. TRAC used a 15 - 3,500 kiloelectron-volt (KeV) photon spectrometer (stabilized on the 1461 KeV gamma peak of potassium-40) based on a sodium-iodide crystal well-type detector, which was housed in a copper-lined, lead shield and held at



Fig. 5. Loading a bottle into the spectrometer detector.

24.0°C. (8,200 channels acquired spectrum were transformed to 165 channels of a constant photopeak width of 3 channels. Because this system has a much better energy stability that the width of those photopeaks, true spectral subtractions are feasible. This reduces the usual problems of peak interferences [6].) Figure 5 shows a sample bottle being loaded into the detector in the lead shield. inside the temperature

controller. The lid on the shield and temperature controller is swung open to the left.

TRAC obtained a "background" spectrum from a background water sample, extracted from sediment collected from the Hanford side of the river, 6 miles upstream of Vernita; See Fig. 2, near the front of this report.

TRAC subtracted that background spectrum from each sample spectrum. TRAC then subtracted the potassium-40 (K-40) activity in each sample spectrum (according to its gamma peak area), thus eliminating the K-40 contribution to those spectra. Then, TRAC similarly subtracted natural thorium and natural uranium spectra equal to their respective activities. TRAC compared the resulting sample spectra to references in TRAC's spectrum library. Residual peaks in the final spectra were then listed for later identification.

#### Step #4: review of work-in-progress.

After the contract laboratory reported the alpha activity data from its first batch of 10 samples, TRAC compared those data to the thorium and uranium activities TRAC had calculated from its gamma analyses in Step #3. This comparison identified a sample handling error at the contract laboratory. Consequently, alpha data for 9 of the 10 samples in the first batch of samples submitted to the contract lab were discarded. (They were reported as mishandled ("mh") in the results of this study. After the handling procedure was corrected, TRAC sent the second batch of thirty bottles of filtered water samples to the lab for alpha analysis by EPA Method 900.0.

#### Step #5: post-analysis of gamma spectra and alpha results.

TRAC compared the gamma and alpha data and discovered that radium was the heaviest (*farthest up the decay chains*) radioactive element in the water samples. TRAC then changed the gamma post-analyses to subtracting a Ra-224 reference spectrum instead of (as previously, in Step #3) subtracting the complete thorium spectrum [9].

Decay factors for Ra-224, from the dates of sample preparation to the dates of alpha and gamma analyses, were applied. Practically all the Ra-224 (3.7 day halflife) in the water samples had decayed before the EPA 900.0 alpha analyses.

TRAC then reviewed the set of still unidentified, residual photopeaks in the 77 gamma spectra (including references, backgrounds, and recounts) that had been acquired. That review revealed a frequently occurring photopeak at 440 KeV. The possibility that the 440 KeV photopeak might actually be the 439 KeV peak in the natural potassium-40 (K-40) spectrum had already been precluded by the spectral subtraction of the K-40 component in Step #3. The possibility that the photopeak might be due to europium-152 in the samples was also ruled out, because the water samples tested negative for europium-152.

TRAC searched reference libraries of gamma emissions for radioactive decays close to 440 KeV and found only one likely candidate: Bi/Po-213. When radioactive bismuth-213 (Bi-213) atoms decay into polonium-213 (Po-213) atoms, about 16% (—the emission "intensity" used in this analysis—) of the newly formed Po-213 nuclei are in an excited state. That state has a nuclear energy 440 KeV above the normal state for the

polonium nucleus. When the excited Po-213 nucleus relaxes to its normal state, a 440 KeV photon is emitted [10]. The emission of this photon from Po-213 is the candidate explanation for the previously unidentified, residual peaks in the set of gamma spectra.

About sixteen percent of Bi/Po-213 decays emit a "countable" 440 KeV photon that fits into the artificial uranium-233 (U-233) decay chain, as shown underscored in Fig. 6, at the top of the next page.

TRAC checked reference spectra for interferences with the 440 KeV photopeak, to avoid false positive reports. The acceptance criterion for energy calibration of gamma spectra had to be tightened to  $\pm 0.2$  channel about the K-40 peak at Channel 112.7 in the Constant-Photopeak-Width (CPW) transformed spectra to avoid false positive reports. Twenty seven of the 77 acquired spectra were thus rejected outright. All other spectra were accepted into this study for their intended purposes. An additional acceptance criterion was placed on the 440 KeV peak to further avoid false positive reports: The CPW transformed spectra were required to have both a maximum height within the Channel 65-67 counting region and a positive count for that region. (This second acceptance criterion was consistent with the concept of "detected" versus "undetected" for a radionuclide that has not been previously reported in the particular situation.) The Results of this study list No Detects ("nd") or Peak Location ("pl") for samples failing the latter acceptance criterion.

Fig. 6. Uranium-233 radioactive decay chain, with halflives (struck through), and alpha ( $\alpha$ ) and beta ( $\beta$ ) emissions (bolded).

U-2	233 <del>-159,000 years</del> > <b>C</b>	X Th-229 -7300 years >	> <b>α</b> Ra-225* <u>15 days</u> → <b>β</b>
C	<b>x</b> < <del>0.03 seconds</del> At-2	17 <b>α</b> < <del>-5 minutes-</del> Fr-2	221 <b>Q</b> < <del>-10 days-</del> Ac-225 <- <sup>+</sup>
ł	—> <u>Bi-213 -46 minute</u>	$p_{s} > \beta P_{0} - 213 = 0.00 s_{0}$	econds > $\alpha$ Pb-209 -3 hours > $\beta$
		(stable bismuth) B	i-209 < <sup>†</sup>
where:	U = uranium Ac = actinium Bi = bismuth	Th = thorium Fr = francium Po = polonium	Ra = radium At = astatine Pb = lead

\* The decay of Ra-225 itself emits a beta particle. Four alpha particles emissions and another beta emission occur later in the decay chain. The halflife of the partial decay chain, beginning with Ra-225 and ending in stable Bi-209, is 25 days. TRAC applied the radioactive decay law to correct Bi/Po-213 measurements at the date of gamma analysis to Ra-225 activity both at the date of sample preparation and the at the date of alpha analysis.

At the end of Step #5, TRAC validated the data with consistency and realism checks. Some of those checks appear in the Discussion, beginning on Page 27. Note that a completely independent analysis, using different analytical techniques, is required to prove the detection of Ra-225 that is reported in this study.

## Results

Narrative Results: Explorations concerning Hanford's historic discharges into the Columbia River.

The 8 original, plutonium-production reactors at Hanford passed treated river water "once through" their piles (cores) of graphite to cool the fuel rods within. The cooling water was held briefly to allow decay of short-lived radioactivity and some heat dissipation, before being discharged back into the river.

The cooling water discharges back into the river were less than clean. That was evidenced by the installation of strainer grates, called "grizzlies," over the outfall pipes to stop large debris from entering the river [11]. Despite such efforts to limit Hanford's releases, the pollution of the Columbia River was already becoming a regional and national concern by 1960 [2].

Hanford facilities had several other pipes, chutes, and overflows that discharged wastewaters into the Columbia River. In 1975, the Department of Energy (DOE) applied to the Environmental Protection Agency (EPA) for a permit for 13 of these discharges, plus a permit for the secondary cooling water from N-Reactor [12]. DOE suggested that Hanford's only noxious discharges into the Columbia River had been from the original, once-through reactors. DOE believed it had fixed that problem by shutting those outdated reactors down by 1971.

The notion that Hanford's main impact on the river had come from its old oncethrough reactor discharges was challenged by a 2002 report [6]. A long plume of europium-152 of Hanford origin was reported in riverbed sediments downstream of an old Columbia River ferry crossing at the D-Reactor Area [with remains of a terminal on the Hanford side of the river, at North 46° 41.830', West 119° 32.764'] [13]. See Fig. 7, below.

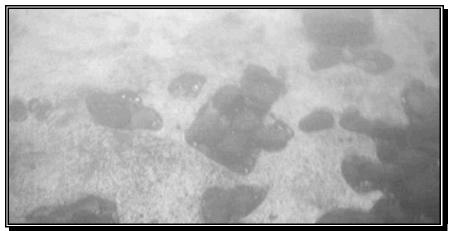


Fig. 7. Underwater concrete approach to D-Ferry crossing [at North 46° 41.839' to 41.850', West 119° 32.777' to 32.785'].

That 2002 report suggested that Hanford's main, presentday impact on the Columbia Riverbed might be radioactive waste that had been dumped into the river from Hanford's D-Ferry crossing. Critics said the reported europium-152 had come out of the old reactor discharge pipes.

Field work for the present study thus began with inspection of the riverbed and the long forebeach of D-Island, which is exposed at low river stages. TRAC found vents

still in place, on top one of the old D/DR-Reactor discharge pipes.

A sediment sample was collected next to the most radioactive vent from that discharge pipe. The cross-pipe on top of that vent is shown in Fig. 8.

That sample was analyzed, and the results are included in this report. The



Fig. 8. Vent on top of D/DR-Reactor discharge pipe, upstream of D-Island.

location of that Sample #2y1013b is shown in Fig. 9, on the next page [14]. Figure 9 shows the location of D-Ferry crossing, upstream of the D/DR-Reactor discharge pipes. (DR-Reactor was "D-" Reactor's "Replacement" reactor.) Locations of sample collection and other features on the river were determined by the Global Positioning System, using a Garmin GPS 12 on WGS 84 datum. Table 2 lists coordinates for each sample. The other sample locations shown in Fig. 9 are discussed, along with their radiological results, beginning on Page 27.

Before the laboratory and analytical phases of this study began, TRAC pursued implications of the discovery of the D-Ferry crossing. Historical documents were reviewed to learn how Hanford's ferry crossings fit into Hanford's transportation facilities and operations.

The blueprint for site design and layout for Hanford's original once-through reactors had been strictly functional [3]. Anticipating the potential hazards of producing plutonium on an industrial scale for the first time, the reactors were required to be almost independent of each other. They were spaced at least one mile apart so a catastrophe at one reactor would not disrupt any other. Each reactor area was "designed virtually identical."

Hanford's three original, World War II reactor areas were B, D, and F. The welldeveloped barge terminal and ferry crossing for F-Reactor (and later, H-Reactor) is now the White Bluffs Boat Launch [at North 46° 42.138', West 119° 32.473']. See Fig. 10, on the page after next. TRAC compared its observations of the F-Ferry and the D-Ferry crossing locations to old Hanford Site maps. TRAC sought the "virtually identical," river crossing location at B-Reactor Area. The only prospect was the northward extension of the Route 6 leg of the Army

Loop Road, on the west side of B-Reactor Area. TRAC found that ferry crossing on 6 September 2002. The south, Hanford-side terminus [North 46° 38.428', West 119° 39.966'] is marked by earthen mounds, possibly the remains of old approach caissons, and a 12inch intake pipeline for B-Area. The dredged face for that terminal can still be seen with underwater television. The approach to the B-Ferry crossing from the north, Wahluke

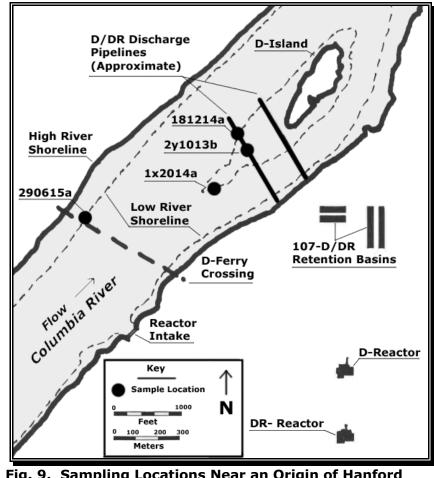


Fig. 9. Sampling Locations Near an Origin of Hanford Radioactivity in the Riverbed, with sample numbers.

side [North 46° 38.581, West 119° 39.814] has been covered with boulders, but is still visible.

TRAC sampled sediment downstream of the north terminus of the B-Ferry crossing [Location +3.6 opp, Sample #290711a] and included the analytical results in this report.

Discovery of the B-Ferry crossing barge-and-tug terminal, only 3.6 miles downstream of Vernita, raised concerns that some Hanford wastes might possibly have been barged far upstream and dumped, to lessen their contamination of Hanford's reactor intake waters. Sampling at Vernita —Hanford River Mile Zero— might not be far enough upstream to assure non-elevated, radiological *background* conditions for this study. After sampling 22.5 miles upstream of Vernita and analyzing sediments for gamma radioactivity, TRAC concluded a sample location [North 46° 37.526', West 119° 51.998', at Location –6.0 Han, Sample #292009a+b] 6 miles upstream of Vernita would suffice as background for the present study.



GAP submitted a request under the Freedom of Information Act for records relating to Hanford's bargeand-tug operations, dredging of the Hanford Reach, and related topics. DOE responded with a list of hundreds of documents, mostly photographs. A few, relevant captions are included in Table 1, below.

Figure 10. White Bluffs / F-Ferry Crossing, looking east at what is now a public boat launch, opposite Hanford.

Doc. Date	Key Words or Description	Doc. Number
0901/1949	HANFORD FERRY, HANFORD SITE, PATROL EXERCISE,	348-49-NEG
	PHYSICAL PROTECTION AND SECURITY, TANK	
01/26/1953	HANFORD FERRY, RAN BY ARMY	5698-1-NEG
08/04/1954	FIRST SET OF TWO PUREX TANKS FROM BARGE TO SETUR	2 3007-NEG
08/06/1954	AQUATIC BIOLOGY LAB AND DREDGING EQUIPMENT ON	10513-NEG
	BARGE	
09/04/1954	PUREX PROCESS CELL EQUIPMENT REMOVAL FROM	DDTS-GENER-
	BARGE	ATED-3211
01/07/1955	<b>REACTOR PLANT MODIFICATION - 100-B AREA</b> ,	11725-4-РНОТО
	BARGE-MOUNTED CRANE	
01/01/1956	SMALL RIVER BOAT AND DREDGE USED TO SCOOP	9-4-NEG
	ALGAE-COVERED ROCKS AND OTHER MATERIALS FRO	M
	THE RIVER BOTTOM TO TEST FOR RADIOACTIVITY	
01/25/1956	190-DR OUTFALL STRUCTURE - LOOKING NORTHWEST AT	
	DREDGING OPERATION THROUGH ISLAND -55% COMPL	
01/01/1961	BARGES WITH TUG BOATS ON THE COLUMBIA RIVER 35	500-3-СN-РНОТО
01/24/1961	100-N AREA PHOTOS - 181N RIVER WATER PUMPHOUSE	
	DREDGING FOREBAY	
09/06/1965	REACTOR OPERATIONS, AERIAL, RIVER, REACH, DREDGE	40322-8CN

These photo captions provide a certain perspective of Hanford's extensive barge operations on the Hanford Reach.

The history of Hanford Site Security provides another perspective. As the Cold War escalated in the 1950s, Hanford Site was ringed with 16 antiaircraft batteries of 90 mm and 120 mm guns. By the late 1950s, the Army replaced those antiaircraft artillery (AAA) batteries with 8 Nike Ajax and then, later with Nike Hercules missile sites.

The Army used barge-and-tugs at the Hanford Reach ferry crossings to support its AAA and missile defenses on the Wahluke Slope, on the north side of Hanford, and near White Bluffs, to the northeast [15].

Some of the AAA and missile defense structures have been preserved for historical interest [**3**]. A blockhouse [at North 46° 41.834', West 119° 33.576'], on the north side of the river, opposite D-Reactor, was apparently part of an AAA site supported from the D-Ferry crossing [between North 46° 41.830', West 119° 32.764' and North 46° 41.994', West 119° 33.098']. The blockhouse, with N-Reactor in the background, is shown in Fig. 11.



Fig. 11. Blockhouse remaining, across from D/DR-Reactors, looking upstream, with N-Reactor in the left background.

The Army's historic barge-and-tug operations, that are documented on the Hanford Reach, provided the following services for Hanford Site:

- transport of heavy reactor and processing equipment
- support of dredging and construction of river facilities
- support of defense facilities on the side opposite Hanford

The experimental results of this study will add another possible service to this list of varied and extensive barge-and-tug services provided on the Hanford Reach, supporting Hanford Site's mission for production of special nuclear materials:

• disposal of certain, heavy liquid radioactive wastes into the river

**Experimental Results:** Explorations concerning Hanford's historic discharges into the Columbia River.

Table 2 reports the radiological results of three primary radioactivities: radium-224, radium-225, and alpha according to EPA Method 900.0. A summation of the Ra-224 activity plus four times the Ra-225 activity is also tabulated as "non-volatile alpha." That summation takes into account that the decay product of Ra-224 is inert radon (Rn-220) gas, which might escape before later radioactive decays occur along the decay chain, which would reduce alpha doses to biota that had accumulated Ra-224.

The three primary radionuclides reported in Table 2 came from three different analytical procedures, described in the Methods section. Their presentations differ

accordingly. The uncertainties ( $\pm$  values) accompanying the data reflect confidence in the analyses. For Ra-224 and Ra-225, the " $\pm$ " value is one standard deviation counting uncertainty. For EPA 900.0 alpha, the " $\pm$ " value is two standard deviations counting uncertainty.

The unit of radioactivity used in this report is picocuries per liter: pCi/L. One picocurie per liter equals radioactive decay per minute in a pint (half liter) of water.

Table 2 presents radiological data in the downstream direction. Figure 2 (with the Problem Statement, at the beginning of this report) shows general locations along the Hanford Reach of the Columbia River. Results are tabulated by Hanford River Mile (HRM) location. HRM zero is at Vernita Bridge. Upstream of Vernita, HRMs are approximated and are displayed as minus (–) numbers. The location information includes position across the river from the Hanford side of the river, as noted at the bottom of each sheet of Table 2, beginning on the next page.

-22.5 opp at Beverly	-22.5 opp	10.0	
at Beverly	11	-19.0 opp	<b>-6.0</b> opp
at Deveny	duplicate	basalt xing	
<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>
8.5±1.9	ob	16.±68.	-3.6±2.0
8.5±1.9	ob	-0.5±2.3	-3.6±2.0
pl	ob	4.2±16.3	nd
(4)	(95)	(5)	(4)
(~50*)	(~50*)	(mh)	(mh)
$0.0{\pm}0.0$	ob		
2.0±1.1*	2.0±1.1*	mh	mh
(nd)	(nd)	(nd)	(nd)
50.873'	50.873'	48.397'	37.710'
57.010'	57.010'	55.391'	51.929'
2y0915a	2y0915b	171417a	171418a
	8.5±1.9 pl (4) (~50*) 0.0±0.0 2.0±1.1* (nd) 50.873' 57.010' 2y0915a	8.5±1.9         ob           pl         ob           (4)         (95)           (~50*)         (~50*)           0.0±0.0         ob           2.0±1.1*         2.0±1.1*           (nd)         (nd)           50.873'         50.873'	$\begin{array}{c cccccc} 8.5\pm 1.9 & \text{ob} & -0.5\pm 2.3 \\ \hline \text{pl} & \text{ob} & 4.2\pm 16.3 \\ \hline (4) & (95) & (5) \\ \hline (-50^*) & (-50^*) & (\text{mh}) \\ \hline 0.0\pm 0.0 & \text{ob} & \\ \hline 2.0\pm 1.1^* & 2.0\pm 1.1^* & \text{mh} \\ \hline & & & \\ \hline (\text{nd}) & (\text{nd}) & (\text{nd}) \\ \hline 50.873' & 50.873' & 48.397' \\ \hline 57.010' & 57.010' & 55.391' \\ \hline 2y0915a & 2y0915b & 171417a \\ \hline \end{array}$

#### Alpha Radioactivity Extracted from Hanford Reach Sediment

\* Combination of results from two sample fractions. See Discussion.

	<i>——— downstream ———&gt;</i>			
Hanford River Mile Location:	<b>-6.0</b> Han	<b>-3.0</b> Han	+3.6 opp	+7.0 Han
Notes:	bkg check	* reference	barge xing	ds KE
	<u>pCi/L</u>	<u>pCi/L</u>	pCi/L	<u>pCi/L</u>
Non-Volatile Alpha Activity:	ob	54.±41.	84.±66.	93.±46.
Ra-224, by gamma analysis:	ob	15.3±5.9	5.1±2.0	1.3±3.4
Ra-225, by gamma analysis:	nd	9.8±8.7	19.8±16.1	23.0±10.7
(days to gamma analysis of Ra, above):	(59)	(10)	(4)	(7)
(days to EPA 900.0 alpha test, below):	(mh)	()	(106)	(mh)
EPA 900.0 alpha, calc. from gamma:			1.2±1.0	
EPA 900.0 alpha, as measured:	mh		7.7±1.0	mh
<b><u>Reference</u></b>				
(Eu-152 in sediment: pCi/g):	(nd)	(nd)	(nd)	(nd)
Latitude: North 46° + minutes:	37.526'	37.756'	38.550'	39.455'
Longitude: West 119° + minutes:	51.998'	48.053'	39.737'	35.773'
Sample/analysis number:	292009c	172208a	290711a	1x0110a

\* Nonconforming sample: Sediment preparation did not include removal of suspendable fraction.

#### location:

#### —— code key —

- number Hanford River Mile; see Fig 2.
- Han <u>Han</u>ford side of riverbed
- toH riverbed, side of island toward Hanford
- awH riverbed, side of island <u>aw</u>ay from <u>H</u>anford
- opp riverbed, side <u>opp</u>osite Hanford
- ds <u>d</u>own<u>s</u>tream of

- <u>data</u>:
- mh <u>mishandled sample: settled not shaken</u>
- ob <u>out of bounds calc: decay factor >10</u>
- nd <u>no detection</u>: peak count <0
- pl <u>peak location: nonconforming</u>
- no EPA 900.0 alpha analysis performed
- pCi/L radioactivity: <u>picocuri</u>es per <u>l</u>iter

Table 2 (Cont'd) Sheet 2 of 6.	downstream			>
Hanford River Mile Location:	+ <b>9.1</b> Han	+10.0Han	+10.5opp	+10.6toH
Notes:	N-Springs			ds D Intake
	pCi/L	pCi/L	pCi/L	pCi/L
Non-Volatile Alpha Activity:	54.±71.	6.9±5.9	1.5±2.8	168.±66.
Ra-224, by gamma analysis:	-5.9±2.0	6.9±5.9	1.5±2.8	4.9±1.9
Ra-225, by gamma analysis:	15.0±17.2	nd	nd	40.9±15.9
(days to gamma analysis of Ra, above):	(4)	(10)	(6)	(4)
(days to EPA 900.0 alpha test, below):	(mh)	()	(81)	()
EPA 900.0 alpha, calc. from gamma:			$0.0{\pm}0.0$	
EPA 900.0 alpha, as measured:	mh		1.3±0.8	
Reference				
(Eu-152 in sediment: pCi/g):	(nd)	(nd)	(nd)	(0.5)
Latitude: North 46° + minutes:	40.886'	41.496'	41.992'	42.053'
Longitude: West 119° + minutes:	33.895'	33.204'	33.089'	32.590'
Sample/analysis number:	172912a	182307a	290615a	1x2014a

#### Alpha Radioactivity Extracted from Hanford Reach Sediment (Cont'd)

	downstream			
Hanford River Mile Location:	+10.7awH	+10.7toH	+10.9awH	+11.0toH
Notes:		pipe vent	on D Island	at D Island
	pCi/L	pCi/L	pCi/L	pCi/L
Non-Volatile Alpha Activity:	76.±44.	ob	0.7±2.5	26.±41.
Ra-224, by gamma analysis:	3.1±4.1	ob	0.7±2.5	13.3±4.8
Ra-225, by gamma analysis:	18.3±10.1	nd	nd	3.3±9.0
(days to gamma analysis of Ra, above):	(8)	(85)	(5)	(9)
(days to EPA 900.0 alpha test, below):	()	(mh)	(83)	()
EPA 900.0 alpha, calc. from gamma:			$0.0{\pm}0.0$	-
EPA 900.0 alpha, as measured:		mh	1.3±0.5	
<b><u>Reference</u></b>				
(Eu-152 in sediment: pCi/g):	(nd)	(3.3)	(0.4)	(0.6)
Latitude: North 46° + minutes:	42.183'	42.134'	42.208'	42.122'
Longitude: West 119° + minutes:	32.502'	32.472'	32.328'	32.377'
Sample/analysis number:	181214a	2y1013b	271813a	182309a

#### location:

#### - code key

- number Hanford River Mile; see Fig 2.
- Han <u>Han</u>ford side of riverbed
- toH riverbed, side of island toward Hanford
- awH riverbed, side of island <u>aw</u>ay from <u>H</u>anford
- opp riverbed, side opposite Hanford
- ds <u>d</u>own<u>s</u>tream of

<u>data</u>:

 $mh \qquad \underline{m} is \underline{h} and led sample: settled not shaken$ 

- ob <u>out of bounds calc: decay factor >10</u>
- nd <u>no detection: peak count <0</u>
- pl <u>p</u>eak <u>l</u>ocation: nonconforming
- no EPA 900.0 alpha analysis performed
- pCi/L radioactivity: picocuries per liter

Table 2 (Cont'd) Sheet 3 of 6.	3 of 6. <i>downstream —</i> >				
Hanford River Mile Location:	+11.0Han	+11.1opp	+11.1awH	+11.1awH	
	pCi/L	pCi/L	pCi/L	pCi/L	
Non-Volatile Alpha Activity:	6.4±4.9	ob	-5.3±4.9	ob	
Ra-224, by gamma analysis:	6.4±4.9	ob	-5.3±4.9	ob	
Ra-225, by gamma analysis:	nd	nd	nd	nd	
(days to gamma analysis of Ra, above):	(9)	(16)	(9)	(17)	
(days to EPA 900.0 alpha test, below):	()	(52)	(80)	()	
EPA 900.0 alpha, calc. from gamma:		$0.0{\pm}0.0$	-0.0±0.0		
EPA 900.0 alpha, as measured:		1.0±0.6	1.0±0.6		
<u>Reference</u>					
(Eu-152 in sediment: pCi/g):	(nd)	(nd)	(1.4)	(0.6)	
Latitude: North 46° + minutes:	42.052'	42.590'	42.372'	42.432'	
Longitude: West 119° + minutes:	32.295'	32.246'	32.238'	32.146'	
Sample/analysis number:	182308a	181215a	182414a	181216a	

## Alpha Radioactivity Extracted from Hanford Reach Sediment (Cont'd)

		— downs	tream ——	>
Hanford River Mile Location:	+11.4awH	+12.0Han	+12.5awH	+15.0Han
Note:	at D-Island			
	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>
Non-Volatile Alpha Activity:	>50.±32.	3.4±4.8	ob	-1.4±4.3
Ra-224, by gamma analysis:	ob*	3.4±4.8	ob**	-1.4±4.3
Ra-225, by gamma analysis:	12.5±8.0	nd	nd	nd
(days to gamma analysis of Ra, above):	(22)	(9)	(17)	(8)
(days to EPA 900.0 alpha test, below):	()	()	(52)	(81)
EPA 900.0 alpha, calc. from gamma:			$0.0{\pm}0.0$	-0.0±0.0
EPA 900.0 alpha, as measured:			1.5±0.6	$0.4{\pm}0.7$
<b><u>Reference</u></b>				
(Eu-152 in sediment: pCi/g):	(0.4)	(0.5)	(nd)	(nd)
Latitude: North 46° + minutes:	42.705'	42.909'	43.622'	42.433'
Longitude: West 119° + minutes:	31.862'	31.733'	31.289'	28.887'
Sample/analysis number:	172914a	191711a	172915a	190616a

\* The ob activity is 134±59. \*\* The ob activity is 116±23, allowing an EPA 900.0 alpha calculation.

	code key		
<u>locatio</u>	<u>n</u> :	<u>data</u> :	
number	Hanford River Mile; see Fig 2.	mh	mishandled sample: settled not shaken
Han	Hanford side of riverbed	ob	out of bounds calc: decay factor >10
toH	riverbed, side of island toward Hanford	nd	<u>no</u> <u>d</u> etection: peak count <0
awH	riverbed, side of island <u>away</u> from <u>Hanford</u>	pl	peak location: nonconforming
opp	riverbed, side opposite Hanford	_	no EPA 900.0 alpha analysis performed
ds	downstream of	pCi/L	radioactivity: <u>picocuries</u> per <u>liter</u>

Table 2 (Cont'd) Sheet 4 of 6.		>		
Hanford River Mile Location:	+15.0Han	+15.4Han	+16.0Han	+16.3toH
Note:	duplicate			
	<u>pCi/L</u>	pCi/L	<u>pCi/L</u>	pCi/L
Non-Volatile Alpha Activity:	ob	ob	-1.3±6.0	ob
Ra-224, by gamma analysis:	ob	ob*	-1.3±6.0	ob**
Ra-225, by gamma analysis:	nd	nd	nd	nd
(days to gamma analysis of Ra, above):	(57)	(44)	(10)	(17)
(days to EPA 900.0 alpha test, below):	(81)	(59)	()	(56)
EPA 900.0 alpha, calc. from gamma:	-0.0±0.0	$0.0{\pm}0.0$		$0.0{\pm}0.0$
EPA 900.0 alpha, as measured:	$0.4{\pm}0.7$	3.7±0.8		1.9±0.7
<u>Reference</u>				
(Eu-152 in sediment: pCi/g):	(nd)	(nd)	(1.2)	(nd)
Latitude: North 46° + minutes:	42.433'	42.162'	41.699'	41.498'
Longitude: West 119° + minutes:	28.887'	28.581'	27.940'	27.149'
Sample/analysis number:	190616b	172916b	1823x_a	170214a

#### Alpha Radioactivity Extracted from Hanford Reach Sediment (Cont'd)

\* The ob activity is 1391±4482, allowing an EPA 900.0 alpha calculation. \*\* The ob activity is 6±23.

	downstream				
Hanford River Mile Location:	+18.0Han	+19.3Han	+ <b>20.0</b> Han	+20.3Han	
Note:		ds F-Reactor			
	pCi/L	pCi/L	pCi/L	pCi/L	
Non-Volatile Alpha Activity:	ob	16.±39.	1.2±7.5	6.3±6.8	
Ra-224, by gamma analysis:	ob*	4.4±6.9	1.2±7.5	6.3±6.8	
Ra-225, by gamma analysis:	nd	2.8±8.0	nd	nd	
(days to gamma analysis of Ra, above):	(25)	(11)	(11)	(11)	
(days to EPA 900.0 alpha test, below):	(71)	(73)	(72)	()	
EPA 900.0 alpha, calc. from gamma:	$0.0{\pm}0.0$	0.8±2.3	$0.0{\pm}0.0$		
EPA 900.0 alpha, as measured:	3.1±0.8	5.3±0.8	$2.2 \pm 0.8$		
Reference					
(Eu-152 in sediment: pCi/g):	(0.3)	(nd)	(nd)	(nd)	
Latitude: North 46° + minutes:	40.190'	39.481'	39.199'	38.920'	
Longitude: West 119° + minutes:	27.344'	25.889'	25.065'	25.785'	
Sample/analysis number:	182310b	190607a	182311a	152915a	

\* The ob activity is 225±99, allowing an EPA 900.0 alpha calculation.

#### location:

#### \_\_\_\_\_ code key \_\_\_\_

- number Hanford River Mile; see Fig 2.
- Han <u>Han</u>ford side of riverbed
- toH riverbed, side of island <u>toward H</u>anford
- awH riverbed, side of island <u>away</u> from <u>H</u>anford
- opp riverbed, side <u>opp</u>osite Hanford
- ds <u>d</u>ownstream of

- data:
- mh <u>mishandled sample: settled not shaken</u>
- ob <u>out of bounds calc: decay factor >10</u>
- nd <u>no detection: peak count <0</u>
- pl <u>p</u>eak <u>l</u>ocation: nonconforming
- no EPA 900.0 alpha analysis performed
- pCi/L radioactivity: <u>picocuri</u>es per <u>liter</u>

Table2 (Cont'd) Sheet 5 of 6.		>		
Hanford River Mile Location:	+ <b>20.6</b> awH	<b>+21.0</b> awH	+21.1toH	+22.0awH
	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>
Non-Volatile Alpha Activity:	47.±50.	-0.3±6.0	14.9±6.1	39.±43.
Ra-224, by gamma analysis:	5.3±3.6	-0.3±6.0	14.9±6.1	4.0±6.2
Ra-225, by gamma analysis:	10.4±11.7	pl	nd	8.7±9.2
(days to gamma analysis of Ra, above):	(7)	(10)	(10)	(10)
(days to EPA 900.0 alpha test, below):	(39)	(63)	()	(57)
EPA 900.0 alpha, calc. from gamma:	12.±14.	-0.0±0.0		4.9±5.2
EPA 900.0 alpha, as measured:	1.1±0.8	$2.7{\pm}0.7$		1.8±0.7
Reference				
(Eu-152 in sediment: pCi/g):	(nd)	(nd)	(0.2)	(nd)
Latitude: North 46° + minutes:	38.780'	38.289'	38.241'	37.967'
Longitude: West 119° + minutes:	25.182'	24.626'	24.765'	24.655'
Sample/analysis number:	1906x_a	172114a	190611a	190613a

Alpha Radioactivit	y Extracted from Hanford Reach Sediment (	Cont'd)

	downstream				
Hanford River Mile Location:	+23.0Han	+25.0opp	+25.0Han	+26.0Han	
	pCi/L	pCi/L	<u>pCi/L</u>	pCi/L	
Non-Volatile Alpha Activity:	45.±43.	-0.8±2.0	16.±34.	14.4±5.7	
Ra-224, by gamma analysis:	15.0±6.0	-0.8±2.0	5.0±1.9	14.4±5.7	
Ra-225, by gamma analysis:	7.6±9.2	nd	2.8±8.0	nd	
(days to gamma analysis of Ra, above):	(10)	(4)	(4)	(10)	
(days to EPA 900.0 alpha test, below):	()	(mh)	(mh)	(74)	
EPA 900.0 alpha, calc. from gamma:				$0.0{\pm}0.0$	
EPA 900.0 alpha, as measured:		mh	mh	-0.1±0.7	
Reference					
(Eu-152 in sediment: pCi/g):	(1.1)	(0.6)	(1.7)	(0.7)	
Latitude: North 46° + minutes:	37.077'	35.844'	35.771'	34.850'	
Longitude: West 119° + minutes:	24.612'	22.897'	23.074'	34.356'	
Sample/analysis number:	171508a	171511a	171510a	182312a	

#### location:

– code key -

- number Hanford River Mile; see Fig 2.
- Han <u>Han</u>ford side of riverbed
- toH riverbed, side of island <u>toward H</u>anford
- awH riverbed, side of island <u>away</u> from <u>H</u>anford
- opp riverbed, side opposite Hanford
- ds <u>d</u>own<u>s</u>tream of

#### <u>data</u>:

- mh <u>mish</u>andled sample: settled not shaken
- ob <u>out of bounds calc: decay factor >10</u>
- nd <u>no d</u>etection: peak count <0
- pl <u>p</u>eak <u>l</u>ocation: nonconforming
- no EPA 900.0 alpha analysis performed
- pCi/L radioactivity:  $\underline{p}ico\underline{c}ur\underline{i}es$  per <u>l</u>iter

Table 2, Sheet 6 of 6.		>		
Hanford River Mile Location:	+27.0Han	+28.0Han	+28.0Han	+32.0toH
Note:			duplicate	
	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>	<u>pCi/L</u>
Non-Volatile Alpha Activity:	15.±40.	6.0±3.8	ob	11.4±8.9
Ra-224, by gamma analysis:	4.6±5.8	6.0±3.8	ob	11.4±8.9
Ra-225, by gamma analysis:	2.5±8.6	nd	nd	nd
(days to gamma analysis of Ra, above):	(10)	(8)	(73)	(12)
(days to EPA 900.0 alpha test, below):	(75)	()	()	()
EPA 900.0 alpha, calc. from gamma:	0.6±2.1			
EPA 900.0 alpha, as measured:	0.8±0.6			
Reference				
(Eu-152 in sediment: pCi/g):	(0.6)	(1.1)	(1.1)	(nd)
Latitude: North 46° + minutes:	34.356'	33.973'	33.973'	31.159'
Longitude: West 119° + minutes:	21.192'	20.296'	20.296'	16.445'
Sample/analysis number:	190516a	193014a	193014b	193018a

		>		
Hanford River Mile Location:	+ <b>38.0</b> Han	+ <b>40.0</b> Han	+ <b>45.6</b> Han	Gable Mtn
Note:				ref. *
	pCi/L	pCi/L	pCi/L	pCi/L
Non-Volatile Alpha Activity:	ob	ob	15.3±5.0	7.3±7.1
Ra-224, by gamma analysis:	ob**	ob***	15.3±5.0	7.3±7.1
Ra-225, by gamma analysis:	nd	nd	nd	nd
(days to gamma analysis of Ra, above):	(34)	(13)	(9)	(10)
(days to EPA 900.0 alpha test, below):	()	()	(93)	()
EPA 900.0 alpha, calc. from gamma:			$0.0{\pm}0.0$	
EPA 900.0 alpha, as measured:			1.3±0.6	
Reference				
(Eu-152 in sediment: pCi/g):	(nd)	(0.4)	(nd)	(nd)
Latitude: North 46° + minutes:	26.335'	24.386'	18.876'	37.004'
Longitude: West 119° + minutes:	16.237'	16.090'	15.601'	31.565'
Sample/analysis number:	1x1913b	1x1915a	181316a	181410a

\* Nonconforming reference, sand sample: Preparation did not include removal of suspendable fraction.

----- code key -

**	The c	b ac	tivitv	is	178±553.

#### location:

#### <u>data</u>:

mhmishandled sample: settled not shakenobout of bounds calc: decay factor >10

\*\*\* The ob activity is 35±10.5.

Han <u>Han</u>ford side of riverbed

number Hanford River Mile; see Fig 2.

- toH riverbed, side of island <u>to</u>ward <u>H</u>anford
- awH riverbed, side of island <u>away</u> from <u>H</u>anford
- opp riverbed, side opposite Hanford
- ds <u>d</u>own<u>s</u>tream of

- nd <u>no d</u>etection: peak count <0
- pl <u>peak location: nonconforming</u>
- no EPA 900.0 alpha analysis performed
- pCi/L radioactivity: <u>picocuries per liter</u>

## Discussion

The exploratory aspect of this study sought an understanding of the connections between historical Hanford operations and presentday radioactivity in riverbed waters of the Hanford Reach.

The first result of this study was recognition of the importance of the Army's barge-and-tug operations on the Columbia River, in support of Hanford's Cold War mission to produce special nuclear materials for nuclear weapons. See the Results for more detail.

A report in 2002 had suggested that the Army's barge-and-tug operations might have also served as a means for large-scale radioactive waste disposal into the Hanford Reach [6]. Government agencies criticized that suggestion. They ascribed Hanford's europium-152 in the riverbed to the widely publicized pollution that had been discharged from Hanford's long-decommissioned reactors.

One objective of the present study was to distinguish precisely whether a broad pattern of radioactive pollution in the riverbed originates at the D-Reactor discharge pipes, Fig. 8, or if it originates just upstream of those discharge pipes, close to the old D-Ferry crossing. D-Reactor is shown in Fig. 12.



**Fig. 12. D-Reactor, looking north, across the river to Wahluke Slope** [DOE archive].

The distinction is as follows: The flow of the river carries most everything, including radioactivity, downstream. If Ra-225 radioactivity from Hanford is found upstream of the D-Reactor discharge, then the radioactivity must have entered the river upstream of that discharge.

The distinction between riverbed contamination coming either from the D-Reactor discharge pipes or from the D-Ferry crossing depends on their locations in the river. Sample locations are mapped in Fig 9. The results are as follows (from Table 2, Results):

#### Table 3. Outcome: Origin of Hanford Radioactivity in Riverbed.

Location	Sample #	<b>Ra-225</b> [pCi/L]
just downstream of ferry crossing, side opposite Hanford	290615a	nd
downstream of ferry crossing, upstream of discharge pipe	1x2014a	41.±16.
at discharge pipe vent	2y1013b,a	a nd*
near discharge pipe	181214a	18.±10.

<sup>k</sup> The No Detect (nd) of Sample #2y1013b analysis was checked against an analysis of #271013a that is not formally reported because it had failed the K-40 peak quality control criterion. The #271013a analysis was found to be nil (pl) for Ra-225.

TRAC compared Table 2 to locations of old Hanford structures upstream of the D-Reactor discharge pipe for other possible sources of the Ra-225 contamination in the riverbed. No likely candidates were found.

TRAC concludes that the <u>origin</u> of this Ra-225 contamination in the riverbed is <u>upstream of the D-Reactor discharges</u>. The outcome of this test is significant to greater than 95% confidence, based on gamma counting results for Sample #1x2014a.

#### Increasing radioactivity in riverbed water, from historic U-233 production.

After the Second World War, Hanford began to produce uranium-233 for tactical battlefield nuclear weapons —"mini-nukes" [6]. The *Davy Crockett* was one such weapon. See Fig. 13, on the next page.

As soon as radioactive U-233 was produced in Hanford's nuclear reactors, it began to decay. The U-233 decay chain is shown in Fig. 6. Part of that decay chain is summarized, as follows:

U-233 <u>159,000 years</u> Th-229 <u>7300 years</u> Ra-225 <u>25 days</u> ...  $4\alpha + 2\beta$ ... -> Bi-209

Uranium-233 has a halflife of 159,000 years; so half the U-233 in the riverbed will have decayed away after 159,000 years.

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Fig. 13. Davy Crockett Rocket, deployed 1961 to 1972, also fitted for bazookas; explosive equal to 1,000 tons of TNT. [Display at National Atomic Museum]

The fraction of Hanford's U-233 that decays into Th-229 <u>in one year</u> is 0.00043%. [The equation is:  $1 - (1/2)^{(1)}(1) = 0.0000044$ . The "^" symbol means that what follows is the exponent of what precedes. See the Glossary for more explanation.] This is the annual rate that Th-229 is added to the riverbed, because the U-233 that decays away becomes Th-229.

Let's suppose that Hanford's waste U-233 entered the riverbed in the early 1960s. Then about 40 years have passed since the U-233 began to decay in the riverbed. The amount of Th-229 that has been added to the riverbed (from U-233 decay) over these last 40 years is: 40 times the annual addition of Th-229:  $40 \times 0.00044\% = 0.017\%$ .

Because the passage of these 40 years is so much less than the 7,300 year halflife of Th-229, only a tiny percentage of the Th-229, that has already been created by U-233 decay, has yet decayed away. {The percentage of added Th-229 that has decayed away (relative to the amount of U-233 in the riverbed) over those 40 years, is about  $0.017\% \times [1 - (1/2)^{(40 \text{ years}/7,300 \text{ years})] = 0.000064\%$ . The 0.017% added Th-229 per year is 265 times this amount of Th-229 that decays away per year.}

#### Th-229 is increasing 265 times as fast as it is decaying away.

Because the annual addition of Th-229 (from decay of U-233) is 265 times greater than the annual loss (by decay of the accumulated Th-229), Th-229 is accumulating in the riverbed at almost the same rate it was at the beginning: about 0.017% of U-233 is added to the Th-229 in the riverbed each year.

The radioactivity of Th-229 in the riverbed is, thus, 40 times what it was about 40 years ago. The radioactivity of the decay product of Th-229, namely Ra-225, is also 40 times what *it* was 40 years ago, because the decay chain from Ra-225 comes into radioactive equilibrium with Th-229 in the time of *its* halflife: 25 days.

After 4,000 years will have passed, the amount of Ra-225 in the riverbed will be about one hundred times the present amount of Ra-225. (4000 years is 100 times the 40 years that have already passed. The amount of Ra-225 in the riverbed will have increased by the same 0.017% of U-233 in the riverbed, in each of those years.)

#### Ballpark estimate of the amount of U-233 in the riverbed

*—Roughly how much U-233 currently resides in the Hanford Reach riverbed to account for the Ra-225 reported in the Results?* 

For this estimate, assume the U-233 currently in the riverbed has been there for 40 years. Further assume the average value of the Ra-225 detections (5 pCi/L, from Table 2) applies to a *slice* of Hanford Reach riverbed water, say about 100 Km long, one Km wide, and 1 m deep. [The calculation is 5 pCi/L X (10^-12 Ci/pCi) X 100,000 m X 1,000 m X 1 m X (10^6 L/m^3) = 0.5 Ci of Ra-225. This calculation neglects exchange of water between the riverbed and the river above, and so might be an under-estimate.] Then about half a curie of Ra-225 would presently be in the riverbed water of the Hanford Reach.

How much U-233 would it take to create 0.5 Ci of Ra-225 over 40 years? The answer is on the order of ten kilograms. (Here's the ballpark calculation, in metric units for ease: Assume Ra-225 is in decay equilibrium with Th-229, the presentday U-233 activity must be the presentday Ra-225 activity of 0.5 Ci multiplied by the 265 factor, calculated above. That is 130 Ci of U-233 in the riverbed. The original definition of one curie was the radioactivity of one gram of Ra-226, having a halflife of 1,600 years. That halflife is 1% of the 159,000 year halflife of U-233. So 100 grams of U-233 have the same activity as one gram of Ra-226, namely one curie. 13,000 grams of U-233 have an activity of 130 Ci. 13,000 grams = 13 kg. This answer is rounded to 10 kg.)

From the perspective of aquatic biota living in the riverbed, 10 kg of U-233 probably was not much when it arrived about 40 years ago. But as time passes and

## 10 kilograms of U-233 in the riverbed would account for the Ra-225 reported in this study.

alpha radioactivity in riverbed water steadily increases, 10 kg of U-233 will come to be a lot.

Another perspective on 10 kg of U-233 is its volume: Ten kilograms of metallic U-233 would have a volume of half a liter (one pint). One pint of U-233, spread over miles of riverbed sediments does not seem like much.

Yet another perspective of 10 kg of U-233 is comparison to Hanford's total production of U-233, which is classified. One run, of the "jumbo" KE- and KW-Reactors in 1968, involved delivery of 460 kg of U-233 to the Atomic Energy Commission [16]. If we assume Hanford's total production of U-233 was roughly twenty of those jumbo reactor runs, Hanford's total output of U-233 might have been roughly 10,000 kg of U-233. Ten kilograms in the riverbed would then represent about 0.1% of all the U-233 Hanford produced (= 10 kg / 10,000 kg).

The implication of the present study that a small fraction of one percent of all Hanford's U-233 product somehow found its way onto the surfaces of sediment grains in the Columbia Riverbed demands some plausible explanation. Why would so much, readily dispersible U-233 have been disposed into the river? How could such disposal have occurred?

We now take a closer look at the patterns of Ra-225 in the riverbed to gain an understanding of disposal means that might account for these observations. Then this Discussion will move on to consider large-scale processes at Hanford that involved kilogram quantities of U-233, recognized challenges that might have necessitated disposal of kilogram quantities of U-233 as waste, reasons to dispose of that U-233 waste to some unusual location, other than to underground tanks or to the soil (cribs or trenches), and means for that disposal.

#### Patterns of Ra-225 in the riverbed

The central radiological result of this study is **41.±16.** pCi/L of Ra-225 [at location +10.6toH, Sample # 1x2014a]. This result identifies the origin of a plume of riverbed pollution, downstream of the D-Ferry crossing but upstream of the D-Reactor discharge. With the present data, that plume of Ra-225 activity in riverbed water can already be seen for one mile downstream to HRM 11.4 [Location +11.4awH, Sample #172914a].

That plume downstream of the D-Ferry crossing is distinct from a lesser positive Ra-225 result of 20.±16. pCi/L [Location +3.6 opp, Sample #290711a], obtained 7.9 miles upstream of the D-Ferry crossing. That lesser positive result is just downstream of the B-Ferry crossing. These positive Ra-225 results, just downstream of two Hanford ferry crossings, along with the results of historical investigations reported in the Results, evidence a pattern of Hanford waste disposals from the barge-and-tug transportation systems operated by the Army Engineer Corps for DOE.

Downstream of the D-Ferry crossing (at HRM +10.5), the Army Engineer Corps used the F-Ferry crossing (at HRM +17.9) on the east end of the White Bluffs to support AAA and missile sites [15]. See Fig. 10. The radiology of the riverbed near the White Bluffs / F-Reactor ferry crossing is obscured by silt in the river, from the sagging and sliding of the bluffs into the river. TRAC did not detect Ra-225 along this stretch of the Hanford Reach, perhaps because the silt might have diluted Hanford's waste.

Sample 190607a [Location +19.3 Han], downstream of F-Reactor at HRM +19, provides the next, although weak, evidence of Ra-225 in the riverbed:  $Ra-225 = 3.\pm 8$ . pCi/L.

Continuing farther downstream, Table 2 provides evidence of Ra-225 along the six and a half mile stretch of the river from HRM +20.6 to HRM +27. This evidence is summarized in Table 4, below.

HRM Location	<u>Sample #</u>	<b>Ra-225</b> [pCi/L]
+20.6, Hanford side of mid slough channel	1906x_a	10.±12.
+22.0, Hanford side of main river channel	190613a	9. ±9.
+23.0, Hanford side of river	171508a	8. ±9.
+25.0, Hanford side of main river channel	171510a	3. ±8.
+27.0, Hanford side of river	190516a	2. ±9.

Table 4. Cluster of Ra-225 Detections Downstream of F-Reactor.

The individual, positive results in Table 4 are weak, so details of the pattern of Ra-225 are unclear. But taken together, these 5 positive results suggest a plume of Ra-225 extending past the east side of F-Slough and downstream below Hanford Slough. Negative results from the east side of the river [Location +25.opp, Sample 171511a] are attributed to sediment dilution with silt from East White Bluffs. This pattern is not defined well enough to be sure its upstream origin is the White Bluffs F-Ferry crossing rather than the F-Reactor discharge pipe.

TRAC found two other positive results, just downstream from two Hanford Reactor discharge pipes. See Table 5:

Table 5. Ra-225 Detections Downstream	of KE/KW- and N-Reactors.
---------------------------------------	---------------------------

HRM Location	<u>Sample #</u>	<b>Ra-225</b> [pCi/L]
+7.0, K-Springs, Hanford side of river	1x0110a	23.±11.
+9.1, N-Springs, Hanford side of river	172912a	15.±17.

Either or both of these positive results might be attributable to localized, radioactive groundwater seepages from the reactor areas into the riverbed, instead of flows from the old reactor cooling water discharge pipes. The Ra-225 result from K-Springs is significantly positive; the result from N-Springs is not statistically significant.

#### Possible sources of the plumes of Ra-225 in the riverbed

The Ra-225 data in this study suggest roughly 10 kg of U-233 is in the riverbed of the Hanford Reach. The patterns suggest that most of that U-233 entered the river at old Hanford ferry crossings and was readily *dispersible* rather than solid particulate. Public documents suggest those ferry crossings provided terminals for barge-and-tug operations that played an important part of Hanford's transportation system.

There are at least three types of waste streams from Hanford's U-233 processing that are consistent with the colloidal nature of the U-233 reported in the riverbed:

• Disposal of uncategorized system flushes while changing over between processes, either from Pu-239 production to U-233 production, or from U-233 production back to Pu-239 production.

For U-233 to be "clean" enough for tactical nuclear weapons (Fig. 13) Hanford limited impurities exactingly. To meet a requirement of no more than 0.5% of U-238 residue contaminating Hanford's U-233 product, Hanford's PUREX separations plant had to be flushed out for 6 weeks or longer [17], before beginning a U-233 production run. The flushing processes used industrial chemicals and "cold" non-irradiated thoria (the raw material for U-233 production) before a "hot" U-233 production run. The same sort of flushing with industrial chemicals and "cold" non-irradiated uranium (the raw material for Hanford's plutonium-239 production) must have preceded conversion of PUREX back to plutonium production after completion of a U-233 production run. That is, the "hot" thoria and U-233 still in the works at the tail end of each U-233 production run had to be flushed out. Those left over dregs of each U-233 separations run through a Hanford processing plant are one candidate for uncategorized waste now found in the riverbed.

• Avoidance of fluoridating the processing plant's waste disposal systems.

One glitch in disposing of U-233 process wastes at Hanford was the undesirability of those wastes. The

problem comes in the <u>dissolving</u>: Uranium metal will dissolve just fine in nitric acid, but thorium [the source material from which U-233 was separated] won't. Thorium just sits there in nitric acid because a thin, invisible film of oxide develops on it. Moore discovered you must add fluoride to the nitric acid to act as a catalyst, and the thorium dissolves just fine. But, fluoride is bad as a waste product; you don't want this in your waste [18].

The easy alternative might have been to route more of the troublesome U-233/thoria processing waste to cribs in central Hanford. However, once thorium and U-233 were indeed dissolved in fluorided nitric acid for processing, the thorium and U-233 were more environmentally hazardous:

# The principlal liability from any accidental release of thorium nitrate to the environs will result from the contamination of ground water in the vicinity of a spill and exposure of persons in the vicinity using the water [19].

Hanford discharges of contaminated water to the soil dominated groundwater flow under the site during Hanford's production years. During the early 1960s, Hanford's liquid waste disposals to the ground averaged about 200 gallons per second [**20**]. In comparison, the flow of the Columbia River through the site is about 1,000,000 gallons per second. The arithmetic is that the Columbia River offered 5,000 times the carrying capacity for liquid wastes that were unsuitable for disposal to groundwater. Fluorided nitrate wastes are a possible source material for river disposal.

#### • Clearing stoppages in separations processes.

Hanford processing facilities did not handle the U-233-thorium mix well. The material "caused plugging and other equipment and contamination problems within PUREX [20]." Where would the plumbers have put the waste they had to remove to clear stoppages?

Any of these three scenarios might account for the mass of U-233 evidenced in the riverbed by the detections of Ra-225 in Table 2 of this report. Any such source of U-233 in a liquid form would account for the presentday colloidal nature of radioactive contamination on riverbed sediment grains.

Hanford's known problems in separating U-233 from thorium could explain both the reason for disposal in the river and the dispersed nature of U-233 in the riverbed.

#### **Biological effects in the riverbed**

In 1998, the Washington State Department of Ecology reported signs that sediments from D-Island are significantly "ecotoxic" to freshwater amphipods living on those sediments [21]. The Department of Ecology concluded that contaminants "other than those identified" by Hanford scientists are responsible for the high mortality of amphipods living on Hanford Reach sediments.

In 2001, University of Idaho researchers reported an unusually high incidence of chromosomal anomalies in fall chinook salmon spawning in the Hanford Reach [5]. Salmon hatchlings (*alevin*) live in riverbed water, among the gravels while they absorb their egg sacs; see Fig. 1. The environmental stresses that are responsible for the high incidence of chromosomal anomalies in the wild salmon spawning in the Hanford Reach have not been identified.

Radium mimics biologically essential calcium and concentrates in the bodies of freshwater organisms [22]. Alpha activity from radioactive decay of concentrated Ra-225 might be responsible for chromosomal anomalies in Hanford Reach alevin. Such damage would show up in the adults returning to the Hanford Reach to spawn.

#### Summary of points raised in the Discussion

This study raises four points toward understanding Hanford's contribution to radioactivity in riverbed water:

(1) "Plumes" of Hanford's artificial radioactivity contaminate the riverbed for miles downstream of old ferry crossings. The extensive, wartime operations on the Hanford Reach need fuller disclosure and evaluation to better understand what is in the riverbed. (2) The important radioactivity in the riverbed is in a "colloidal" form, adhering to particles of sediment. This sticky material holds long-lived "actinides" (like U-233), until they decay to radioactive radium. Artificial U-233 is probably the main concern in riverbed colloids. U-233 could be analyzed by alpha spectrometry of colloids removed from fine sediments from the Hanford Reach.

(3) Artificial Ra-225, from U-233 decay, is probably the main radiological concern in riverbed water of the Hanford Reach.

(4) The amount of Ra-225 in riverbed water is increasing every year because of radioactive "in-growth." Depending on the actual dates of U-233 waste disposal into the riverbed during the Cold War era, the rate of Ra-225 increase in the riverbed is in the range of 2% to 4% annually.

## Ra-225 is increasing 2 – 4%, annually.

This rate of increase is linear, rather than compounding.

## Conclusions

- 1. Two or three plumes of artificial radium-225 (Ra-225) have been located in the Hanford Reach riverbed, extending downstream a few miles from old ferry crossings. Barge-and-tug terminals at those ferry crossings had served the transportation sector of wartime operations at Hanford Site.
- 2. Barges were probably the platforms from which radioactive waste from Hanford separations processes were disposed into the Columbia River, close to the ferry crossings. The separations processing waste was probably disposed into the river because it did not meet requirements for on-site disposal to underground tanks or to the ground.
- **3.** The dense liquid U-233 wastes disposed into the river probably settled into the riverbed sediments. Some of the U-233 was relatively insoluble in riverbed water and has remained over the decades, adhering to sediment grain surfaces. The amount of U-233 in the riverbed is estimated very roughly to be 10 kilograms.
- 4. Uranium-233, in the riverbed, is decaying into radioactive radium-225, which dissolves into riverbed water. The average value of detections of Ra-225 in this laboratory study is 5 pCi/L. Four alpha particles are emitted as Ra-225 steps through its radioactive decay in 25 days. Thus, the average alpha activity in the laboratory experiment is 20 pCi/L.
- 5. The amount of Ra-225 in riverbed water is increasing at a linear rate of 2 to 4%, annually (not compounding). The radioactive equilibrium activity of Ra-225 in the riverbed is more than one hundred times the current activity. Ra-225 activity in the riverbed is increasing because an intermediary isotope (thorium-229) between U-233 and Ra-225 has such a long halflife: 7,300 years. Thorium-229 is slowly "growing in" by radioactive decay, and Ra-225 is following suit.
- 6. Radium-225 is of concern in Columbia Riverbed water because radioactive radium mimics biologically essential calcium and is concentrated in fish. Radium decays emit alpha particles which are capable of damaging biological control molecules such as genes.

## Recommendations

1. The reported detection of radium-225 (and U-233) in material adhering to the surfaces of sediment particles in the Hanford Reach should be confirmed by independent means. Alpha spectrometric or mass spectrometric techniques could provide such a confirmation.

#### **Depending on that confirmation:**

- 2. An *independent* Columbia River Comprehensive Impact Assessment (CRCIA) of the Hanford Reach should be performed with emphasis on radiology and toxicology in the riverbed ecosystem.
- 3. If the results of that assessment confirm that increasing radioactivity of Ra-225 in riverbed water threatens long-term health of riverbed species, methods of mitigation should be evaluated. For example, it might be feasible to "block" radium uptake by sowing calcium (possibly as marble chips) into the riverbed. The biological implications of any remediation should be carefully examined before any remediation is undertaken.
- 4. The great disparity between the results of this study and the Department of Energy's present strategy for restoring the Columbia River corridor by 2012 should be addressed at a high organizational level.
- 5. Until the radiological and toxic reality of old Hanford operations is under control, new wastes should not be added to the problem.

Glossary	
actinide	- the name of the group of elements having atomic numbers from 89 through 104, all having chemical properties similar to the element
1 .	actinium.
alevin	- first hatchling stage of salmon. Alevin live in the riverbed gravels while they absorb their yolk sacs.
alpha	- a high-speed helium nucleus (two protons and two neutrons) emitted
aipila	from certain nuclear decays.
background	<ul> <li>the actual level of radioactivity at some location, minus the radioactivity contributed by a facility like Hanford. Background radioactivity is the level of radioactivity that would exist around Hanford if Hanford did not contribute radioactivity to its surroundings.</li> </ul>
beta	- a high-speed electron emitted from certain nuclear decays.
channel	- See "spectrometer."
colloid	- a large molecule having a weak electric charge
DOE	- U.S. Department of Energy. Hanford Site is owned and operated by
	DOE, as its Richland ("DOE/RL") Washington office.
EPA	- U.S. Environmental Protection Agency.
gamma	<ul> <li>a photon emitted from a nucleus during radioactive decay. A radioactive decay is usually thought of involving some main emission, such as release of an alpha particle or a beta particle. A photon is often emitted also, matching the total energy before the decay to the total energy after the decay. The region of the energy spectrum in which most of these photons that emitted from nuclei undergoing radioactive decay is in the range of 50 – 2,000 KeV. Radioactive decays of nuclei are often accompanied by changes in the orbits of electrons surrounding the decaying nuclei. Some of those energy changes are in the x-ray range of the spectrum: 2 – 100 KeV.</li> </ul>
GAP	- the Government Accountability Project. See inside front cover.
g, gram	- One 28th of an ounce. One thousand grams is 2.2 pounds.
Hanford Reach	<ul> <li>51 mile stretch of the Columbia River passing through Hanford Site, in southeastern Washington, now the Hanford Reach National Monument.</li> </ul>
HRM	- Hanford River Mile. Miles posted on white "<"-shaped signs, located above the highwater line, on the Hanford side of the river, with Mile zero at Vernita Bridge and increasing downstream.
KeV	- <u>kiloe</u> lectron- <u>v</u> olt, a unit of energy. One KeV is the energy an electron gains as it drops across 1000 volts. The energy an electron gains as it drops across a spark made across the terminals of a 12 volt car battery is $0.012$ KeV.
L, liter	- 1.06 quarts.

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North	- North Latitude. Latitudes at Hanford Site are 64° north of the equator. The additional minutes northward are listed.
pCi, picocurie	- a measure of radioactivity. One picocurie is one nuclear decay in 27 seconds.
photon	- one unit of electromagnetic mass/energy. In the range of decay energies of atomic nuclei, the x-rays and gamma-rays emitted behave like particles, called photons.
polar	- compounds having a displaced electric charge, akin to the poles of a magnet. Polar molecules stick together and to other molecules —also akin to the magnet analogy.
spectrometer	- as used in this report, a device that measures the energy (in KeV) of photons (from a sample) impinging on a photon detector. That energy corresponds to a particular <i>channel number</i> . Continued:
spectrum	- See spectrometer. The relative number of photons accumulated in every energy channel of a spectrometer is the energy spectrum of a sample
TRAC	- The RadioActivist Campaign. See About the Author.
West	- West Longitude. Longitudes at Hanford Site are 119° west of the prime meridian. The additional minutes westward are listed.
^	- exponential. Example: $(2)^{(3)} = 2^3 = 8$ .

## Appendix - Algal Mat Data

The study plan included sampling algal mat on the rocks in the Hanford Reach riverbed and analyzing the algae for artificial, photon-emitting radioactivity. The purpose was to see whether algae at the bottom of the Hanford Reach food chain are contaminated with radioactive waste associated with uranium-233 in the riverbed.

TRAC found insufficient algae to scrape off the bottom sides of rocks in the riverbed. Therefore, the plan was modified, and algae were scraped off the upper surfaces of cobbles and boulders in the riverbed. The algae were rinsed over a 2 mm sieve to remove fine sediments.

These algal samples were probably more subject to influences of the river proper than of the riverbed.

Results were negative. Values for natural uranium (as U-238) and cesium-137 (Cs-137) are listed in Table A, on the next page. Cesium-137 is a product of nuclear fission. The values in Table A are attributed to worldwide fallout from atmospheric testing of nuclear weapons in the 1950s and 1960s.

These negative results invite further investigation of other, more useful biological sample media.

Table A.	<i>downstream &gt;</i>			
Hanford River Mile Location:	-22.5opp	<b>-6.0</b> Han*	- <b>6.0</b> Han*	-6.0 opp
	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)
Natural Uranium (as U-238)	3.1	1.4	2.1	2.1
Cs-137	0.15	0.14	0.13	0.21
Reference		split*	split*	
Latitude: North 46° + minutes:	50.873'	37.526'	37.526'	37.715'
Longitude: West 119° + minutes:	57.010'	51.998'	51.998'	51.962'
Sample/analysis number:	2y0914a	292010a	292010b	292011a

#### Gamma Radioactivity in Upper Algal Mat

\* Sample #292010 was split before analyses "a" and "b".

	<i>downstream &gt;</i>			>
Hanford River Mile Location:	– <b>1.0</b> Han	-1.0 opp	+1.0Han	+1.0 opp
	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)
Natural Uranium (as U-238)	2.5	2.8	2.6	2.8
Cs-137	0.10	0.17	0.17	0.11
<b><u>Reference</u></b>				
Latitude: North 46° + minutes:	38.076'	38.195'	38.788'	38.978'
Longitude: West 119° + minutes:	45.645'	45.667'	42.855'	42.965'
Sample/analysis number:	290707a	290708a	290709a	290710a

	<i>downstream &gt;</i>			
Hanford River Mile Location:	+3.6opp	+10.5opp	+10.6toH	+10.7awH
	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)	pCi/g(dry)
Natural Uranium (as U-238)	2.3	1.9	2.1	2.3
Cs-137	0.08	nd	0.17	0.22
Reference				
Latitude: North 46° + minutes:	38.550'	41.992'	42.058'	42.182'
Longitude: West 119° + minutes:	39.737'	33.089'	32.569'	32.472'
Sample/analysis number:	290712a	290616a	2906XVa	292014a

– code key -

location: number Hanford River Mile; see Fig 2.

Han <u>Han</u>ford side of riverbed

toH riverbed, side of island toward Hanford

awH riverbed, side of island <u>away</u> from <u>Hanford</u>

opp riverbed, side <u>opp</u>osite Hanford

<u>data</u>: nd

nd pCi/g(dry) <u>no d</u>etection: peak count <0 radioactivity: <u>picocuri</u>es per gram dry weight

## **References and Notes**

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- [2] M.S. Gerber, D.W. Harvey, et al, September 2002, "The Manhattan Project and Cold War Eras, plutonium production at the Hanford Site, December 1942-1990," <www.hanford.gov/doe/culres/mpd/sec5.htm>, pp.6-7.
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- [4] T.M. Poston, R.W. Hanf, at al, September 2002, *Hanford Site Environmental Report* for Calendar Year 2001, PNNL-13910, Pacific Northwest National Laboratory, Richland, WA, p.1.1.
- [5] J. Nagler, J. Bouma, et al, January 2001, *Environmental Health Perspectives*, "High Incidence of a Male-Specific Genetic Marker in Phenotypic Female Chinook Salmon from the Columbia River," vol. 109(1), pp. 67-9. Salmon carcasses were sampled from upstream of Vernita.
- [6] N. Buske, August 2002, *Hanford Radioactivity in Salmon Spawning Grounds --quality, extent, and some implications*, Government Accountability Project, Seattle, WA. That report describes the sediment sampling procedure, pp. 12-13, and sediment samples that are the source materials for this study. Radiological analyses of the sediments are included in that report. Appendix 1 is a working summary of the history of uranium-233 production at Hanford. The gamma analytical technique is described in App. 2.
- [7] Columbia River water was collected from Vernita, near the upstream end of the Hanford Reach, in December 2002 and passed through 8 μm filter paper.
- [8] H.L. Krieger and E.L. Whittaker, August 1980, Prescribed Procedures for Measurement of Radioactivity in Drinking Water, Sec. 1, EPA-600/4-80-032, National Technical Information Service, Springfield, VA. Environmental Protection Agency, 7 December 2000, Federal Register, Vol. 65, No. 236, "National Primary Drinking Water Regulations; Radionuclides; Final Rule, Part II," 40 CFR Parts 9, 141, and 142. See pp. 76709 - 76712, with Table I-1.
- [9] Dust blown off Hood Canal, Washington was collected on an air filter and then analyzed for gamma radioactivity twice, 24 hours apart. The difference spectrum (after CPW transformation and routine 5-point smoothing) was a surrogate spectrum of Ra-224 by descendant Pb-212, assuming equilibrium of the decay chain beginning with Ra-224. See Fig. 14, below.

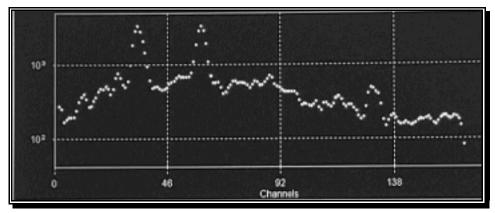


Fig. 14. Ra-224 energy spectrum used in this study, CPW-transformed, log scale.

The activity of the Ra-224 spectrum (assuming equilibrium with Pb-212) was obtained by matching the Pb-212 peak areas to TRAC's equilibrated thorium decay spectrum.

- [10] The level scheme for Bi-213 decay to Po-213 is shown in C.M. Lederer and V.S. Shirley, eds., 1978, *Table of Isotopes*, 7th ed., Wiley-Interscience, New York, NY, p.1363; and partly confirmed with the NUDAT database at <www.nndc.bnl.gov>. Intensity = 16% for the 440 KeV emission is given by R.C. Weast, ed, 1988, *CRC Handbook of Chemistry and Physics*, CRC Press, Inc., Boca Raton, FL, p. B-411.
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- [12] \_\_\_\_\_, December 1975, *Waste Management Operations, Hanford Reservation*, ERDA-1538-Vol. 2, National Technical Information Service, Springfield, VA, p.II.1-D-3.
- [13] Discovery of the D-Reactor ferry/barge crossing is described in [6], pp.26-30.
- [14] The base map for this sketch is S.J. Hope and R.E. Peterson, September 1996, *Chromium in River Substrate Pore Water and Adjacent Groundwater: 100-D/DR Area, Hanford Site, Washington*, BHI-00778, Rev.0, Bechtel Hanford, Inc., Richland, WA, Figure 4-1. Two GPS positions were used to place the sampling locations onto the map and to revise the approximate positions of the D/DR discharge pipelines, as follows: A location ten meters offshore of the middle of the D/DR Intake structure was positioned at [North 46° 41.679', West 119° 32.902']. The location of the TLD station at the upstream end of D-Island is the location of Sample #271813a [+10.9awH, at North 46° 42.208', West 119° 32.328'].
- [15] R.H. Ruby and J.A. Brown, 1974, *Ferryboats on the Columbia River, including the bridges and dams*, Superior Publishing Company, Seattle, WA, p. 87, caption to photo of "The Tug, *Doris.*"
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- [17] M.K. Harmon, 29 September 1965, "Thorium-Uranium-233 Processing," General Electric Company, Richland, WA, 5 pp.

- [18] \_\_\_\_\_, 16 November 2000, Partial Response to "Freedom of Information Act Request (RL 2000-0077)" dated 7 June 2000, thorium-related documents found in Box No. 145544, handwritten notes "early 1950s."
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- [21] D.A. Delistraty and J. Yokel, 1998, Bull. Environ. Contam. Toxicol., vol. 61, "Ecotoxicity of River and Spring Sediment Along the Hanford Reach," Springer-Verlag New York, Inc., New York, NY, pp. 754-761.
- [22] \_\_\_\_\_, March 1976, Effects of Ionizing Radiation on Aquatic Organisms and Ecosystems, Technical Reports Series No. 172, International Atomic Energy Agency, Vienna, Austria, 131 pp. From Table V, the range of natural Ra-226 in whole, freshwater fish is 1.0 - 3.5 pCi/g(wet) = 1000 - 3500 pCi/L(wet). The range of natural Ra-226 in fresh water is 0.01 - 3 pCi/L, from Table IV. Mid-values are 2000 pCi/L and 0.2 pCi/L, respectively. The biological concentration factor for radium in freshwater fish is, thus, on the order of (2000 pCi/L)/(0.2 pCi/L) = 10,000. Five picocuries per liter of Ra-225 in riverbed water might then translate into 50,000 pCi/L in salmon alevin in the riverbed. Because Ra-225 decay products are not volatile, the alevin might absorb most of the four alpha particles resulting from the decay of Ra-225. If so, the alevin might have an internal alpha activity on the order of 200,000 pCi/L.

#### About the Author

Norm Buske directs The RadioActivist Campaign (TRAC). He has masters degrees in physics from the University of Connecticut and in oceanography from the Johns Hopkins University. Norm has received a certificate of honor for his scientific and technical investigations of the environmental consequences of nuclear weapons production in the United States and Russia. He has conducted non-governmental, in-field, radiological investigations around nuclear weapons facilities since 1983. He operates TRAC's in-house radiological laboratory.

The RadioActivist Campaign, based in Belfair, Washington is a scientific campaign of the Tides Center of San Francisco. TRAC's mission is to measure and publicize radioactivity around nuclear facilities to promote site accountability and public safety. TRAC publicizes its radiological results through reports and brochures, by presentations to schools, public-interest organizations, and governmental agencies, and by producing educational videos for distribution with public-interest organizations.

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## About This Report

This technical study was conducted under a contract with the Government Accountability Project (GAP). For four years, GAP's Nuclear Oversight Campaign and scientist Norm Buske have conducted independent scientific investigations —focused radiological sampling and analyses— that are revealing significant radioactive and chemical contamination throughout the Hanford Site and in the Columbia River shore.

Buske's important discovery of previously unreported radioactive contamination of the Hanford Reach riverbed where critical salmon stocks spawn cries out for the government to conduct comprehensive testing of the Hanford Reach riverbed. Buske's work augments GAP's traditional methodology, which involves working with whistleblowers to reveal problems and wrongdoing at nuclear sites.

GAP has focused on Hanford for 15 years and set up an office in Seattle in 1992 to monitor the site more closely. Over the past 11 years, GAP has helped to develop several components of a citizen oversight model, including inside information (GAP's whistleblowers), a structure for alternative dispute resolution for employees (the Hanford Joint Council), a forum enabling government, corporate, and citizen interaction (the Hanford Advisory Board), and a coalition of citizen's groups (the Hanford Public Interest Network), and the new Hanford Roundtable.. Tom Carpenter is the Director of GAP's Nuclear Oversight Campaign.

GAP's mission is to protect the public interest by promoting government and corporate accountability through advancing occupational free speech and ethical conduct, defending whistleblowers, and empowering citizen activists. Founded in 1977, GAP is a non-profit, public interest organization and law firm.