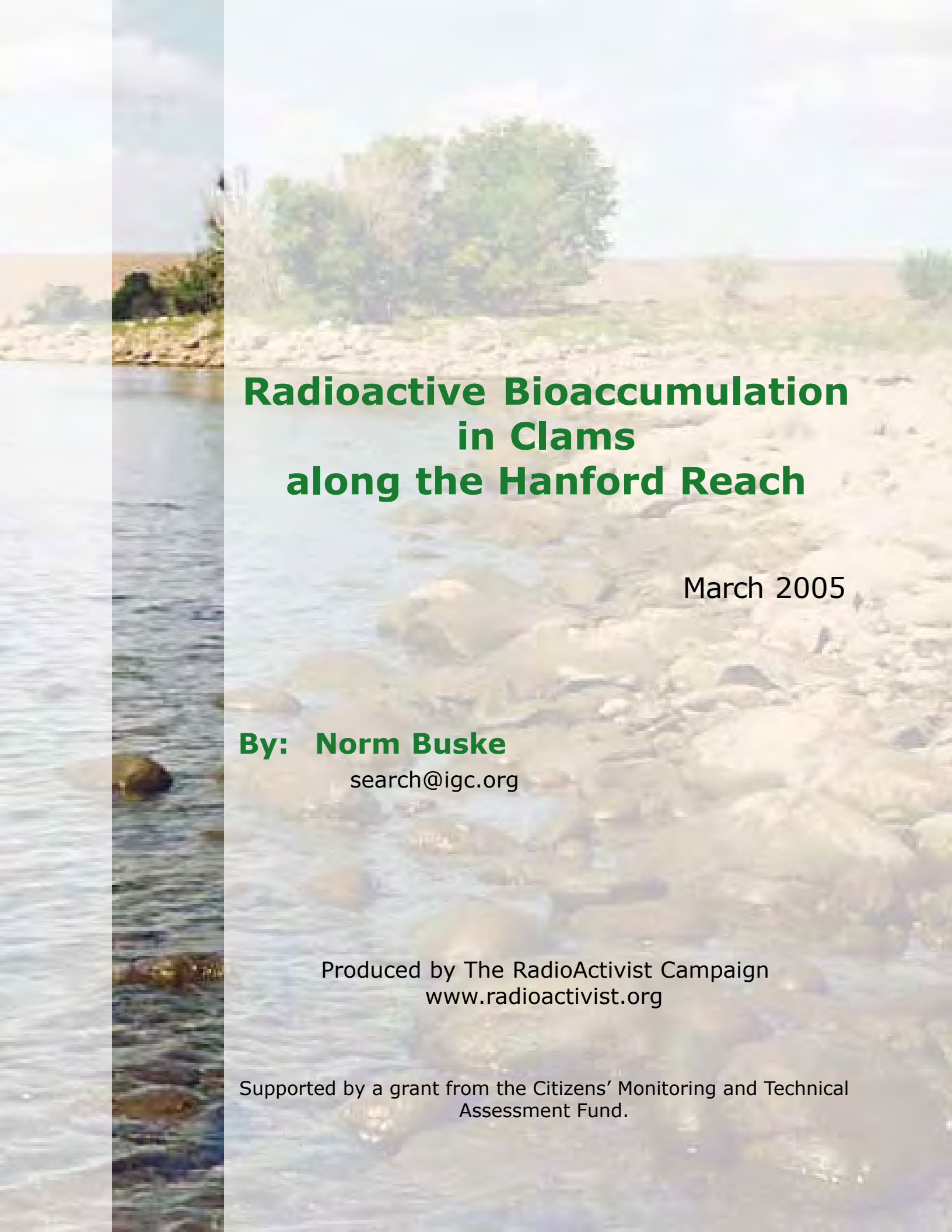

Radioactive Bioaccumulation in Clams along the Hanford Reach

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Produced by The RadioActivist Campaign
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Radioactive Bioaccumulation in Clams along the Hanford Reach

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TRAC is a scientific project of the Tides Center of San Francisco. TRAC measures radioactivity around nuclear facilities and reports the results and implications to the public. In 2003-04, TRAC measured radioactivity around four DOE sites: Hanford in eastern Washington, LANL in north central New Mexico, LLNL in California, and the Savannah River Site, in southern South Carolina.

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CONTENTS

SUMMARY

Public concern drives clean-up of the Hanford Site in eastern Washington state, especially where radioactive groundwater seeps from the site into the river. In September 2004, The RadioActivist Campaign (TRAC) collected riverbed water and Asian clams from six locations along the Hanford Reach:

- **Vernita** (upstream background location, by a shoreline spring)
- **N-Springs** (which drain old N-Reactor waste disposal trenches)
- **D-Island** (sampled upstream of the D- and DR-Reactor discharge pipes)
- **F** (sampled near a spring below F-Reactor)
- **Hanford Townsite** (sampled at a spring fed from central Hanford)
- **300 Area** (sampled near a spring at "Location 9")

Asian clams (*Corbicula sp.*) are a recognized indicator of Hanford's biological impact on the Columbia River ecology. TRAC collected about 200 clams from each location, and shucked these clams into a flesh fraction and a shell fraction. From each of the 6 locations, TRAC analyzed one clam flesh sample, one shell sample, and one riverbed water sample, for both short- and long-lived radioactivity.

Based on these analyses, the present study finds that the biological effects of strontium-90 seeping from N-Springs into the river have been under-reported in official monitoring. This finding raises concern for the adequacy of official Hanford Site monitoring.

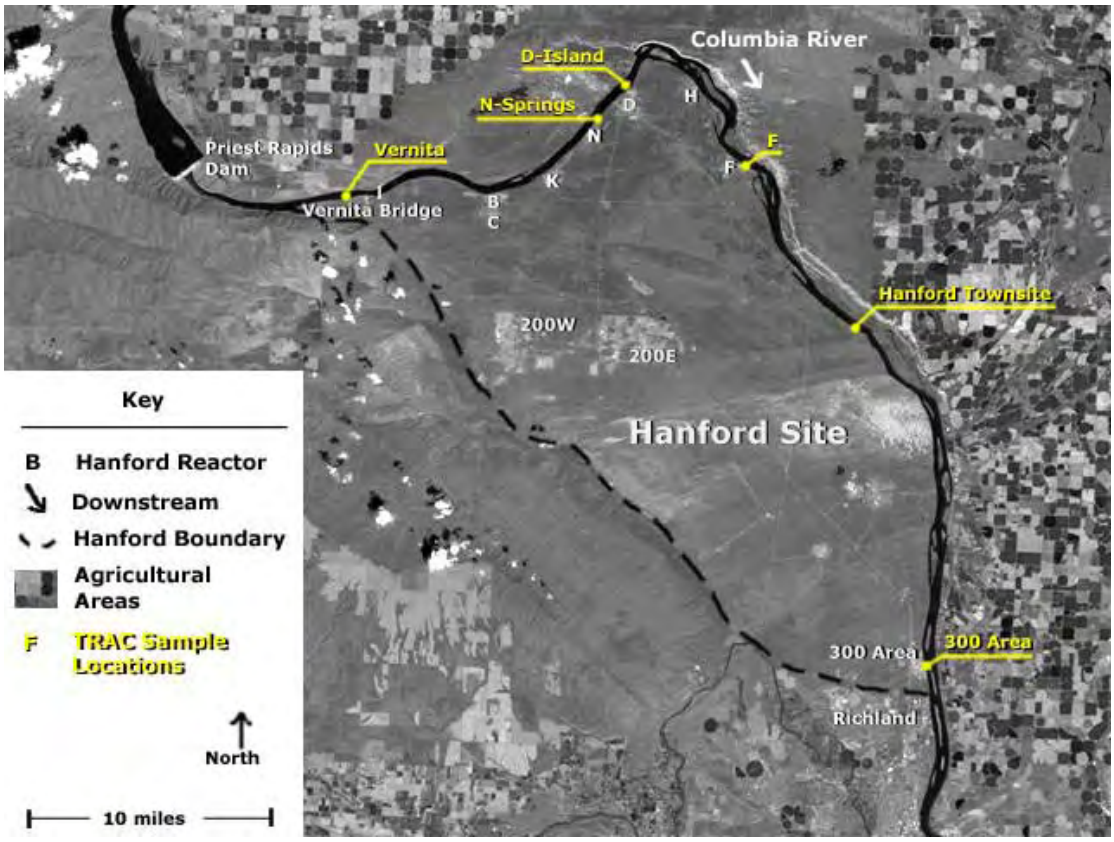
This study confirms official reports that uranium seeping from Hanford's 300 Area contaminates nearby aquatic biota.

This is the first report of radioactive radium isotopes (radium–226 and radium–228) from the 300 Area contaminating aquatic biota at Hanford. Although both radium isotopes are on the official Module-3 checklist of radioactive contaminants for aquatic biota, the monitoring agencies have failed to measure and report radium entering the river from the 300 Area. The impact of 300 Area on nearby river biota has thus been systematically under-reported, by omission.

Previously unmeasured and unreported radium contributes 90% as much alpha radioactivity to sensitive riparian and aquatic organisms near the 300 Area as does uranium, which has been officially measured and reported. Radium is probably the radioactive element of greatest concern entering the river from the 300 Area. Radium in clamshells near the 300 Area is more than four times background level.

On the positive side, TRAC is pleased ***not*** to report any radium, already seeping from the active waste disposal units in central Hanford Site, into springs discharging into the river at the old Hanford Townsite.

This study is an independent check of radiological reporting by the Hanford Site operator (the U.S. Department of Energy through its Pacific Northwest National Laboratory) and regulators (through the Washington state Department of Health).



Sample Locations, along the Hanford Reach of the Columbia River

INTRODUCTION and DRIVERS

Radioactive and toxic wastes make Hanford Site in Eastern Washington the most contaminated site in North America. Those radioactive and toxic wastes are byproducts of production of nuclear weapons materials during World War II and the Cold War.

Assessing and evaluating Hanford's wastes and remediating them by stabilization or clean-up is a billion-dollar-a-year investment of public resources. Underlying this investment is the doctrine of *rational objectivism*. According to this doctrine, what we are doing makes sense, both qualitatively and quantitatively. At the decision-making level, Hanford Site management relies on *quantitative risk assessments*.

After more than a decade of Hanford clean-up, real progress is now appearing on many efforts, and a world-class vitrification project is in-the-works. Yet there are concerns for the quality of expedited clean-up and for Hanford's growing role as site for new waste disposals.

In 2004, Hanford Action of Oregon, a public-interest group, contracted The RadioActivist Campaign (TRAC) to conduct an independent check of Hanford Site clean-up.

TRAC selected six locations along the 50-mile Hanford Reach of the Columbia River for sampling:

- Vernita, at the upstream end of the Hanford Reach, as a background location.
- N-Springs, near N-Reactor, where strontium-90 enters the river.
- D-Island, near where radioactive waste might have been dumped into the river.
- F shoreline, at a routine monitoring location.
- Old Hanford Townsite, as the location where groundwater seeps from massive, present and planned, waste disposals at central Hanford.
- 300 Area, near a spring routinely monitored for uranium contamination.

See the sample location map at the beginning of this section.

Traditional monitoring of Hanford's impact on the Columbia River environment has focused on fairly well known contaminants-of-Hanford-origin (Napier 1995, Table 9.1). The traditional checklist of radioactive Contaminants of Concern was based on radionuclides reported in Hanford groundwater within 150 meters (500 feet) of the Columbia River; on reported radionuclides in Columbia River water, sediment, and soil; and on "Continued Public Interest." Over the past 15 years, monitoring has focused more on biota sampled at shoreline springs along the Hanford Reach. Asian clams (*Corbicula sp.*) collected in and near those springs, have been used as an ecological indicator (Patton 2003).

The concept of this study was the measurement of *bioaccumulation*, both in Asian clam flesh and clam shells, in comparison to measurements of corresponding radioactivity in riverbed water at the same location and depth in the riverbed from which the sampled clams were collected. To realize this concept, TRAC modified the technology emplacing water *sampling tubes* in a riverbed (Appendix B). Unfortunately, TRAC did not account for the effect of a rising river level versus a falling river level, while the river was relatively low in September 2004. TRAC attributes negative radiological results in riverbed water samples to unfavorable conditions at the times of sampling, except at Vernita and D-Island.

METHODS:

Riverbed water collection. Ordinarily, samples of riverbed water are pumped from perforated plastic tubes driven into the riverbed with Geoprobe® drive-point probes. A riverbed water sample is thus pumped from a vertical section of perforated tubing.

Asian clams are observed in the riverbed of the Hanford Reach, from the bottoms of the cobblestones that line the fast-flowing river, into the coarse gravels, about 10 cm (4 inches) lower. TRAC reasoned that a perforated sampling tube, emplaced horizontally, would more precisely sample water in this clam habitat, than a tube driven vertically. Thus, TRAC designed, built, and employed a *spring-auger* system to install perforated sampling tubes horizontally, close to the top of the coarse gravel stratum in the riverbed.

TRAC's spring auger system is described in Appendix A. However, the key to success in riverbed water sampling seems to be to pump water samples only during low river levels, *when the river level is falling*.

Pumping water from the shallow riverbed into a 20-liter container. The perforated end of the sampling tube is in the riverbed, underwater. This sample is from D-Island. The river level has begun to drop, as can be seen from the still-wet rocks at the shoreline.

Sample preparation. TRAC pumped twenty-liter (5 gallon) samples of riverbed water from each sampling location. These water samples were coarse filtered (Whatman 2V Grade, 8 μm) to remove suspendable sediment that had been pumped with riverbed water. The water samples were then quiescently evaporated in a microwave oven. (Microwaves heat the bulk of the water. Microwaved water can be evaporated quickly

without boiling, and so without spray formation.) The water samples were evaporated to paste, that was then bulked to a standard, 52-gram analytical geometry.

TRAC collected about 200 clams from each of the six locations. The clams were then refrigerated or frozen to preserve them until they could be shucked. Visibly empty shells were discarded. Before shucking, the clams were rinsed with tap water to remove sediment grains. TRAC shucked the clams into "flesh" and "shell" sample fractions. Shells found empty during shucking were discarded.

Clams shucked into flesh, shell (and empty discards in cup) fractions.

Flesh and shell fractions were weighed ("wet") and then dried in a conventional oven to 95°C (<100°C). The dried samples were then weighed ("dry"). Flesh samples were bulked to the standard 52-gram analytical geometry. Shell samples were crushed and a fraction was taken to make the standard analytical geometry.

Analytical setting. TRAC believes that radiological analyses in the service of the public interest should not produce any radioactive or toxic wastes. TRAC accepts only *environmental samples* that screen below four times background radioactivity. TRAC derives all new radioactive reference materials from natural sources, without exceeding sample acceptance criteria.

TRAC processes environmental samples in a residential venue. As with national laboratories, such as the Los Alamos National Laboratory, the underlying concept is that lab processes and the lab have to be *cleaner than* their ordinary surroundings. That is, TRAC's lab is protected from ordinary background radioactivity in its location outside Belfair, Washington. (Belfair has half the background radioactivity of Hanford. The air

entering TRAC's lab is filtered to remove natural lead-212 (Pb212) in aerosols blown from nearby Hood Canal.)

Sample analysis. Each sample was analyzed in TRAC's highly stabilized, photon spectrometer for at least two different counts, each of 23-hours duration. The first count was about two weeks after sample collection; the second count was about a month after the first count.

TRAC analyzed each sample for gamma radioactivity in a 3 to 3,000 kilo-electron-volt (KeV) photon spectrometer (stabilized on the 1461 KeV gamma peak of potassium-40) based on a sodium-iodide crystal well-type detector, which was housed in a copper-lined, lead shield and held at 24.0°C. (See photo at left.). 8,200 channels acquired spectrum were then transformed to 165 energy channels, each of a *constant-photopeak-width* of 3 channels. (See examples of spectra with their photopeaks on pp. 8-10.) Because this system has a much better energy stability than the energy width of those photopeaks, *true* spectral subtractions are feasible. This reduces the usual problems of peak interferences.

TRAC's photon spectrometer system has certain advantages, along with disadvantages, compared to other radiological analytical systems. Advantages include enhanced sensitivity over a wide range of energies from 3 to 3000 KEV, practicality of true spectral subtractions sequentially, and intermediate and final results that are visual instead of merely numerical. Disadvantages include wide photopeaks and exacting requirements needed to overcome a multitude of nonlinear effects in the analog sodium-iodide detector and its photomultiplier.

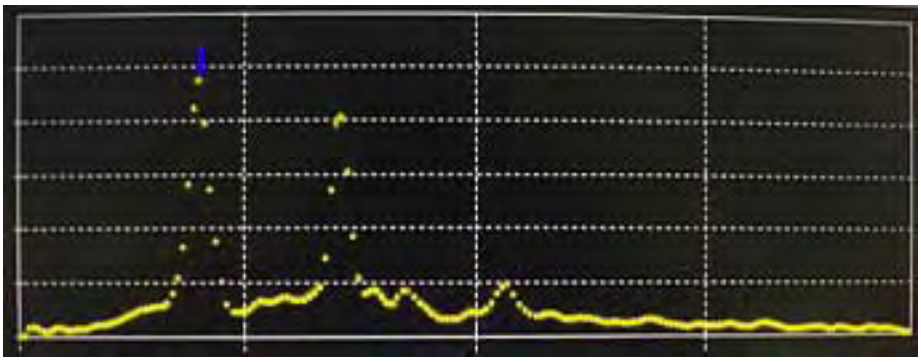
The present study seeks Hanford radioactivity, above background. This is nominally achieved spectrometrically by subtracting a photon spectrum obtained from a sample at Vernita, upstream of Hanford, from the spectrum of a comparable test sample collected anywhere along the Hanford Reach. The *difference* spectrum, that is the first step of the analysis, presents a visual indication of the photon energies in the test sample, *above background*. Then standard and reference spectra are sequentially subtracted according to the intensities of their peaks in the test spectrum that is being analyzed. The activity (above background) of a subtracted radionuclide in the sample is the fraction of

the reference spectrum that has been subtracted, multiplied by the activity of that reference material.

TRAC's spectrometric results are calibrated against (old) certified standards as well as both old and new reference materials, below:

certified standards	**reference materials**
natural uranium	natural radium (from the Oyler mine)
cobalt-60	natural uranium minus natural radium
strontium-90	natural thorium (from Coleman gas lantern mantles)
cesium-137	lead-212 (from aerosols)
europium-152	natural thorium minus natural lead-212 (above)
technitium-99	beryllium-7 (from rainfall)
	potassium-40 (in reagent potassium chloride)
	americium-241 (from smoke detectors)
	blank (or *background*)

The 5 most important reference spectra used in these full-spectral subtractions follow.



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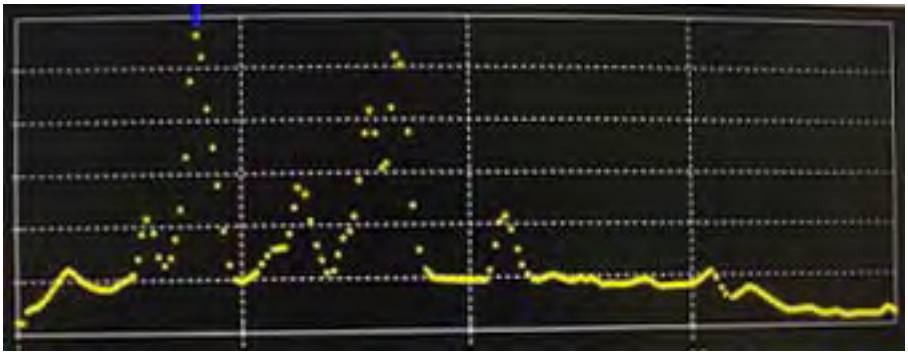
Intensity (by energy) spectrum of "natural lead-212-with-progeny".



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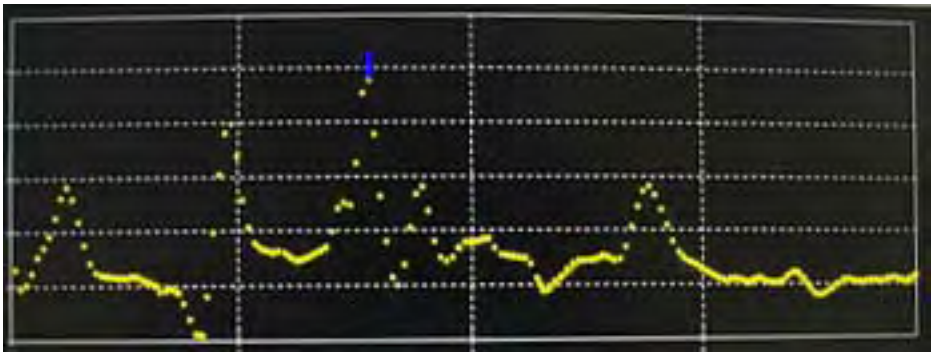

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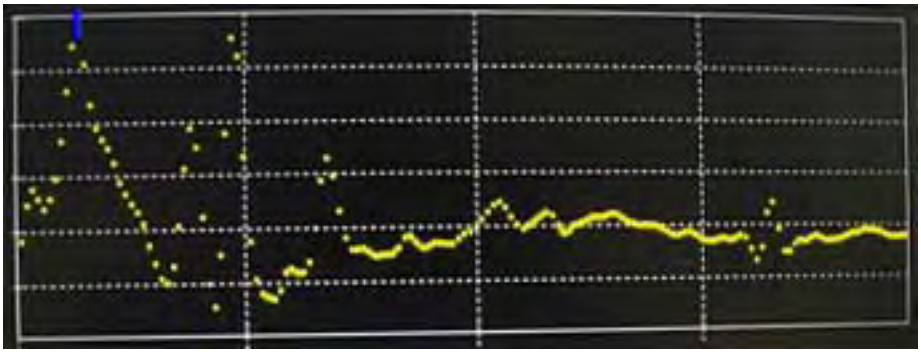
Intensity (by energy) spectrum of "natural-thorium minus natural-Pb212-with-progeny (NotPb)".



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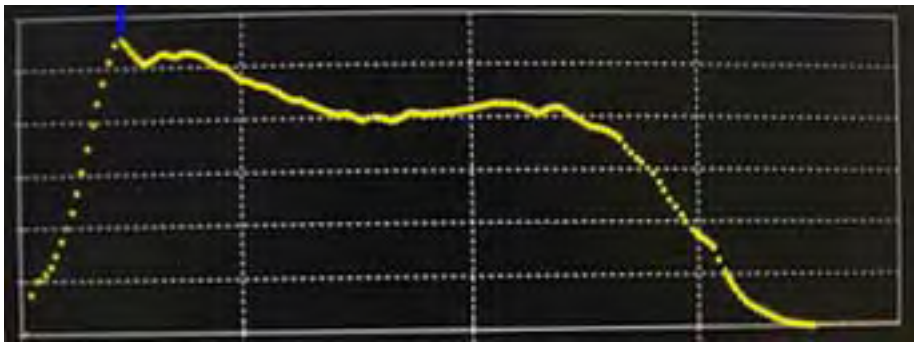
Intensity (by energy) spectrum of "natural-uranium minus natural-radium-with-progeny (NotRa)".



0 energy ——>

Intensity (by energy) spectrum of "strontium-90-with-progeny".

The intensity zero level in these spectra is an arbitrary vertical level on linear intensity scales. That arbitrary zero intensity is close to the spectral height at the high energy end (right side) of each spectrum.

Notice that some regions of the third and fourth displayed spectra, above, have less energy (are lower) than the arbitrary zero intensity level. These negative regions are attributed to secular *dis*equilibria in the decay chains of the reference materials. Equilibrium in decay chains that have a radon gas intermediary depends on the degree to which the radon gas is retained in the specimen or released to the atmosphere.

This question of equilibrium and negative intensity regions in subtracted spectra illustrate a practical limitation to the technique of spectral subtraction. A comparable limitation of such spectral subtractions is (nonrandom) negative reports, as in the top row in Table 1 and in Appendix B, Section 5 of this report. In practice, such limitations do not detract seriously from survey-type applications like this study.

After the analyst completes the spectral subtractions, any residual photopeaks peaks are noted. Those residuals are later checked against libraries of photopeaks, for identification.

TRAC obtained "background (BKG)" spectra from the water, clam flesh, and clam shell samples collected at Vernita.

To screen the five sets of water, clam flesh, and clamshell samples collected along the Hanford Reach, TRAC subtracted those Vernita background spectra from each sample spectrum of the same sample medium. TRAC then subtracted the potassium-40 (K40) activity in each sample spectrum (according to its gamma peak area), thus eliminating the K40 contribution to those spectra. Then, TRAC similarly subtracted natural thorium and natural uranium reference spectra, according to their respective photopeak activities in the spectrum being analyzed. TRAC compared the resulting sample spectra to references in TRAC's spectrum library. Residual peaks in the final spectra were then noted for later identification.

TRAC's full-spectrum, subtraction technique is most effective for analysis of short-lived radionuclides, with half-lives between a few days and several months. A test sample is counted soon after collection "a", and then recounted, a few weeks later "b". The background sample is likewise counted close to times "a" and "b". The first *difference spectrum*, with which the analysis for short-lived radioactivity begins, may be expressed mathematically as:

first *difference spectrum* = (sample spectrum "a" – sample spectrum "b")
— (Vernita background spectrum "a" – Vernita background spectrum "b")

This double subtraction technique identifies short-lived in-growth as effectively as it identifies short-lived, radioactive decay.

After a sample is prepared and sealed from air, radon released from the natural decay chains is trapped. The corresponding, natural thorium (Th232) and uranium (U235

and U238) decay chains (along with their photopeak spectra) then begin to grow in. The first difference spectrum (spectrum "a"– spectrum "b") thus contains *negative* photopeaks for samples which have *above background* radium-224 (thorium-232 decay chain), radium-223 (U235 decay chain), or radium-226 (U238 decay chain).

Results of screening against Vernita background are presented in Appendix B. After this screening, the sample spectra-of-interest were re-analyzed against a blank spectrum to provide absolute results (instead of results-above-background). Based on those results, TRAC re-counted several samples and performed other analyses to assure data quality adequate for the purposes of the present study.

TRAC archives the analyzed samples. These archived samples are available for checks by other laboratories. TRAC provides sample descriptions and locations to allow any interested party to check TRAC's results independently.

An analytical concept. The radioactive decay of any atomic nucleus releases one or more photons that account for the energy difference between the nucleus before decay and the final products of that decay. If the nuclear decay releases a particle such as an electron (beta particle), an anti-electron (positron), a helium nucleus (alpha particle), a proton, or a neutron; part of the kinetic energy of the released particle is emitted as photons of characteristic energies, as the particle comes to rest. That is to say, even a "pure" particle decay with no immediate gamma emissions, emits a characteristic photon spectrum resulting from energy accounting, secondary interactions, and other effects.

A sufficiently broad-band photon spectrometer can potentially detect almost any such radioactivity. However, the sensitivity of the analysis depends on, (1) the efficiency of the detector in comparison to the intensity of the photon energy spectrum of the decay, (2) on interferences in the regions-of-interest in the spectrum, and (3) on post-analysis of residual photopeaks in a spectrum. Some radioactive decays, such as hydrogen-3 (tritium) and carbon-14 have kinetic energies effectively below TRAC's cutoff at 3 KeV. Other analyses, such as strontium-90 require spectral form fitting of several decay effects.

A pivotal feature of TRAC's photon spectrometry is the acquisition of sample spectra of 3 - 3000 KeV into 8000 spectral *channels*. The raw spectra are then transformed to spectra of constant photopeak width (FWHM=3 channels), yielding transformed spectra of 165 channels over that energy range.

RESULTS

In September 2004, TRAC collected riverbed water and Asian clams from six locations, as follows. "HRM" is Hanford River Marker distance (in nominal miles) downstream of the Vernita Bridge. [GPS coordinates are on WGS 84 datum]:

Vernita (background, "BKG"). Sampled one kilometer (half mile) upstream of Vernita Rest Area, 50 meters (m = 3 feet) upstream of the slough on the south side of the river, in a broad spring in the cobble shore. [North 46.63460°, West 119.74915°]

N-Springs. Sampled at the downstream end of a stretch of large boulder rip-rap covering the main flow of N-Springs, located about HRM 9.1, close to "HEIS Location 100 N Spring-2". [North 46.68019°, West 119.56614°]

D-Island. Sampled at about HRM 10.7, on the Hanford (SE) side of the long spit, that is visible at low river levels, extending upstream from D-Island. [North 46.70145°, West 119.54172°]

F. Sampled in a small embayment on the west side of the river, below F-Reactor, at about HRM 19.1. [North 46.65935°, West 119.43833°]

Hanford Townsite. Sampled in the main seepage, in a depression just below the vegetation line on the west side of river at about HRM 28.1. [North 46.56529°, West 119.33688°]

300 Area. Sampled on the west side of the river, about 5 m (meters) upstream from the main "Location 9" sampling pump station, about 4 m downstream of "SESP Spring 42.2" at HRM 42.2. [North 46.372175°, West 119.27184°]

The four radionuclides reported here, general information about them (and their maximum permissible limits for drinking water in parentheses) are listed, below:

Reported radionuclides

- strontium-90 (Sr90), beta emitter, 29-year half-life.
  - a product of fission of uranium, bone seeker (<8 pCi/L*).
- radium-226, alpha emitter, 1,599 year half-life.
  - the fifth progeny of natural uranium-238 decay, bone seeker (<5 pCi/L**).
- radium-228, beta emitter, 5.8-year half-life.
  - the first progeny of natural thorium (Th232) decay, bone seeker (<5 pCi/L**).
- uranium (U), alpha emitter, >200,000 year half-lives.
  - a natural mixture of U234, U235, and U238; bone seeker (<22 pCi/L***).

* 1 pCi = one picocurie = one radioactive disintegration in half a minute. To convert from pCi to Bq (becquerels), multiply by 0.037. "L" = liter.

** The maximum permissible drinking water limit is Ra226+Ra228 < 5 pCi/L.

*** To convert radiological units (pCi/L) to chemical units (µg/L), **for uranium**, divide by (0.70 pCi/µg). The maximum drinking water limit is 30 µg/L.

Positive screening results that did not pass quality checks are not reported here. See Appendix B for the screening results and other sample information.

Background (Vernita) results and sample results that screened above-background appear in the Analytical Results Table 1, below:

Asian clams (*Corbicula sp.*)

Table 1. ANALYTICAL RESULTS [pCi/g(dry)].

Flesh	**Strontium-90**	**Radium-226**	**Radium-228**	**Uranium**	**(wet/dry*)**
Vernita (BKG)	--	*0.48*	*–0.08*	*0.24*	*(7.5)*
N-Springs	16.	--	--	--	(9.7)
D-Island	--	--	--	--	(7.5)
F	--	--	--	--	(7.1)
Hanford Townsite	--	--	--	--	(7.0)
300Area	--	--	0.37	0.74	(9.9)
Shell					
Vernita (BKG)	--	*0.14*	*0.06*	*0.036*	*(1.02)*
N-Springs	290.	--	--	--	(1.09)
D-Island	0.5	--	--	--	(1.10)
F	--	--	--	--	(1.08)
Hanford Townsite	--	--	--	--	(1.09)
300 Area	--	0.56	0.38	0.86	(1.10)

* "(wet/dry)" is the dimensionless ratio of wet weight to dry weight of the sample. To convert dry weight results to wet weight, divide the sample result by the wet/dry value.

DISCUSSION

Results of the present study are readily inter-comparable with results presented by Poston (2004) . Routine monitoring of the Hanford Reach shoreline is reported for most of the locations of the present study. The Hanford Site Public Safety and Resource Protection Program (SESP) and the Washington State Department of Health also sample at some of these locations.

This screening study was designed, executed, and reported to provide a summary check on the reliability of official monitoring programs and to detect and highlight some weaknesses. This study is far too limited to address most aspects of monitoring the Hanford Reach. For one example, the samples and analyses in this study are insensitive to uranium-233, reportedly in the riverbed (Buske 2003). Analysis in the present study of riverbed water from D-Island was expected to reveal radium-225, but did not.

The results of this study are indicative rather than comprehensive.

This study reveals three problems in official monitoring of radioactivity in Hanford Reach biota: (1) sampling and reporting strontium-90 at N-Springs, (2) identifying, analyzing, and reporting radium isotopes at 300 Area, and (3) estimating alpha doses and setting alpha dose limits for sensitive aquatic and riparian biota. These problems are discussed, below:

Problem (1) Strontium-90 at N-Springs.

Groundwater contaminated with strontium-90 (Sr90) from old liquid waste disposal trenches near N-Reactor seeps into the Columbia River at N-Springs. Terrestrial and aquatic organisms living at N-Springs bioaccumulate Sr90, which mimics calcium, an essential nutrient. In 2003, the average activity of Sr90 entering the river from a contaminated spring [Well 199-N-46; see Table 3.2.4 (Poston 2004)] was 4,100 pCi/L. That was 500 times the maximum allowable contamination in drinking water.

In September 1997, the Hanford Site Surface Environmental Surveillance Project (SESP) and the Washington State Department of Health (WDOH) sampled terrestrial and aquatic biota at N-Springs, and analyzed the samples for Sr90 (Van Verst 1998).

Van Verst (1998, Table 6.1) estimated a maximum dose to a muskrat living at N-Springs of 0.9 rad/day. That dose to a muskrat would exceed the present, interim requirement (DOE Order 5400.5) for control of liquid waste discharges from Hanford: 0.1 rad/day for terrestrial biota. Van Verst reported one sample of caddisflies that measured Sr90 = 65 pCi/g(dry). That result was described as “unexpected”; “additional caddisfly samples may be required to confirm that unusual finding (Van Verst 1998, p. 7.1).” The authors did not publish a dose estimate for the caddisflies.

Van Verst did not report any Asian clams at N-Springs proper (their Site 3), and so reported Sr90 only in clams collected from the upstream (their Site 1) and downstream (their Site 5) ends of N-Springs. At their Site 1, Van Verst reported Sr90 = 1.5 pCi/g(dry)

in clam shells sampled in 1997. That was a tiny fraction of Sr90 = 270 pCi/g(dry) that had been reported in 1991 (p. 4.12).

The 1991report for Asian clam shells from N-Springs is almost the same as the result of Sr-90 = 290 pCi/g(dry), in Table 1 of the present study.

For dose calculations, clam shells are a "tissue" (Mod-3 2002, Sec. 2.2.1). A tissue is a uniform aggregate of similar material forming one of the structures of an organism. For dose calculations, the radionuclides of interest are assumed to be uniformly distributed throughout the tissue, and the tissue is large enough to absorb the energy of the radioactive decay. With serious reservations discussed in Problem (3), below, beta doses to tissues can be estimated according to Mod-3 (2002). The internal dose factor for Sr90 is assumed to be 0.000058 rad/day per pCi/g(wet) (Mod-3 2002, Table 2.4). The Sr90 dose to TRAC's sample of clam shells from N-Springs is then estimated, with serious reservations, as:

0.000058 {(rad/day)/[pCi/g(wet)]} x 290 pCi/g(dry) / 1.09 (wet/dry) = **0.015 rad/day**.

That is 5 times Van Verst's (1998) Sr90 dose estimate to clams at N-Springs: **0.0029 rad/day**.

The concern here is how easily the seemingly conservative factors used by Van Verst (1998) are lost by a single follow-up measurement.

These considerations suggest there are probably some populations of some yet unidentified biota that are adversely affected by the seepage of Sr90 from N-Springs.

(2) Radium at 300 Area.

TRAC collected about 150 live clams from Vernita and 296 live clams from 300 Area (by Location 9), generally in 0.5 to 1.0 m water depth. TRAC's results accord with the results of Patton (2003, Table C.5), for water depths 0.5 to 1.0 m.

Patton (2003) reported uranium in clam flesh and clam shells in different water depths and in springs, both at Vernita (background) and at 300 Area Location 9. At 300 Area Location 9, Patton reported much higher uranium contents in shallow water close to the spring at Location 9. Their averaged results for Location 9 are thus weighted toward higher uranium contents than TRAC's. Patton's (2003, Table 4.7) summary results compare to TRAC's uranium measurements, at the top of the next page.

Table 2 shows that, at 300 Area Location 9, uranium is much more concentrated in clam shells than in clam flesh.

Table 2. Comparison of Uranium in Clams. [pCi/g(dry)].

Flesh	TRAC	Patton (2003, Table 4.7)*	
		median	maximum
Vernita (background)	0.24	0.14	0.29
300Area (Location 9)	0.74	1.97	4.67
Shell			
Vernita (background)	0.036	0.048	0.076
300 Area (Location 9)	0.86	2.92	7.73

*Converted from chemical units [µg/g] to radiological units [pCi/g] by multiplying by 0.69pCi/µg for natural uranium.

In Table 3, below, the values in Table 2 for uranium in shells from 300 Area Location 9 are divided by the values for shells from Vernita. These ratios are the uranium accumulation factors for 300 Area Location 9 in comparison to background.

Table 3.

Uranium Concentration Factors in Clam Shells	TRAC	Patton (2003) median	Patton (2003) maximum
300 Area Location 9 / Vernita (BKG):	**24.**	**61.**	**102.**

Uranium in clam shells at 300 Area Location 9 is between 24 and 102 times background, depending on the exact location of the clams next to the spring at Location 9.

These huge multiples of uranium activity in clam shells near 300 Area Location 9 can be compared to the U.S. Department of Energy's (DOE's) level of tolerance for environmental, radioactive samples removed from Hanford Site for scientific study. DOE has allowed environmental samples screening to pass no more than **four times background** radioactivity to be removed from the site, for the purpose of public-interest analysis (DOE 2002). The factors of uranium-above background in Table 3 range from (24/4 =) 6 to (102/4 =) 25. By DOE's own rationale,

Uranium contamination of clam shells at 300 Area Location 9 is intolerable by a factor of 6 to 25.

Radium (Ra226+Ra228) contributes about 90% as much radioactivity-above-background in TRAC's clam shells from 300 Area Location 9 as uranium contributes:

Above background in clamshell at 300 Area Location 9 [pCi/g(dry)], from Table 1:

radium [(0.56–0.14) + (0.380–0.06)] = 0.74
uranium (0.86–0.036) = 0.82

Furthermore, if the alpha dose-modifying factors for radium and uranium are assumed to be the same, then one unit of radium activity produces about 35% more alpha dose than does one unit of uranium activity (Mod-3 2002, pp. 17 - 18). That is, the above-background dose in clam shells from Location 9 is about (0.90 x 1.35 = 1.22) 22% greater than the above-background dose contribution of uranium. This consideration suggests that if clamshell is treated as an indicator medium for sensitive biota near 300 Area,

Radium is probably the radioactive contaminant of greatest concern entering the river from 300 Area.

Neither Ra226 nor Ra228 is difficult to detect in "gamma scans" such as TRAC performs. Ra226 is evidenced by intensive photopeaks of progeny lead-210, lead-214, and bismuth-214. Ra228 is evident in the intensive photopeaks of decay product actinium-228.

Since 1991, concern for radium isotopes in drinking water has been a regulatory concern for the Environmental Protection Agency, in formulating National Primary Drinking Water Standards (EPA 2000). As already mentioned, the Department of Energy, in its annual environmental monitoring report for Hanford Site takes notice of the Primary Drinking Water limit of Ra226+Ra228 < 5 pCi/L.

Both Ra226 and Ra228 are listed among the 23 radionuclides to be screened through Mod-3 (2002) for impacts on aquatic biota in the Columbia River. Public assurances of low dose to aquatic biota in the Columbia River rely on all substantially contributing radionuclides to have been screened as not-of-concern. Failure of official monitors to measure and report radium at the springs at 300 Area casts the whole system of quantitative risk management at Hanford into doubt.

(3) Assumptions for Estimating Doses.

In its "Graded Approach For Evaluating Radiation Doses to Aquatic and Terrestrial Biota (Mod-1 2002)," DOE attempts to accommodate the differing biological sensitivities, in differing taxonomic groups, to radiation. DOE notes that many taxonomic groups of aquatic organisms are more resistant to radiation than are many terrestrial groups.

DOE somewhat arbitrarily sets a dose limit of 1.0 rad/day for aquatic animals and 0.1 rad/day for terrestrial animals. This introduces a conceptual problem, because dose limits are set on location instead of on taxonomic group. The conceptual problem becomes serious at the shoreline of the Columbia River. The riparian zone is the visually *green band* on the shore, between high water levels and low water levels.

DOE defines "Riparian Organisms are those organisms related to, living, or located on the bank of a natural watercourse (as a river) or sometimes of a lake or a tidewater." DOE sorts riparian animals into the more protected class of terrestrial animals in its Biotic Dose Calculator (Mod-3 2002, Sec. 3.2).

DOE's graded approach is nominally somewhat conservative in lumping riparian animals into the more protected terrestrial group. But "the user is allowed to modify the lumped parameter [technical term omitted here] to a more site-representative value (Mod-3 2002, p. M3-29)." In practice, official monitoring programs have taken this flexibility in DOE's Biota Dose Calculator to calculate doses for many Riparian Organisms as if they were aquatic organisms. Through that contrivance, no doses to organisms living in the riparian zone exceed the aquatic limit of 1.0 rad/day. See Patton (2003, Sec. 6 and App. E). Poston (2004, Table 5.0.5) lumped the RESRAD-BIOTA screening of the entire Hanford riverbank into the less protected aquatic class. That treatment of the riparian riverbank allowed a screening value of 0.79 to pass. If the riparian zone at 300 Area were protected as DOE claims, then that screening value would have exceeded the limit of 0.1 and failed the test.

DOE designed Mod-3 (2002, p. M3-1) to (quote:) "protect 'all biota' everywhere". In order to assure that dose limits had not been arbitrarily set too high, so some biota might be inadequately protected, DOE's "**graded approach for evaluating compliance had to allow site users to examine and revise, if appropriate, the screening limits to more realistically reflect the conditions at their site.**" In practice, that flexibility has allowed Hanford Site "users" to set screening levels high enough to pass.

Mod-3 dose estimates require the size of the dosed "tissue" to be much greater than the penetrating depth of the radiation responsible for that dose. For alpha radiation, the size of the maximally affected "tissue" is small. This concern has been quantified for alpha-emitting radon gas and its alpha-emitting decay products in human lung tissues (Eisenbud 1997, p. 28):

> **For radon and its [alpha-emitting] decay products, the depth of penetration in tissue is only a few tens of micrometers. ... The dose calculated in this way is orders of magnitude higher than when it is assumed the energy is absorbed by the whole [tissue].**

In other words, the size of the appropriate *region of concern* for alpha dose calculations is ***less than*** tens of micrometers (μm). The diameter of a human hair is about 50 μm. Thus, the maximally exposed "tissue" for calculating alpha doses to sensitive organisms is likely much smaller than the diameter of a hair!

If cellular structures of microscopic size are *tissues,* for the purpose of alpha dose calculations: **Exactly what micro-structure in precisely which organism near 300 Area is the most sensitive, in terms of the legal requirement for Columbia River water quality?**

DOE addresses this question by introducing a "dose-modifying factor" Q with a default value of Q=20 (Mod-3 2002, p. M3-17) into the Biota Dose Calculator. This "provides the capability" for the user to modify (= change) the value of Q. Thus, DOE's graded approach for evaluating alpha radiation doses to aquatic and terrestrial biota invites the site user to calculate almost any alpha radiation dose by "appropriately" altering the dose-modifying factor. Although the environmental data base for dose calculations is objective, the alpha dose calculations, themselves, are subjectively adjustable rather than objective. Therefore, the calculated doses are subjective rather than objective.

This subjectivity arises from the requirements the Biota Dose Assessment Committee (BDAC) imposed on the general methodology for DOE's graded approach for dose evaluations. The method had to be (Mod-3 2002, Sec 1.1):

based on existing data
simple
defensible
user-friendly
useful for evaluating combined doses from water and sediment or soil
broadly applicable to both aquatic and terrestrial species
logical and consistent as a departure point for in-depth analyses and evaluations

The BDAC did not require the method to yield objective results. Thus,

Official calculations of alpha doses are subjective rather than objective.

DeBruler (2003) has raised other, broader concerns with official calculations of doses to Columbia River biota at the 300 Area. Dunning (2005) has expressed concerns that there has been "no real science" in establishing the BDAC levels, and they are not truly protective of populations. He recommends that the BDAC threshold values be adjusted downward by a factor of at least 100.

Consequently, DOE's alpha dose estimates for Hanford Site, calculated according to Module 3, lack technical credibility.

In order to protect the most sensitive biota dependent on Columbia River waters, the dose modifying factor for alpha radiation would have to be adjusted "orders of magnitude" in the direction that would fail Hanford Site in terms of legal compliance with Columbia River water quality regulations.

CONCLUSIONS and RECOMMENDATIONS:

1. **Radiological monitoring and dose calculations of the Hanford Reach are not protective of aquatic biota. At N-Springs, maximally dosed biota have probably not been sampled adequately, and a high measurement has been discounted as "unexpected". Near the 300 Area Location 9 spring, the highest values of uranium contamination have been unreported through the artifices of not converting chemical measurements to their radiological equivalents and then estimating their importance.**

2. **Radiological monitoring of aquatic biota near 300 Area has failed to follow the "Candidate Sets" checklist approach for evaluating radiation doses to aquatic and terrestrial biota at Hanford. This procedural failure has led to an overall failure to identify, measure, and report radium-226 and radium-228 in aquatic and riparian biota affected by the contaminated springs along the 300 Area shoreline of the Columbia River.**

3. **Previously unmeasured and unreported radium contributes about 90% as alpha radioactivity to clam shells near 300 Area Location 9 as does uranium, which has been officially measured and reported.**

4. **Releases of radium and uranium from the shoreline springs at Hanford's 300 Area are indicated by clamshell data to be substantially unprotective of the most sensitive, nearby aquatic and riparian biota.**

5. **Official dose limits on and calculations for internal alpha radioactivity in aquatic and riparian biota are not protective. A crucial problem is that official users of the Biota Dose Calculator can and do adjust internal factors, obtaining dose calculations that pass screening requirements.**

6. **Unprotective biases of Hanford Site monitoring should be corrected, in order to manage the site well and to obtain an end state of clean-up that serves the public interest.**

Appendix A. SAMPLING TUBE EMPLACEMENT

TRAC sought to emplace sampling tubes for collection of riverbed water at the general depth of the habitat for Asian clams, under the Hanford Reach. The goal was to co-sample riverbed water and clams, to measure directly bioaccumulation of radionuclides.

Six riverbed water sampling tubes were installed laterally with a spring-auger in September 2004. The installations were difficult because the spring was too flexible and the crank mechanism not rugged enough. Not withstanding these deficiencies, the installations were completed, and the concept proved attractive.

Spring auger (left) after completion of sampling tube installation (right)

TRAC pumped 20-liter water samples during relatively low river stages during September 2004. Four of these 6 samples were collected with the river rising. Shoreline springs were not flowing at the times of any of these water collections. Analytical results were negative, suggesting that intruding river water had been sampled instead of water representative of the riverbed. In the future, shallow riverbed water samples should be collected at low river level, when the shoreline springs are flowing.

Parts List for Spring-Auger for Riverbed Sampling Tubes

1. Cobra™ No. 80200 drain cleaning tool with self storing poly canister.
   * Bore shaft to 1/2 inch to receive larger 12.7mm spring auger.
   * Remove thumbscrew holder. Saw two slots 3/4 inch into end of auger shaft. Replace thumbscrew holder with 1/2-inch hose clamp to secure spring.

2. Cobra™ No. 44030 drain cleaning tool.
   * Cut most of expanded end off steel spring. Pull spring from handle.
   * Cut 12.7mm, 1.1 meter long, steel spring auger from handle.
   * Bend auger end to 30 degree angle with clamping pliers. Cut end to form cutter.

3. PTFE thread seal tape, 1/2". Wrap clockwise around auger spring to reduce sediment intrusion when auger bends around rocks. Rewrap for each installation.

4. Stainless steel hex-head metal screw 10 x 3/4.

5. Polyethylene tubing: 1/4" O.D., 0.040" wall. ~100 (1.5mm) perforations for 30 cm, behind screw. Load 12 meters into canister.

6. Polyethylene tubing: 5/8" I.D, 3/4" O.D. x 1.2m, with slot over thumb screw, as sheath for auger.

7. Pump: Guzzler® Model 07090-10, diaphragm hand pump, 7 strokes/gallon, lift 12 ft. Wetted parts: Delrin® plastic and Buna N diaphragm. Bore inlet to epoxy in 3/8"x1/4" Lasco (Santa Fe Springs, CA 90670) brass bell reducer (17-9275) with teflon taped-in 1/4"x3/16" Lasco male hose barb (17-7711).

Appendix B. SAMPLE SCREENING

Samples were screened for radioactivity "above background", meaning greater radioactivity than in "background" material of the same medium, collected from Vernita. The Screening procedure is described in the Results section.

1. Sample Key

"BKG"	=	sample of same medium, collected from Vernita
"Water"	=	riverbed water sample
"Clams"	=	number of live Asian clams sampled, rinsed before shucking
"Flesh"	=	Asian clam sample, shucked fraction
"Shell"	=	Asian clam sample, shell fraction
"g(wet)	=	wet weight of sample. A small fraction of shell samples was used.
"w/d"	=	wet weight/dry weight, dried to 95°C.
"s"	=	sample collection date: year.month.day
"a"	=	first analysis counting date: year.month.day
"b"	=	second analysis counting date: year.month.day
"c"	=	third analysis counting date: year.month.day
"#"	=	sample designation number

2. Sample Collection and Analysis Information, By Location

Vernita (background, "BKG").

Location: One km upstream of Vernita Rest Area, 50 m upstream of slough on south side of river, in broad spring in cobble shore.
[North 46.63460°, West 119.74915°]

Water: #491015. 21,170 g(wet).
s=4.09.10. a=4.10.17. b=4.10.15. c=5.01.09.

Clams: Not counted, about 150.

Flesh: #490413f. 202 g(wet). 7.5 w/d.
s=4.09.04. a=4.09.09. a'=4.09.18. b=4.10.20.

Shell: #490413s. 34.1 g(wet). 1.02 w/d.
s=4.09.04. a=4.09.08. b=4.10.06.

Sample Collection and Analysis Information, By Location (continued)

N-Springs.

Location: At downstream end of a stretch of large boulder rip-rap covering the main flow of N-Springs. About HRM 9.1. Close to "HEIS Location 100 N Spring-2". [North 46.68019°, West 119.56614°]

Water: #492407. 20,220 g(wet).
s=4.09.24. a=4.10.17. b=4.11.27.

Clams: 110 (limited by availability and sampling schedule).

Flesh: #4911.10f. 186 g(wet). 9.7 w/d.
s=4.09.11. a=4.09.30. b=4.10.25. c=5.01.08.

Shell: #491110s. 32.9 g(wet). 1.09 w/d.
s=4.09.11. a=4.10.01. b=4.10.26.

D-Island.

Location: On the Hanford (SE) side of the long spit, visible at low river stage, extending upstream from D-Island. About HRM 10.7.
[North 46.70145°, West 119.54172°]

Water: #492508. 20,510 g(wet).

Clams: 202.

Flesh: #491112f. 247 g(wet). 7.5 w/d.
s=4.09.11. a=4.09.26. b=4.10.21, c=5.01.04.

Shell: #491112s. 36.9 g(wet). 1.10 w/d.
s=4.09.11. a=4.09.27. b=4.10.22. c=5.01.01.

F.

Location: In a small embayment on west side of river, below F-Reactor, about HRM 19.1. [North 46.65935°, West 119.43833°]

Water: #492409. 20,880 g(wet)
s=4.09.24. a=4.10.02. b=4.11.12. c=4.12.21.

Clams: 225.

Flesh: #491115 f. 238 g(wet). 7.1 w/d.
s=4.09.11. a=4.09.20. b=4.10.16. c=4.12.20.

Shell: #491115s. 35.9 g(wet) 1.08 w/d.
s=4.09.11. a=4.09.16. b=4.10.14. c=4.12.22.

Hanford Townsite.

Location: In main seepage, in depression just below vegetation line on west side of river. HRM 28.1. [North 46.56529°, West 119.33688°]

Water: #492411. 20,140 g(wet).
s=4.09.24. a=4.10.03. b=4.11.20. c=4.12.19.

Clams: 209.

Flesh: #491210f. 232 g(wet). 7.0 w/d.
s=4.09.12. a=4.09.21. b=4.10.19. c=5.01.10.

Shell: #491210s. 32.8 g(wet). 1.09 w/d.
s=4.09.12. a=4.09.25. b=4.10.21. c=5.01.05.

Sample Collection and Analysis Information, By Location (completed)

300 Area.

Location: On the west side of the river, about 5 m upstream from the main "Location 9" sampling pump station, about 4 m downstream of "SESP Spring 42.2" at HRM 42.2. [North 46.372175°, West 119.27184°]

Water: #492413. 19,950 g(wet).
s=4.09.24. a=4.10.07. b=4.11.26. c=5.01.02.

Clams: 296.

Flesh: #491213f. 318 g(wet). 9.9 w/d.
s=4.09.12. a=4.09.28. b=4.10.23. c=5.01.07.

Shell: #491213s. 37.2 g(wet). 1.10 w/d.
s=4.09.12. a=4.09.29. b=4.10.24. c=5.01.03.

4. Screening Key

"Short"	=	sample (spectrum "a" – spectrum "b") minus BKG (spectrum "a" – spectrum "b"); except for Vernita, where: "Short" = BKG (spectrum "a" – spectrum "b").
"Long"	=	sample (spectrum "b" or "b"+"c") minus BKG (spectrum "b"); except for Vernita, where: "Long" = BKG (spectrum "a' "+"b", "b", or "b"+"c")
"Pb"	=	lead-212 (with progeny); in Results as Ra224
"NotPb"	=	natural thorium (with progeny) minus "Pb"; in Results as Ra228
"Ra"	=	natural radium (with progeny); in Results as Ra226
"NotRa"	=	natural uranium (with progeny) minus "Ra"; in Results as U238
"~"	=	spectrum subtraction inconsistent
"--"	=	| Not detected |

Units: pCi/kg(wet), either absolute or > BKG (Vernita), as noted

Note: Results in Table 1 of the Results of this study are based on absolute analysis, not these screening analyses above-BKD.

5. Screening Results

Water [pCi/kg(wet)]

Short	Pb (Ra224)	Ra (Ra226)	NotPb (Ra228)	NotRa (U238)	Other
Vernita BKG [Absolute]	0.11	--	–0.048	--	--
N-Springs [>BKG]	--	--	--	--	--
D-Island [>BKG]	--	--	--	--	--
F [>BKG]	--	--	--	--	--
Hanford Townsite [>BKG]	--	--	--	--	--
300 Area [>BKG]	--	--	--	--	--
Long					
Vernita BKG [Absolute]	–0.095	–0.34	0.095	0.098	0.038 Cs137
N-Springs [>BKG]	0.10	0.22	--	0.22	--
D-Island [>BKG]	0.072	0.46	--	0.082	--
F [>BKG]	0.096	0.022	--	0.030	--
Hanford Townsite [>BKG]	0.18	0.26	--	0.072	--
300 Area [>BKG]	0.11	--	--	0.22	--

Flesh [pCi/kg(wet)]

Short	Pb (Ra224)	Ra (Ra226)	NotPb (Ra228)	NotRa (U238)	Other
Vernita BKG [Absolute]	13.	–22.	--	--	--
N-Springs [>BKG]	–18	–20.	--	--	--
D-Island [>BKG]	–20.	--	--	--	--
F [>BKG]	–13	--	--	--	--
Hanford Townsite [>BKG]	–13	40.	--	--	--
300 Area [>BKG]	–13	48.	–49.	--	--
Long					
Vernita BKG [Absolute]	47.	64.	–10.	16.	--
N-Springs [>BKG]	--	--	--	--	1600. Sr90
D-Island [>BKG]	--	--	--	--	12. U233
F [>BKG]	--	--	25.	--	--
Hanford Townsite [>BKG]	--	--	--	--	--
300 Area [>BKG]	–16.	–26.	38.	20.	3. Cs137

Screening Results (completed)

Shell [pCi/kg(wet)]

Short	Pb (Ra224)	Ra (Ra226)	NotPb (Ra228)	NotRa (U238)	Other
Vernita BKG [Absolute]	74.	–326.	--	70.	--
N-Springs [>BKG]	--	--	--	--	--
D-Island [>BKG]	--	150.	--	--	--
F [>BKG]	–13	--	--	--	--
Hanford Townsite [>BKG]	--	530.	--	--	--
300 Area [>BKG]	--	--	--	--	--
Long					
Vernita BKG [Absolute]	46.	142.	55.	18.	--
N-Springs [>BKG]	--	--	--	92.	268,000. Sr90
D-Island [>BKG]	80.	30.	--	--	407. Sr90, 102. Cs137
F [>BKG]	--	--	--	--	--
Hanford Townsite [>BKG]	--	20.	--	--	--
300 Area [>BKG]	40.	~100.	209.	396.	6. Cs137

Note 1: Some positive results of this screening test did not pass follow-up quality control checks and quality assurance, and so were not reported in Table 1 of Results.

Note 2: Analyses for absolute values in Table 1 are separate from these screening values.

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