Data Report

of a Radiological Study Around the Oak Ridge Reservation, Tennessee in November 2004

06 May 2005. REV.2

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Background

The Oak Ridge Reservation (ORR) is a complex of nuclear weapons-related facilities owned by the U.S. Department of Energy (DOE) and operated by contractors. The main clusters of facilities are the Y-12 National Security Complex (Y-12), the Oak Ridge National Laboratory (ORNL), and the East Tennessee Technology Park (ETTP). ORR was established in the early 1940s at the beginning of the Manhattan Project that produced the world's first three nuclear explosions: at Trinity Site in New Mexico, and at Hiroshima and Nagasaki in Japan. ORR continues to produce nuclear materials and conduct research and development for nuclear weapons. Wartime and subsequent work at ORR involves radiological and hazardous materials. See www.ornl.gov for reports and supporting data. The DOE Oversight Division of the Tennessee Department of Environment and Conservation (TDEC) and other government agencies monitor and regulate activities at ORR. ORR is almost entirely within the City Limits of Oak Ridge, Tennessee.

TRAC, a scientific project of the Tides Center of San Francisco, responded to citizen concerns by conducting this independent sampling around ORR in November 2004 and independent analysis of those samples. This Data Report presents the analytical results of that study, radionuclide by radionuclide. These radiological results have been updated, since REV.0 of 08 March 2005. These results are summarized from the perspective of radioactive contaminant pathways, along with conclusions and recommendations, in TRAC's final report, "Radioactive Pathways --Oak Ridge, Tennessee," May 2005.

This study is supported by a grant from the Citizens' Monitoring and Technical Assessment Fund. See www.mtafund.org.

Narrative

TRAC staff reviewed official reports by the Oak Ridge Operations of the DOE and by TDEC to prepare for this independent radiological investigation around ORR. The Chamber of Commerce publication, "Oak Ridge, Tennessee, A Citizen's Guide to the Environment (www.orcc.org)," introduced other citizen concerns.

TRAC listened to community concerns at a public meeting at the Oak Ridge Civic Center, on 01 November 2004. Those expressions of concern helped TRAC define questions to be addressed by this sampling around ORR and the radiological analysis of the samples.

TRAC began collecting samples from the environment around ORR on 02 November 2004. TRAC updated ORR Operations daily, on the sampling progress and on arrangements to avoid problems, particularly with site security. After TRAC sampled aquatic moss from an unfenced, unposted stretch of Bear Creek, next to Rt. 95, DOE's Operations Office denied TRAC access to ORR, including all unfenced and unposted areas. TRAC's scientific study was thus banned from areas ordinarily accessible to scouts, hunters, and other members of the public. TRAC altered its sampling program to use a boat to access creeks flowing from ORNL into the Clinch River.

The City of Oak Ridge's Environmental Quality Advisory Board invited TRAC to meet with the Board and concerned citizens on 04 November. TRAC described the work-in-progress, answered questions, and listened to concerns.

TRAC completed sampling on 06 November and returned to its base to prepare and to analyze the 15 samples. TRAC analyzed the samples in its wide-band, photon spectrometer that is variably sensitive to about a hundred radionuclides. Several radionuclides are not routinely detectable by TRAC's method, including: tritium (H-3), carbon-14, and plutonium. Radionuclides that are ordinarily reported by TRAC often provide an indication of overall radiological conditions, allowing definition of follow-up work with additional radionuclides or toxics included in the analytical plan.

TRAC completed the radiological analyses on 11 February 2005. The first version of this data report was uploaded on March 6, 2005. That version included a section, "Defining Questions —and Preliminary Comments." Those preliminary comments were later modified into the implications, conclusions, and recommendations of the final report. The preliminary comments regarding lead-210 (Pb-210) were entirely changed in the subsequent interpretation.

The radiological results of the study appear in Tables 1 and 2, on the next two pages. Those results are summarized in the "Results of Independent Radiological Monitoring Around Oak Ridge" in TRAC's final report. The summarization is described later in this Data Report.

Table 1. Sample Numbers, Locations, and Media

Ref. Number	Sample*	North** Lat. [deg.]	West** Long. [deg.]	LOCATION	DESCRIPTION	Sample Medium
BKG	391819	47.40310	122.94957	Hood Canal, WA	high background reference, from composition roof	terrestrial moss
1.	4y0209	36.00168	84.24924	East Poplar Creek	Scarboro, beyond end and north of Woodbury Lane	clam shell
2.	4y0210	36.00168	84.24924	East Poplar Creek	"	green grass
3.	4y0214	36.00760	84.24916	Woodland	creek-side, W of Manhattan and Northwestern intersect	green grass
4.	4y0411	35.91759	84.33585	Rt. 95, Bethel V. Rd.	east shoulder of Rt. 95, north of Bethel Valley Road.	green grass
5.	4y0414	35.99381	84.26739	Scarboro, Oak Valley	old road, west of Hampton Rd., north of Baptist Church	terrestrial moss
6.	4y0212	35.99815	84.26574	Scarboro, trib. creek	north side of Tuskegee Dr., opposite South Fisk Ave.	aquatic moss
7.	4y0309	35.94374	84.34515	Bear Creek (Rt. 95)	west side of Rt. 95, a quarter mile N of Bear Creek Rd.	aquatic moss
8.	4y0610	35.99760	84.22353	U. Tenn. Arboretum	on boulder, east of creek, 1/4 mile NW of visitor center	terrestrial moss
9.	4y0410	36.00066	84.22262	Cattail Spring	south of Union Rd., E of driving range, S of residence	liverwort
10.	4y0510	35.89667	84.33305	White Oak Creek	on Clinch River side of discharge weir, from main flow	surface water
11.	4y0511	35.89667	84.33305	White Oak Creek	as above, from both sides of mouth of creek	sorrel
12.	4y05y2	35.90151	84.34170	Creek (between 11, 13)	at creek mouth, north side of Clinch River	privet fruit
13.	4y05y1	35.90248	84.34366	Melton Creek Mouth	at creek mouth, north side of Clinch River	privet fruit
14.	4y0512	35.89795	84.33575	Jones Island Creek	at creek mouth, north side of Clinch River	privet fruit
15.	4y0514	35.91638	84.41463	Clinch Ri. @ ETTP	north shore, next to old power house	privet fruit

* Sample Number code is time of sample collection: |Year|Month|DayDay|HourHour|. "4y0209" = 2004, November 02, 09:00 am. Hours "y1" and "y2" are between 11:00 am and 12:00 noon.

** Global Positioning System datum: WGS 84.

In Table 2, on the following page, results are reported in units of picocuries/kilogram(wet weight) [= pCi/kg(wet)]. These units allow direct comparisons of radioactivities of samples of different materials (media). Noting that one liter of water has a mass of one kilogram, results are directly comparable to Drinking Water Standards for radionuclides. Furthermore, bioaccumulation factors are simple ratios of radioactivities in biota to radioactivities in their water substrate, without any unit conversions.

One pCi/kg(wet) is one nuclear decay per minute, in a pound of wet sample (for example, a pint of water).
To convert pCi/kg(wet) to pCi/g(dry), multiply by 0.001 X wet/dry.
To convert pCi to Becquerels [Bq], multiply by 0.037.

± values are one standard deviation, counting uncertainty.
Blank space: Radionuclide not detected.

Key to Table 2 For conversion factors, see Page 3.

Sr-90	Strontium-90 (half-life = 29 years) is a product of nuclear fission —a beta particle-emitting bone-seeker.
Cs-137	Cesium-137 (half-life = 30 years) is a product of nuclear fission —a beta particle emitter that mimics potassium.
excess Pb-210	Lead-210 in excess of equilibrium with U-238 decay chain (half-life = 22 years) is a decay product of uranium through a radon intermediary —a beta emitter, with alpha-emitting progeny, that accumulates in bones.
Ra-226	Radium-226 (half-life = 1599 years) is an intermediate decay product of U-238 —an alpha particle-emitting bone-seeker.
U-238	Uranium-238, the main isotope of natural uranium (half-life = 4.5 billion years) —an alpha particle-emitting bone-seeker.
Thorium	Natural thorium and decay progeny (half-life = 14 billion years) —an alpha particle-emitting bone-seeker.
Pb-212	Lead-212, a radioactive intermediary of thorium decay (half-life = 11 hours) —See "***excess*** Pb-210", above.

Table 2. Radiological Results of Independent Monitoring Around Oak Ridge

Reference No.	Location	Sample Medium	washed?	wet/dry	pCi/kg (wet) Sr-90	Cs-137	***excess*** Pb-210	Ra-226	U-238	Thorium	Pb-212
BKG	Hood Canal, WA	terrestrial moss	no	6.98			994. ±53.	198.		83.	82.
1.	East Poplar Creek	clam shell	yes	1.17	840.		–2. ± 9.				
2.	East Poplar Creek	green grass	no	5.06			12. ± 2.		54.	93.	
3.	Woodland	green grass	no	5.90			11. ± 2.	25.			
4.	Rt. 95, Bethel V. Rd.	green grass	no	5.41		1. ± 4.	20. ± 2.	29.			
5.	Scarboro, Oak Valley	terrestrial moss	yes	5.93			402. ± 3.	107.	8.		35.
6.	Scarboro, trib. creek	aquatic moss	yes	7.07		8. ± 3.	19. ± 2.	511.	28.	214.	189.
7.	Bear Creek (Rt. 95)	aquatic moss	yes	7.72	403.	17. ± 3.	25. ± 2.	903.	469.	505.	260.
8.	U. Tenn. Arboretum	terrestrial moss	yes	5.04		39. ± 4.	685. ± 3.	159.	18.	17.	34.
9.	Cattail Spring	liverwort	yes	23.0			131. ± 1	86.		33.	24.
10.	White Oak Creek	surface water	8µm*	--*	47. ±8.	35. ± 0.	0.03 ± 0.03		0.4	0.9	0.4
11.	White Oak Creek	sorrel	no	13.2	2550.	1270. ±10.	1. ± 3.		19.	55.	42.
12.	Creek (between 11, 13)	privet fruit	no	4.11	856.		1. ± 2.		7.		16.
13.	Melton Creek Mouth	privet fruit	no	4.09	~81.	18. ± 4.	6. ± 2.		7.		
14.	Jones Island Creek	privet fruit	no	3.47			9. ± 3.				5.
15.	Clinch Ri. @ ETTP	privet fruit	no	3.52		12. ± 5.	–6. ± 3.		8.	62.	
EPA Drinking Water Standards, as references [pCi/kg(wet)]:					**8.**	**200.**	**15.**	**3.**	**21.**	**5.****	**15.**

* 21.79 kg (= 21.79 liters) surface water sample from mouth of White Oak Creek, coarse filtered (8 µm, Whatman 2V paper) to remove debris.

** EPA standard is for Ra-228 decay product, here assumed in secular equilibrium with parent thorium.

See End Notes on next page.

END NOTES for Table 2 (above):

- The relatively high-activity background "BKG" results are presented for comparison to *natural* radioactivity, unaffected by ORR.
- EPA drinking water standard values are presented for convenient reference comparisons only. These standards do not apply to vegetation or other biota.
- Preliminary Comments interpreting *Excess* Pb-210 in REV. 0 of this Data Report are corrected here.

Implications of Radionuclide Measurements

This section provides the interpretive connection between the data in Table 2, output from TRAC's radiological analyses, and the radiological results presented in the final report, "Radioactive Pathways --Oak Ridge, Tennessee [**www.radioactivist.org/ORRreports.html** (May 2005)]."

Radiological data in Table 2 are organized by radionuclide in columns in the original order of this Data Report: fission products, decay products of U-238, U-238, thorium, and a decay product of thorium (Pb-212). The final report is organized by kind of pathway: airborne, surface water, groundwater, and transportation accident. This section connects those two organizations of the radiological results. The Discussion section of the final report interprets the radiological results by pathway.

In the final report, the ratio of (excess Pb-210) / U-238 [listed as "Pb/U"] and Cs-137 are both treated as indicators of the origin of radioactivity: ORR or natural. *Excess* Pb-210 is not tabulated in the final report.

The intermediate product of thorium decay, Pb-212, is close enough to secular equilibrium with thorium in Table 2 that Pb-212 is not split out from thorium in the final report.

The other four radionuclides in Table 2, —Sr-90, U-238, Ra-226, and thorium— are the direct radiological *results* of this study, in the final report.

Strontium-90

General: Sr-90 is a product of nuclear fission with a half-life of 29 years, releasing beta particles. Sr-90 is a radioactive bone-seeker that mimics calcium, an element that regulates cellular functions and is a building material for bones. In contaminated regions having low calcium, Sr-90 is taken up into the food chain as a calcium substitute. Sr-90 retention in bones is limited by its 29-year radioactive half-life. Sr-90 can cause cellular malfunctions that can lead to cancer.

Implications of this study: A surface water sample from the weir at the mouth of White Oak Creek had 47 pCi/kg(wet), which is five times the maximum allowed in drinking water. This value is consistent with ORR perimeter monitoring reports of surface water.

Vegetation (sorrel) on the Clinch River shore, at the mouth of White Oak Creek, below ORNL, contained 2550 pCi/kg(wet) Sr-90. This is 300 times the reference value of 8 pCi/kg(wet). The calculated ***bioaccumulation*** factor for Sr-90 is 2550/47 = 54. This indicates how Sr-90 bioaccumulates in the food chain. TRAC measured this shoreline plume of Sr-90 in shoreline vegetation as far as 0.7 miles downstream from the White Oak Creek weir. TDEC notes that, "Watercress is naturally high in calcium, ... so it is likely that strontium (beta emitter) would be bioaccumulated as well." But TDEC has not analyzed for Sr-90 in its biological monitoring program.

Clam shells from East Fork Poplar Creek had Sr-90 at 100 times the reference value. This activity seems too high to have bioaccumulated from Y-12 Groundwater Treatment Facility 512 water discharges, averaging only 0.32 pCi/kg(wet) in 2003.

Cesium-137

General: Cs-137 is a product of nuclear fission with a half-life of 30 years, releasing a beta particle. Cesium has little or no ordinary biological role. In potassium-poor environments, cesium is taken up into the food chain as a substitute for potassium. Although Cs-137, like any radionuclide, can cause cancer, the relatively high reference value of 200 pCi/kg(wet), suggests lesser hazard than Sr-90. In this study, the low levels of Cs-137 measured are one indicator of the presence of artificial radioactivity.

Implications of this study: Cs-137 levels found in this study indicate contamination from ORR in the sample. Seven of 15 environmental samples that TRAC collected from publicly accessible areas around ORR tested significantly positive (>2 standard deviations) for Cs-137. Cs-137 levels found in this study are an indicator of the origin of contamination and not a direct concern for environmental quality or public health.

Excess **Pb-210**

General: Lead-210, with a half-life of 22 years, is an intermediate product of decay of natural U-238. Pb-210 is a beta-particle emitter. A product of Pb-210 decay is polonium-210 (Po-210). Po-210 emits alpha particles which are a significant contribution to the total radiation dose associated with radon exposure.

Lead has no biological function and is usually not taken up by biota. When lead does enter the human body, it accumulates in bones and other organs, where it resides for up to ten years. Elemental lead causes cancer and birth defects. Radioactive lead adds to those consequences.

Implications of this study: In samples in this study, Pb-210 indicates the origin of radioactivity in the U-238 decay chain: *natural* origin vs. *ORR* origin. The basis of this indication is that the U-238 decay chain in separated uranium is blocked by thorium-230 (Th-230), having a half-life of 75,400 years. Thus, subsequent products of decay are relatively depleted until thousands of years have passed, allowing Th-230 to grow-in toward equilibrium with uranium. Therefore, a low ratio of Pb-210 to U-238 is a direct indication of U-238 of ORR origin.

The situation with Pb-210 is complicated ORR's use of pure radium-226 (Ra-226), with a half-life of 1599 years. Ra-226 is the first decay product of Th-230. The use of Ra-226 *jumps* the decay block provided by Th-230.

After Ra-226 decays, the radioactive nucleus decays to Pb-210 in only four days. Then the 22-year halflife of Pb-210 itself retards build-up of Pb-210 to secular equilibrium, by about those 22 years.

TRAC's radiological analysis of a sample includes subtraction of the photon spectrum of Ra-226 with decay progeny including Pb–210. This subtraction is according to the intensity of photopeaks in the sample spectrum. After completion of the main analysis, the residual counts in the photopeak at 46.5 KeV in the residual sample spectrum are listed as "*Excess* Pb-210," in Table 2.

"*Excess* Pb-210" takes into account (by subtraction) the amount of Ra-226 in a sample. As already mentioned, separated uranium (of ORR origin) has almost no Pb-210 content, because of the Th-230 block. Thus, the ratio of *Excess* Pb-210 to U-238, in Table 2, is a measure of relative separation of uranium from its decay products in the sample. This ratio is called "Pb/U" in the Results of Independent Radiological Monitoring Around Oak Ridge, in the final report. In samples in which U–238 is not detected, blanks in Table 2, the Pb/U ratio is listed as >30 if *Excess* Pb-210 >0, and <0 if *Excess* Pb-210 <0.

The samples analyzed in this study were collected close to ORR, with the exception of the BKG sample. The Pb/U results fell into two categories: >30 and <1. Pb/U >30 is an indication of U-238 of natural (not ORR) origin, for the reasons described, above. Pb/U <1 is likewise an indication of U-238 of ORR origin, for samples in this study.

Radium-226

General: All four radium isotopes are radioactive. Ra-226 is most abundant radium isotope because of its relatively long half-life, 1599 years. Radium-226 of ORR origin is of particular concern because Ra-226 is the base material for intense sources of neutrons (PbBiPo–214 α into $BeT_2 \rightarrow$ ~1 Ci/μg). Such intense neutron sources can be used to produce *blocked* isotopes, such as curium-250, on an industrial scale, for micro-nuclear weapons. Ra-226 of ORR origin thus evidences new pollution from a new generation of weapons production already contaminating surrounding waters.

The manager of DOE's Portsmouth/Paducah Project Office published quantitative, environmental evidence of DOE's new use of Ra–226 dated February 24, 2005 ["Additional Information Regarding Radiation Levels Associated with Off-Site Creek Foam," PPPO-01-277-05, W.E. Murphie, letter to V. Colley]. On November 19, 2003, the operator of the Portsmouth Gaseous Diffusion Plant (GDP) had sampled foam from a stream emerging from the plant. They were checking a statement by Sergey Pashenko, at a press conference the day before, that radioactivity in the water foam was "at least 100 times higher than normal background levels." DOE confirmed Pashenko's result with gross beta values of 300 and 1,010 pCi/kg(wet) [= pCi/L] in two samples. DOE reported that the beta source was not technetium-99.

Pashenko, Director of Siberian Scientists for Global Responsibility, based in Novosibirsk, Russia, had measured energetic beta emissions from a foam sample, using a Geiger counter with a "fine-screens" method [S. Pashenko, et al., "A Citizen's Guide to Monitor Radioactivity Around the Energy Department's Nuclear Facilities," ISAR (May 2005), in press, Fig. 17]. Pashenko noticed in-growth of radon (Rn-222) and attributed the extreme beta activity to "uranium," which is a main product of the Portsmouth GDP.

However, the ***separated uranium*** (U-234) decay chain is immediately blocked by long-lived (75,400 year half-life) thorium-230, before radon is produced by radioactive decay. Therefore, Pashenko's attribution of "uranium" as the source material, may be understood to be generic rather than specific. The specific source of Rn-222 is Ra-226. The energetic beta that Pashenko detected [in his Fig. 20] was evidently the 3,270 KeV beta particle emitted by bismuth-214, in the Rn-222 decay chain.

Historically, after ORR has begun to produce special nuclear materials, much of the production has been farmed out to other facilities in the U.S. nuclear weapons complex, including the Portsmouth GDP. DOE's Oak Ridge Operations still has programs at the Portsmouth GDP.

Radium-226 decays to radon, an inert gas that emanates from the Ra-226 source. The radon is easily separated and captured as a secondary source of short-lived, radioactive decay products. An industrial process that recovers Rn-222 and progeny is called a *radon plant*. Rn-222 progeny release energetic alpha (**α**) particles and gamma (**γ**) radiation that yield neutrons (**n**) upon impact with beryllium nuclei. The main reactions are: $\alpha + \text{Be-9} \rightarrow \text{C-12} + \mathbf{n}$; $\gamma + \text{Be-9} \rightarrow 2\alpha + \mathbf{n}$.
Similarly, neutrons are released by alpha particles impacting tritium (H-3, **T**) nuclei, as in beryllium (tritium)hydride.

Ra-226 has been officially reported at Y-12, both in contaminated groundwater in the EMWMF Area of the Bear Creek Regime and in exit pathway spring water in the East Fork Poplar Creek Regime. In TRAC's study, Sample 7, aquatic moss from Bear Creek tested highest for Ra-226 at 903 pCi/kg(wet). The gross beta activity of that sample, due to radium content is estimated at 2,090 pCi/kg(wet). If the bioaccumulation factor for the beta emitters averages 50, then the contribution of the Ra-226 decay chain to gross beta in Bear Creek water would be about 40 pCi/kg(wet).

Ra-226 is a radioactive bone-seeker that mimics calcium, an element that regulates cellular functions and is a building material for bones. In contaminated regions having low calcium, Ra-226 is taken up into the food chain as a calcium substitute. Radium is retained in bones for about 40 years. Ra-226 can cause cellular malfunctions that can lead to cancer.

Implications of this study: Two indicators of Ra-226 of ORR origin are Cs-137 > 0 and Pb/U < 1 (see below). Sample BKG indicates a high background value of Ra-226 that is of natural origin.

Samples 3, 4, 5, 8, and 9 evidence natural levels of Ra-226 in vegetation samples around Oak Ridge. Sample 7, aquatic moss from Bear Creek, measured Ra-226 = 903 pCi/kg(wt), attributable to ORR. This is consistent with the known contamination of Bear Creek.

Sample 6, aquatic moss from a tributary to East Fork Poplar Creek, in Scarboro, measured 511 pCi/kg(wet). This is more than half the Ra-226 content of comparable moss from Bear Creek. Most of the Ra-226 in Sample 6 is attributable to Y-12. This positive result is attributed to production special materials for a new generation of nuclear weapons.

Uranium-238

General: Natural uranium is 99.3% U-238, with a half-life of 4.5 billion years. When natural U-238 is bombarded with neutrons, it is *neutron-activated* to plutonium-239, which is a fissile radionuclide used in nuclear weapons.

The main decay chain of natural U-238 includes 9 elements, including both alpha and beta emitters. Half-lives of the intermediate products of U-238 decay range from a tiny fraction of a second to 246,000 years for U-234. The decay chains include alpha emitting and beta emitting radionuclides. Uranium has no biological function and is rebuffed by biota. When uranium does enter organisms, it is concentrated in bones, where it is retained about one year. Products of uranium decay are often of greater concern than the uranium itself.

Over the last 6 decades, ORR has processed uranium in many ways. Some of these processes have released uranium into air and surface waters around the site. Uranium is usually considered the radiological contaminant of greatest concern around ORR.

Implications of this study: The two indicators of U-238 of ORR origin are Cs-137 > 0 and Pb/U <1 (see below)

Sample 5 yielded the highest content, in vegetation, of U-238 attributable to naturally occurring uranium: 8 pCi/kg(wet). Samples 3, 6, 7, and 11 had higher U-238 content, in all cases attributable to ORR.

By far the highest level of U-238 was 469 pCi/kg(wet) in aquatic moss from Bear Creek, Sample 7. This accords with the recognized problem of uranium contamination of Bear Creek and on-going remediation programs for that problem.

The ratio of U-238 levels of Sample 11 compared to Sample 10 (19./0.4 = 48.) indicates the magnitude of U-238 bioaccumulation, of ORR origin.

The U-238 content [28 pCi/kg(wet)] of Sample 6 is primarily attributable to ORR, as indicated by Pb/U<1 and significantly positive Cs-137.

Thorium

General: Natural thorium is almost entirely Th-232, with a half-life of 14 billion years. The decay chain of thorium contains 8 elements, including both alpha and beta emitters. Thorium is used at ORR in ways similar to U-238 and Ra-226. See the descriptions, above.

Like uranium, thorium has no biological role and is somewhat rebuffed from biota. However, thorium attaches to a variety of sites, with retention times ranging from 60 to 200 years.

Implications of this study: The two indicators of U-238 of ORR origin (Cs-137 > 0 and Pb/U < 1) also infer ORR-origin to thorium.

Sample 9 and the BKG sample evidence natural levels of thorium in comparable vegetation: up to 83 pCi/kg(wet). The ratio of thorium levels in Sample 11 compared to Sample 10 (55./0.9 = 61.) indicates the magnitude of thorium bioaccumulation, of ORR origin.

Pb-212

General: Lead-212, with a half-life of 11 hours, is an intermediate product of decay of natural thorium. Pb-212 is the first, intense photon-emitting radionuclide following the intermediate decay product, radon-220, in the thorium decay chain. Thus, the abundance of Pb-212 and its subsequent decay products indicates how much Rn-220 has de-gassed from the thorium decay chain, on one hand. On the other hand, an abundance of Pb–212 in excess of secular equilibrium indicates accumulation along the thorium decay chain.

See "*Excess* Pb-210" for a description of properties of lead.

Implications of this study: None.

Pb/U (as in the final report)

General: Pb/U is the ratio of lead-210 in *disequilibrium-excess* to parent uranium–238, in a sample. The "disequilibrium excess" is relative to radium-226 (with its decay products in the U-238 decay chain) subtracted from U-238 in the sample spectrum. Pb/U is an indicator of the source of U-238 in some environmental samples.

If the source of uranium is natural, Pb/U often has a high value (>30) in vegetation and other biological samples. Non-actinide decay products, like Pb-210, accumulate preferentially in many environments. If the source uranium comes from the processing of uranium that has been separated from other elements, Pb/U has a low value (<<1) in vegetation and other samples. The decay chains of separated uranium are *blocked* by long-lived actinide intermediate decay products, like thorium-230 with a half-life of 75,400 years. Thus, subsequent products of

radioactive decay, including Pb-210 with a half-life of 22 years, have grown-in to no more than minimal levels, over the first six decades of the atomic age. The results for Samples 2, 10, 11, 12, and 13 evidence this general situation.

The first, long lived product if the decay chain beginning with Ra-226 is Pb-210, with its 22 year half-life. Thus processes involving recently separated Ra-226 are relatively deficient in Pb-210. Two decades must pass before Pb-210 grows in approaching equilibrium. Until then, Pb-210 is depleted from a Ra-226 process flow. In terms of TRAC's radiological analysis, a *deficit* of Pb-210 remains after the Ra-226 spectrum (with progeny) is subtracted.

The amount of Pb-210 *deficit* from a radon plant operation diminishes over time, as Pb-210 grows in. The results for Sample 5 most clearly illustrate this general situation.

Therefore, releases of both refined uranium and refined radium from ORR entail low (or even negative) *excess* Pb-210, in comparison to natural materials in which the U-238 decay chain is usually closer to secular equilibrium. All samples of biota from around ORR presumably contain some naturally occurring U-238, Ra-226, and Pb-210. The question is the relative contributions of both natural and ORR sources. The ratio of

excess Pb-210 / U-238 = "Pb/U"

provides one indication of the dominant origin of U-238 and Ra-226 measured in a sample. These values appear in the final report. In cases where U-238 is not detected, Pb/U >30 is reported if *excess* Pb-210 >0, and Pb/U <0 is reported if *excess* Pb-210 <0. These reports are consistent with TRAC's U-238 detection levels and are conservative in the sense of possibly erring on the side of ascribing radioactivity of ORR origin to natural sources.

Implications of this study: Generally, Pb/U <1 indicates a sample contaminated from ORR. In particular, Pb/U<1 in a sample from around Oak Ridge indicates the U-238 in the sample is primarily of ORR origin. Comparison of U-238 levels of natural origin and of ORR origin, in similar sample media, from around Oak Ridge suggests how relatively influential ORR uranium-related processes are on the surroundings.

The unambiguous split between Pb/U <1 and >30 added to the merit of this indicator of origin of radioactivity in the U-238 decay chain.

Low Pb/U (<1) together with positive Cs-137 provide two radiological indicators of contamination of ORR origin.

Sample 8 had the only inconsistent results for these two indicators: Pb/U = 38 and Cs-137 = 39 pCi/kg(wet). The primary origin of the radiological content of Sample 8 is thus inconclusive.