Hanford Radioactivity in Salmon Spawning Grounds

--quality, extent, and some implications

REPORT
to the
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Summary

Sixty percent of the Columbia Riverbed where important salmon stocks spawn is contaminated with previously unreported radioactivity from old Hanford Site, nuclear weapons production wastes.

This radioactive contamination of the Hanford Reach riverbed evidently results from disposal of solid radioactive waste from the still semi-secret thorium-to-uranium233 production at Hanford, for tactical nuclear weapons. Possible remnants of Hanford's old solid waste disposal system have been discovered next to D-Reactors.

The magnitude of the long-term radiological threat to the salmon stocks remains undetermined.

Public oversight of Hanford needs to be re-invented if management of Hanford Site is to become realistic and clean-up is to become effective and meaningful for the long term.

TABLE OF CONTENTS

Introduction....1
Figure 1. Location Map....2
Objective....5
Problems....5
Method....5
Results....10
Figure 2. Reference Sediment Collection....10
Figure 3. Suffix Convention....11
Table 1. Reference Sediments....13
Table 2. Miscellaneous Samples....18
Table 3. Dried Mulberry Leaves....21
Table 4. Upper Algal Mat....22
Discussion and Implications....23
Figure 4. D-Reactors....24
Figure 5. Remains of Ferry Crossing....25
Conclusions and Recommendations....29
Appendix 1 - Working Summary: Thorium at Hanford....30
Appendix 2 - Validation....32
Figure 6. Spectrum Acquisition....33
References And Notes....35

Introduction

Most of the non-hatchery, fall chinook salmon (*Oncorhynchus tshawytscha*) from the Columbia River spawn in the rocky bed of the 50-mile long stretch of the river called the Hanford Reach. These salmon are an important regional resource for commercial, tribal, and sport fisheries [1 --references are at the end of this report].

The Hanford Reach begins 6 miles upstream of Vernita Bridge and extends 50 miles downstream to Richland, Washington. This stretch of the Columbia River free-flows through the U.S. Department of Energy's Hanford Site.The Hanford Site produced the plutonium for the first nuclear explosion, Trinity, and the Nagasaki bomb that heralded the end of World War II. Plutonium along with other nuclear weapons materials for the Cold War were produced in 9 nuclear reactors next to the river.

The location of the Hanford Site is shown in Fig. 1, on the following page. "Hanford River Miles" (HRMs) are marked along the top and right edges of the inset, with dashed lines and numbers every 5 miles. Mileposts of these HRMs are situated along the shore of the Hanford reactor side of the Hanford Reach. HRM Zero is at Vernita Bridge. There are no mileposts upstream of Vernita Bridge, and so the upstream-most 6 miles of the Hanford Reach have only extrapolated HRM locations.

The old reactor areas are designated by a single letter on the location map. There are two reactors, each, in the B, D, and K areas.

Major salmon spawning areas are shown as "Area #" in the figure.

The Hanford Reach of the Columbia River is accessible by the public. As the U.S. Department of Energy (USDOE) cleans up the river corridor, it will be turned over to the public as the Hanford Reach National Monument.

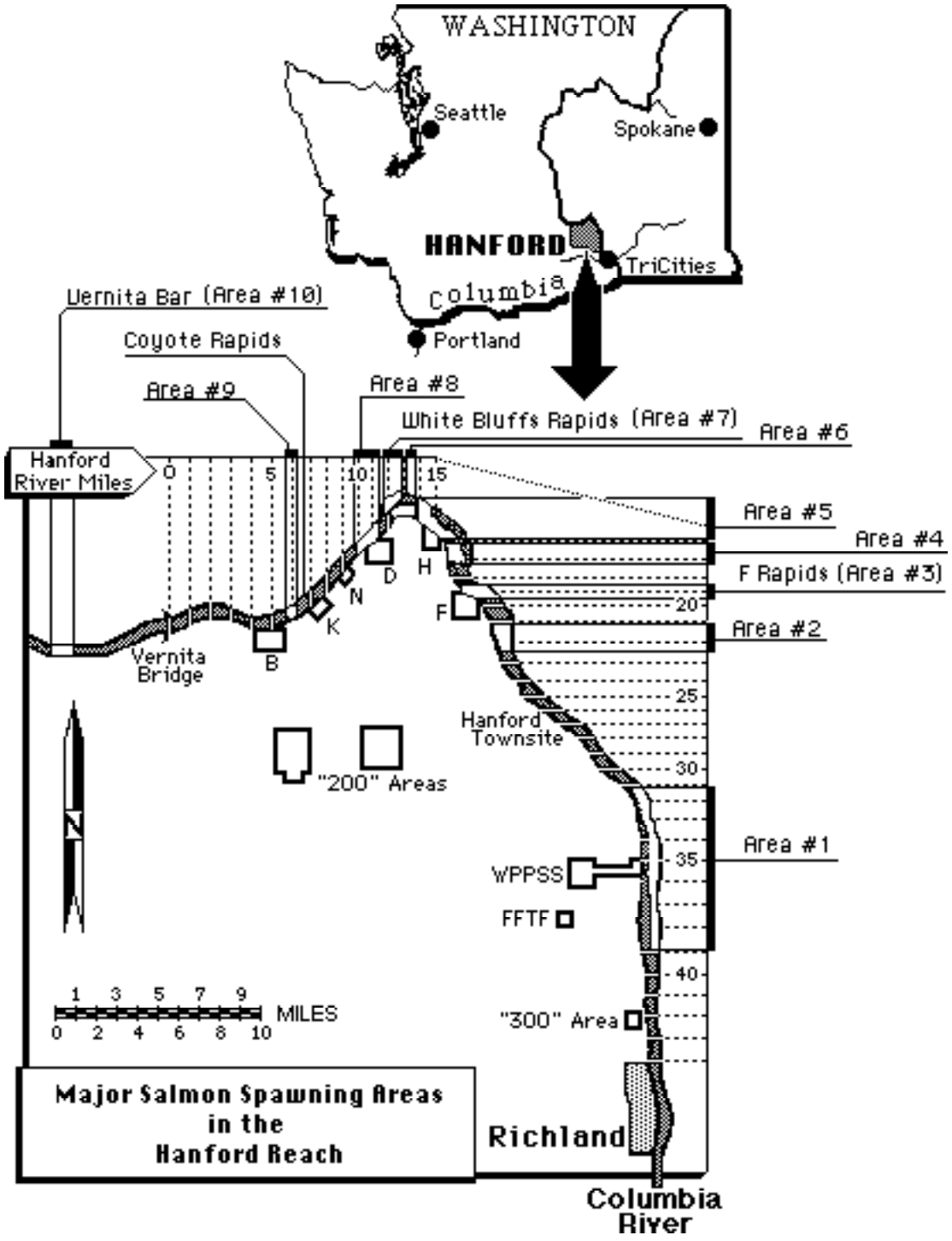

Figure 1. Location Map, with major fall chinook salmon spawning areas.

At a public conference on Hanford and the Columbia River, sponsored by the Government Accountability Project (GAP) in Portland, Oregon in October 1998, concern was expressed for the long-term strength (or weakness) of the salmon stock, which relies on natural spawning in the Hanford Reach riverbed.

After that conference, GAP began research to assess what, if any, effect Hanford operations might have on the salmon that spawn in the Hanford Reach.

For the first two years, GAP research focused on identifying the nature of candidate impacts of Hanford on the salmon and on ranking such candidates. This work involved review of published documentation of Hanford radioactivity and toxic chemicals, measurements of radioactivity seeping into the river from contaminated Hanford groundwater, and an initial, direct measurement of radioactivity in water in the riverbed [2].

The result of that work was a focus both on radioactive waste in the riverbed and on the newly hatched *alevin* of the fall chinook salmon --the life stage between the eggs laid in nests (*redds*) in the riverbed and the salmon fry that swim down the Columbia to mature in the Pacific Ocean.

The newly hatched alevin might be particularly susceptible to riverbed contaminant exposure, because these alevin remain within the riverbed throughout the winter-spring months of this phase of growth absorbing their yolk sacs, before they swim away as fry [3]. Thus, GAP's early work sought to determine whether riverbed water wherein the alevin live is generally more contaminated (1) from groundwater contamination seeping from Hanford Site into the riverbed or (2) from slow release of contamination that has resided in the riverbed for decades.

This question of the origin contamination reached a pivot with the discovery of thorium at ten-times background in mulberry leaves, downstream of F-Reactor (HRM 22 - 24) late in 1999 [4].

Thorium (Th) is a naturally occurring, radioactive element, like uranium (U), that can be irradiated in a nuclear reactor to produce fissile material for nuclear weapons. The special product of thorium irradiation is "clean U233", which is used in tactical nuclear weapons that can be deployed on the battlefield.

Based on the 10-times background thorium activities in some mulberry leaves, GAP requested under the Freedom of Information Act (FOIA) Hanford's thorium disposal history. US DOE's responsive declassification of documents invites a new vision of Hanford's Cold War mission; see Appendix 1.

In short, the declassified documents portray weapons production programs at Hanford having been far more diverse than the public has yet been informed. The crux of this lack of information is the prospect that a still semi-secret weapons production program at Hanford might continue to threaten the health of salmon spawning in the Hanford Reach riverbed. This raises

broad questions of governmental openness regarding Hanford Site management and of public oversight over this arguably most contaminated site in the Western Hemisphere.

Meanwhile, the technical question turned to the *character* of the thorium uptaken by mulberry trees on the shore of the Hanford Reach between HRM 22 and 24. By the beginning of the present study under the MTA Grant, radiological evidence suggested the high thorium in these Hanford mulberry leaves comes from riverbed sediments in the Hanford Reach, rather than from Hanford groundwater. So the technical effort began to move toward the riverbed where the alevin live.

While a procedure for sampling an *effective* reference sediment was being developed in this study, another study from University of Idaho researchers provided first evidence that chinook salmon spawning in the Hanford Reach are affected by some unidentified environmental stress. James Nagler, et al, reported that 80% of apparently (phenotypically) female salmon spawning in the upper Hanford Reach are sex-changed (genotypical) males, according to one genetic marker [5]. That report attracted national attention. But continuing research has muddied the scientific waters. Nagler now says, "We have some interesting observations, but I think it will be a number of years before we hammer out what is going on here [6]."

Thus two new scientific questions arose by early 2001: Are the salmon spawning in the Hanford Reach subject to some environmental stress causing them to change sex? What are the extent, character, and origin of radioactivity in the riverbed of the Hanford Reach where the salmon spawn?

These two questions challenged decades-long assurances by the Hanford Site operator, the U.S. Department of Energy, that there is no radioactivity even approaching safe drinking water standards anywhere near salmon redds, and the Hanford Reach salmon are "doing pretty good [7]." State of Washington Water Quality Standards for the Hanford Reach of the Columbia River require radioactivity to be as low as practically attainable and in no case shall exceed the EPA-570/9-76-003 drinking water regulations. The spirit of the applicable regulations is to suppose that if the river water is radiologically good enough for people to drink, everywhere in the Hanford Reach, that should be good enough to protect the biota living in the Hanford Reach. Thus, the inference that Hanford Reach waters are be "safe" makes a certain sense, but in practice, USDOE obtains permits for its violations of the applicable regulations, if USDOE is called to account.

The question of applicable ownership of and regulatory limits on water quality in the riverbed where the salmon spawn is presently unresolved and little addressed.

An unusual sidelight of this work has been Moon Callison's on-going production of a video documenting this challenge. *Sex, Salmon, Secrecy* has program advisors from different sides of the growing controversy and is sharpening the issues in front of her camera.

Objective

The objective of this study is to characterize the extent and intensity of radioactivity entering the Hanford Reach riverbed water. This involves developing appropriate sampling procedures, sampling, and analyzing samples from the entire length of the Hanford Reach, and interactive reviews with the site operator. This effort seeks initial understanding of the main impacts on salmon alevin and other biota living in the riverbed.

Problems

The preliminary problem confronting the present study was development of a sampling procedure to yield *reference* material meeting the following requirements:

- consistent
- representative and indicative
- cost-effective and non-hazardous
- stable and archivable

This problem was exacerbated by lack of sufficient knowledge at the outset. Both the relevant character of Hanford Reach sediments and the radionuclides of actual concern were poorly known. Thus, development of an adequate sampling procedure was a main concern of this study. This problem was solved by trial.

As this procedural problem was slowly solved, radioactive contamination of the Hanford Reach riverbed was found to be much more extensive than anticipated. Thus, the sampling program had to be expanded by a factor of ten, and the number of required analyses had to be increased beyond prior laboratory capability.

These problems were addressed by scaling the effort up and by taking a variety of risks.

Method

The conceptual basis for this technical study stems from the following: The Hanford Site operator, the U.S. Department of Energy (USDOE), has sufficient financial resources from Congressional appropriations and sufficient technical resources from its on-site, national laboratory, the Pacific Northwest National Laboratory (PNNL), to assess tractable technical problems adequately. This concept juxtaposes with a nebulous concept of public oversight of Hanford Site, as part of our peacetime democratic process. The interest here is in technical oversight in the public domain. Which translates as: fully independent, technical inquiry. But technical inquiry of what? How? By whom? Why? With what resources? With what accountability? To whom?

These conceptual issues are raised here, but not answered. The concept here is of independent technology focused on what might both be missed by US DOE's technical means and what the public might care most about. So the present work begins with statements by the interested public that the public cares about the health of the salmon. Which leads to a focus on

potential Hanford impacts on salmon, impacts that might have been studied by government agencies.

This study looks at radioactivity, possibly of Hanford origin, that might contaminate the Hanford Reach riverbed water wherein the salmon alevin spend their phase of life. When the salmon fry emerge from the riverbed and swim to the Pacific, they might carry with them some effects of radiological stresses during their alevin days.

After the salmon mature in the ocean, they return to the Hanford Reach to reproduce. University biologists find 80% of the spawning females to be genetically confused males. The environmental factor or factors causing this confusion are yet unknown.

The technical starting problem for this study is to define both the likely radionuclides of concern for the salmon and a sample medium in which to analyze those radionuclides.

The magnitude of this starting problem can be appreciated by looking ahead to the final results of this work: Namely that Hanford's contamination of the riverbed water where the salmon alevin live might be dominated by alpha decay radioactivity from Hanford's U233 product. Alpha radioactivity is extremely harmful to biota, but correspondingly difficult to analyze by ordinary radiological methods [8].

This leads to one technical difficulty: The relevant photon (x-ray, gamma-ray, and other emissions yielding photons in this energy band) radioactivity of the riverbed water is surprisingly dominated by short-lived lead-212 radioactivity, more or less in disequilibrium with the rest of the "natural" *thorium decay chain* of radioactivity. This surprising dominance of short-lived radioactivity in disequilibrium in a decay chain introduces several more technical problems.

That is to say, concern for salmon spawning in the Hanford Reach riverbed is probably not for thorium itself, but rather with product and byproduct, artificial radioactivity that happens to be more difficult to detect by ordinary means.

By the mid-1960s, Hanford researchers and engineers had developed a range of production methods for U233 having between 2.5 and 300 parts per million (ppm) contamination of U232 [9]. "Clean" U233 having only a few ppm contamination of highly radioactive U232 turned out to be cheaper for Hanford to produce than "dirty" U233 with up to 300 ppm contamination. The reason was that "clean" U233 was produced from *recycled* thorium which had more contaminants removed each time it was passed around the Hanford U233-production cycle, and the recycling process was cheaper than the purchase price of purified new thorium.

The technical problem for this study partly boils down to details of the radioactive natural thorium decay chain in comparison to the decay chain from artificial U232 inadvertently produced by irradiating thorium. The natural thorium decay chain is symbolized as:

Th232 -> Ra228 -> **Ac228** -> Th228 -> Ra224 -> Rn220 ->
Po216 -> **Pb212** -> ... -> Pb208

--where:	Th	symbolizes	thorium	of isotopic weight	232 or 228
	Ra	"	radium	"	228 or 224
	Ac	"	actinium	"	228
	Rn	"	radon	"	220
	Po	"	polonium	"	216
	Pb	"	lead	"	212 or 208

and "->" means "decays to".

Lead-208 is stable and so ends the radioactive decay chain of natural thorium-232.

This decay chain is not in equilibrium in the riverbed, apparently because the noble gas radon-220 is selectively leached from sediments into the riverbed water. This disequilibrium complicates analysis and reporting of the thorium decay chain.

More importantly, a troublesome contaminant of Hanford's U233 production, U232, decays into the natural thorium chain at Th228, symbolically 018]:

U232 -> Th228 -> Ra224 -> Rn220 -> Po216 -> **Pb212** -> ... -> Pb208

Notice that natural thorium (Th232) decays through Ac228 and then through Pb212; whereas artificial U232 decays through Pb212, without passing through Ac228.

Suppose there is some mixture of thorium with U232. The thorium part of this mixture yields some Ac228 and some Pb212, per the first decay chain, above. The U232 part of this mixture contributes Pb212, per the second decay chain displayed, but no Ac228 is contributed. So the decay chain of the mixture has a lower Ac228/Pb212 ratio than would be expected from only a thorium progenitor. But the Ac228/Pb212 ratio would exceed the zero value from pure U232 decay.

This introduces a technical possibility of identifying U232 in the riverbed by quantifying overabundance of artificial+natural **Pb212** in comparison to natural only **Ac228**. For the purpose of an initial screening of reference sediment samples in the present study, a preliminary measure of this

"Pb212 Excess"

has been applied to the radiological analyses of reference river sediments to look for the possible presence of U232, as an indication of the possible presence of U233 product.

If the radioactive waste in the Hanford Reach riverbed comes from Hanford's "clean" U233 recycling process and had only 8 parts per million (ppm) U232/U233, the U233 would have 50 times the (alpha) radioactivity of contaminant U232 [11]. Thus, any evidence of U232 is a warning for U233.

Elevated thorium activities are themselves another possible indicator of solid waste from Hanford's thorium-to-U233 production runs. Data from larger production runs in 1968 - 69 have

average yields of U233/Th = 0.0019 by weight [12]. This corresponds to a ratio of radioactivity of U233/Th = 170. Thus, any detectable, artificial elevation of thorium in the riverbed would signal grave concern for very difficult-to-detect U233 activity in salmon spawning areas.

Both "Pb212 Excess" and relatively high values of thorium in the riverbed are thus seen to be interesting but weak indicators of solid radioactive waste in the riverbed from Hanford's thorium-to-U233 production campaigns.

What is essential for the present study is crystal clarity of radiological evidence: whether a sediment sample from the Hanford Reach is truly contaminated with solid radioactive waste from Hanford's thorium-to-U233 production campaigns or merely has a lot of natural thorium that might have naturally accumulated in some stretches of the Hanford Reach.

Although alpha-emitting, artificial U232 and U233 *might* prove to be the radionuclides of greatest concern in the Hanford Reach riverbed, for the present study some unequivocal, easily measured, quantitative indicator of radioactive waste from Hanford's thorium-to-U233 campaigns is needed. This unequivocal indicator is

europium-152

The main reason is as follows: The usual resource mineral for thorium extraction is monazite sands, consisting of thorium with other *rare earth* phosphates, including natural europium (Eu) [13].

Natural europium is an almost equal mix of isotopes 151 and 153, weighing in at an atomic weight of 151.96. Both these natural europium isotopes are hundreds of times more easily neutron-irradiated to Eu152 and Eu154 than natural Th232 is irradiated to U233 [14]. So traces of europium in thorium *target rods* placed in Hanford reactors, yielded readily detectable Eu152 and Eu154. Allowable europium impurity content of thorium feed stock was specified <0.5 ppm, presumably for the very purpose of limiting co-production of contaminants like Eu152 [15].

During thorium feedstock recycling, the europium content was probably decimated, in accord with the concept of Hanford's "clean" U233 campaigns. But the europium was evidently not eliminated, and so Eu152 and Eu154 must have been cycled around too.

Unfortunately, the author has yet to discover in declassified documents the Eu152 content of the irradiated thorium discharged from Hanford's production reactors. So a calculation of U233/Eu152 activity in irradiated reactor discharge or in the waste streams is not yet feasible. Eu152 is thus only a qualitative indicator of irradiated thorium and the presence of U233 of Hanford origin in the Hanford Reach riverbed. But this indication awaits quantification.

Eu152 has a radioactive halflife of 13.5 years, in comparison to 8.6 years for Eu154. Consequently, even now, decades after deposition of europium isotopes were neutron-activated in Hanford's thorium-to-U233 campaigns, Eu152 remains readily detectable by ordinary photon

spectrometry [16]. Refined thorium that has been neutron activated to U233 in Hanford reactors is thus readily identifiable by the presence of easily detected Eu152.

It is also noteworthy that Eu152 is not a fission product and is not reported in world-wide fallout from historic atmospheric weapons testing.

Even though europium is a minor contaminant in uranium ores, europium isotopes were not reported in fallout from the Chernobyl reactor accident in Russia on 26 April 1986 [17]. Likewise, Eu152 is a minuscule component of Hanford's plutonium production wastes, yielding less than 1/10,000 of the radioactivity of either Sr90 or Cs137 fission products [18]. As Sr90 and Cs137 are reported in the present study and do not overwhelm Eu152 activity where Eu152 is detected, the Eu152 reported in the riverbed of the Hanford Reach unequivocally comes either from Hanford's thorium-to-U233 campaigns or from some more exotic and yet unidentified process.

There is no reason to suppose any origin of the detected Eu152 more sinister than Hanford's still semi-secret thorium-to-U233 campaigns, and so this minimum attribution is assumed.

Having identified Eu152 as the unequivocal, preferred indicator of Th-to-U233 waste in the riverbed of the Hanford Reach, if Eu152 is detected, then the Eu152 distribution might allow location of the source of this contamination of the riverbed. In particular, no Eu152 would be found upstream of the source of Eu152, because the river carries everything downstream with the flow. Thus, the source of Eu152 contamination should be discoverable at the upstream end of any Eu152 contamination pattern.

Thus, we have in Eu152 an unequivocal indicator of radioactive waste in the riverbed from Hanford's thorium-to-U233 campaigns. Meanwhile we have a prospect to screen "Pb212 Excess" for possible presence of U232 and a first indication of U233 product. Finally, we can compare activities of thorium and uranium and other fission and activation products to seek other patterns and inferences.

From these methodological considerations comes the prospect of having several indicators of the nature of radiological contamination in the Hanford Reach riverbed.

Another aspect of study methodology is selection of a sample collection procedure and, thus, sample medium as surrogate for riverbed water wherein the alevin live. This aspect has already been mentioned.

For both physical reasons and requirements of scientific reference samples, both large gravel and cobble fractions and fine silt fractions were eliminated from sediment samples, reported in Table 1. Details of the sampling method for reference sediments appear in the next section of this report.

The procedure of hand removing cobbles and sieving out gravels larger than 2mm, followed by suspension and pouring off silt yielded a remarkably constant sample medium, from diverse areas of the Hanford Reach having such visually distinct sediments.

Results

As a scientific study, the work reported here is unusual in its orientation toward a particular objective. Scientific studies are usually less focused. This difference affects interpretation of the technical results.

To interpret the results of a highly focused study, it is useful to consider the objective and method, rather than simply letting the data *speak for themselves*, as suffices with less focused work. Thus, the reader is invited to look to the sections of this report describing the Objective, the Method, and a Discussion and Implications of the Results presented here.

The central results are the "reference" sediment data in Table 1. Briefly, sediment sampling locations were selected for the purpose of scoping the extent and character of radiological contamination of Hanford origin along the Hanford Reach riverbed. Thus, sampling was sparse where little or no artificial radioactivity was found, and sampling was intensive where such radioactivity was detected, in order to delineate its character and boundaries.

Sediment samples were collected from the river side of the shoreline during low river levels which prevailed in 2001. Boulders and cobbles were discarded by hand and finer sediments were passed through a stainless steel, 2mm U.S.A. Standard Test Sieve; see Figure 2.a. This eliminated coarse sediments which have a relatively small surface-to-volume ratio and so would not much affect the radiochemistry of riverbed pore water. The resulting sediment samples were repeatedly agitated in river water in a Ziploc bag at their collection location, and the suspended silts were poured off, until the water above the sample was relatively clear; see Figure 2.b.

Figure 2. Reference Sediment Collection.

Fig.2.a. Sample, sieve, and sieving.

Fig.2.b. Pouring off suspended sediment fraction

The purpose of this floatation was to eliminate the variability of the fine fraction of the sediments and thus to enhance the replicability of study results. The effect of this floatation is clearly and intentionally to reduce the Hanford influence and so to introduce a measure of conservatism (under-stating Hanford's influence on the riverbed) to the results. The sample was dried <100C for 24 hours and about 30g was counted for photons for at least 24 hours in a standardized 125ml container in a highly stabilized, sodium-iodide, well-type detector, with a photon energy window from 15 KeV to 2800 KeV [19].

Locations of samples in Table 1 are indicated by approximate "Hanford River Mile," as seen on the mileposts along the Hanford side of the Hanford Reach. Precise locations are GPS latitude and longitude (read from a 12-satellite instrument) near the right side of Table 1. As every latitude in the study area is between 46 and 47 degrees North and every longitude is between 119 and 120 degrees West, the corresponding headers are "N46°" and "W119°", and the additional minutes of latitude and longitude are recorded to 3 decimals.

The convention for the suffixes to the HRM location designations in the tabulated results is shown in Figure 3.

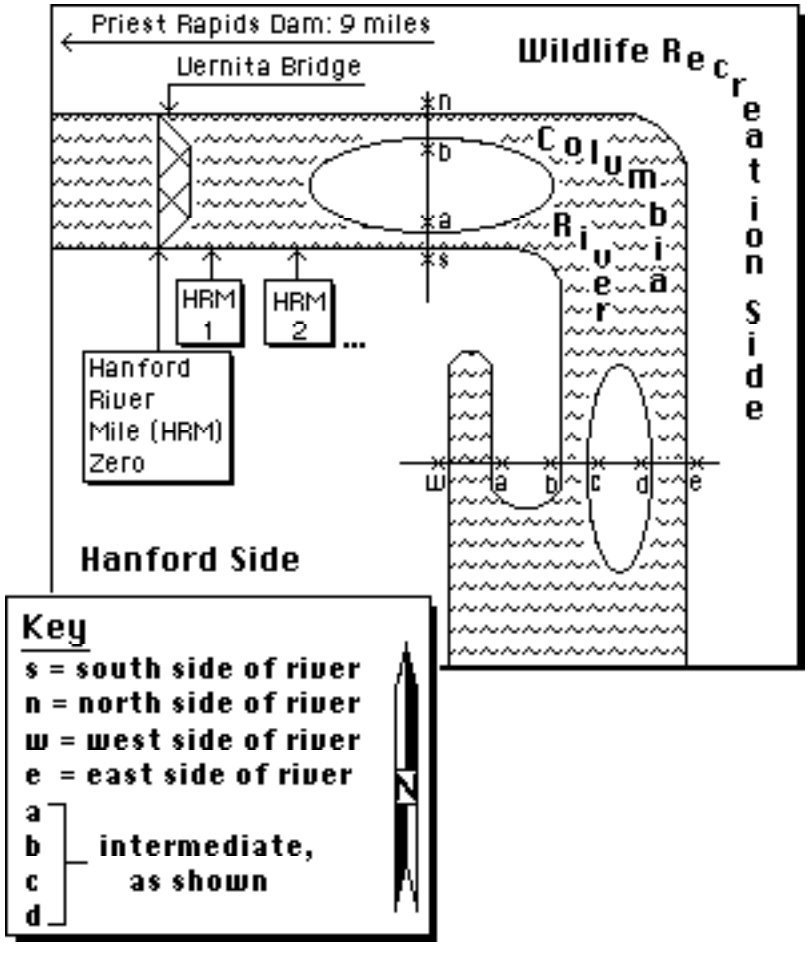

Figure 3. Suffix Convention, for HRM locations in tables in this report.

As seen in Fig. 3 and as appears in the tables of results, suffixes “s” and “w” are from the Hanford side of the river. Suffixes “n” and “e” indicate the side of the river opposite Hanford facilities. Other suffixes are intermediate, for islands and peninsulas, as shown, above.

One “Reference Material” datum is presented at the beginning of Table 1. The remainder of the data are Columbia River sediments and are presented in downstream order. Geographic features at the designated location are noted in the middle of the table. Underscored features followed by a colon indicate the beginning of the described feature.

Results are presented for the following radionuclides:

natural thorium as	“Th”
natural uranium as	“UNat”
cesium-137 as	“Cs137”
europium-152 as	“Eu152”
cobalt-60 as	“Co60”
strontium-90 as	“Sr90”
indication of U232 as	“Pb212 Excess’ %”

In every case, where a sample was collected (as indicated by a Sample No.) a period “.” standing alone or the space filled with a location description indicates “no detect” for the radionuclide in the table header.

The layout of Table 1 invites the reader to scan the Hanford Reach data in the downstream direction and to visually pick out patterns of riverbed contamination. The underlying logic is:

Contamination is carried downstream in the river and can be accumulated and dispersed there in the riverbed by various processes, but contaminants cannot be carried in the upstream direction by the river.

The left side of Table 1 is a sort of pictograph, with more samples and more detail in stretches of the Hanford Reach in which there is radiological change and thus focal interest in the processes that might have been and might yet be involved.

“Sample No.” refers to the designation of the archived sample. Sample number is a code for the time of sample collection, beginning with “1”, indicating the year 200**1**; followed by a single character for the month, with “x” = October; followed by one or two characters for the hour of sample collection and occasionally a final letter indicating a special count of a specially processed or recounted sample.

Table 1. REFERENCE SEDIMENTS

		Picocuries / Gram (Dry)							+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Sr90**	**"Pb212 Excess" %**	**Lat N46°**	**Long W119°**	**Sample No.**
Reference Material Sand Drifting from Gable Mtn.		2.4	1.1	.	.	.	.		37.004	31.565	181410
Columbia River Sediments:											
Upstream Basalt Outcrop	-19.0e	2.8	2.2	.	.	.	.	-24	48.397	55.391	171417
	-06.0n	2.1	1.5	.	.	.	.	-00	37.710	51.929	171418
	-03.0s	3.5	2.0	.	.	.	.	-23	37.756	48.053	172208
Vernita Bridge	00s								38.598	44.087	
	Downstream of B- and C-Reactors										
	04.1s	2.7	1.3	.	.	.	.	-07	38.397	38.423	172910
	05s	2.4	1.1	.	.	.	.	-02	38.549	37.632	1x0108
	Downstream of K-Reactors										
	07s	3.7	0.97	0.10	.	.	.	-18	39.455	35.773	1x0110
	N-Springs, Downstream of N-Reactor										
	08.9s	1.4	0.68	.	.	.	4.5	-37	40.681	34.113	171513
	09s	1.8	0.87	0.05	.	0.16	1.8	+15	40.812	33.970	1x01x1
	09.1s	2.0	0.79	0.08	.	0.50	5.4	-62	40.886	33.895	172912
	10s	2.0	0.96	0.31	.	.	.	-20	41.496	33.204	182307
	10.4s	1.9	0.80	.	.	.	.	-30	41.779	32.802	1x2008
Remains of Ferry	10.5s								41.830	32.764	
	10.5s	2.0	0.96	0.08	.	.	.	-29	41.830	32.749	1x2010

Table 1. REFERENCE SEDIMENTS

		Picocuries / Gram (Dry)							+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Sr90**	**"Pb212 Excess" %**	**Lat N46°**	**Long W119°**	**Sample No.**
	D-Island										
	10.7a	2.2	1.2	.	0.50	.	.	-35	42.053	32.590	1x2014
	10.7a	2.2	1.2	0.09	0.09	0.14	.	+02	42.063	32.548	191708
	10.8b	2.5	0.79	.	.	.	.	-18	42.183	32.502	181214
	11.0a	2.3	0.43	0.10	0.63	2.2	.	-67	42.122	32.377	182309
D-Reactors Outfall Header	11.0s								42.033	32.364	
	11s	2.2	0.77	0.06	.	.	.	-26	42.052	32.295	182308
	11.1n	2.5	1.7	.	.	.	.	-47	42.590	32.246	181215
	11.1b	2.0	1.4	0.28	1.45	.	.	-09	42.372	32.238	182414
	11.1b	2.0	1.2	0.48	0.64	.	.	-19	42.432	32.146	181216
	11.4b	2.8	0.93	0.10	0.38	.	.	-09	42.705	31.862	172914
	12s	4.2	2.2	0.31	0.47	.	.	-20	42.909	31.733	191711
	White Bluffs Rapids										
	12.5b	2.1	0.74	.	.	.	.	+06	43.622	31.289	172915
	13s	2.2	1.3	.	.	.	.	-41	43.112	30.844	191712
	15w	1.9	1.3	.	.	.	.	-20	42.433	28.887	190616
	Downstream of H-Reactor										
	15.4w	3.2	1.4	.	.	.	.	-28	42.162	28.581	172916
	15.6w	2.5	1.1	0.23	0.43	.	.	-32	41.982	28.379	190614
	16w	2.1	1.2	0.26	1.15	.	.	-15	41.699	27.940	1823x
	17w	2.1	0.53	.	0.25	.	1.3	-05	41.098	27.207	190514
	18w	1.7	0.90	.	0.26	.	.	+23	40.190	27.344	182310

Table 1. REFERENCE SEDIMENTS

		Picocuries / Gram (Dry)							+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Sr90**	**"Pb212 Excess" %**	**Lat N46°**	**Long W119°**	**Sample No.**
	Downstream of F-Reactor										
	19w	2.0	0.88	.	0.28	.	1.2	-08	39.593	26.342	1823xi
	F-Rapids										
	19.3w	3.8	1.2	.	.	0.05	.	-52	39.481	25.889	190607
	19.7w	3.0	1.0	.	.	.	1.4	-17	39.370	25.342	190609
	20w	3.5	1.6	.	.	.	.	-36	39.199	25.065	182311
Black Sand Drifting Into F-Slough	20.3w	1.5	0.77	.	.	.	.	-27	38.920	25.785	152915t
	21.0e	3.1	1.2	.	.	.	.	-29	38.328	24.312	172115
	21d	2.0	0.69	.	.	.	.	-12	38.289	24.626	172114
Lower F-Slough	21.1w	1.7	0.94	.	.	.	.	-48	38.257	25.168	1607c
	21.1c	2.6	1.2	0.03	0.25	.	.	-44	38.241	24.765	190611
	22.0c	3.1	0.73	.	.	.	.	-28	37.967	24.655	190613
	Downstream of F-Slough										
	22w	2.5	1.4	0.20	.	.	.	-13	37.825	24.937	171507
	23w	4.6	0.92	0.51	1.1	.	.	-34	37.077	24.612	171508
	23w	4.8	1.7	0.56	0.63	.	.	-35	37.077	24.612	171508(b) (recount after 4 months)
	24w	2.7	1.4	0.08	0.43	.	.	-24	36.380	23.903	171509
	25.0e	5.2	2.1	0.10	0.59	.	.	+34	35.844	22.897	171511
	25w	3.2	1.6	0.32	1.7	.	.	+27	35.771	23.074	171510
	26w	3.7	1.4	.	0.72	0.10	.	-59	34.850	22.109	182312

Table 1. REFERENCE SEDIMENTS

		Picocuries / Gram (Dry)							+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Sr90**	**"Pb212 Excess" %**	**Lat N46°**	**Long W119°**	**Sample No.**
	27w	2.8	1.4	0.05	0.64	.	.	-47	34.356	21.192	190516
	28.0e	3.4	1.8	.	.	.	.	-26	34.153	20.108	193016
Hanford Townsite Seep	28w	4.2	1.8	0.22	1.1	.	.	-23	33.973	20.296	193014
	32.0a	4.2	1.5	0.16	.	.	.	-20	31.159	16.445	193018
	37.4c	2.3	0.79	0.06	0.42	.	.	-39	26.774	15.890	812313
	38w	6.0	2.0	.	.	.	.	-29	26.335	16.237	1x1913
	40w	3.9	1.9	0.21	0.39	.	.	-12	24.386	16.090	1x1915
	300 Area										
	42w	2.1	0.69	0.03	0.13	.	.	-51	22.713	16.392	1x1916
	42.8w	2.1	1.7	.	.	.	.	-78	21.918	16.152	1x1919
Richland Drinking Water Intake	45.6w	4.4	1.2	.	.	.	.	-15	18.876	15.601	181316

*HRM = Hanford River Mile. No decimal point in designation means sample at the posted mile.

Several individual samples were collected or processed for special purposes. These results are presented in Table 2, on the next page, and discussed in the following section, Discussion and Implications.

These samples were individually adjusted to conform with the specified mass (32g total) and volume of the standard 125ml counting bottle. Water samples were quiescently evaporated by microwaves, to a paste on plastic film. The film was bagged and bulked to the specified mass and geometry for counting.

As in Table 1, the results are arranged in a downstream order, with salient features noted in the middle of the table. This layout invites the reader to scan the Hanford Reach data in the downstream direction and to visually pick out possible patterns

Table 2. MISCELLANEOUS SAMPLES

		Picocuries / Gram (Dry) Except / Where Noted							+ Minutes		
Reference to Location	**Location HRM***	**Medium**	**Th**	**UNat**	**Co60**	**Sr90**	**Cs137**	**Eu152**	**Lat N46°**	**Long W119°**	**Sample No.**
Vernita Bridge	00s								38.598	44.087	
	N-Springs, Downstream of N-Reactor										
	08.9s	Yellow Dock	.	.	.	110.	0.13	.	40.681	34.113	153015
	09.1s	Milfoil	0.37	1.3	.	3.5	.	.	40.886	33.895	172913
	Downstream of H-Reactor										
	15.8w	Fishfry	0.07	.	.	.	0.01	.	41.839	28.152	191614w
	Downstream of F-Reactor										
	20.1w	Molehill	3.1	1.7	.	.	0.77	1.7	39.122	25.990	112611
	23w	Ref. Sediment	4.8	1.7	.	.	0.56	0.63	37.077	24.612	171508
	23w	Synthetic H20	5.6/L	.	6.8/L	.	7.7/L	0.63	37.077	24.612	171508-s
	23w	Synthetic H20	1.0/L	.	2.5/L	.	12./L	0.63	37.077	24.612	171508-h
	23w	Yellow Dock	0.43	0.46	.	.	0.29	.	37.077	24.612	172113
	Downstream of Hanford Townsite										
	28.0w	Seep Water	0.07/L	0.74/L	0.81/L	1.1/L	.	.	33.952	20.277	131111

Table 2. MISCELLANEOUS SAMPLES

		Picocuries / Gram (Dry) Except / Where Noted							+ Minutes		
Reference to Location	**Location HRM***	**Medium**	**Th**	**UNat**	**Co60**	**Sr90**	**Cs137**	**Eu152**	**Lat N46°**	**Long W119°**	**Sample No.**
	Downstream of WPPSS										
	36.0w	Puddle Water	.	0.06/L	.	.	.	.	27.997	15.871	181412

*HRM = Hanford River Mile. No decimal point in designation means sample at the posted mile.

Mulberry trees grow along much of the Hanford shore near the highwater level. The roots of these trees often reach down to groundwater, and the leaves of the mulberry trees provide an easily sampled indicator of radioactivity and toxic chemicals in the groundwater seeping into the Columbia River from the Hanford shore.

Mulberry trees have an affinity for calcium, so they uptake strontium because strontium chemically mimics calcium. Mulberry leaves provide a convenient, biological reference material for the present study which focuses on riverbed sediments on the other side of the Hanford shoreline from mulberry trees.

As in the first two tables, the results are arranged in a downstream order, with salient features noted in the middle of the table. This layout invites the reader to scan the Hanford Reach data in the downstream direction and to visually pick out patterns.

The single sample collected from the north side of the river 0.4 miles upstream of Vernita Bridge yielded unexpected, positive results for Cs137 and Co60. No explanation is presented.

One mulberry tree *downgradient* of K-Reactors was sampled twice in 2001. With low river levels during 2001, increasing Sr90 activities were found. The second sample was collected jointly with USDOE on 14 August to encourage the site operator to address implications of such Sr-90 contamination downgradient from spent fuel basins which whistleblowers have reported to be leaking.

No mulberry leaves were collected from the N-Springs area, because those trees have been repeatedly cut down and the stumps treated with herbicide by USDOE .

These results can be compared to the sediment results in Table 1 to appreciate the tendency of terrestrial flora to reject the radioactive elements of the thorium and uranium decay chains [20].

Table 3. DRIED MULBERRY LEAVES

		Picocuries / Gram (Dry)						+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Sr90**	**Lat N46°**	**Long W119°**	**Sample No.**
	-0.4n	0.10	.	0.08	.	0.03	.	38.375	44.635	171420
Vernita Bridge	00s							38.598	44.087	
	Downstream of K-Reactors									
	6.9s	0.09	.	.	.	.	18.7	39.339	35.851	153012
	6.9s	.	.	0.06	.	.	44.4	39.339	35.851	181409
	Downstream of N-Reactor Springs									
	10.4s	0.10	0.26	0.02	.	.	.	41.779	32.802	1x2009
D-Island	10.9a	0.05	.	0.01	.	.	.	42.133	32.403	191710
	Downstream of H-Reactor									
	15.4w	0.04	.	.	.	.	.	42.145	28.625	172917
	15.6w	0.04	.	.	.	0.04	.	41.982	28.379	190615
	Downstream of F-Reactor									
	19.6w	0.06	.	.	.	.	.	39.374	25.860	190608
	20.5b	0.05	.	.	.	.	.	38.780	25.182	1906x1
	20.7w	0.04	.	.	.	0.03	.	38.587	24.664	172116
Hanford Townsite Seepage	28.0w	.	.	.	.	.	.	33.911	20.245	181411
	38w	0.06	0.16	.	.	.	.	26.318	16.250	1x1914
300 Area	42.5w	0.02	0.16	.	.	.	.	22.331	16.298	1x1917

*HRM = Hanford River Mile. No decimal point in designation means sample at the posted mile.

Positive results for the fishfry sample (#191614w) in Table 2 suggested that Hanford-origin radioactivity might somehow intrude into the mainstream biota of the Columbia River. To begin to address this possibility, samples of mixed algae and silt were gently scraped off the upper surfaces of the top cobbles and boulders at several sampling locations.

The samples were dried to <100C, crumbled and bagged to the standard mass and geometry for radiological counting for at least 24 hours.

These mixed algal samples were observed to include a wide range of silt content. This variation in silt content between samples from different areas of the Hanford Reach implies that the sample medium is not reasonably constant, and the results are not suited for inter-comparison.

The silt content of algal samples was dramatically greatest on the east side of the river downstream of White Bluffs.

Considering the great differences in silt content, the radiological results in Table 4 for the algae are remarkably uniform. The uniformity of Cs137 values suggests a possible source of fallout from historic atmospheric testing of nuclear weapons. That might also account for the unidentified interference at 19 KeV that rejected Sr90 from reporting in these data. The nil result for the sample from the east side of the river, opposite the Old Hanford Townsite is attributed to an overwhelming contribution of silt from erosion of White Bluffs.

Table 4. UPPER ALGAL MAT

		Picocuries / Gram (Dry)					+ Minutes		
Reference to Location	**Location HRM***	**Th**	**UNat**	**Cs137**	**Eu152**	**Co60**	**Lat N46°**	**Long W119°**	**Sample No.**
	-06.n	3.7	1.8	.	.	.	37.712	51.943	192917
Vernita Bridge	00s						38.598	44.087	
	05s	3.5	1.9	0.15	.	.	38.549	37.632	1x0107
	07s	3.4	2.0	0.05	.	.	39.455	35.773	1x0109
	09s	3.6	1.9	0.22	.	.	40.812	33.970	1x01x
	10s	3.7	1.1	0.21	.	.	41.492	33.209	1x0111
D-Island	11.1a	2.5	1.4	0.03	.	.	42.122	32.375	191709
	12s	4.5	1.6	0.28	.	.	42.911	31.728	1x0112
	23w	5.3	2.2	0.05	.	.	37.079	24.607	193011
	25w	3.9	2.1	0.12	.	.			
	28.0e	4.2	1.9	.	.	.	34.153	20.108	193015
Hanford Townsite Seep	28w	3.4	1.9	0.21	.	.	33.973	20.296	193013

*HRM = Hanford River Mile. No decimal point in designation means sample at the posted mile.
An unidentified interference at 19 KeV precludes Sr90 report for these algal samples.

Discussion and Implications

See the Method and Results sections of this report for logical and factual bases for the following discussion .

Table 1 reveals 3 main patterns of artificial radioactivity in the riverbed of the Hanford Reach:

<1> Strontium-90 contamination of the riverbed from N-Springs continues downstream for a few tenths of a mile, on the Hanford reactor side of the river.

<2> A pattern of europium-152 contamination begins at the upstream end of D-Island at HRM 10.7 and continues downstream to HRM 42. This pattern is interrupted at rapid stretches of the river, where erosion might have scoured away or deposition might have deposited over sediments containing Hanford contaminants. Similarly, passage of the river below HRM 42 into the pool for McNary Dam might be associated with recent sedimentation covering old Hanford-origin contaminants.

<3> Within Pattern <2>, there is an indistinct pattern of relatively high thorium, relatively high Eu152, and positive "Pb212 Excess" on both sides of the river at HRM 25.

Pattern <1> is attributable to seepage from N-Springs, still coming from the now defunct 1301N and 1325N trenches near the shore. This radioactive contamination is essentially local and therefore likely of little concern for the general viability of the Hanford Reach riverbed habitat.

Pattern <2> is attributed to solid radioactive waste in the riverbed, remaining decades after Hanford's thorium-to-U233 production campaigns ended. This radioactive contamination of the riverbed is extensive --from about HRM 10.5 to about HRM 42-- or about 60% of the length of the Hanford Reach. This extensive pattern is likely of great concern for the general viability of the Hanford Reach riverbed habitat, as will be discussed shortly.

Pattern <3> might be attributed either to some unidentified phenomenon that concentrates contaminated sediments in the stretch of the river below F-Reactor or to some yet unidentified, historic source of contamination there. Pattern <3> cannot be easily evaluated by radiological methods because of the complexity of erosion and deposition occurring in that stretch of the river by F-Slough. As a practical matter for the purpose of this report, Pattern <3> is considered an uncharacterized feature within Pattern <2>.

The Eu152 that fingerprints Pattern <2> is a neutron activation product of naturally occurring europium-151. Natural europium accompanies natural thorium in minerals from which thorium is extracted and purified. Europium impurities resist chemical separation and elimination from thorium [13]. Europium-151 has an affinity for neutrons. Therefore, when thorium is loaded into target rods in a nuclear reactor for neutron activation to produce fissile U233 for weapons or power applications,

Eu152 is also produced as a waste byproduct, having a halflife of 13.5 years. Thus, Eu152 remains for decades as an easy way to identify flag for radioactive waste from thorium-to-U233 production.

Hanford's history of U233 production remains clouded by military secrecy and destruction of documents. Work-in-progress to reconstruct Hanford's relevant U233-production history is summarized in Appendix 1 of this report.

The upstream end of Pattern <2> is distinct. The Eu152 flag is first detected at the very upstream end of D-Island at HRM 10.7. But this pattern apparently does not reach the Hanford shore until downstream of HRM 11 which is the location of the D-Reactors outfall structure. That is, Pattern <2> begins upstream of the D-Reactors outfall and probably near mid-river.

Inasmuch as water and sediments move only downstream, Pattern <2> of radioactivity cannot have originated from D-Reactors outfalls. Likewise, the nearest upstream reactor, N-Reactor, is two miles upstream. Because the reference sediments sampled are sands lying beneath the cobbles and boulders that line the bottom of the Hanford Reach, Pattern <2> could not be so pervasive in the lower stretches of the Hanford Reach without exhibiting any radiological evidence upstream, closer to any conceivable N-Reactor source.

Figure 4. D-Reactors

These radiological considerations suggested an old, mid-river source of thorium campaign wastes, just upstream of D-Island. Prudent operation of D-Reactors would have prohibited intentionally dumping radioactive waste just upstream of or into the D-Reactors intake at HRM 10.2.

...There are two "D-Reactor**s**" in Hanford's 100-D Area: "D-Reactor" operated between 1944 and 1967 and "DR-Reactor" operated between 1950 and 1964. D-Reactor was Hanford's second operational reactor, following B-Reactor's start-up by only a few months. DR-Reactor had the shortest operating life of any Hanford weapons-material production reactor and was the first Hanford production reactor to be shut down.

That same sort of prudent operation that would prohibit dumping radioactive waste into D-Reactors intake would prohibit dumping radioactive waste where it would be taken in to any Hanford reactor intake or, for that matter, the Richland Drinking Water Intake downstream, at HRM 45.6. This prudence reveals three relatively favorable locations for what is sometimes called "midnight" disposal of solid radioactive waste into the river:

(a) Near the upstream end of the Hanford Reach, with as much as 10 miles of river to catch and dilute wastes before reaching the B-Reactor intake. The problem confounding waste disposal into the upper Hanford Reach is that it might have been quite visible from public roads and accesses.

(b) Just downstream of D-Reactors' intake, allowing 4 miles for catchment of solids and dilution, before H-Reactor's intake.

(c) Just downstream of F-Reactor, allowing 31 miles before 300-Area intakes and then 3 more miles to the Richland Drinking Water Intake.

These considerations of presumed prudence by Hanford reactor operators, together with the distinct radiological evidence of historic dumping of solid radioactive waste into the middle of the river just upstream of D-Island, allowed a prediction to be tested on 20 October 2001: The shoreline of the Hanford reactor side of the river was carefully searched at low water, beginning at D-Reactors intake and continuing downstream a few hundred meters to the upstream end of D-Island, looking for remains of whatever structure had presumably been used to transport solid radioactive waste from Hanford's thorium-to-U233 campaigns in to the middle of the river for convenient disposal.

Remains of concrete piers and a severed, **1-1/2** inch steel, load-bearing cable were discovered at the low water shore at [46° 41.830' North, 119° 32.764' West] midway along the examined shoreline, downstream of D-Reactors intake and upstream of D-Island in the river [21]. Offshore, the riverbed appeared to be partly paved. See Fig. 5.

Figure 5. Remains of Ferry Crossing.

a. Looking south at old river crossing, with concrete block on right.

b. Close-up of concrete block.

c. One-and-a-half inch steel cable.

D. .Concrete riverbedding

Near the high water line, an orange-painted marker of rebar was found driven into the ground. Remains of old access roadways and approaches were also noted.

The north shoreline of the river was then searched for remains from the other end of an old river crossing. No structural remains were found on the wildlife recreation side of the river, opposite D-Reactors, but another rebar marker was found driven into the north shore opposite, at [46° 41.994' North, 119° 33.098' West].

These remains at D-Reactors are consistent with the mapped location of the Wahluke Ferry crossing, which had operated in various ways since about 1880. The Wahluke crossing used the KITTY-GRANT ferryboat before November 1943, when the crossing was closed to the public by the Atomic Energy Commission (AEC), as the agency took possession of Hanford Site [22].

Some of the Hanford Site ferry crossings taken by the AEC used cables to keep the ferries from drifting downstream and used the river current to tack back or forth across the river. Other Hanford Reach ferries used tugboats for power or had inboard engines. The Manhattan District Corps of Engineers operated at least two of the pre-existing ferry crossings to support anti-aircraft emplacements on the side of the river opposite Hanford Works [23].

Upstream of what now appears to be remains of the Wahluke Ferry crossing, there is an electric power cable crossing the river to a concrete blockhouse opposite D-Reactors [21]. Maybe the AEC used the pre-existing Wahluke Ferry crossing to support military defense on the north side of the river, opposite D-Reactors.

Despite accumulating evidence that solid radioactive waste from Hanford's still somewhat secret U233-production campaigns was dumped from the Wahluke Ferry crossing into the middle of the Columbia River, site management denies any such dumping practice [24]. Yet the official history of Hanford Site reveals clearly enough the operational mindset of the nuclear weapons production era:

> **...[S]pecial precautions for U and Th as radioactive substances were not taken in 321 Building. Solutions, scraps and other substances containing U and Th were handled and disposed as ordinary process wastes. Some U and Th entered the sanitary sewer system from personnel who contacted these substances [25].**

This suggests a certain tolerance by Hanford management for *natural* elements like thorium and uranium, as long as their radioactivity and toxicity did not seem to pose immediate health threats to workers.

This suggestion turns the initial questions around: Rather than asking how Hanford officials could possibly have justified dumping solid U233 production wastes into the Columbia River, the questions turn to: Why not? Where would the best dump sites be? How could the dumping have been done fastest and cheapest?

A ferry crossing located downstream of D-Reactors intake and upstream of D-Island seems, in retrospect, a pragmatic means, easily modified for disposal of some "natural" thorium and uranium solid wastes.

The next question is: What kind of irradiated (Eu-52 contaminated) thorium waste was dumped into the river? No answer has yet been found in the documents provided by USDOE.

One candidate for the source material of the solid radioactive waste dumped into the river is the ruined contents of thorium- (as *thoria*) -filled target elements that failed during their irradiation in the reactors. Some of those target rods failed because cooling water leaked into the rods [26]. The purity of the thoria contents was thus compromised in uncontrolled and different ways. It might have been cheaper to dispose of the failed rod contents instead of custom re-purifying the partly activated thoria.

The technical question then turns to consideration of what U233-production radionuclides might have been in the failed Th target rods, along with Eu152 which is so easily detected by photon spectrometer, and flags U233-production waste products in most of the Hanford Reach riverbed.

To begin exploration of what other, more difficult to measure, artificial radioactivity might prove important in the Hanford Reach riverbed, disequilibrium of the thorium decay sequence was examined with an eye to detecting excess U232, which contaminated the U233 produced from thorium.

A preliminary analysis to identify possible U232 contamination in the sampled, reference sediments of Table 1 was tried during the present study and applied to the radiological results from the middle of the effort; see Method. This yielded a few positive results in Table 1 (as "+" values of "Pb212 Excess").

Although this technique needs to be refined before any firm conclusions can be drawn, the possibility that a few parts per million of U232 contamination in U233 product might have been detected by aberrant disequilibrium in the thorium decay chain warrants further study. If such detection is confirmed, the yet unreported alpha radioactivity of U233 in the Hanford Reach riverbed would be staggering.

This raises a question of what radionuclides in the Hanford Reach riverbed might be ecologically important and yet missed in the single-pass analysis of the present study.

As a concrete step toward answering that question, the reference sediment sample from HRM 23w was agitated in distilled water, and the supernate ("synthetic H2O") water was analyzed for radioactivity, as a water sample, with results in Table 2 as Sample No. 171508-s. This process was repeated with results for Sample No. 171508-h. Such "synthetic H2O" derived from stable reference sediment samples provide a preliminary indicator of radioactivity in Hanford Reach riverbed waters in which salmon alevin live.

The replicate extraction and analysis yielded interesting results, relative to the "ref. sediment" radioactivities. Apparently some radionuclides, like Co60 and Eu152 are dissolved readily in comparison to others like uranium (UNat) and Cs137.

This begs the question of how much U233 might still be in the Hanford Reach riverbed.

If the regulatory limits of artificial radioactivity released into the *surface waters* of the Hanford Reach of the Columbia River also apply either legally or as levels of concern for the riverbed waters where the salmon alevin live, then a relevant standard is

15 pCi/L

for gross alpha radioactivity [27].

As there are 6 alpha decays in the thorium decay chain, the derived values (5.6 pCi/L and 1.0 pCi/L) for thorium in "synthetic H2O" (in Table 2) would correspond to 34 and 6 pCi/L of alpha radioactivity, which are already comparable to this limit of 15 pCi/L.

Radiological analysis of riverbed water needs further development to characterize the radiological content of the Hanford Reach riverbed where the salmon alevin live.

Conclusions and Recommendations

1. **Sixty percent of the riverbed of the Hanford Reach of the Columbia River is contaminated with solid, radioactive waste from Hanford's still semi-secret thorium-to-uranium-233 production campaigns. This artificial radioactivity of Hanford origin is flagged by readily detectable europium-152, which accompanies uranium-233 production in nuclear reactors.**

2. **This radioactive contamination of the Hanford Reach riverbed probably results from disposal of solid radioactive waste directly into the middle of the Columbia River, just upstream of the D-Reactors outfall. Remains of an old river crossing at the radioactive source location might be relics of the old radioactive waste disposal system.**

3. **Despite billions of dollars spent, supposedly for cleaning up the most contaminated site in the Western Hemisphere, and millions of pages of documents declassified, the old culture of Hanford secrecy for nuclear weapons production, supposedly for "national security" remains intact. This secrecy extends off-site into the public domain of the Hanford Reach riverbed, where the salmon spawn and are still more or less threatened by old Hanford radioactivity.**

4. **Quantification of the radionuclides of greatest concern for salmon spawning in the Hanford Reach riverbed awaits further work. In the interim, public oversight of Hanford must be re-invented if remaining secrets from Cold War nuclear materials production are to be managed effectively, and even cleaned up.**

5. **A focus on difficult-to-detect radionuclides of concern in the Hanford Reach riverbed is long overdue. Uranium-233 from Hanford's thorium-to-U233 production campaigns is clearly of concern. The Hanford Site operator and government regulators should address the radiology of the riverbed in a meaningful way.**

Appendix 1 - Working Summary: Thorium at Hanford

-a collaborative memo: initial research by Pat Lavelle, 10 November 1999
FOIA request by Tom Carpenter, 07 June 2000
document review by Norm Buske, 29 January 2001
document review by Alison Marti, 02 December 2001

This is a summary work-in-progress describing Hanford's thorium-to-uranium233 production and consequent waste disposal, based on partial review of already public documents and documents still being published in response to a GAP request under the Freedom of Information Act (FOIA).

Brief citations of source documents appear in text.

Relevant records of thorium-to-U233 production and waste disposal have reportedly been destroyed: "Although extensive amounts of useful data were generated during the SPR [Single Pass Reactor] program, significant portions of this information have been destroyed." [UNI-1400, 11/05/79, p.4] Given the record of thorium document destruction and the on-going declassification, it seems much of the record is not yet public. Thus some of the history must be established indirectly.

First, a little background on thorium and Hanford's production of uranium-233 therefrom:

An undated, hand-written document describes the two main uses of U233 resulting from thorium irradiation in Hanford's reactors:

"a) **Bomb** (just like U235 or Pu239)" --The document notes that both U233 and Pu239 have smaller critical mass than U235 and so "could make a smaller bomb (Might fit in an artillery shell or something)." The main attraction of U233 over Pu239 is the potentially "lower rad. level [of U233] than Pu". That is, U233 powered ordnance could be handled more easily with acceptable radiation exposure to a soldier carrying a tactical nuclear weapon.

"b) **Power** - Reactor Fuel" --The same document describes this as demonstration work, with the U233 product "sent to Oak Ridge Isotopes Div. for sale." "Oak Ridge tried some thorium oxide pellets as reactor fuel, but project just died." From other information, we know the power interest was in the thorium-breeder-reactor cycle tested at USDOE's Shippingport reactor.

The distinction between "bomb" and "power" applications is thus seen in Hanford's effort to minimize contaminants and make Hanford's U233 product less radioactive and so more advantageous over "clean" plutonium, competing for tactical weapons having low enough radiation for personnel to use on a battlefield.

"PROGRAM OBJECTIVE. The objective of the overall program is to establish Hanford as the lead site for the production of clean U233... PROGRAM VALUE. The clean U233-thorium program has a high value in regard to the possible future operations at Hanford. It currently holds promise of providing a significant alternative product for the plant. It is also essential that we demonstrate our competence and capability to adapt to this product. If we cannot demonstrate this capability, it could be construed as an evidence of a lack of flexibility and versatility [U233-Thorium Program Letter - Chemistry Department, 7/15/65]."

Much of the FOIAed thorium information relates to "clean" thorium production and thus "bomb" use. "Past use of U233 has been limited by the buildup of contaminant U232 in the final product. ... A unique advantage of the Hanford production reactors is a soft neutron flux which minimizes the formation of the contaminant U232 and permits the production of a relatively pure U233 which can be handled with little or no shielding [DUN-2409, 4/07/67, p.2]."

Hanford's U233 production program consisted of both reactor "core" and "fringe" loadings. Fringe loadings were at the outside of the reactor, absorbing neutrons that were otherwise uselessly lost. The fringe loadings seem to have been justified for reactor "shield protection" and required about 30 tons of thorium per year in the mid-1960s [DUN-1349, 7/21/66; DUN-2197, 3/09/67; and DUN-3034, 8/28/67, pp.3-4]. "These specifications set [irradiation] exposures [in the reactors] at four to six weeks for core loadings and six to nine months for fringe loadings. The U233 produced from the scheduled loadings of the program, when blended, will contain less than 5 ppm U232 [DUN-1040, 4/15/66, p.4]."

Some FOIAed documents refer to kilograms of U233 produced, while others refer to tons of thorium source material. Typically, about 1.3 Kg of U233 was produced by irradiating one ton of thorium in a Hanford reactor [DUN-5866, 5/29/69].

Laboratory testing at Hanford for thorium's potential use in reducing pile reactivity began shortly after Hanford Atomic Products Operations (HAPO) received a small amount of the material in 1945 [HW-31222, 3/26/54].

The first "Production Test" run yet identified by a FOIA document, produced 30 Kg of U233 (from ~23 tons thorium) for delivery from H-Reactor by July 1, 1955 [HW-30989, 3/11/54]. Documentation of delivery for AEC's subsequently requested U233 production run has not yet been found in Hanford documents [HAN-53744, 2/15/54, D.F. Shaw to W.E. Johnson, "U233 Production"].

For the period between September 1954 and July 1965, we have yet obtained no specific thorium-to-U233 documents. Therefore, our information for this period of Hanford production history is largely based on indirect evidence from the FOIA documents. For example, one technical review document states that by 1958, “approximately one per cent of the [Hanford reactor] neutrons were absorbed in the irradiation of such materials as thorium to make U233,” and other special, non-Pu239 products [HW-78100, 6/27/63, p.23].

Indirect evidence of early production of U233 at Hanford comes from storage and transfer records. After construction in 1952, the 241-WR Vault (Tanks 006,007,008, and 009) was used to store 60% thorium nitrate solution. During the time of thorium nitrate storage, “seepage of liquids through cracks in the wall separating the hot and cold sides of the vault was observed. Ultimately, the thorium nitrate solution in these tanks was removed and the last flushes of these tanks were transferred to underground storage tanks in 1980” [WHC-SD-EN-ES-040, 5/18/94].”

The 241-WR Vault "was used for storage of uranyl nitrate hexahydrate, nitric acid, and tributyl phosphate in support of the uranium recovery operations (1952-1958), and stored thorium nitrate solution (1952-1976) in support of the REDOX and PUREX processes [DOE-RL 1992b]. The vault was deactivated in 1976." This document discusses an undocumented contamination incident that occurred in the early 1960s when a tank overflowed and filled its cell [DOE-RL 1992b]. The tank was pumped out and "then floated loose from its base, rupturing its lines, jumpers, and mechanical connections. A *significant cleanup effort* was required to return the facility to operational status."

A report, from 1968, notes large shipments of thoria received by Douglas United Nuclear's Production Fuels Section [DUN-4475, 7/12/68]. Every month between July 1968 and August 1969, hundreds of thousands of pounds of virgin thoria powder came to Hanford; the arithmetic mean of shipments received was 500,051 pounds.

A thoria delivery schedule from 1968 indicates 170 tons of thoria required for fiscal year 1969, with 24 tons required for each of fiscal years 1971 and 1972, and 20 tons for fiscal year 1973 [DUN-4737, 9/18/68].

Hanford reportedly produced the U233 fuel for the third core of USDOE's Shippingport Atomic Power Station, which was loaded into the reactor in 1976 [28]. This core was operated successfully as a Light Water Breeder Reactor (LWBR) between September 1977 and October 1982. This is the reported example of the "b) Power - Reactor fuel" application of Hanford's thorium-to-U233 program, cited at the beginning of this appendix.

When DOE began to scope N-Reactor for renewed U233 production from thorium in 1978, the effort was "very cumbersome" because of previous data destruction: "Although extensive amounts of useful data were generated during the SPR [Single Pass Reactor] program, significant portions of this information have been destroyed [UNI-1400, 11/05/79, p.4]."

Appendix 2 - Validation

The fundamental validity of the results presented in this report is based on description of the reference source materials and on their availability for independent replicate sampling. The site operator, USDOE, is capable of such sampling and so these results are open to technical refute. The counted samples have been archived and so can likewise be recounted by any interested party. Likewise, the author continues to invite USDOE representatives to joint sample the riverbed of the Hanford Reach, this invitation having so far been declined.

The general nature of photon spectrometry, such as employed for the present study is well known. The results have been replicated by independent radiochemical means on several occasions. The technique employed in this study, with calibrations and result comparisons, is deemed adequate and appropriate for the intended survey purpose of this study. This particular spectrometer has a good operational record spanning a decade, and it has been refined or upgraded annually; Fig. 6

.An important feature of the public-interest use of this spectrometer in its system context is development of "non-hazardous", user-friendly procedures and processes. This begins with restriction on sample radioactivity to no more than 4 times background by screening. In a few cases, this greatly restricts the weight of samples collected from the public-accessible, open environment. In the few cases in which weight of environmental samples has to be restricted, special handling is required. Sample preparation is either oven drying to <100C or quiescent, microwave evaporation of water onto plastic film, followed by least adjustment to the standard geometry for spectrometric analysis.

Figure 6. Spectrum acquisition.

For the low detection levels reported in this study, potential for contamination of laboratory equipment is a far greater concern than contamination of personnel. The detector is thus protected from routinely immobilized samples by at least two layers of containment.

At this level of radiological precaution, special attention is paid to charged particles of dried sample material. This problem is adequately solved by attention to the problem, by minimizing sample size, by crushing samples only as needed to conform to the sample geometry, by containment, and by step-forward processes.

A corresponding concern for a study like this, in which the thorium decay chain is of analytical importance, is the presence of natural Pb212 in ambient air at the laboratory location in Belfair WA, on the west side of Puget Sound, on Hood Canal. Although background radioactivity is only two-thirds background in the previous location in Spokane WA, dust containing Pb212 requires aggressive air filtration at the present location.

As part of the validation of this study, there is a possibly important point made with these considerations: Analytical study of the publicly accessible surroundings of nuclear weapons facilities need not, and maybe should not, be hazardous or toxic. Conceptually: A public-interest study *should* set an example of innocuous openness for the governmental subjects of the study to learn from and to learn to emulate, by eliminating the hazards they manage in the name of the public.

This rather philosophical consideration has been incorporated fairly consistently in the field work and sampling, sample handling and preparation, and lab analysis.

The general analytical approach of the spectrometry employed in this study is conservative in the sense of returning false negative results in preference to false positives. This conservative bias is initiated by analyzing only radionuclide peaks in ordinarily negative-count spectral regions. This means that a count for a radionuclide that is truly not present yields a result that is as negative as the spectral region counts negative. This is not a random effect, so the usual interpretation of counting statistics does not apply. The actual magnitude of this conservative bias depends on the interferences present in a particular

sample, after previous spectral subtractions have reduced the spectral peaks of the predominant radionuclides present. As the negativity of a region of interest is unquantified after known radionuclides have been subtracted, no correction can be applied. Negative results are actually "not detected".

Other conservative features are built into the analysis to reduce false-positive reports.

A primary feature of this spectrometer for survey purposes is detection of a range of spectra of radionuclides that are not listed as photon emitters. Strontium-90 and technetium-99 are two important examples (of pure beta emitters) that ordinarily require radiochemical analyses and thus thwart simple screening. The utility of a non-chemical, single-pass, inclusive radiological analysis for survey or screening work is evident.

The sensitivity of a single-pass analysis is, however, very dependent on the radionuclide of interest. Some sensitivity values for the present study are as follows:

Sensitivity (for 1500-minute sample acquisition)

radionuclide	counts/pCi
Th	200.
UNat	40.
Co60	50.
Sr90	2.
Tc99	0.02
Cs137	400.
Eu152	40.
Am241	lost to interferences in this study

The range of sensitivity to radionuclides of interest exceeds a factor of 10,000.

This huge range would preclude detection of radionuclides like Tc99 except that the detector can be highly stabilized, and the broad photon peaks of the sodium-iodide detector allow *true* spectral subtractions of blanks and reference radionuclides. As a consequence, by sequential subtraction of stabilized referential spectra, phenomenally low detection levels of some particular radionuclides are feasible, depending on the actual interferences present in a particular sample.

The radiological analyst appreciates the implications of this approach: In essence, it is an expert system rather than a typical, defined-procedure system. The validity of such an expert system hinges to an unusual degree on replicability, as mentioned at the outset.

The spectrometer is partly recalibrated (at 2 points on a Cs137 source) daily. A final *blank* comparison to an initial blank shows the spectrometer has not been contaminated during this study: Radionuclides of interest count appropriately negative, both forward and backward in time.

Other ordinary checks indicate the reported data are appropriate for the public-interest objective of the present study.

References and Notes

[1] D.D. Dauble and D.G. Watson, 1990, *Spawning and Abundance of Fall Chinook Salmon (Oncorhynchus tshawytscha) in the Hanford Reach of the Columbia River, 1948-1988*, PNL-7289, Pacific Northwest Laboratory, Richland WA. According to Glen Spain [verbal communication], Conservation Director of the Institute for Fishery Resources, 80% of the fall chinook salmon that spawn in the Columbia River spawn in the Hanford Reach.

[2] There is an extensive Hanford literature consisting of annual reports, special studies, and summaries. One excellent reference is [1]. An early reference for the present line of study is, N. Buske and L. Josephson, 1989, *Water and Sediment Reconnaissance of the Hanford Shoreline*, Hanford Reach Project; now Project207 of the Tides Center, Belfair WA. Concern for hexavalent chromium pollution of Hanford Reach salmon spawning grounds is documented in, S.J. Hope and R.E. Peterson, 1996, *Pore Water Chromium Concentration at 100-H Reactor Area Adjacent to Fall Chinook Salmon Spawning Habitat of the Hanford Reach, Columbia River*, BHI-00345 (Rev.1), Bechtel Hanford, Inc., Richland WA. See also, S.J. Hope and R.E. Peterson, 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. A sample of riverbed water, collected from a dug well in a sandbar, was dominated by lead-212 in disequilibrium in the thorium decay chain, having a radioactive halflife of only 10.6 hours. This unexpectedly short halflife for a dominant radionuclide, in disequilibrium, became a consideration partly defining subsequent work in this project.

[3] In Ref. [1], D.D. Dauble and D.G. Watson, 1990, p.1.1.

[4] N. Buske, October 1999, *Thorium Springs at Hanford: Implications for Salmon Spawning in the Hanford Reach of the Columbia River*, Government Accountability Project, Seattle WA. Thorium activities measured about 1 pCi/g(dry), in comparison to background activities of 0.02 to 0.1.

[5] J. Nagler, J. Bouma, et al, January 2001, "High incidence of a male-specific marker in phenotypic female chinook salmon from the Columbia River," *Environmental Health Perspectives* 109 (11).

[6] D. Hansen, 8 November 2001, "Tests find more cases of salmon sex change," *The Spokesman-Review*, Spokane, WA.

[7] Dana Ward, 3 August 2001, Interview transcribed at <www.reelmoon.org/trans/dw/html>. See also Ref. [1], p.5.9.

[8] See, M. Eisenbud, 1987, *Environmental Radioactivity* (3rd ed), Academic Press, San Diego, Appendix, for a fairly specific introduction to concerns for uranium contamination. In this regard, notice that U233 is ten thousand times as radioactive as natural uranium.

[9] W.K. Woods, 22 April 1965, "Production of clean uranium-233, Research and Engineering Irradiation Processing Department, General Electric, Richland WA, pp.1-2.

[10] The basis for Hanford production of "'clean' U233, relatively free from the bothersome[ly radioactive] isotope U232" is documented by the Staffs of the Irradiation Processing Department and Chemical Processing Department, 27 June 1963, *Hanford Reactor and Separations*

Facility Advantages, HW-78100, Hanford Atomic Products Operation, Richland WA, p.33. This document also mentions more advanced, nuclear weapons production possibilities: "...[T]he use of Hanford reactors is especially attractive for the first stage of a curium-244 campaign [p.4]."

[11] The 8 ppm figure is typical of the "clean" U233 process, for example, J.P. Schmidt, 15 August 1968, "Production test authorization 149, large scale thoria wafer irradiation," DUN-4462, Douglas United Nuclear, Inc., Richland WA, p.6. The alpha activity ratio of 50 is due to the long halflife of U233, 159,200 years in comparison to only 70 years for U232. Radioactivity is inversely proportional to halflife.

[12] T. Prudich, 29 May 1969, "Irradiated thoria," DUN-5866, Douglas United Unclear, Inc., Richland WA, tables for KE Basin and KW Basin discharges. The halflife of U233 is 159,200 years, and the halflife of Th232 is 14,000,000,000 years.

[13] C.A. Hampel (ed), 1968, *The Encyclopedia of the Chemical Elements*, Reinhold Book Corporation, New York NY, p.713.

[14] F.W. Walker, J.R. Parrington, et al. (rev.), 1989, *Nuclides and Isotopes*, 14th ed., General Electric Company, San Jose CA, pp. 36-37.

[15] W.M. Mathis, 21 July 1966, "Thoria delivery schedule," DUN-1349, U.S. Atomic Energy Commission, Douglas United Nuclear, Inc., Richland WA, p.2.

[16] Four clean peaks in the acquired spectrum from certificated reference source T18070 of Eu152, Product Code EFR.121, Source Number 7D235, Calibration No. 0146, by Amersham International plc, 1 November 1989, are used for Eu152 analysis. This analyzed spectrum is subtracted from the *working* spectrum to continue analysis for other radionuclides in a sample.

[17] Ref. [8], Table 14-6, p.381.

[18] Energy Research and Development Administration, December 1975, *Waste Management Operations, Hanford Reservation,* ERDA-1532-Vol.2, UC-70, reproduced by National Technical Information Service, Springfield VA, Table III-D-2. This table provides waste radioactivities under different N-Reactor loading conditions.

[19] This wide spectral window is realized by transformation of each, 4000-channel acquired spectrum to 250 channels output spectrum having the property of constant photo-peak width. Adequate sensitivity is achieved by true blank and reference spectra subtractions, thus progressively eliminating quantitatively most interferences.

[20] The elements in the uranium and thorium decay sequences are described as having "None" biological role and being more or less dangerous due to radioactivity, carcinogenicity, and/or teratogenicity; see, J. Emsley, 1991, *The Elements*, 2nd ed, Clarendon Press, Oxford.

[21] A stranded steel "load-bearing" cable is distinct from an electrical cable which contains insulated, stranded electrical conductors. An electrical cable emerges from the north side of the Columbia River at [46° 41.994'N, 119° 33.098'W], upstream of the load-bearing cable crossing, leading to a concrete block house in the wildlife recreation area, opposite D-Reactors.

[22] R.H. Ruby and J.A. Brown, 1974, *Ferryboats on the Columbia River, including the bridges and dams*, Superior Publishing Company, Seattle WA , map and p.90. This reference was brought to the author's attention by John Warner of Coos Bay OR, <jww@webenet.net>. Mr Warner had worked on a ferry at Vernita in 1962 and previously rode barges up and down the Hanford Reach around 1960. He has described from those recollections a cable strung high over the Columbia River close to D-Reactors.

[23] Ref. [22], caption to the photo, "The tug, *DORIS*, and barge, *Mary*."

[24] J. Lynch, 09 December 2001, "Hanford watchdog makes bark heard," *The Oregonian*, Portland OR, archived at <www.oregonlive.com/news/oregonian/index.ssf?/xml/story.ssf/html_standard.xsl?/base/news/1007729759134551 73.xml>. "Hanford officials say there was no such crude dumping practice."

[25] _____, *300 Area History, Hanford Site*, <Http://www.hanford.gov./history/300area/300-4th.htm#300-4-2>.

[26] "The majority of thoria [target rod] failures is a result of water entering the target element through some type of closure weld defect [DUN-1010, 4/15/66, p.12]." "Whereas the cause of the three types of failures is thought to result from water entry into the fuel [target] element, the actual failure mechanisms are not thoroughly understood [p.9]." This suggests variability in the chemistry of the thoria contents after failure, as borne out by analyses [Tables IV and VI]. Unfortunately, no europium data are included. This report depicts advanced thorium-to-U233 process control technology as fully operational by the mid-1960s.

[27] T.M. Poston, R.W. Hunt, et al, (eds), September 2000, *Hanford Site environmental Report for Calendar Year 1999*, PNNL-13230, UC-602, Pacific Northwest National Laboratory, Richland WA, p.C.3 and references.

[28] D.E. Robertson, C.W. Thomas, et al, June 1993, *Radionuclide Characterization of Reactor Decommissioning Waste and Neutron-Activated Metals*, NUREG/CR-5894, Office of Nuclear Reactor Regulation, U.S. Nuclear Regulatory Commission, Washington DC.

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