Hanford Radioactivity in Salmon Spawning Grounds

-quality, extent, and some implications

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

This radioactive contamination of the Hanford Reach riverbed evidently results from disposal of solid radioactive waste from still semi-secret uranium-233 production at Hanford, for tactical nuclear weapons. Possible remnants of the old waste disposal system have been discovered next to Hanford's 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.

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Preface

This Preface is a contextual chronicle of the progress of the technical work that is the subject of this report. The context is the relationship between this technical investigation, as it progressed, and the owner/operator of the subject Hanford Site, the U.S. Department of Energy (USDOE).

This Preface addresses whether or not USDOE and other governmental agencies overseeing Hanford Site have already had opportunity to confirm/refute the results of this study by their own technical means of replicate sampling, sample preparation, and analysis. The answer to this question is important because the validity of the results reported herein relies entirely on independent replicability of the reported results by any interested party possessing adequate technical means.

This Preface specifically addresses opportunities USDOE has already had to confirm or refute the results reported herein. Opportunities for the Washington Department of Health's Division of Radiation Protection and for the Nez Perce Tribe to have confirmed or refuted these results can be inferred from the documents in App. 3.

The investigative work reported herein responds to public concern for the longterm strength of the salmon stock that spawns in the Hanford Reach of the Columbia Riverbed. In response to this public concern, GAP began to investigate radiological contamination of the riverbed of the Hanford Reach in 1999.

Before the present study, contamination of Hanford origin in the Columbia Riverbed where salmon spawn was presumed to have two origins: (1) radioactive materials discharged from Hanford's old, "once-through" nuclear reactors, and (2) contaminated groundwater from Hanford seeping up into the riverbed. Since the mid-1990s, USDOE has developed a *pump-and-treat* program to remove an industrial waste, hexavalent chromium, from Hanford groundwater seeping up into the riverbed near Hanford's D- and H-Reactors.

In 1999, GAP began to investigate contaminated groundwater seeping into the river. The first candidate radionuclide of Hanford origin selected for consideration was strontium-90 (Sr90), a fission product of Hanford's old plutonium production mission. During the unusually high river stages of 1999, GAP's investigator sampled leaves from mulberry trees growing along the Hanford shoreline as surrogates for groundwater the roots of the mulberry trees tap into. (Mulberry leaves bio-accumulate Sr90 and provide more replicable analytical results than groundwater seepage itself, the contamination levels of which are sensitive to changes in river flow.)

Mulberry leaves collected along the Hanford Reach shoreline provided new radiological information: Previously unreported seepages of Sr90 were discovered near the K-Basins; thorium ten-times background levels was found downstream of F-Slough, just downstream of Hanford's old production reactors.

On the basis of that thorium anomaly discovered in mulberry leaves, GAP published preliminary draft findings. Then in June 2000, GAP requested from USDOE Hanford's records of waste disposals from thorium-related programs.

USDOE replied to GAP's request for documents with titles to over 50,000 documents relating to: production of uranium-233 from thorium at Hanford, U233 processing campaigns at Hanford's PUREX facility, and waste disposals on site. GAP then requested copies of the seemingly most pertinent documents and began to sort out Hanford's history of U233 production.

Meanwhile, GAP's in-field work focused on identifying and characterizing radioactive wastes in the riverbed, seemingly flagged by the anomalous thorium in mulberry leaves downstream of F-Slough. The in-field work had several false starts until the middle of 2001.

GAP invited USDOE to jointly sample riverbed sediments, in order to assure that government agencies and GAP were in analytical accord. On 14 March 2001, GAP proposed to USDOE to use a new agreement for technical access to Hanford as the context for jointly sampling and analyzing sediments in the riverbed of the Hanford Reach. USDOE, site contractor Pacific Northwest National Laboratory (PNNL), and GAP tentatively scheduled joint sediment sampling for sometime during the last ten days of June 2001. That date was later pushed back to the end of August to coincide with PNNL's regular sampling schedule.

GAP received no scheduling information from USDOE or PNNL and checked back with USDOE on 27 August 2001. USDOE's Dana Ward informed GAP's Norm Buske that,

"The sediment sampling has been done for this year. The schedule was pushed forward to accommodate the analytical lab to distribute sample load. We are planning to do some additional sampling in the river related to chromium monitoring some time in September or October. ... If you would send me a sample plan we may be able to accommodate you to some extent."

On 29 August, Buske submitted a description of proposed sampling in reply and asked,

"How does USDOE want to relate to what is being discovered in the riverbed? If USDOE wants to be actively involved (rather than to be reactive to what is published) now is probably a good time...."

Later that day, Ward replied,

"I will have to pass your question on to my supervisors prior to giving you our answer. Therefore, expect a slight delay before full response is made to your question...."

GAP finished its radiological exploratory investigations in November 2001, and Buske drafted this technical report at the end of December 2001. GAP sent copies of the December draft and a February 2002 revised draft to agencies and other reviewers for their comments. As review comments returned, Buske edited the report drafts.

By May 2002, USDOE had not responded to GAP's question: "How does USDOE want to relate to what is being discovered in the riverbed?

On 30 May 2002, a reporter for the Tri-City Herald newspaper faxed to GAP's Tom Carpenter a 7-page letter of criticism from the Nez Perce Tribe, dated 28 May 2002. That letter of criticism had already been distributed; see App. 3 of this report.

On 31 May 2002, the Tri-City Herald published the story of the Nez Perce criticism of this draft report: "Nez Perce denounce maverick scientist's Hanford waste theories." (See <www.tri-cityherald.com/news/2002/0531/story4.html>.)

In mid-June 2002, Buske received a 7-page copy of "Comments on 'Hanford Radioactivity in Salmon Spawning Grounds' (ERS 02-506)" from the Division of Radiation Protection of Washington Department of Health, dated 24 May 2002, addressed to Interested Parties; see App. 3.

Thus, criticism of this technical report has reached the public ahead of this report itself.

Introduction

Most *wild*, 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 and Notes 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 flows freely without any dams 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, was produced in 9 nuclear reactors on the banks of the Columbia 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 in Fig. 1, with dashed lines and numbers every 5 miles. Mileposts of these HRMs are situated on 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 upstreammost 6 miles of the Hanford Reach are extrapolated HRM locations.

The old reactor areas are designated in Fig. 1 by a single letter on the location map. There are two reactors, each, in the B, D, and K areas, and one reactor in each of the other alphabetical areas.

Major salmon spawning areas are shown as "Area #" in Fig. 1, with bars showing the length of these spawning areas along the Hanford Reach.

The Hanford Reach of the Columbia River is accessible by the public. The U.S. Department of Energy (USDOE) is cleaning up the river corridor and turning it over to the public as the *Hanford Reach National Monument*.

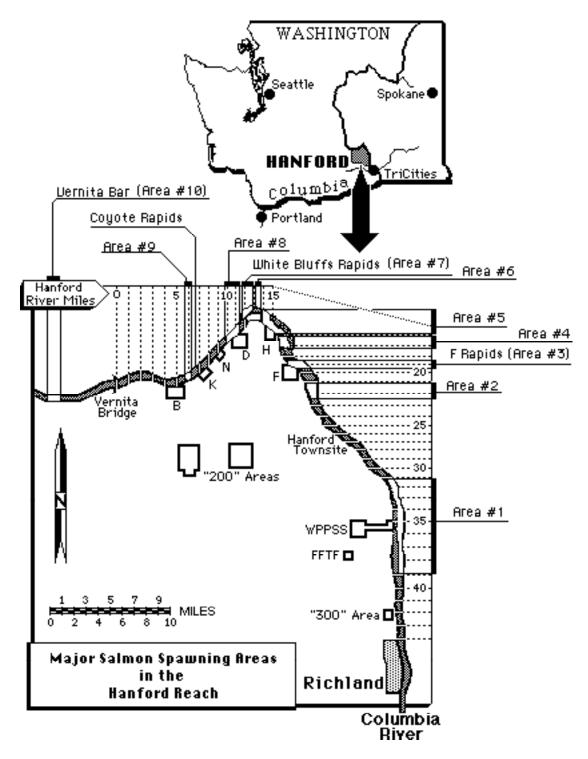


Figure 1. Location Map, With Major Fall Chinook Salmon Spawning Areas.

At a conference on Hanford and the Columbia River, sponsored by the Government Accountability Project (GAP) in Portland, Oregon in October 1998, members of the public expressed concern for the long-term strength of the salmon stock that relies on natural spawning in the Hanford Reach riverbed. After the Portland conference, GAP began to research what, if any, effect historic Hanford operations might still yet have on the wild 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. That 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 interstitial pore water in the riverbed [2].

As a result of that work, GAP focused 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 River to mature in the Pacific Ocean.

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

This question of the origin of potential contamination reached a pivotal point with the discovery of thorium at ten-times background in mulberry leaves, downstream of F-Reactor (between HRM 22 and 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 finding of 10-times background thorium in a few samples of mulberry leaves, GAP requested under the Freedom of Information Act (FOIA) Hanford's thorium disposal history.

USDOE's responsive declassification of documents invites a new vision of Hanford's Cold War mission; see Appendix 1. In short, Hanford's recently declassified documents portray weapons production programs more diverse than has yet been generally reported to the public. One of Hanford's larger production programs is now known to have been production of clean U233 for tactical nuclear weapons.

This new information raises the prospect that wastes dumped into the Columbia River decades ago, from a still semi-secret weapons production program, might still continue to threaten the health of salmon spawning in the Hanford Reach riverbed. This prospect raises broad questions of governmental openness regarding Hanford Site management and of public oversight over this arguably most contaminated site in the Western Hemisphere.

At the beginning of the present study, radiological evidence suggested the high thorium in Hanford mulberry leaves collected between HRM 22 and 24 had derived from riverbed sediments in the Hanford Reach, rather than from Hanford groundwater. The technical investigation thus began to move toward the riverbed where the alevin live.

While a procedure for sampling an *effective* reference sediment was being developed for this study, a completely different study by researchers with the University of Idaho provided first evidence that Chinook salmon spawning in the Hanford Reach might possibly be affected by some yet unidentified environmental stress. James Nagler, et al, reported that 80% of apparently (phenotypically) female salmon spawning in the upper Hanford Reach may be sex-changed (genotypical) males, according to one genetic marker [5]. These results for "Wild Columbia River" salmon were compared with results for fish of hatchery origin from the Columbia River watershed. The hatchery fish did not show phenotype switching.

Nagler's report attracted national attention. But continuing research has not yet been definitive. Nagler more recently said, "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]." In particular, Nagler's results did not pinpoint salmon phenotype switching exclusively to the Hanford Reach alone. Other wild populations of salmon in the Columbia River watershed might well be subject to environmental stresses of various kinds with consequential phenotype switching. In that case, Nagler's results would not be specific to the Hanford Reach, and any inference of a potential association with historic Hanford operations and the chromosomal anomaly would be weakened.

Some possible causes of the sex reversals have been suggested by Nagler and others: \bullet Environmental estrogens from detergents, pesticides, or other chemicals from domestic sewage processing, industrial operations, or agriculture. \bullet Water temperature fluctuations, possibly resulting from operation of the Priest Rapids Dam, upstream. \bullet Retarded migration of salmon fry downstream to the Pacific Ocean. \bullet Accelerated sloughing of White Bluffs into the river, apparently caused by irrigation northeast of Hanford Site, and increasing siltation of the riverbed. As yet, there is no *direct* evidence that radioactivity of Hanford origin in the Hanford Reach riverbed is causing the chromosomal anomaly reported by Nagler.

Thus, two new technical questions arose by early 2001: Are the salmon spawning in the Hanford Reach subject to some yet-unidentified environmental stress causing them to switch phenotype? 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, USDOE, 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 supposes that if river water everywhere in the Hanford Reach is radiologically good enough for people to drink, that should be good enough to protect the biota living in the Hanford Reach. Thus, the inference that Hanford Reach waters are "safe" makes a certain sense, but in practice, USDOE obtains permits for its violations of the applicable regulations.

Questions of applicable ownership of and regulatory limits on quality of water in the Hanford Reach riverbed where the salmon spawn are presently only beginning to be addressed. The flavor of such concerns is evidenced by the controversy that drafts of this report have already stirred; see Appendix 3.

A sidelight of this study has been Moon Callison's production of a video documenting thiss work-in-progress. *Sex, Salmon, Secrecy* has program advisors from different sides of the growing controversy.

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, collecting and processing samples, analyzing samples from the length of the Hanford Reach, and interactive reviewing with the government agencies. This work is one step toward characterizing the radiology of the Hanford Reach riverbed. This line of analytical work parallels other, independent investigations of toxicology and biology. When all these lines of investigation are followed and put together, the public will gain an initial understanding of the main impacts on salmon alevin and other biota living in the Hanford Reach of the Columbia River.

Problems

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

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

This problem of replicability 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 problem of this study, eventually solved by trial and error.

As this procedural problem was slowly solved, radioactive contamination of the Hanford Reach riverbed was found to be much more extensive than had been 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. These problems and their solutions during this course of this study rendered this study unusually exploratory in nature. The exploratory nature of this study continues into this report, which is partly a chronology of the exploration as this study developed and partly a report of final results.

The dual nature of this report, both as a chronology of a technical exploration and as a publication of results required by grant contract, makes for confusing reading. Adding to the confusion, this report has been written to be accessible to the reading public that is concerned for the salmon, for the river, and for Hanford clean-up; while the controversy is somewhat technical, as can be seen from the specialized comments in App. 3.

An additional problem for this study was the presence of Pb212 in airborne dust at the lab location next to Hood Canal. This dust has been suppressed by air infiltration at the lab entrance.

Method

The conceptual basis for this technical study stems from the following: The Hanford Site operator, 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 means that the usual processes for independent technical study validation boil down here to provision of enough information about sampling and sample processing to allow USDOE and other government agencies and major stakeholders to themselves independently collect and analyze replicate samples and so to confirm or refute the results reported here.

This concept juxtaposes with a nebulous concept of public oversight of Hanford Site, as part of our peacetime democratic process. Because Hanford Site is operated and overseen by government agencies having all the technical resources of the government available, the science-in-the-public-interest reported here can dispense with some of the expensive accreditations and other checks usually attending technical investigations.

The concept here is of independent technical inquiry of that which government agencies might overlook by their own technical means and what the public might care most about. The results of this independent work are truth checked by the replicability of sampling.

The present work thus began with public statements of concern for the health of the salmon. This led to a focus on potential Hanford impacts on salmon, impacts that might somehow have yet been overlooked 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 under the Hanford Reach.

After the salmon mature in the ocean, they return to the Hanford Reach to reproduce.

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 historic U233 production. 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 concern for thorium itself, but rather with product and byproduct, artificial radioactivity that happens to be more difficult than thorium 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]. Surprisingly, this "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.

Part of 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:

where:	Th	symbolizes	thorium	of isotopic weight	232 or 228
	Ra	"	radium	· C	228 or 224
	Ac	"	actinium	۲۵	228
	Rn	"	radon	۲۵	220
	Ро	"	polonium	۲۵	216
	Pb	"	lead	۲۵	212 or 208
1 44 > 44		1 4 - ??			

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

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.

A zero-order trial assumption was made in regard to the disequilibrium of the thorium decay chain in the Hanford Reach riverbed. This assumption was that in sieved reference sediments from the riverbed, the natural disequilibrium might be relatively invariant. That is the geographic variations in disequilibrium might be indicative of artificial perturbation.

Such a poorly controlled indicator can have no more meaning than to prompt further investigation or to abandon it. As will be seen in the Results, little merit is attached to this indicator.

The interest in possibly identifying some artificial perturbation in the thorium decay chain arose from the fact that a troublesome contaminant of Hanford's U233 production, U232, decays into the natural thorium chain at Th228, symbolically:

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 possible 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. At the accuracy of the present study, this indication has little merit.

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 would be a warning flag 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, some unequivocal, easily measured, quantitative indicator of radioactive waste from Hanford's thorium-to-U233 campaigns is needed for the present investigation. This unequivocal indicator, in the Hanford Reach riverbed, upstream of the McNary Dam pool, is

europium-152

A reason for this indication is the close association of europium with thorium in ores: 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 radioactive contaminants like Eu152 [15].

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.

Reviewers of this report have suggested other possible origins of the Eu152 used here as an unequivocal indicator of wastes from Hanford's U233 production. These possibilities are mentioned here:

1. Uranium fuel rod waste. Both Eu151 and Eu153 are fission products of U235 decay. These decay products formed in the fuel rods of Hanford's plutonium production reactors. These decay products were then neutron activated to Eu152 and Eu154. This possible origin of Eu152 has several scenarios. One possible pathway might have involved loss of radioactive material from ruptured fuel rods having been released from the discharges of the old, once-through reactors into the river. There have been anecdotal reports of workers chipping concrete off the old reactor discharge faces to reduce europium contamination levels.

However, Eu152 is only a tiny fraction of a reactor's neutron activation products. Europium isotopes were not reported in fallout from the Chernobyl reactor accident in Russia on 26 April 1986 [17]. Likewise, Eu152 was 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]. Both these fission products live more than twice as long as Eu152. Therefore, either Sr90 or Cs137, or both, would overwhelm Eu152 in the riverbed if the source were ruptured uranium fuel rods.

Other scenarios involving waste processed from Hanford's plutonium production runs encounter the same consideration of abundance of Eu152 in the Hanford Reach riverbed, namely comparable to or greater than either Sr90 or Cs137. That is to say, the mere fact that Eu152 turns out to be a radioactive indicator in the riverbed precludes uranium fuel as a credible candidate source material.

2. Europium control rod waste. Europium has been used as a neutron absorber in reactor control rods [13]. Neutron activated material might conceivably have been leached from europium-containing control rods and then discharged from Hanford's reactors. Such a control rod source of Eu152 in the riverbed would introduce plumes of riverbed Eu152 downstream of each of the old reactor discharges. However, the Eu152 contamination pattern does not show elevated activity immediately downstream of *any* reactor discharge. Therefore, control rods are eliminated as a candidate source of the extensive Eu152 found in the Hanford Reach riverbed.

3. Atmospheric fallout. Eu152 is not a fission product and is not reported in world-wide fallout from historic, atmospheric testing of nuclear weapons.

4. Other Hanford source. There were many programs at Hanford that have not yet been disclosed. There is a possibility that some unidentified, completely secret production program was the source of the Eu152 still remaining in the riverbed. This possibility is discounted on the basis of Occam's Razor: There is no reason to suppose any origin of the detected Eu152 more secretive than Hanford's still semi-secret U233 production campaigns.

Having identified Eu152 in the riverbed as an indicator of U233 production waste in 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 could 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 with Eu152 an unequivocal indicator of radioactive waste in the riverbed from Hanford's thorium-to-U233 campaigns. Meanwhile we have a prospect to weakly screen "Pb212 Excess" for possible presence of U232 and possibly a first tenuous 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, a well-defined sample medium as surrogate for riverbed water wherein the alevin live. This aspect has already been mentioned.

For both physico-chemical reasons and technical requirements replicable 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.

The analysis of these reference samples relied on "survey quality" photon spectrometry. This has been justified on the bases of logic and economy.

The logic is that the USDOE and Hanford regulatory agencies are well equipped to check results by their own means. Sample locations and collection methods are described in this report. All samples are archived for re-analysis. Furthermore, GAP has repeatedly

invited USDOE to jointly sample the riverbed to confirm results; as chronicled in the Preface.

The feasibility of independent, replicate sampling by the site operator and regulatory agencies relaxes usual requirements for certifications for published *scientific* reports. Instead, the format and quality of a *technical* report seems appropriate in the present case.

Analysis by photon spectrometry allows many radionuclides to be identified with a single instrumentational pass, without any need for chemical preparation or toxic materials. Such a single pass analysis is economical. A few details of the spectrometric method are described in Appendix 2.

Results

This is a report of a technical study that was exploratory in nature and subject to special considerations described in the other sections of this report. The most important consideration is the opportunity for the site operator and for regulatory agencies to replicate the results reported here. That opportunity is described in the Preface.

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 the character and boundaries of that radioactivity.

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 in tray, sieve above, and shovel.



Fig.2.b. Pouring off suspended sediment fraction.

The purpose of this floatation was to reduce the analytical variability introduced with the fine fraction of the sediments and thus to increase replicability of study results. The suspendable sediment lost during sediment collection and sieving cannot be controlled readily. Therefore, the suspendable fraction is practically eliminated by flotation to diminish this reference material variability and to improve replicability of results obtainable by subsequent, independent sampling. (A single check during development of this procedure suggested a third of thorium-related radioactivity was lost by flotation of the suspendable fraction of the sediment.)

This flotation procedure for the purpose of improving replicability is likely to reduce the Hanford influence and so to introduce a measure of conservatism (under-stating Hanford's influence on the riverbed) to the results.

Each reference sediment sample was dried <100C for 24 hours, and about 30g (in a 32-gram geometry) 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 15KeV to 2800KeV [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 0.001'.

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

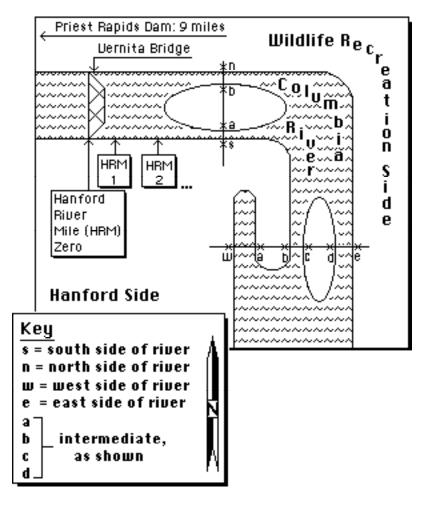


Figure 3. Suffix Convention, for HRM locations in tables in this report.

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 for Columbia River sediments and are presented in downstream order. The upstream beginnings of geographic features are noted in the middle of the table.

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 2001; followed by a single character for the month, with "x" = October; followed by two numbers for the day of the month, and then two digits for the hour of sample collection, and occasionally a final letter indicating a special count of a specially processed or recounted sample:

Sample No. = YMDDHH(x).

Table 1. REFERENCE SEDIMENTS											
		Picocuries / Gram (Dry)								inutes	
Reference to Location	Location HRM*	Th	UNat	Cs137	Eu152	C060	Sr90	"Pb212 Excess" %	Lat N46°	Long W119°	Sample No.
Reference Material		111	Unat	C3157	Luisz	000	5170	EACCSS /0	1140	w11)	
Sand Drifting from Gable Mtn.		2.4	1.1						37.004	31.565	181410
Columbia River Sediments:		2.1	1.1	•	•	•	•		57.001	51.505	101110
Upstream Basalt Outcrop	-19.0e	2.8	2.2		•			-24	48.397	55.391	171417
	-06.0n	2.0	1.5	•	•			-00	37.710	51.929	171418
	-03.0s	3.5	2.0		•	•	•	-23	37.756	48.053	172208
Vernita Bridge	00s	0.0		•	•				38.598	44.087	112200
· · · · · · · · · · · · · · · · · · ·	1	1	Downstr	eam of P	- and C-Re	actors	I	1			1
				Calli UI D.	- anu U-N		1	07	20.207	20.422	172010
	04.1s	2.7	1.3	•	•	•	•	-07	38.397	38.423	172910
	05s	2.4	1.1	•	•	•	•	-02	38.549	37.632	1x0108
			Dow	nstream o	of K-React	ors					
	07s	3.7	0.97	0.10	•		•	-18	39.455	35.773	1x0110
		N	-Springs	, Downsti	ream 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
	•		•	D-Isl	and				•		
	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

				Pice	ocuries / G	ram (Dr	y)		+ Mi	nutes	
Reference to Location	Location HRM*	Th	UNat	Cs137	Eu152	C060	Sr90	"Pb212 Excess" %	Lat N46°	Long W119°	Sample No
	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
			1	White Blu	ffs 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
	1		Dow	vnstream	of H-Reac	tor					1
	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
	·		Dov	vnstream	of F-React	tor					
	19w	2.0	0.88	•	0.28	•	1.2	-08	39.593	26.342	1823xi
				F-Ra	pids						
	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
Slough	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
	1	1		wnstream	of F-Slou			1	-	1	1
	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

Hanford Radioactivity in Salmon Spawning Grounds

August 2002

Page

	Location HRM*			Pice	+ Minutes						
Reference to Location		Th	UNat	Cs137	Eu152	C060	Sr90	"Pb212 Excess" %	Lat N46°	Long W119°	Sample No.
											171508(b)
											(recount after
	23w	4.8	1.7	0.56	0.63	•	•	-35	37.077	24.612	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
	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 individual samples were adjusted to conform with the specified mass (32g total) and volume of the standard 125ml counting bottle. Water samples were quiescently evaporated by microwave, to a paste on plastic film. The paste-laden 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. MISCELLANEOU	S SAMPLES										
Reference to Location		<u>Pic</u>	cocuries /	Gram (D	oted	+ M i					
	Location HRM*	Medium	Th	UNat	C060	Sr90	Cs137	Eu152	Lat N46°	Long W119°	Sample No.
Vernita Bridge	00s								38.598	44.087	
			-Springs	, Downstr	eam of N-	Reactor				1	1
	08.9s 09.1s	Yellow Dock Milfoil	0.37		•	110. 3.5	0.13	•	40.681	34.113 33.895	153015 172913
	09.18	MIIIOII			•	1	٠	•	40.880	33.893	1/2915
	1			nstream o	of H-React	tor	<u> </u>		1	1	1
	15.8w	Fishfry	0.07	•	•	•	0.01	•	41.839	28.152	191614w
		•			of F-React	or			-	•	
	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
			Downstr	eam of H	anford To	wnsite					
	28.0w	Seep Water	0.07/L	0.74/L	0.81/L	1.1/L	•	•	33.952	20.277	131111
			Do	wnstream	of WPPS	S					
	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].

			Pico	curies / C		+ Mi				
	Location							Lat	Long	
Reference to Location	HRM*	Th	UNat	Cs137	Eu152	C060	Sr90	N46°	W119°	Sample No
	-0.4n	0.10	•	0.08	•	0.03	•	38.375	44.635	171420
Vernita Bridge	00s							38.598	44.087	
		Downs	tream of]	K-Reacto	ors					
	6.9s	0.09	•	•	•	•	18.7	39.339	35.851	153012
	6.9s	•	•	0.06	•	•	44.4	39.339	35.851	181409
]	Downstrea	um of N-R	eactor S	prings					
	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
		Downs	stream of	H-React	or					
	15.4w	0.04	•	•	•	•	•	42.145	28.625	172917
	15.6w	0.04	•	•	•	0.04	•	41.982	28.379	190615
		Down	stream of	F-React	or					
	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.

Table 4. UPPE	ER ALGA	L MA	T				
		Pic	ocuries	/ Gram	+ M	inutes	
			<u>(Dry)</u> *	**			
Reference to	Location				Lat	Long	
Location	HRM*	Th	UNat	Cs137	N46°	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

Considering the great differences in silt content, the radiological results in Table 4 for the algae are remarkably uniform.

* HRM = Hanford River Mile. No decimal point in designation means sample at the posted mile. **Neither Co60 nor Eu152 were detected in any of the samples in Table 4.

An unidentified interference precluded Sr90 report for these algal samples.

The uniformity of Cs137 values in Table 4 suggests a possible Cs137 origin from fallout from historic atmospheric testing of nuclear weapons. That might also account for

the unidentified interference 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.

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, non-detection of Eu152 downstream of 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 U233 production campaigns ended. This radioactive contamination of the riverbed is extensive, from about HRM 10.5 to about HRM 42. This is 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 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



Figure 4. D-Reactors.

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.

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-Reactors" 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.

The same sort of prudent operation that would have prohibited dumping radioactive waste into D-Reactors intake would have prohibited dumping radioactive waste where it would be taken into any Hanford reactor intake or, for that matter, the Richland Drinking Water Intake downstream, at HRM 45.6. Consideration of such prudence in comparison to the Hanford Site map, Fig. 1, reveals three relatively favorable, potential locations for historic "midnight" disposal of solid radioactive waste into the Hanford Reach:

(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. However, any such waste disposal into the upper Hanford Reach would have been relatively visible from public roads and accesses.

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

(c) Just downstream of F-Reactor, allowing 31 miles before 300-Area intakes and then 3 more miles before 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 into the middle of the river for convenient disposal.

Remains of concrete piers and a severed, 1-1/2-inch diameter 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, on the next page.

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 at the far 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].

Figure 5. Ferry Crossing Remains Next To Predicted Location Of Dumpsite.



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.

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, Fig. 6, on the next page. The ferry cable might be seen at the lower left of the photo.

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 preexisting ferry crossings to support anti-aircraft emplacements on the side of the river opposite Hanford Works [23].

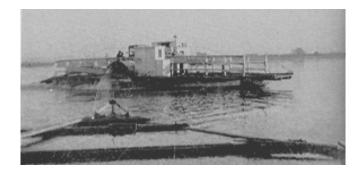


Figure 6. The Old KITTY-GRANT Ferryboat At The Wahluke Ferry Crossing Before The AEC Took Possession Of Hanford Site In 1943 [22].

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 managers continue to deny any such dumping practice [24]. Yet the official history of Hanford Site reveals 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-152 contaminated) U233production waste was dumped into the river? No answer has yet been found in the documents thus far 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 weakly 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. These results seem to suggest that some radionuclides, like Co60 and Eu152, might be dissolved from sediments more readily than others like uranium (UNat) or Cs137. This suggestion is somewhat contrary to experience of Co60 being relatively insoluble from sediments and Cs137 being relatively soluble. These results invite follow-up study.

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 riverbed of the Hanford Reach National Monument, where salmon spawn are still more or less threatened by old Hanford radioactivity.
- 4. The radionuclides and the toxicants of greatest concern for salmon spawning in the Hanford Reach riverbed await further identification and quantification. The Hanford Site operator and government regulators should begin expediently to address the radiology and toxicology of the riverbed comprehensively. In the interim, public oversight of Hanford must be reinvented if remaining secrets from Cold War nuclear materials production are to be opened to rational site management, so Hanford Site might then even be really cleaned up.
- 5. Effective clean-up of Hanford Site demands a new focus on difficult-to-detect radionuclides and toxicants in the Hanford Reach riverbed. Solid wastes from Hanford's U233 production campaigns are a primary concern.

Appendix 1 - Working Summary: U233 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 appendix summarizes work-in-progress to describe Hanford's thorium-touranium233 production and consequent waste disposal, based on partial review of already public documents and documents still being declassified in response to a GAP request under the Freedom of Information Act (FOIA).

Brief citations of selected source documents appear in the text, below.

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 [28].

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 U233 information relates to "clean" U233 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 documents obtained through GAP's FOIA request 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, specific thorium-to-U233 documents have not yet been provided by USDOE. 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 monthly shipments was 500,051 pounds.

A thoria delivery schedule from 1968 required 170 tons of thoria 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 core was loaded into the reactor in 1976 [28]. That reactor core was operated successfully as a Light Water Breeder Reactor (LWBR) between September 1977 and October 1982. That 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 validity of the results of this study is based on the scientific principle of replicability. That is, the samples and procedures are specified sufficiently that interested parties can check the results by their completely independent technical means. The main stakeholders at Hanford Site either have this technical means themselves, or they have access to independent contractors capable to replicating the results. USDOE has the means through its on-site contractor, Pacific Northwest National Laboratory (PNNL).

The U.S. Environmental Protection Agency (EPA) has the technical means through its own laboratories, such as its Las Vegas laboratory. The Washington Department of Ecology has the technical means through arrangements with the radiological laboratory of the Washington Department of Health (DOH) Division of Radiation Protection. In addition to these three agencies who are the "Tri-Party" of agencies that oversee Hanford clean-up, other stakeholders, such as the Nez Perce Tribe, have technical expertise and access to independent radiological laboratories. (See App. 2.)

The fundamental basis for the sample replicability of this study is description of sample collection and preparation procedures and listings in the Results of sample locations by high-resolution Global Positioning System (GPS). In this regard, GAP has made a special effort to invite and engage USDOE in joint sampling, so USDOE might through PNNL confirm the results of this report; see the Preface. Furthermore, all but one sample (No. 181410 in Table 1) have been obtained from public access along the Hanford Reach, so any independent party can replicate the results without having to obtain access to Hanford Site.

In case analytical doubts might arise, the samples analyzed for this report have been archived and are available for re-analysis by any interested party. This retention of samples provides a secondary level of the replicability that is the basis of the analytical validity of this study and its results.

The author of this study provides a tertiary level of replicability by opening his laboratory to inspection. The Washington Department of Health, the Yakama Nation, and a Hanford whistleblower have availed themselves of the opportunity to inspect this laboratory and its procedures. In 1989 and 1990, the author participated in joint sampling with the USDOE/PNNL, the States of Oregon and Washington, EPA once, and one tribal representative, confirming laboratory inter-comparability of results.

Until the present time, as this report is being finalized 7 months after draft copies were circulated to parties interested in Hanford clean-up:

There have been no requests by any party for additional information to allow replication of reported results nor any requests for access to the archived samples nor any requests for laboratory inspection to examine procedures.

The replicability described above is <u>the</u> basis for the technical validity of the results presented in this report.

Inasmuch as the spectrometric technique is slightly unusual, a brief description is provided here, as an introduction to the reader unfamiliar with the analysis.

The analytical technique employs a 3x6-inch, 125mL well-type, sodium-iodide (NaI) x-ray detector in a high-purity lead shield lined with copper. The sample matrix geometry is low density (32g/125mL) to minimize self-absorption of x-rays in the sample.

Ordinary problems of low resolution and of non-linearities are overcome by procedural attention to many technical details. Detector temperature is maintained to +/-0.1C. The detector is two-point energy calibrated daily. These stabilizations allow samples to be acquired routinely for 25 hours.

Detection of high-energy gamma rays in the long-acquisition spectra is enhanced by "constant photopeak width" transformation of the acquired spectra. This CPW transformation contracts the broad, high-energy peaks and thereby improves the signalto-noise ratio as the square-root of the CPW contraction. CPW transformation improves detection by a factor of 6 at 2500KeV.

Finally, the usual problem of broad-peak interferences in NaI detectors is overcome by taking advantage of what is usually considered a problem with NaI detectors: broad photopeak width. This feature allows true, full spectra subtractions of important radionuclides and whole decay chains for which the laboratory has reference standards, substandards, or other adequate source materials. These subtractions always begin with "Blank" which is the spectrum of the lead-shield loaded with a sample bottle filled with inert material comparable to the standard counting geometry. The second subtraction is the potassium-40 spectrum, because K40 is ubiquitous in environmental samples, and the 1460.8KeV emission is relatively free of interferences. Thereafter, the operator determines the order of spectral subtractions based on the peaks occurring in an actual sample spectrum.

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. 7.

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 7. 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, the following considerations make a point: 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	<u>net, peak counts/pCi</u>
Th	200.
UNat	40.
Co60	50.
Sr90	2.
Tc99	0.02
Cs137	400.
Eu152	40.
Am241	lost to interferences in this study
	-

After K40 and Cs137 spectra are subtracted according to the counts in their main gamma peaks, Sr90 is counted on an unlisted x-ray peak at 30KeV. Tc99 is counted on a bremsstrahlung x-ray peak. Notice the great range of detector sensitivity to radionuclides of interest for this study: >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.

A radiological analyst might appreciate 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.

Other ordinary checks indicate the reported data are appropriate for the publicinterest objective of the present study.

Appendix 3- Controversy About This Report

Events leading to publication of this report are chronicled in the Preface and the Introduction. This appendix includes review comments by the Washington Department of Health, Division of Radiation Protection, and by the Nez Perce Tribe, along with replies to those comments. A newspaper article based on the Nez Perce Tribe criticism is also included. This controversy has also been uploaded at <u>www.radioactivist.org</u>.

On 24 May 2002, the Washington Department of Health, Division of Radiation Protection, published Comments on a draft of this report. Those comments follow, within sidebars, with replies by Norm Buske and page numbers in this final report inserted *[italics in brackets]*:

	ERS 02-506						
May 24, 2002							
то:	Interested Parties						
FROM:	Debra McBaugh						
SUBJECT:	COMMENTS ON <i>Hanford Radioactivity In Salmon Spawning Grounds</i> by Norm Buske						
The Division of Radiation Protection has been involved in monitoring the radiological impact of Hanford on the Columbia River for over 40 years. Our annual reports document the higher levels and wider variety of radioisotopes that entered the River in the '60s and '70s, while the single pass production reactors were operating, as well as the decreases as those reactors went off-							

reactors were operating, as well as the decreases as those reactors went offline. As the restrictions due to national security lessened, our sampling became more extensive, spreading into more river related media and getting closer to the sources. Presently we collect water, sediment, water plants, and fish from the Hanford reach. We collect these samples in conjunction with other organizations so as to act as a quality assurance check on their generally larger sampling programs.

[I have participated in this joint sampling. Inclusion of the larger rocks is meaningless from a radiochemical standpoint becuase of their large volume to surface ratios, and because they decimate the analytical appearance of Hanford's impact on the riverbed.]

The Division acts as technical advisors to the Washington State Department of Ecology, thus allowing the State to better represent the interests and concerns of the people of Washington through the Tri-Party agreement. While the Division recognizes that the legacy of weapons production at the Hanford site evokes emotional responses regarding the contamination that remains, as well as the future designation of the land, the Division advocates using technically sound decisions concerning priorities and techniques when dealing with the Hanford issues that pose a potential threat to the public and the environment.

[The Division's participation is indeed valuable. Other points of view that differ from the agencies' perspectives are not necessarily "emotional".]

Interested Parties ERS 02-506 Technical comments on *Hanford Radioactivity In Salmon Spawning Grounds* Page 2

Over the decades there have been thousands of samples of a number of media collected from the Columbia River. They have been subjected to a wide range of analysis techniques, capable, in aggregate, of detecting and identifying any radioisotope. While it is possible, even likely, that our understanding of the radiological nature of the Hanford Reach will continue to be refined, it is extremely unlikely that a major source of ionizing radiation contaminating a major portion of the Hanford Reach has gone unnoticed.

[The large "rock" fraction of Hanford Reach sediments must be excluded to yield meaningful radiological analyses.]

In the last 40 years the Federal agencies responsible for the operation, and now the clean up, of Hanford have become progressively more open with the people of the State of Washington on what has and is currently happening at Hanford. This has been done through agreements with State agencies such as the Department of Ecology and the Division of Radiation Protection, and through select groups of interested, informed stakeholders such as the Hanford Advisory Board. Attempting to "re-invent" the system at this late date is likely to disrupt and delay clean up efforts, without producing any realistic expectations that decisions from the re-invented system would be any better, or even as good.

Our detailed comments are attached. If you have any questions, please contact Mike Brennan at (360) 236-3253 or e-mail at Mike.Brennan@doh.wa.gov.

Attachment [begins on the following page:]

Technical comments on *Hanford Radioactivity in Salmon Spawning Grounds* by Norm Buske

On page 4 Mr. Buske cites Nagler et al in their study which found evidence of sex alteration in wild salmon. The press release concerning the study clearly states:

"The researchers ruled out radiation as a possible cause of the apparent sex reversal but suggested environmental contaminants that mimic hormones or water temperature changes could be the culprits."

In telephone conversations and emails Dr. Nagler has reiterated this position, saying that there is nothing known about the interaction of radiation and fish that would support the idea that radiation is causing the sex change. Dr. Nagler is interested in doing laboratory experiments that would definitively rule out radiation as a possible cause, not because he believes it is the cause, but as the simplest way of putting this speculation to rest.

[I relied primarily on Nagler's scientific report rather than press statements, telephone calls, e-mails, or personal appearances. At the same time, I wanted readers of my report to know that the scientific conclusions might not yet be as clear as might have been imagined from Nagler's initial report:]

Mr. Buske also says, "But continuing research has muddled the scientific waters." This is a disconcerting statement to come from someone who claims to be a scientist. It says that he prefers incomplete information that can be misinterpreted to more complete information that may not support his position. Such an attitude strikes at the heart of science and it puts his scientific objectivity in question.

[Thank you. I've changed the text to make clearer the need for additional research.]

On page 4 Mr. Buske says, "Thus two new scientific questions arose by early 2001: Are the salmon spawning in the Hanford Reach subject to some environmental stress that is 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?"

It should be noted that nowhere in this paper does Mr. Buske *expressly* state that these two questions are linked, nor that he believes they are linked. He offers no discussion at all of how radiation might be responsible for the sex change in salmon.

[Science progresses step by step. With my draft report that is the subject of this review by the Division, there is now a theory that might account for sex change of salmon spawning in the Hanford Reach, if any such sex change exists. One step that other investigators are already studying involves clarification of the sex change question. In my report, I have begun to address another evident question: "What are the extent, character and origin of radioactivity in the riverbed of the Hanford Reach where the salmon spawn?" These two questions are part of a logical "theory" that connects U233 production at Hanford through solid radioactive waste disposal into the river at the old Wahluke Ferry crossing, with Eu152 fingerprinting U233 byproduct wastes in the riverbed, with potential effects, possibly on salmon alevin living in riverbed pore water.]

On page 5 Mr. Buske states there is a need for independent technical inquiry.

The Department of Health, through the Division of Radiation Protection, provides just such an independent technical check on the activities of the various contractors at the Hanford site. We engage in split and co-located sampling and evaluate environmental data gathered at and around the site.

[The public seems best served by more "independent" investigators having greater independence.]

The technical discussion within the paper is somewhat difficult to follow. It appears that Mr. Buske took some sediment samples and decided that Pb212 was in dis-equilibrium from the Th232 chain that it is a part of, in particular with Ac228. No analytical results for Ac228 or Pb212, the two isotopes Mr. Buske claims are not in equilibrium, are included in the report, therefore it is not possible to assess the validity of this contention. It is possible that what Mr. Buske interprets as dis-equilibrium is actually due to some aspect of his sampling and/or analysis. Ac228 has a half-life of 6.13 hours, and Pb121 has a half life of 10.6 hours. Both decay via beta decay. Both are usually considered naturally occurring isotopes, as the isotope at the head of the decay chain, Th232, is naturally occurring. Mr. Buske does not discuss how he eliminated any possible chemical and/or mechanical factors in the environment that might be responsible for this dis-equilibrium.

By way of background, several of the reactors at Hanford were used at various times to irradiate Th232 to make U233, which had weapons applications. When U233 is produced a small amount of U232 is also produced. The procedures used at Hanford produced U232 on the order of 10 parts per million when compared to U233. U232 is over 2000 times more radioactive than U233 (by weight) due to its shorter half-life (72 years for U232 compared to 158,500 years for U233).

[Declassified records reveal that all 9 of Hanford's production reactors produced U233.]

Mr. Buske speculates the "excess" Pb212 is due to the decay of U232, as U232 has a decay chain that includes Pb212, but does not include Ac228.

[I was testing for possible indicators of Hanford U233 byproduct wastes in the riverbed and concluded that both thorium itself and "Pb212 Excess" are "interesting but weak" indicators. My contract for this work requires me to report all results, including dead ends and weak leads such as these.]

He introduces a term "Pb212 Excess", which he does not define, but could be a ratio of Ac228 to Pb212. In Table 1 it is in a column labeled "Pb212 Excess" %", with numbers that have "+" or "-" signs in front of them. This makes this apparently key factor difficult to understand.

["Pb212 Excess" was an unproductive lead, rather than a "key factor".]

It would be useful if Mr. Buske calculated how much U232 would be required to generate the Pb212 Excess that he believes he found. It is quite possible that such a calculation would show that an impossibly large amount of U232 was needed, and thus one or more of the underlying assumptions are incorrect. If, for example, in order to produce the Pb212 Excess 8 grams of U232 needed to currently be in the Hanford Reach, there would have to be 1,000 kilograms of U233 in the same part of the river. It is difficult to believe that it would not have been detected. Given the pathway that Mr. Buske proposes later in the paper, this would, in all likelihood, require a number of tons of irradiated targets to be in the River, without factoring in the insolubility of the targets, or that decades have passed since the campaigns ended, which means even more targets would have had to have been in the River originally.

[I don't believe I yet have enough information to make this calculation. Anyone is free to calculate however they please.]

On page 7 [9] Mr. Buske says "Elevated Thorium levels are themselves another possible indicator from Hanford's thorium to U233 production runs." It is unfortunate that Mr. Buske did not speculate on the differences in the chemical and physical natures of thorium that has been processed and entered the river via dumping or target failure vs. naturally occurring thorium in its mineralized form. This might have shed some light on the source of the thorium that was found. It seems reasonable that processed thorium would be quite distinct from natural thorium, which is incorporated in the minerals that make up grains of sand.

[I did speculate in this regard and concluded that more analytic work is needed before reporting speculations.]

Mr. Buske's sampling technique (described on page 10 [10-12]) of repeatedly agitating the sample in water and discarding the suspended particles could, arguably, have concentrated the larger, denser particles that one might suspect contain natural thorium (thorium is about as dense as lead). His technique was similar in effect to gold panning, which uses agitation in water to allow the denser particles to be retained while discarding the less dense ones. This means that the results of his analysis, given in picocuries/gram (dry) in table 1 are not representative of the actual concentrations in the sediment.

[Thank you. My draft included a typo. Correction might have addressed some of this concern.]

Mr. Buske uses a ratio of 170 to 1 for the relationship of radioactivity between U233 and Th232 from the production runs. While this number ignores many important factors that would have to be considered in attributing the ratio in the finished fuel rods to a ratio in the river, it does illustrate a weakness in Mr. Buske's argument: for each 1 pCi/g of elevated thorium due to Hanford that he assumes is in the sediment, there should be 170 pCi/g of U233. While U233 is not particularly easy to identify by gamma spectroscopy, it is difficult to believe that in the thousands of sediment samples that have been taken the supposedly high levels (potentially hundreds of pCi/g, using Mr. Buske's numbers) of it has NEVER been noticed. And it has not.

[U233 is actually difficult to detect by photon means because its main emission, a thorium L-x-ray at 3.3% efficiency, has interferences with the same x-ray produced by natural uranium decays. Thorium L-x-rays have an energy of about 19.5KeV, which was below the LLD of my detector in 2001. With new equipment in 2002, it might be feasible to sort out the U233 by subtraction. Until such development is proven, U233 detection at the anticipated environmental activities probably requires alpha spectrometry. I don't know if anyone has ever done those analyses and so would have noticed U233.]

On Page 8 [10] Mr. Buske changes direction, describing a rationale for using Eu152 as an indicator for U233. This is based on the idea that Eu151 was a contaminate in the Th232 target rods, and that after it had been activated to Eu152 it entered the River by the same path as the supposed U233 contamination. Mr. Buske notes that it is not possible with the information at hand to calculate a U233/Eu152 ratio for reactor discharge or the waste streams. It is quite possible that given all the uncertainties and changing

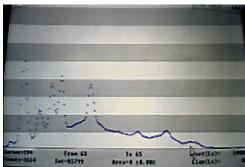
conditions, a useful ratio could never ever be developed, even if there were a link between the two.

While the production reactors were running at Hanford, Eu152 was discharged to the River, and to other locations where it entered the ground water. However, based on comparisons between the Eu152 levels from the single pass reactors compared to the N reactor, it appears likely that the Eu151 that was being activated dissolved in the river water used for cooling, and did not come out of the fuel and/or targets. Be that as it may, it is unlikely that any Eu152 remains in the Hanford Reach from that time.

[Eu152 remaining from Hanford's historic materials production era is routinely reported in Hanford Reach sediment samples, as described in the text and mentioned in the Nez Perce criticism; see below. The question of the origin of this Eu152 in the Hanford Reach riverbed still does demand more attention.]

Mr. Buske believes that Eu152 can be used as a surrogate for U233 because Eu152 is easily detectable by photon spectrometry. Unfortunately, the primary gamma peak for Eu152 has the same energy (1408.0 KeV) as Bi214, part of the U238 chain. Mr. Buske does not discuss how he dealt with this problem. If it was not corrected for, all of the conclusions that rely on his Eu152 results are fatally flawed.

[Please notice my Ref. 16. The 1408KeV peak in the Eu152 standard spectrum is the right peak above the cursor arrow, in Fig. 8, below. That peak was not used for Eu152 analysis because of interferences. The three large peaks at the left of the spectrum tested clean of interferences and were used for the Eu152 results.]



[Figure 8. Eu152 photon energy spectrum.]

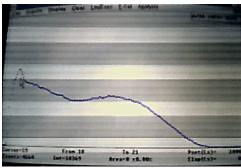
On pages 10 [11-] and 11 [-15] Mr. Buske describes his sampling technique, explaining that his intention was to reduce the Hanford influence in order to "...introduce a measure of conservatism..." Unfortunately, after quite possibly increasing the concentration of native Th232 himself, he then assumes the increase is due to Hanford activity. At very least Mr. Buske

should have split several samples, treating one half with his novel technique, and treating the other in a more standard manner. If nothing else, it would have told him if there was any value in washing out the fine particles.

[Thank you for identifying a typo. Hopefully, I've clarified my intention and the procedural conservativism in the fix.]

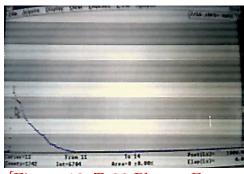
Mr. Buske used photon spectrometry to analyze his samples, but he includes columns for Th, UNat, and Sr90. It would be useful for him to define how he used photon spectrometry to quantify these, especially Sr90, which has no gamma peak, being a pure beta emitter. On page 32 [36] in Appendix 2 – Validation he acknowledges that Sr90 and Tc99 are not normally analyzed by spectrometry, but does not go into detail as to how he is able to do it.

[Sr90 decay is accompanied by an unreported x-ray at about 30KeV. A Sr90 spectrum is shown in Fig. 9, below, with the 30KeV peak above the cursor arrow. This peak is in a relatively "quiet" x-ray spectral region, allowing the good sensitivity of the pure beta emitter, as reported in App. 2. The Division of Radiation Protection has jointly sampled at N-Springs and elsewhere and reported comparable Sr90 results from their conventional techniques.]



[Figure 9. Sr90 Photon Energy Spectrum.]

[The Tc99 spectrum is shown in Fig. 10, on the next page. The analytical peak is subject to x-ray interferences, yielding the low sensitivity reported in App. 2.]



[Figure 10. Tc99 Photon Energy Spectrum.]

The data tables starting on page 13 [15] are not complete. The data for the various isotopes does not include Lower Limits of Detection, nor estimates of errors. The "Pb212 Excess" is not supported by data for Pb212 and Ac228, as noted above, nor is the meaning of the "+" or "-" in front of the number explained.

[See App. 2 – Validation.]

Mr. Buske implied there is a pattern in the data on table 1, but it is not obvious. Assuming his results are accurate, the positive results for Eu152 do start at D Island, but there is no apparent correlation to Th or UNat, which one might expect if Eu152 were an indicator of the supposed U233/U232 contamination (and assuming U233/U232 was connected to Th232 targets and reacted chemically like natural U). If anything, the data disproves his contentions.

[Any reader is welcome to evaluate the pattern that is the main result of this report. The reported pattern allowed prediction of the location of historic dumping of solid radioactive waste into the middle of the river, downstream of D-Reactors intakes and upstream of D-Island. Remains of the Wahluke Ferry crossing were thus discovered, based on this prediction. Such verifiable predictions are at the heart of science.]

Tables 2, 3, and 4 seem to be on a tangent. They give results for samples that are not connected with main subject of this report.

[True. My contract required me to report all results, even those tangential.]

The sample numbers Mr. Buske uses are interesting, as, according to his explanation of his numbering system (on page 12 [15]), the sample number includes the year, month, and hour of collection, but not the day.

[Thank you for identifying a typo. I've corrected the text, noting the sample numbering system is YMDDHH.]

The "Discussion and Implications" section (page 21 [24]) discusses the data from the tables, and what he believes it means. He places much importance on what he describes as a pattern of Eu152 contamination. The results that he lists on Table 1 correspond to as few as four counts in a 1500-minute counting period (based on the table on page 32 [38]). Without Lower Limit of Detection and Counting Error data, it is impossible to say if the results in the table represent actual detection. Additionally, as was discussed above, there is a possibility that naturally occurring Bi214 might have been misidentified as Eu152. Neither Th nor "Pb212 Excess" track with the supposed Eu152 contamination, even though in the body of the report Mr. Buske builds a case that all three should be connected with the supposed isotope of concern, U233.

[The results are in "pCi/g" while the sensitivity units are counts per "pCi". To compare the two, multiply the sample results by the sample mass: ~30g. See previous replies regarding the other matters.]

Mr. Buske assumes that solid radioactive waste from the Th232/U233 process was dumped in the river. From page 24 [28]: "This suggestion turns the initial question around: Rather than asking how Hanford officials could possibly have justified dumping solid U233 production wastes into the Columbia River, the question turns to: Why not?" The "suggestion" he refers to is an excerpt from a 300 Area history that states that special precautions for U and Th as radioactive substances were not taken in 321 Building. He goes from a statement that special precautions were not taken to assuming that large amounts, at least hundreds, perhaps thousands, of pounds of target rods that were likely hot enough to require special handling, were loaded into vehicles, driven through probably 30 plus miles of desert dotted with disposal sites, to a ferry selected because they calculated that dumping there was less likely to interfere with reactor operations. Mr. Buske does not attempt explain to why anyone would choose this involved disposal method over any number of much easier, more secure, methods which were more readily available. Nor does he speculate as to why no identifiable pieces have ever shown up, given that waste must be exposed to the currents of the river for this scenario to work at all, and would therefore have the possibility of washing downstream.

[There are anecdotal reports of pieces having turned up. The present report might stimulate a closer search for debris.]

On page 25 [29] Mr. Buske discusses his "synthetic H2O" samples, in which two sediment samples were agitated in distilled water, and the water processed and analyzed. He then takes the thorium result, (5.6 pCi/L) and multiplies it by 6 (the number of alpha decays in the Th232 decay chain). The result of 34, he states, is above the Safe Drinking Water limits of 15 pCi/L gross alpha. This implies either that Mr. Buske has a fundamental misunderstanding about how decay chains work, or that he believes that the Th232 decay chain is in equilibrium *despite the fact that the chain not being in equilibrium is why he constructed this whole U232 argument in the first place.* Additionally, the Safe Drinking Water Act is not intended to imply that any organization, even the Department of Energy, is responsible for altering the physical and chemical properties of water so that other parties such as Mr. Buske can not make things dissolve or go into suspension.

In summary:

• There is no scientifically sound reason to assume the sex change in the salmon from the Hanford Reach is due to radiation, and there is sound reason to assume that it is not.

• The speculation that that "Pb212 Excess" indicates the presence of U232 is not well developed, and does not discuss other possible explanations. The results given on Table 1 do not seem to support the contention that there is in fact a "Pb212 Excess" at all.

• The speculation that Eu152 is connected to U232/233 contamination is not well developed, and other possible sources of Eu152 are not explored. The possibility that another isotope has been misidentified as Eu152 can not be discounted.

• The sampling technique for collecting the sediment was non-standard, and may have biased the results. At very least it made the samples nonrepresentative of the sediment in the River, which the young salmon would be exposed to, and which was presumably the purpose of this study.

• The data tables do not include enough information to assess the validity of the results given.

• The data used to claim there is a pattern of contamination is weak and contradicted by other data.

• There are no estimates of how much material would have to be disposed of into the River in order to produce the levels of either Eu152 or "P212 Excess" supposedly found.

• No sound argument is provided to show that large amounts of the material being produced at great expense would be disposed of in the River.

[See my replies, above, to specifics of critical comments. In other regards, this report speaks for itself. The reader is welcome to decide between differing viewpoints regarding this controversy.]

At the end of May 2002, the Nez Perce Tribe distributed by fax the following criticism of an early draft of this report:

MAY. 30 '02 (THU) 15:36 TRI-CITY HERALD NEWS



ENVIRONMENTAL RESTORATION & WASTE MANAGEMENT P.O. BOX 365 · LAPWAI, IDAHO 83540-0365 · (208) 843-7375 / FAX: 843-7378

May 28, 2002

Tom Carpenter Government Accountability Project 1402 Third Avenue, Suite 1215 Seattle, WA 9810

Post-it* Fax Note	7671	Date		# of Pages	
	enter	From	John	Ga	ne
Co./Dept.		Co.	7-6	Here	1
Phone #		Phone	507-	-582	-1512
Fax# 206-29	2-06/0	Fax •			

RE: Review of "Hanford Radioactivity in Salmon Spawning Grounds – quality, extent, and some implications

Mr. Carpenter:

The Environmental Restoration and Waste Management Program (ERWM) appreciates the opportunity to review and comment upon Mr. Buske's report of Hanford radioactivity in salmon spawning grounds. The ERWM Program has reviewed the above-mentioned document and has serious concerns with both the substance of the report and the effect the existence of such radioactivity might have on salmon spawning grounds on the fishery resource throughout the Columbia River Basin.

In 1855, the United States negotiated a treaty with the Nez Perce Tribe. (Treaty of June 9, 1855, with the Nez Perce Tribe, 12 Stat. 957 (1855). In Article 3 of this treaty, the Nez Perce Tribe explicitly reserved to themselves certain rights, including the exclusive right to take fish in streams running through or bordering the Reservation and "the right to fish at all usual and accustomed places in common with the citizens of the Territory." These rights include the right to fish, hunt and gather within area now occupied by the Hanford Nuclear site.

Salmon are integral to the spiritual, physical, and economic health of the Nez Perce people and the fisheries that provide this life and sustenance are revered. As such, potential contamination problems related to salmon and other fish in the Columbia River Basin System are of great concern to ERWM and tribal members. The treaty-reserved interest in these natural resources in the Hanford Reach of the Columbia River and throughout the Tribe's traditional use areas, which have been repeatedly recognized and reaffirmed by Federal courts, would be greatly diminished if the fishery and the habitat that supports it are not protected and preserved.

General Comments

As a Natural Resource trustee for the Hanford area, it is the responsibility and obligation of the ERWM to review these reports in order to provide recommendations and disseminate accurate information to tribal members. To this end, ERWM has consulted with technical experts within and outside the Tribe to ensure that the specific comments detailed below will be of assistance to the Government Accountability Project (GAP).

The ERWM has numerous concerns with the Hanford Radioactivity report. The report is not well-written, thereby causing confusion; is often scientifically misleading; and creates a perception about thorium and europium contamination in salmon and sediments that is not based on a rigorous application of the scientific method. While not addressing grammatical problems per se the ERWM is disappointed that GAP would allow a report to be disseminated without ensuring that it is both well-written and technically defensible.

Moreover, the ERWM Program does not believe that reports of this nature aid service to the public; rather, they foster an atmosphere of mistrust that most agencies, including the Department of Energy (DOE) has been trying to eliminate for years. The ERWM has witnessed dramatic changes in the last few years at Hanford that reflect a positive movement away from old management styles and towards greater transparency. For instance, a Biological Resource Management Plan for the site now exists, requiring mitigation for lost or injured natural resources and a fully functioning natural resource trustee council and a stakeholder advisory board are in place that serves as models throughout the DOE Complex.

While the ERWM certainly welcomes any kind of technical report that refines the ecological and human risk assessment models for the Hanford site or provides additional information on contaminant uptake by natural resources to ensure the adequate protection for both the resource and those that depend on them for sustemance, reports such as this fail to serve these purposes.

Specific Comments:

Concerns With Introduction Section

When reading the Introduction one is struck immediately by the anti-DOE bias that is expressed. This immediately makes a reader wonder if the author can conduct an objective scientific investigation that is not predisposed to a specific point of view.

The 8th Paragraph on page three suggests that thorium activity in mulberry leaves was 10 times background level. Since there is no explanation in the document explaining these high levels we can only assume the author is referring to previous reports that GAP has issued relative to these types of projects. If this is so, we need to call attention to the fact that the scientific community discredited those reports concerning contaminant uptake in leaves (EPA letter to GAP Sept 4, 1999 Re: EPA Perspective on Government Accountability (GAP) Reports).

On page 4 statements are made concerning the assertion that some preliminary research has suggested that salmon are changing sex. This statement is misleading because no salmon have been documented to actually have changed sex. What the research does show is that a genetic marker normally found in males was also found in females (James Nagler, presentation to Nez Perce Tribe on May 21). This has also been documented in situations occurring elsewhere where nuclear waste operations could not be implicated. We, like others are awaiting detailed scientific studies that may shed further light on this phenomenon.

The ERWM Program wishes to note that not all of the contamination found in the Columbia River can be attributed to Hanford nuclear waste operations. A significant amount of dioxins, furans, PCB's, heavy metals, and other contaminants have their origins upriver at aluminum plants, mining operations, agricultural operations, and pulp mills. The ERWM does not rule out the possibility that Hanford contaminants may be impacting the salmon; however, this report must present a more thorough review of the contamination problem. Moreover, the report fails to reveal whether the areas where the sediments were collected have any correlation to fall chinookspawning grounds. This makes the title of this report very misleading. A better title might have been "Radioactivity in Hanford Reach Shoreline Sediments."

The last paragraph on page 4 makes reference to a video that is being produced with no explanation of content or purpose. This paragraph should be deleted.

Concerns With Objective Section

The first sentence is possibly appropriate for the representative statement of an objective except that the term "radioactivity" does not appear to be bounded. Does this study strive to address a suite of radionuclides or is it primarily focused on one, two, or some other smaller number of radionuclides? The second sentence doesn't represent an objective but rather should be reserved for a Methods Section. The third sentence discusses impacts, but does not provide any clarification of the impacts being evaluated.

The study does not state a problem and does not form a hypothesis. Instead, it states an objective, and that is "to characterize the extend and intensity of radioactivity entering the Hanford Reach riverbed water.... This effort seeks initial understanding of the main impacts on salmon alevin and other biota living in the riverbed." It does attempt to do the former, though the results are questionable in part because the method is unclear. But an understanding of the impact of the information on the salmon is presented only through conjecture, not the scientific method.

Concerns With Problems Section

It is difficult to determine the value of this section. Mention is made of the need to develop a sampling procedure. Part of the problem the author states, was the "lack of sufficient knowledge at the outset."

Universally accepted soil and sediment sampling procedures have been utilized by the scientific community for years. Dozens of universities, government agencies and professional organizations conduct this type of work routinely. Hanford scientists and technicians have used these proven methods for years.

The last sentence in this section states that the problems were addressed by "taking a variety of risks." The reader is not provided any explanation of what these risks might be. Were they personal risks, risks associated with data interpretation, or something else? Did these risks potentially compromise the study results? This sentence should be eliminated or the risks should be explained and justified as to there benefit to the study.

Concerns With Methods Section

Generally speaking, this section does not appear to meet the usual requirements of a Methods Section because conceptual issues are raised. There is no discussion of the sampling methods used, information on the study design, and the analytical or statistical methods employed

The first paragraph in the Methods Section is irrelevant and incomprehensible. The paragraph is grammatically incorrect, does not flow well, and obscures the point the author is attempting to make.

The second paragraph is equally confusing. While it appears that the real objective of this report is to document Hanford impacts to salmon, the objective on page five states that the study seeks

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to "characterize the extent and intensity of radioactivity entering the Hanford Reach riverbed water." Is this report studying the impacts to the river or to salmon? They are two totally — different questions and would require completely different methodologies.

Also disturbing is the fact that the ERWM Program was unable to find anywhere in this report where it is documented that salmon had been collected and analyzed to determine potential impacts from any kind of Hanford contaminant.

The problems continue in the fourth paragraph on page nine where a one-sentence aspersion on "Hanford's still semi-secret thorium campaign" is not backed up by any evidence. Moreover, a characterization of this accusation as "sinister" is not appropriate for a scientific report. Again, this appears to be evidence of a clear bias against DOE and raises real questions about the objectivity of this report.

Concerns With Results Section

Much of the information provided in the first 5-7 paragraphs is more appropriate for the Methods section

The first paragraph is not factual. The author asserts that the "work reported here is unusual in its orientation toward a particular objective. Scientific studies are usually less focused." On the contrary, rigorous studies that follow the scientific method are highly focused and totally objective. Hypotheses are clearly stated and rigorously tested using both physical, empirical, and statistical methods. This report suffers both from focus and objectivity.

Starting in the third paragraph the reader is informed that sediment samples were collected but little information is provided on sample methods. All of this information and more precise details should have already been presented to the reader in the Methods Section.

The description of the soil sampling method in the fourth paragraph of page 10 is not well presented nor is there enough detail given where another researcher could duplicate their procedures. The report states, "sampling was sparse where little or no artificial radioactivity was found." What method was being used to determine radioactivity in sediments prior to sampling? How much wet material was sieved from the river and how was the 30 g of material removed for analysis?

We do not comprehend the logic in removing the fine silts and clays from the sample, because there remains a large range of particle size variation in the 2 mm down through coarse silt size that was not used. There seems to be a concern about the variability of size of the fine fraction, but not of the coarse fraction. It is not clear to us why. In fact, there is a concern that the sample medium is "reasonably constant", but never explain just what is meant by "reasonably constant". We infer that a relatively constant grain size is desired in the sample, but we're not convinced that this procedure helped produce any significant consistency in grain size

The reader is not informed in the text concerning the number of samples collected nor given any information about the quality and qualifications of the "independent" analytical laboratory that was used.

Analytical Methods and Accuracy of Radionuclide Values

Based on the limited description of the laboratory equipment that was used to analyze the samples in this study, we are very skeptical that this equipment can measure the extremely low levels that were reported.

As part of our review we have consulted with other scientists who are familiar with the analytical methods and laboratory equipment used in this report. We will only say that there is a considerable amount of doubt in the scientific community that the instruments and methods used have the capability of measuring and producing such low numbers as reported here. In addition, a clear description of analytical instrumentation employed should be available in the Methods Section and is not.

In any kind of scientific study, and especially a study that is conducted at the Hanford Site, it is imperative that an EPA approved independent laboratory analyzes the samples. This ensures that: (1) the researcher can't be accused of bias; (2) approved standard methods are used; and (3) counting errors are reported with each detectable value. Certainly, if DOE were to conduct a radiological study at Hanford and use their own methods in their own laboratories, the GAP and other organizations, including the ERWM Program, would raise serious concerns about the credibility of the results generated from such a study. If we want to hold DOE and its contractors to these high standards then other organizations conducting studies at Hanford should also abide by the same standards.

Two years ago the ERWM Program collaborated with DOE and Bechtel to collect soil and biological samples on the North Slope. An independent EPA certified laboratory in the eastern U.S did all of the data analysis. In our opinion GAP should have utilized a similar laboratory and approach to analyze their samples in order to avoid any questions of bias and objectivity.

The ERWM Program is also concerned with the absence of counting errors associated with each sample result and there isn't any basic statistical information provided such as the standard deviation, standard error, mean, or other variables. Often, when dealing with radionuclide data, the counting error may be greater than the value of a given sample and indeed it is not possible to objectively interpret the data without the counting errors being known. Associated counting errors must be included to provide credibility.

Concerns With Discussion and Implications Section

The report states that 60 % of the riverbed is contaminated with solid, radioactive waste. We see nothing in his paper that supports a claim of 60% of the riverbed. It is clear that 60% of the riverbed was not sampled. It is, however, unclear how many samples were collected, i.e., whether the sixty locations represent sixty samples or multiples of sixty, or multiples adding to sixty.

The assertion that bullet \ll shows a pattern of Eu-152 contamination isn't substantiated in our opinion by the data presented in Tables 1-4. The data for the most part appear to be remarkably consistent and, in fact, don't exhibit a wide range in variation that often occurs when analyzing radionuclides. Simply looking at the data in absence of any associated laboratory and counting errors is not good science and is not an acceptable method for analyzing or interpreting results.

As a result of these concerns, we conducted an independent assessment of the Eu-152 sediment results in Table 1. Making our own graph we quickly ascertained the following: (1) thirty-three of the fifty-six samples collected were "no detect;" (2) nine of the seventeen samples collected downstream from the F reactor were "no detect;" and (3) two of the five samples located the farthest downstream were "no detect." Thus, it appears that six sets of samples (12 individual samples) were essentially collected at the same location (no explanation given) and, in the case where both samples detected radioactivity, the data overstates the influence of europium or other radionuclides. If these six data points were removed, the computed mean value would necessarily decrease, thereby further undermining this study

The author states in the third paragraph about Pattern <2> that this "extensive pattern is likely of great concern for the general viability of the Hanford Reach riverbed habitat and will be discussed shortly." However, we find no evidence from the data that there is an extensive pattern and no evidence is presented about how these levels that are mostly below 1.0 pCi/gm affect the viability of the riverbed habitat.

A Master's thesis study at the University of Idaho (An Investigation of the Origin of Eu-152 in Columbia River Sediment by Gregory Gibbons, July 2000) reported on Eu-152 levels in sediments that were collected and analyzed in the McNary Reservoir. This study concluded that neutron activation of naturally occurring sources of Eu-151 in Columbia River water was the likely origin of the Eu-152 contamination observed at the Hanford 100-Area and in the sediments at McNary reservoir. These results refute the author's assertion that contaminated materials were being dumped into the Columbia River from the Wahluke Ferry.

We are also aware of radionuclide soil concentrations that DOE is proposing to use as a starting point for the ecological risk assessment in the 200-CW-1 and 200-CW-3. These numbers can be found in Table 1-7 of the Draft DQO Ecological Assessment of 200-CW-1 and 200-CW-3 Ou's and 200 North Area Waste Sites (BHI-01629) and represent soil threshold values.

The biota concentration guide in pCi/gm for suropium 152 is 1,400 pCi/gm that is two orders of magnitude above any values in GAP's report. Thorium-228, 230 and 232 BCG's are 2,200, 2,700 and 2,000 pCi/gm respectively. This provides some indication of the bias in attempting to promote concerns at the low concentrations provided in the reviewed report.

Beginning with the sixth paragraph on page twenty-four, the study loses the remainder of its credibility as a result of unfounded accusations unsupported by scientific study. For instance, the assertion is made that accumulating evidence suggests that the Wahluke Ferry was used to dispose of solid radioactive waste. Certainly, if there were enough such evidence, the GAP should have presented it here or in other reports. However, no such evidence exists. The ERWM also fails to see any relevancy in discussing the Wahluke Ferry being used to dump waste into the river, because it is not logical or factual. These types of accusations and statements should never be part of a "scientific study" and is another example that strongly questions GAP's ability to conduct an objective study.

Concerns With Conclusions and Recommendations Section

The ERWM Program does not agree with the first three conclusions. References continue about Hanford's secret thorium campaigns and the assertion that the ferry system was used to dispose of solid radioactive waste. The remains of the ferry system the author refers to show no evidence of being relics of "the old waste disposal system." Further, there is no evidence that salmon are being threatened by old Hanford radioactivity. We would certainly welcome any studies that

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could document any kind of impacts to salmon. This study certainly did not do that and accusations without good scientific evidence do not serve the ERWM Program or the general public any good.

CONCLUSION

In conclusion, the ERWM Program has grave concerns with the credibility and accountability of this study. The ERWM respectfully requests that the GAP withdraw this study and republish it taking into consideration the concerns mentioned above.

Sincerely,

Cc:

Patrick Sobotta ERWM Director

Justin Gould (NPTEC) Doug Dampier (CRITFC) Patty Howard (CRITFC) Keith Klein (DOE) Dana Ward (DOE) Kevin Clarke (DOE) Connie Smith (DOE) Jamie Zeisloft (DOE) Tod Martin (HAB) Larry Goldstein (NRTC Council) Russell Jim (Yakama Nation) Jay McConnaughey (Yakama Nation) Tom Zeilman (Yakama Nation) Stuart Harris (CTUIR) Ron Mitchell (Duratek) Ted Poston (PNNL) Dennis Dauble (PNNL) Brett Tiller (PNNL) Roger Dirkes (PNNL) Larry Gadbois (EPA) Mike Wilson (Ecology) John Price (Ecology) Wayne Soper (Ecology) Ray Johnson (Fluor Daniel) Richard Roos (Fluor Daniel) Ken Gano (Bechtel) Steve Weiss (Bechtel) Susan Hughes (Oregon DOE) Laurie Vigue (Washington Fish and Game) Greg Hughes (USFWS) Al Danielson (Dept. Health) John Stang (Tri-City Herald)

Reply To Nez Perce Tribe Criticism:

June 07, 2002

Patrick Sobotta ERWM Director Nez Perce Tribe P.O. Box 385 Lapwai, ID 83540

<u>Re: Your letter of May 28, 2002, to Tom Carpenter, commenting on my draft</u> <u>report</u>,

"Hanford Radioactivity in Salmon Spawning Grounds - quality, extent, and some implications,"

Mr. Sobotta:

Your expedient publication of the Nez Perce review of my draft report has served the public concern for the health of salmon spawning in the Hanford Reach.

I reply here to two of your issues: (1) Laboratory certifications and public confidence in results. (2) The origin of europium-152 in the Hanford Reach riverbed and its significance. I hope to clarify these two issues now, before the June 10th public meeting of the Hanford Scenarios Task Force on the topic of the Hanford Reach river corridor. I'll address other issues in my final report for which you have provided helpful review comments; thank you.

(1) Laboratory certifications and public confidence in results. You commented that radiological analyses should only be conducted at an EPA approved, independent laboratory. However, the basic requirement for *scientific study* is for replicatability of results rather than for approvals or certifications.

I have adhered to that scientific requirement in my technical study by archiving samples so they can be re-analyzed by any critic and by describing my sampling method and providing precision (GPS) locations of my sample collections in Table 1 of my draft report at <www.radioactivist.org>. Anyone who doubts my analytical results, you are free to check out the radiological facts directly. (2) The origin of Eu-152 in the Hanford Reach riverbed and its

significance.

You say the source of the Eu-152 that I've reported in the riverbed is naturally occurring europium in Columbia River water having been irradiated when it passed through Hanford's old nuclear reactors. That possibility was addressed in the U.S. Fish and Wildlife Service, Upper Columbia River Basin Field Office, October 1999 Assessment Plan: "Hanford Site 100 Area Assessment Plan, Vol. 1: Columbia River Aquatic Resources, <www.hanford.gov/boards/nrtc/100apfin.doc>, p.52. The Assessment Plan explained the presence of Eu-152 in deeper sediments behind McNary Dam, downstream of Hanford, by the fact that radionuclides readily adsorbed onto fine silts and clays that settled to the bottom of the quiescent pool above the dam.

As that Assessment Plan explained, "There are few areas within the Hanford Reach where fine silts and clays are deposited in the mainstream of the river." The mainstream of the Columbia River through the Hanford Reach is too turbulent for the fine particles that had passed through the old reactors to

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settle to the bottom of the river. The physics of the situation therefore demands a different explanation for the Eu-152 found in the Hanford Reach riverbed from the Eu-152 found in McNary Dam pool sediments.*

The actual pattern of Eu-152 in the riverbed turns out to be most interesting. I found *no* concentrations of Eu-152 immediately downstream of *any* of the old Hanford reactor outfalls; see Table 1 of my draft report. If the Eu-152 in the riverbed had entered the river from the old reactor outfalls, as you say, then I would have found more Eu-152 just downstream of at least a few of the old reactor outfalls.

I tracked the Eu-152 in the riverbed back upstream to its source point in the middle of the river, downstream of D-Reactors intake and upstream of D-Island:

upstream of D-Reactors outfalls

I found no Eu-152 in the riverbed near any of the old reactors that are farther upstream: B-, C-, KE-, KW-, or N-Reactor.

Based on my finding of the upstream end of the Eu-152 pattern in the riverbed, I predicted that I could find remnants of whatever structure the old Atomic Energy Commission could have used to transport solid radioactive waste into the middle of the river and dump it there. That prediction led me to search a few hundred meters of Hanford shoreline, downstream of D-Reactors intake and led to my discovery of the remains of the old Wahluke ferry crossing, upstream of the D-Reactors outfalls.

Such predictions-and-confirmations are at the heart of science.

Based only on the early radiological indications from my reported work, Tom Carpenter of the Government Accountability Project, had requested all Hanford documents related to waste disposal from Hanford's old thorium-touranium-233 production campaigns. To its credit, the Department of Energy produced a list of over 50,000 relevant document titles. See, for example, HAN-53744, 2/15/54; HW-78100, 6/27/63, p.23; U-233-Thorium Program Letter -Chemistry Department, 7/15/65. I sketched Hanford's U-233 production history in Appendix 1 of my draft report, as gleaned from hundreds of documents I selected for review on the basis of their titles.

Those documents revealed that still semi-secret uranium-233 production, primarily for mini-nuclear weapons, was a main Hanford operation for much of the facility's history.

The fact that still-secret U-233 production is the source of solid radioactive waste labeled by Eu-152 in the Hanford Reach riverbed is important. It means Hanford is still unwilling to probe its old, but still-secret weapons material production practices, such as solid radioactive waste dumping directly into the Columbia River. It means government agencies are still unwilling to monitor U-233-related radioactivity in the riverbed in a meaningful way. (Notice that U-233 is not even mentioned in the above-referenced <www.hanford.gov/boards/nrtc/100apfin.doc>.)

What this might or might not mean for salmon that spawn in the Hanford Reach National Monument is not yet known.

Hoping this clarifies two of the controversial issues you raised, (signed original) Norm Buske The RadioActivist Campaign <search@igc.org> * I've not yet located your Gregory Gibbons' Masters' thesis reference.

ec: Al Conklin, Arjun Makhijani, Amber Waldref, Art Tackett, Barbara Wise, Betty Tabbutt, Carole Woods, Bill Kinsella, Casey Ruud, Charles Weems, Chris Chamberlain, Chris Chandler, Christina Richmond, Cindy Meyer, Dana Ward, Daniel Simpson, Darrell Fisher, Dave Rowland, Dave Smith, David Cortinas, David Johnson, David Stensel , David Watrous, Deanna Henry, Debra McBaugh, Dennis Faulk, Dirk Dunning, Doug Huston, Fred Miller, Fred Roeck, Gai Oglesbee, Gariann Gelston, Gerald Pollet, Glen Spain, Greg deBruler, Harold Heacock, Helen Wheatley, Jackrabbit News, James Cochran, James Thomas, Jeff Luke, Jeffrey Van Pelt, Jerry Peltier, Jim Curdy, Jim Hagar, Jim Lynch, Jim Trombold, John Stang, Joe Jackson, John Erickson, John Stanfill, Joy Turner, Karen Dorn Steele, Keith A. Smith, Ken Niles, Kenneth Bracken, Kim Ballinger, Kristy Collins, Larry Gadbois, Larry Jecha, Leon Swenson, Linda Josephson, Linda Mays, Lisa Stiffler, Lorraine Eckstein, Lynn Lefkoff, Lynn Porter, Lynn Simms, Madeleine Brown, Mark Beck, Mark Sautman, Marla Marvin, Martin Bensky, Mary Ann Wuennecke, Max Power, Maynard Plahuta, Melinda Brown, Michael Gearheard, Michelle Anderson-Moore, Moon Callison, Nancy Myers, Natalie Renner, Norm Dyer, Paige Knight, Pam Brown, Patrick Sobotta, Penny Mabie, Peter Bengtson, Rick Bond, Rick Leaumont, Robert King, Robert Larson, Robin Klein, Ron Skinnarland, Ross Ronish, Russ Brown, Russell Jim, Ruth Siguenza, Ruth Yarrow, Sergey E.Pashenko, Shelley Cimon, Steve Wiegman, Sue Safford, Susan Babilon, Susan Coburn Hughs, Susan Gordon, Susan Leckband, Takaro Tim, Tammie Holm, Ted Grudowski, Ted M. Poston, Thomas Schaffer, Tim Connor, Todd Martin, Tom Carpenter, Tony Young, Trisha Pritikin, Wade Ballard, Wade Riggsbee, Wanda Munn, William Kinsella, WorkTeam3 of Tides Center

On 31 May 2002, John Stang reported the Nez Perce denunciation of a draft of this report in the *Tri-City Herald* [29]:

<u>Nez Perce denounce maverick scientist's Hanford waste theories</u>

This story was published 5/31/2002

By John Stang Herald staff writer

A Nez Perce review found that gadfly scientist Norm Buske's research on radioactive thorium and europium in the Hanford Reach is too flawed to be valid.

A draft report by Buske "is often scientifically misleading," said a letter this week from the Nez Perce Tribe's Environmental Restoration and Waste Management Program to the Government Accountability Project, a Hanford watchdog organization.

On Thursday, Buske welcomed the criticism, saying: "I stand by the report."

Buske is a Belfair-based maverick scientist and Hanford critic who often works with GAP.

Although his report is not published yet, Buske presented a draft of the study to the Hanford Health Effects Subcommittee in January.

In it, Buske said his studies showed concentrations of thorium and europium in the Columbia River that exceed government guidelines.

He extrapolated those readings to mean Hanford created massive "semi-secret" amounts of uranium 233 for mini atomic bombs. Buske also contended that the thorium and europium threaten salmon. The Hanford Reach is a major salmon spawning area.

For more than 40 years, Hanford's plutonium fueled the Cold War buildup of atomic bombs, but even as secrecy eased in recent years, officials have never indicated that the site produced more than a handful of experimental uranium 233.

Europium and thorium also rank extremely low among the substances that Hanford worries about.

Hanford health committee members expressed skepticism in January about Buske's conclusions. And some wanted this theory to go through a peer review in which outside experts double-check a researcher's work, a standard practice in scientific publishing.

The Idaho-based Nez Perce, who have cultural ties to salmon and the Hanford Reach, sought experts to conduct such a peer review.

In his Tuesday letter to GAP, Patrick Sobotta, head of the tribe's environmental restoration program, wrote that Buske's research appeared biased, did not follow universally accepted scientific procedures, contained little or no information on his methodologies, was unfocused, did not use an independent lab to analyze water samples, and did not back up its conclusions on paper.

"If we want to hold (the Department of Energy) and its contractors to these high standards, then other organizations conducting studies at Hanford should also abide by the same standards," Sobotta wrote.

The Nez Perce said Buske's research did not prove Hanford's radioactivity has harmed salmon, although the tribe does not rule out other studies showing such harm.

The Nez Perce letter and Buske's draft did not address current Hanford efforts to see if seeping nonradioactive chromium might harm newly hatched salmon.

The Nez Perce letter also said that Buske's did not prove his claims that greater-thanexpected amounts of europium and thorium were found.

Buske's "aspersion on 'Hanford's still semi-secret thorium campaign' is not backed up by any evidence," the letter said.

Meanwhile, Buske and GAP attorney Tom Carpenter said his draft paper was not intended to be a full-fledged scientific study.

"We don't have the budget to do a scientific study (of the scale and thoroughness that the Nez Perce said should be done). ... I disagree you can't publish anything on Hanford without a multi-million-dollar study," Carpenter said. Instead, Buske and Carpenter described Buske's draft as something to provoke DOE, Hanford regulators and others into studying the issues it raised. Buske hopes his report will prompt DOE to sample the Columbia River for thorium and europium.

He believes these are dangerous substances that Hanford and its regulators are not seriously addressing.

Buske calls himself a "technical critic" of the "scientific establishment," contending politics usually influence scientific studies. And he characterized the Nez Perce's criticism as partly motivated by politics.

Carpenter and Buske believe Hanford created massive amounts of uranium 233, which flew under the public's and regulators' radars. Buske contended that uranium 233 was intended for small tactical nuclear weapons.

Uranium 233 was examined decades ago for possible use in atomic bombs or as a nuclear fuel. Normally, this nation's nuclear weapons used cores of uranium 235 or plutonium.

In January, DOE provided the Herald with all the references officials said they could find -- a handful of mostly 1963 documents -- that briefly discussed Hanford's experiments to create uranium 233 to use as a fuel or for atomic bomb cores.

However, a 1999 DOE report said uranium 233 was never used for bomb cores or fuel because of technical problems. Buske claimed roughly half of Hanford's production output consisted of uranium 233.

Last year, Carpenter sent a memo to DOE's Hanford Manager Keith Klein, citing a 1968 Hanford report that said the KE and KW Reactors produced 1,012 pounds of uranium 233. And the memo referred to an earlier report calling for creation of 286 pounds of uranium 233.

Carpenter and Buske anticipated that Buske's draft would be criticized, and welcomed that as healthy public debate.

"We expected controversy," Buske said.

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References and Notes

- 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.
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- [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.
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- [13] C.A. Hampel (ed), 1968, *The Encyclopedia of the Chemical Elements*, Reinhold Book Corporation, New York NY, p. 208, 211, 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 p.87, "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/100772975913455173.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.
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- [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.
- [29] J. Stang, 31 May 2002, "Nez Perce denounce maverick scientist's Hanford waste theories," Tri-City Herald, Pasco, WA, <www.tricityhearld.com/news/2002/053/story4.html>.

Acknowledgments

First thanks to Moon Callison for laying out Figure 1, preparing Figure 2, reviewing report drafts editorially, and formatting in PDF. Many thanks also to Tom Carpenter for editorial comments, suggestions, and questions, to Alison Marti for editorial assistance, and to Aileen Luppert for editorial assistance.

A special thank-you to John Warner for his information about the Wahluke Ferry, which solved a mystery.

Thanks to the many independent reviewers of this report, to Mike Brennan for his extensive review comments for the Division Of Radiation Protection of the Washington Department of Health, and to Patrick Sobotta and his reviewers for the Nez Perce Tribe for their comments and conclusion. Finally, thanks to John Stang for first reporting this controversy to the public.

About The Author

Norm Buske has masters degrees in physics from the University of Connecticut and in oceanography from the Johns Hopkins University. Norm holds three patents. He has been a member of the American Association for Advancement of Science, American Physical Society, the American Society for Testing and Materials, American Society of Mechanical Engineers, American Society of Professional Engineers, Institute of Electrical and Electronics Engineers, and National Fire Protection Association. Norm has quit his professional memberships to be completely free to act as an independent critic of USDOE's nuclear facilities. Norm received a Certificate of Honor Award from the Alliance for Nuclear Accountability in 2001 for his "scientific and technical investigations of the environmental consequences of nuclear weapons production in the United States and Russia."

After being arrested in the course of his work on the shore of the Hanford Reach in 1999, Norm was awarded a unique agreement for access to the Hanford site to conduct independent radiological studies. Norm is a member of the Amchitka Technical Advisory Group (ATAG), an alternate member of the Hanford Advisory Board (HAB). Norm has been a member of the HAB's Hanford Scenarios Task Force, where he reported the results of the present study. Consequently, the Task Force concluded that Hanford cleanup should address the riverbed and the salmon alevin as an indicator population.

Norm has conducted non-governmental, in-field, radiological investigations of American and foreign nuclear weapons and nuclear navy facilities since 1983, independently, for Greenpeace, for the Government Accountability Project, and for others. In 1990, Norm prepared jam from strontium-90 tainted mulberries picked from boughs of trees overhanging the Columbia River at Hanford's N-Reactor. He sent bottles of the jam to Washington's governor Gardner and Energy Secretary Watkins to call attention to the threat posed to the river.

Later that year, Norm assembled a spectrometer laboratory on Greenpeace's RAINBOW WARRIOR and sailed to Mururoa. He found cesium-134 contamination in samples of plankton collected from international waters off the French Nuclear Test Site. This began a new round of international protest, which led to the site's permanent closure in 1995.

In 1996, Norm conducted Greenpeace's 25th anniversary study, on Amchitka Island, Alaska, site of the world's largest underground nuclear explosion. Norm discovered americium-241 leaking from the blast cavity into White Alice stream and flowing into the Bering Sea. The next year, Norm provided oversight for USDOE's follow-up study. Greenpeace published the controversial results of that follow-up in "Nuclear Flashback, Part II."

In 2000, Norm participated in a Russian-American technical reconnaissance survey around four Russian nuclear facilities. This recon discovered the largest dumping of radioactive waste into the aquatic environment, which made international news.

In addition to his GAP contract work, Norm is Director of The RadioActivist Campaign (TRAC) of the Tides Center of San Francisco. Norm conducts all in-field scientific investigations at TRAC and runs TRAC's in-house spectrometer laboratory. His forte is acquiring crucial data of radiological concern, from public lands and waters and then bringing the implications to public attention through mainstream media, 35mm slide shows, and public meetings and hearings.

About The Government Accountability Project

The Government Accountability Project is a non-profit organization based in Washington, D.C. and Seattle, Washington. The Project has conducted oversight at Hanford since 1987, through representation of whistleblowers, participation in advisory boards and councils, and environmental sampling. GAP's webpage is -http://www.whistleblower.org