

Environmental Defense Institute

News on Environmental Health and Safety Issues

February 2018

Volume 29

Number 2

What's Up With The Radionuclides in Drinking Water Around Boise, Idaho?

The public drinking water monitoring conducted by the State of Idaho is available on its website but can be cumbersome to access.¹ So, the efforts of non-profit organization called Environmental Working Group (EWG)² to summarize some recent years of public drinking water contamination helps to spot light problems in the nation's drinking water.

EWG has posted online a review of drinking water violations in Idaho between October 2014 and September 2017.³ And for 2010 to 2015 EWG posted data regarding drinking water that was not necessarily over federal limits but exceeded levels understood to adversely impact health.

The prize for bad water in the Boise area could go the Suez water district that serves 214,237 people. Not only does it have elevated levels of radionuclides uranium and combined radium, it also has levels for tetrachloroethylene (also known as "perc") in its water that exceed health guidelines and exceed both state and national averages. These levels do not exceed state or federal maximum contaminant levels (MCLs). But the levels exceed what is healthy to drink based on health rather than industry lobbying interests that sway federal MCL levels upward.

Levels of gross alpha and other radionuclides fluctuate from very low levels less than 3 pCi/L to levels exceeding 30 pCi/L at the Suez public water monitoring supply. Normal basalt aquifer levels probably should not be exceeding much over 3 pCi/L if detectable at all. Uranium and radium monitoring results to contrast systems with low levels to systems with high levels are provided in Table 1.

¹ Idaho Department of Environmental Quality, <http://www.deq.idaho.gov/water-quality/drinking-water/pws-monitoring-reporting/> and <http://www.deq.idaho.gov/water-quality/drinking-water/pws-switchboard/> and find sample results for all counties at <http://dww.deq.idaho.gov/IDPDWW/> where you select your county or drinking water system, select the specific water system. For the specific water system, it may be helpful to select the link at the left called "Chem/Rad Sample/Result by Analyte." Then select the analyte of interest that the well has data for by clicking on its code. This brings up the applicable lab samples that included that contaminant. Note that non-community wells typically sample fewer contaminants and do not sample radionuclides.

² Environmental Working Group at www.ewg.org and see their tap water database at <https://www.ewg.org/tapwater/> Use the map to select the state and an option for seeing water provider's violations in the state can be selected to view a summary of violators (2014 to 2017).

³ U.S. Environmental Protection Agency, Enforcement and Compliance History Online at <https://echo.epa.gov/help/facility-search/drinking-water-search-results-help>

Table 1. Uranium and radium monitoring results to contrast systems with low levels to systems with high levels. All units in picocurie/liter (pCi/L).

Utility Year	Uranium	Ra-226	Ra-228	Combined Ra-226 and -228
Sun Valley				
2011	0.87	0.16 to 2.13	0.55 to 1.07	0.76 to 2.96
2012	1.12	0.22	0.26	0.48 to 3.61
Shelley				
2013	1.4	0.1	0.9	1.0
2014	1.47	0.05 to 0.13	0.23 to 0.72	0.57
2015	1.13 to 1.42	0.09 to 0.12	0.09 to 0.15	0.22
Suez (near Boise)				
2010	ND to 8.04	NS	ND to 1.08	ND to 1.08
2011	ND to 22.11	NS	ND to 0.89	ND to 0.89
2012	ND to 16.08	NS	ND to 1.84	ND to 1.84
2013	ND to 23.45	NS	ND to 1.93	ND to 1.93
2014	ND to 18.76	NS	ND to 2.30	ND to 2.30
2015	ND to 22.11	NS	ND to 1.86	ND to 1.86
Kuna (near Boise)				
2010	10.72 to 16.75	NS	ND	ND
2011	12.73 to 14.74	NS	0.93	0.93
2012	13.4	NS	ND	ND
2013	ND to 14.74	NS	ND	ND
2014	4.69 to 7.37	NS to 1.6	ND to 2.8	ND to 2.9 *

Table notes: Only years with available sample data for 2010 to 2015 included. ND is nondetect and the criteria depend on the sampling detection capability and is variable for samples taken. Some utilities sometimes use 1 pCi/L as the detection limit although other instances use more accuracy. NS is not sampled. The presentation emphasizes the range of values rather than the average value. If only one sample was taken, that value was presented in the table. * For Kuna, the State of Idaho's switchboard data was used rather than EWG database.

For the water districts with low levels of uranium, the levels are below 1.5 pCi/L.⁴ The levels of radium-226 are normally below 0.2 pCi/L and the levels of radium-228 are normally below about 1 pCi/L. But in water districts around Boise, while nondetections indicate sample values 1 pCi/L or less, depending on the specified detection standards, uranium levels may range from “nondetect” to 23 pCi/L. Levels of radium-226 were not sampled and radium-228 levels are generally near 2 pCi/L. The reason for the rises in uranium levels and gross alpha levels needs to be identified. I suspect that the levels are man-made and fluctuate according to airborne radioactivity entering water tanks more than coming from the aquifer.

The levels for radium-228, from the thorium decay series, have typically been substantially higher than the levels of radium-226, from the uranium series, levels despite the high levels of uranium periodically measured. Levels of radium-228 in basalt aquifer of Idaho's southern region can be expected to be above radium-226 levels but not significantly. The levels of radium-228 at Suez near Boise sometimes exceeds 4 pCi/L (in 2008) which should certainly be

⁴ Note that uranium levels in pCi/L can be expressed as micrograms/liter (ug/L) by dividing by 0.67 pCi/ug, an estimate suitable for natural uranium.

considered elevated and odd. The State of California public health goal for radium-228 is 0.02 pCi/L.

For the Suez water district near Boise, the levels of gross alpha vary greatly even though the levels of adjusted gross alpha that subtract radon and uranium don't exceed the federal limit. Unadjusted gross alpha with uranium was 27.7 pCi/L in 1996, with an adjusted gross alpha (subtracting the uranium and radon) of 4.6 pCi/L. On June 21, 2017, gross alpha excluding uranium and radon was 18.7 pCi/L which is quite a remarkably high level. And it suggests that the source is not natural. Don't hold your breath waiting for the State of Idaho Department of Environmental Quality to explain what is happening — or why.

Importantly, there is some evidence that the activity (in curies) of various uranium nuclides don't match the composition of natural uranium nor do they match uranium mill tailings. The uranium has elevated levels of U-234 relative to U-238. This can arise from depleted uranium that includes uranium that has been enriched. The limited uranium composition sampling performed has at least one sample that suggests U-235 enrichment. The 2004 data I review here is from the state's drinking water database.

High gross beta levels have coincided with the high gross alpha readings. The gross beta levels are typically from man-made radionuclides but analysis is needed to identify the radionuclides contributing to the gross beta result have not been performed.

Unfortunately, water districts try to save money and may also be uninterested in finding the source of the radionuclide contaminants, so no testing is conducted to identify the contributors to the elevated levels of gross alpha. The Suez water district has a high number of water monitoring violations many of which pertain to radionuclide monitoring. In Idaho, it seems that monitoring violations are not uncommon in water districts with high levels of radionuclides.

Airborne contamination may be affecting the public water supply water. This could explain the wide fluctuations. Because there are no uranium mines, no nuclear reactors and no enrichment facilities in the Boise area, it would seem that the potential sources of contamination could be from Department of Defense actions such as testing of artillery. Depleted uranium artillery can have interesting origins of materials reprocessed from reactor operations. Depleted uranium of various origins can be blended, adding to the variety of radionuclides present. I would not rule out possible purifying various nuclear components related to nuclear weapons that don't require extensive shielding.

Whatever the source, and no matter the magic of averaging the water district may use, radionuclide ingestion or inhalation is carcinogenic. Everyone but especially pregnant women and children should consider drinking untreated water to be a health risk even if the average contamination values in the drinking water do not exceed federal limits.

Radionuclides in Drinking Water in Ammon, Idaho

Because I know that the Snake River Plain aquifer contamination from the Idaho National Laboratory does not flow toward Idaho Falls or Ammon, I was rather surprised by the elevated radionuclides in the public drinking water near Idaho Falls in Ammon, Idaho.

So, why are radionuclides in the drinking water in Ammon (and other drinking water supplies) elevated? I believe that elevated levels of airborne contamination are entering the above ground water tanks as they cycle up and down.

The radium levels of radium-226 and radium-228 often are below 1 pCi/L each, but the levels for radium-228 levels sometimes exceed 2 or exceed 3 pCi/L.

The levels of uranium are often less than 2 ug/L but in 2010, uranium was sampled at 3.87 ug/L. In 2016, uranium was sampled at 3.6 ug/L with elevated radium-228 at 2.6 pCi/L. Note that this is counter intuitive because radium-226 is associated with uranium-238 decay series while radium-228 is associated with the thorium-232 decay series.

Adjusted gross alpha levels (excluding uranium and radon) range from less than 1 pCi/L to over 10 pCi/L. Gross beta levels are elevated at times, from 5 to 12 pCi/L — also with no identification of the contributing radionuclides. There is typically no analysis of uranium composition (U-234, U-235 and U-238 activity contribution).

Nearby Comore Loma had the following water sampling data in 2006: gross alpha excluding radon and uranium at 8.2 pCi/L and gross beta at 19.6 pCi/L. The annual environmental reporting for the Idaho National Laboratory and surrounding communities had large spikes in gross alpha and gross beta in air monitoring at www.idahooser.com for 2006. The gamma spectrometry of filters showed contributions to airborne radioactivity from the usual emissions and/or soil resuspensions from the INL of cesium-137, strontium-90, plutonium-239, americium-241 and plutonium-238. These are among the most common radionuclides detected that are significant contributors to radiological dose from the INL.

Note that other transuranics such as curium-244 and californium-252 are known to be disposed of to the open-air evaporation pond at the ATR Complex (formerly known as the Test Reactor Area), in addition to radionuclides listed in the previous paragraph and various other radionuclides such as europium-152 and -154 are also disposed of to the evaporation pond from chemical separations processes as well as normal reactor operations effluent. The Advanced Test Reactor is also a large emitter of tritium, argon, xenon, and krypton.

No matter the repeated refrain that based on the airborne monitoring, officialdom just can't say where the radionuclides might have come from, since, shucks, the contamination is spread from Rupert to Rexburg, Carey and Arco to Mud Lake and Sugar City, and Blackfoot to Craters of the Moon.

The airborne contamination settles on soil which is incorporated into crops like lettuce and wheat. And the airborne contamination from the INL is getting into our drinking water despite not arriving there from the flow of the aquifer.

The uranium processes at the INL's Specific Manufacturing Capability (SMC) that has manufactured military tank armor since 1984 from depleted uranium can obviously release depleted uranium. Other INL facilities associated with highly enriched uranium fuels may be releasing uranium that includes U-236 which would elevate decay progeny radium-228. Other INL operations have significant plutonium-239 are always accompanied by various isotopes of plutonium including plutonium-240 which decays to uranium-236. Plutonium-241 and americium-241 decay to uranium-233 that has its own unique decay series that is rarely presented in drinking water radionuclide monitoring literature. Alpha emitters such as uranium and plutonium include various gamma emissions as they decay through a series of decay progeny through alpha and beta decay. See the next article for details of the decay series.

In past years, the annual environmental monitoring funded by the Department of Energy has held a certain pretense of being fair and unbiased. That has never been the case. And unfortunately, the Idaho Department of Environmental Quality seems to be as interested in covering up the source of radionuclide contamination as the Department of Energy is.

Understanding the Radionuclide MCLs in Drinking Water in Idaho

When you find the drinking water data, you want to understand what the contamination levels mean to your health and your family's health. Federal maximum contaminant levels (MCLs) are often not protective of health, especially for children and the developing child in utero. The State of California publishes health goals that provide a better indicator of health significance than federal MCLs for contamination levels in drinking water.

With drinking water monitoring of community wells, there are several radionuclide sampling measurements taken periodically. The radionuclide measurements in public drinking water systems may include gross alpha particle, gross beta, radium-226, radium-228, and uranium. When contaminants levels have been low, the schedules for sampling are relaxed and skip many years at a time between measurements. See Table 2 below for common measurements of radioactivity in public drinking water supplies. For more information about other common contaminants in drinking water, see EDI's December 2017 newsletter.

The radionuclide sampling by the state is created based on federal standards that are largely built around the expectation of natural sources of radioactivity. It is understood that "naturally occurring" levels of radioactivity can be unhealthy.

When drinking water monitoring started in the early 1990s in Idaho, drinking water monitoring of gross alpha-emitter activity and gross beta-emitter activity was often conducted. The gross beta activity is associated with man-made releases but is less often measured now.

Table 2. Radionuclide monitoring typical of state drinking water monitoring programs of community wells.

Code	Analyte	Typical of uncontaminated basalt aquifer	Federal MCL ^a	Public Health Goal ^b	Comment
4000	Gross alpha excluding radon and uranium	Zero	15 pCi/L	Zero	The source of alpha can be radium, thorium, plutonium, or americium. The absence of radium-228 suggests the absence of thorium-232.
4002	Gross alpha including radon and uranium	< 3 pCi/L	See Uranium MCL	See Uranium goal	Gross alpha including uranium would not include gaseous radon. It would include radium-226 which is an alpha emitter. It would not include radium-228 because it is a beta emitter. And it may include radium-224 although typically the radium-224 is not determined.
4100	Gross beta (excluding K-40)	Zero	4 mrem 50 pCi/L	-	Units of mrem or pCi/L may be used. The source of the beta/photon emitter is usually not identified but can be manmade strontium-90, cesium-237, cobalt-60 or plutonium. Proper determination of mrem requires knowing which nuclides are present.
	Strontium-90	Zero	8 pCi/L	0.35 pCi/L	(Sometimes measured and relates to total strontium)
4010	Combined radium-226 and radium-228		5 pCi/L	See radium-226 and radium-228 limits.	Radium ingestion or inhalation can cause lymphoma, bone cancer or diseases of blood formation such as leukemia and aplastic anemia). Radium-224 is typically not regulated and to do so would require gross alpha testing with 48 hours of sample collection
4020	Radium-226		See combined radium MCL	0.05 pCi/L	Detection levels of 1 pCi/L may be too high to discern low levels.
4030	Radium-228		See combined radium MCL	0.019 pCi/L	Detection levels of 1 pCi/L may be too high to discern low levels.
4006	Combined uranium		20 pCi/L	0.43 pCi/L	20 pCi/L would correspond to 30 ug/L if natural uranium.

Code	Analyte	Typical of uncontaminated basalt aquifer	Federal MCL ^a	Public Health Goal ^b	Comment
					Typical conversion using 0.67 pCi/ug assumes natural uranium composition.
4007	Uranium-234		See combined uranium MCL	See combined uranium goal	Uranium-234 is present is natural uranium and non-natural uranium and contributes significantly to activity.
	Radon		Advisory level between 300 and 4000 pCi/L	1.5 pCi/L	No requirement to monitor radon.
4008	Uranium-235		See combined uranium MCL	See combined uranium goal	Uranium-235 concentration is lower in depleted uranium and higher in enriched uranium. Enrichment can range from 3 to 93.5 percent.
4009	Uranium-238		See combined uranium MCL	See combined uranium goal	Uranium-238 concentration is greater in depleted uranium.
	Tritium		20,000 pCi/L	400 pCi/L	

Table notes: Federal maximum contaminant levels (MCLs) set the state and federal levels requiring enforcement are based on EPA's 2012 edition of Drinking Water Standards at oehha.ca.gov/water/phg/allphgs.html. The public health goals in the table are based on California's State Water Resources Control Board 2016 Groundwater Information Sheet on Radionuclides and are not enforceable.

State drinking water programs were not in place during the 1950s through 1970s during those decades of extensive nuclear weapons testing in the United States, mostly at the Nevada Test Site, by the United States outside the continental U.S. over the Pacific Ocean, for example, and by other countries including China, Russia, France, and the United Kingdom. With the tapering off of nuclear weapons testing, one might expect gross beta levels to tend to decrease from the 1990s to today, but that is not necessarily the case. Gross beta is often an indicator of man-made radionuclides in the water but drinking water sampling generally does not identify the specific radionuclides contributing to gross beta results.

Gross alpha monitoring of drinking water is conducted for public drinking supplies. The gross alpha can be due to natural sources of uranium and thorium in the aquifer. But in Idaho, elevated gross alpha levels in drinking water appear to be because of man-made radionuclides that get into the water supplies. Neither the Department of Energy nor the U.S. Geological Survey want you to know that the radionuclide contamination in the aquifer is from reactor operations, spent nuclear fuel reprocessing, or disposal of nuclear wastes.

If the levels in the aquifer are not elevated but the levels in drinking water are then it appears to me that radiologically contaminated air coming into contact with our drinking water could be the cause. But state and Department of Energy funded air monitoring near the INL continually deny that elevated radionuclides are due to the INL to a degree that is very unscientific. Their continued denials would be laughable if it were not so important to our health and to future generations. The radionuclides we're talking about persist in the environment essentially forever — and the nuclear industry is determined to keep adding to it.

Drinking water monitoring often includes sampling of radium-226 and radium-228. Radium-226 is one of the decay progeny of uranium-238. Radium-228 is one of the decay progeny of thorium-232. Both are naturally occurring in rock and soil.⁵ But the levels of radium-226 and radium 228 can also be elevated from radionuclide releases from nuclear operations. The total radioactivity of these combined is not to exceed 5 pCi/L by the federal MCL for combined radium-226 and radium-228, but there should be concern about the levels of combined radium at levels at far lower levels.

Often the amount of uranium in the water is also sampled and it can be expressed in micrograms/liter (ug/L). The assumption of natural uranium composition can be used to convert the mass measurement (micrograms/liter) to activity in pCi/L using 0.67 pCi/ug. Sometimes water sampling will include analysis of the composition of uranium allowing a comparison of the uranium in the water to that of naturally occurring uranium.

Gross alpha levels excluding uranium and radon are called “adjusted gross alpha” and the federal limit is 15 pCi/L. High adjusted gross alpha levels are an indicator of possible man-made radionuclide contaminants such as plutonium. But drinking water programs tend to not to identify the source of the elevated gross alpha in drinking water.

Uranium, plutonium, americium, and other transuranics released to the air by the Idaho National Laboratory can elevate levels of airborne gross alpha contamination. The elevated levels tend to coincide with elevated levels of gross alpha not only in surface water but also in public drinking water supplies that do not use surface water. Trends can be difficult to discern from sometimes infrequently sampled drinking water.

Exceeding federal MCLs can invoke more costly sampling requirements and could trigger the need for water treatment.⁶ Imposing water treatment is also considered costly and water districts have resorted to aggressive data manipulations to avoid exceeding MCLs. In Texas,

⁵ Zoltan Szabo et al., Elsevier Applied Geochemistry, “Occurrence and geochemistry of radium in water from principal drinking-water aquifer systems of the United States,” Vol. 27, Issue 3, March 2012. See Appendix A Supplementary Data regarding aquifers including basalt aquifer in Idaho. The natural occurrence of uranium and thorium is low in Idaho’s basalt aquifer. <https://doi.org/10.1016/j.apgeochem.2011.11.002> and <https://www.sciencedirect.com/science/article/pii/S0883292711004471>

⁶ Here is a concise overview of water treatment technologies for radionuclide removal by David P. Boaz, HydraTech, 2008. https://www.hydratechnm.org/documents/technical_articles/understanding_the_epa_radionuclides_rule.pdf

uncertainty was subtracted from sample data results to lower the reported results.⁷ Data averaging can be used to dilute peak results. In Idaho, many water districts having high radionuclide levels have numerous reporting violations which might be related to attempts to avoid reporting high radionuclide sample results.

Because drinking water monitoring is complex, it might sound more comprehensive than it is. Public water supply sampling requirements have evolved based on expectations that are not necessarily correct and on economics. The result is a patchwork of sampling that may provide some clues but will leave various radionuclides unidentified in community wells. Non-community wells where people work but don't live, don't require any monitoring of radionuclides.

Radium-224 sampling was not required in part because of the expectation that the thorium decay series that includes radium-228 and radium-224 as decay progeny was expected to be less prominent than the uranium-238 decay series. That assumption has not panned out. Radium-228 is often higher than radium-226, and radium-224 levels when further researched can equal or exceed the radium-228 levels which do require sampling.

The U.S. Geological Survey reported in 2001 that "Conventional monitoring procedures, which do not require analysis of gross-alpha-particle activities in time to account for the contribution of short-lived isotopes such as Ra-224, could lead to false indications of compliance."⁸ The report noted that although Ra-224 was not typically measured, it generally occurs in ratios near 1:1 with Ra-228. Gross alpha particle screening of 3 pCi/L of Ra-226 can wrongly indicate that there is no significant Ra-228 (a beta particle emitter).

The USGS report also pointed out the absence of lead-210 and polonium-210 sampling may be missing this source of radiation but the report indicated that the levels would be expected to be below 3 pCi/L. Polonium-210 binds to hemoglobin and there have been concerns that it may be an underestimated public health risk.⁹

Tritium levels are not sampled presumably because the federal MCL would not be exceeded; yet reasonable public health goals may be exceeded. See EDI reports on the aquifer including

⁷ Bill Walker, Editor in Chief and Wicitra Mahotama, Environmental Analyst, "170 Million in U.S. Drink Radioactive Tap Water. Trump Nominee Faked Data to Hide Cancer Risk," January 11, 2018. https://www.ewg.org/research/170-million-us-drink-radioactive-tap-water-trump-nominee-faked-data-hide-cancer-risk#.WnO_vedG2Um See also the interactive map to find radionuclide contamination in public drinking water in your area.

⁸ Michael J. Focazio et al., U.S. Geological Survey, "Occurrence of Selected Radionuclides in Ground Water Used for Drinking Water in the United States: A Reconnaissance Survey, 1998," Water-Resources Investigations Report 00-4273, 2001. <https://pubs.usgs.gov/wri/wri004273/pdf/wri004273.pdf>

⁹ James A. Jacobus, Minnesota Department of Health, Fact Sheet RP099, <http://www.health.state.mn.us/divs/eh/risk/guidance/dwec/radionpostsetac.pdf>

“Tritium at 800 pCi/L in the Snake River Plain Aquifer in the Magic Valley at Kimama: Why This Matters.”¹⁰

Radionuclides in our drinking water in southeast Idaho include naturally occurring radionuclides, past nuclear weapons testing fallout and resuspension of historical releases from the INL in the soil. The extent that the radionuclides in our drinking water are from continuing airborne releases from the INL is not something that INL monitoring programs are going to discuss given the denials that INL is the source of elevated airborne radionuclide contamination.

Understanding the Man-Made Radionuclides in Drinking Water in Idaho

Most of the literature about public drinking water monitoring will discuss the natural decay chain of uranium-238 and of thorium-232.¹¹ Then there seems to be a reliance on assuming that the uranium, thorium, radon, and radium are from naturally occurring uranium and thorium in rock and soil, even when the levels have been increased above natural occurring levels.

To understand the radionuclides in Idaho’s drinking water, it becomes necessary to also understand the uranium-235 and uranium-233 decay series. It also becomes important to understand the processes that create actinides¹² in a nuclear reactor, such as neptunium and plutonium and none of this is discussed in much of the information about radionuclides in drinking water available from state and federal agencies.

First of all, it is helpful to understand the composition of naturally-occurring uranium provided in Table 3. When high levels of gross alpha occur in the drinking water (greater than 15 pCi/L), additional sampling to determine the amount of uranium per liter will be conducted. If the uranium levels are high, additional analysis of the uranium composition may be conducted.

¹⁰ Tami Thatcher, Environmental Defense Institute Special Report, “Tritium at 800 pCi/L in the Snake River Plain Aquifer in the Magic Valley at Kimama: Why This Matters,” 2017. www.environmental-defense-institute.org/kimamareport.pdf

¹¹ Michael F. Arndt, “Evaluation of Gross Alpha and Uranium Measurements for MCL Compliance,” Water Research Foundation, 2010. <http://www.waterrf.org/PublicReportLibrary/3028.pdf>

¹² Actinides include uranium, thorium, actinium, and protactinium as well as transuranics. Transuranics include radionuclides such as plutonium, americium, neptunium and other radionuclides with a higher number of protons than uranium. Uranium always has 92 protons; but the number of neutrons varies with isotope. Atomic mass number is the rounded sum of protons and neutrons in an atom. The most prevalent naturally occurring nuclide of uranium is uranium-238, with 92 protons and 146 neutrons. The atomic “mass number” for U-238 is 238; the “atomic number” is 92.

Table 3. Natural uranium composition.

Nuclide	Mass percent	Kg	Curie
U-234	0.005	38	23.4
U-235	0.714	5045	1.1
U-238	99.279	70,063.9	23.4
	100 %	70,572.2	47.9

Kg (kilogram) and curie amounts assumed for 1000 Megawatt reactor.

When the activity contribution from nuclides of uranium such as U-234, U-235 and U-238 don't match natural uranium, you may be able to determine the man-made process the uranium came from by comparing to the uranium compositions in Table 4. In Idaho, we don't have much mining or milling uranium waste. But we do have military depleted uranium processes at the Idaho National Laboratory and apparently elsewhere in the state. And the INL has extensive nuclear reactor and fuel reprocessing and storage that involve highly enriched uranium (HEU) nuclear fuel from Department of Energy research reactors and Naval fleet spent fuel.

For example, if the source of uranium was from the use of highly enriched uranium, the presence of U-236 is an indicator that can be identified by gamma spectrometry.

Table 4. Variations of man-made uranium compositions

Nuclide	Mass percent	Kg	Curie
Uranium Mill Tailings			
U-234	0.001	0.9	5.57
U-235	0.25	175.6	0.37
U-238	99.749	70,044	23.34
	100 %	70,220.5	29.34
Depleted uranium (variations dependent on source and blend)	Mass percent	Kg	Curie
U-234	0.0008	-	-
U-235	0.2 (0.32 to 2 percent)	-	-
U-236	0.003 (0 to 64 percent)	-	-
U-238	99.79 (also variable)	-	-

Nuclide	Mass percent	Kg	Curie
Highly Enriched Uranium (fresh)	Mass percent	Kg	Curie
U-234	0.84	2.9	17.9
U-235	93.5	328.9	0.7
U-238	5.66	19.9	0.0006
	100 %	351.7	18.606
Spent HEU	Mass percent	Kg	Curie
U-234	0.1	0.1	0.3
U-235	21.8	17.4	0.0
U-236	55.8	44.6	2.8
U-237	0.0	0.0	0.9
U-238	22.3	17.8	0.0
	100 %	79.8	4.2
Spent thorium cycle U-233 breeding	Mass percent	Kg	Curie
U-232	0.0	0.1	2307
U-233	55.4	187.6	1778
U-234	23.2	78.7	487
U-235	9.5	32.3	0.1
U-236	11.5	39.0	2.5
U-237	0.0	0.0	0.6
U-238	0.3	1.0	0.0
	100 %	338.8	4,575

Kg (kilogram) and curie amounts assumed for 1000 Megawatt reactor. Source of mass percent, kg and curie levels: Anthony V. Nero, Jr, *A Guidebook to Nuclear Reactors*, University of California Press, 1979.

When reviewing drinking water radionuclide sampling, understanding how the actinides can contribute to the “natural” decay series of uranium-238 and of thorium-232, and the decay series of uranium-235 and of uranium-233 can be quite helpful. And it also becomes necessary to understand not only the fission products resulting from reactor operation, but also the radionuclides formed by successive neutron absorption that occurs in the neutron rich environment of an operating nuclear reactor.

Four decay series are presented in Tables 5 through 8 below:

the uranium-238 decay series known as the uranium series;

the thorium-232 decay series known as the thorium series;

the uranium-235 decay series known as the actinium series, and

the uranium-233 decay series which is man-made and remains officially nameless.

I have included these decay series tables here for three reasons: (1) unless you have a degree in radiochemistry, you need to have the names of the nuclides spelled out along with their short-hand symbol identifier (such as U, Pu, Np), (2) it is difficult to locate decay series that are complete with man-made decay chains feeding in, and (3) it is important to understand the specific decay series that a radionuclide belongs to as you study drinking water, lung count results and environmental radionuclide emissions data.

These decay series show the man-made actinides that may also decay through the same series in grey. The decay series depict alpha decay as progressing downward and reducing the atomic mass by 4. Beta decay by electron emission is depicted as progressing upward diagonally to the right. Beta decay flips a neutron into a proton and stays at the same atomic mass. Isotopes of the same chemical element have the same number of protons but can have variable numbers of neutrons and variable atomic mass. The half-lives of the various radionuclides range from millions or billions of years to milli-seconds.

Along with alpha and beta decays at various energy levels, gamma photon emissions of various energy levels can also occur which can be detected by gamma spectrometry.

So, while uranium, thorium and plutonium are thought of primarily as alpha particle emitters, gamma radiation is also emitted and decay progeny may emit beta particles rather than alpha particles along with gamma radiation at various energy levels measured in kiloelectron volts (keV).

Weak or low energy gamma emissions require less shielding than higher energy gamma emissions. Uranium decay progeny of Th-231, Th-234 and Pa-234, all beta emitters, have high specific activity in curies per gram that require some protection of workers.

Sources of uranium-238 include natural soil and rock sources, mill tailings, depleted uranium, reactor fuel melting from reactor accidents, and spent fuel reprocessing. Sources of uranium-234 decay progeny can include man-made plutonium-238 that is present in various materials and processes at the INL.

Sources of thorium-232 include natural thorium-232 in rock and soil. Sources of thorium-232 can also include man-made plutonium-240 and uranium-236 resulting from neutron capture in a reactor.

Table 5. Uranium-238 decay series.

Californium	Cf-250 *						
Curium	Cm-246 *		Cm-242				
Americium	↓	Am-242	↓				
Plutonium	Pu-242	↓	Pu-238				
Neptunium	↓	Np-238 / [^]	↓				
Uranium	U-238		U-234				
Protactinium	↓	Pa-234 / [^]	↓				
Thorium	Th-234 / [^]		Th-230				
Radium			Ra-226				
Radon			Rn-222				
Polonium			Po-218		Po-214		Po-210
Bismuth			↓	Bi-214 / [^]	↓	Bi-210 / [^]	↓
Lead			Pb-214 / [^]		Pb-210 / [^]		Pb-206 (stable)

Table notes: Alpha decay downward reduces the atomic mass by 4; beta decay upward diagonally to the right flips a neutron to a proton and stays at the same atomic mass. In the table, arrow symbols downward are used to show the progression of some alpha decays if there was space to show the arrow. Movement upward and to the right is shown by /[^] which is a lame keyboard attempt to look like an arrow. Man-made actinides are shown in grey.

* Decay series to Cf-250 and Cm-246 not shown which include Cm-250, Pu-246, Am-236 and Bk-250.

Sources of uranium-238 include natural soil and rock sources, depleted uranium, reactor fuel melting from reactor accidents, and spent fuel reprocessing. Sources of uranium-234 decay progeny can include plutonium-238.

Table 6. Thorium-232 decay series.

Californium	Cm-252		Cf-248				
Curium	Cm-248		Cm-244				
Americium	↓		↓				
Plutonium	Pu-244		Pu-240				
Neptunium	↓	Np-240/ [^]	↓				
Uranium	U-240/ [^]		U-236				
Protactinium			↓				
Thorium			Th-232		Th-228		
Actinium			↓	Ac-228/ [^]	↓		
Radium			Ra-228/ [^]		Ra-224		
Radon					Rn-220		
Polonium					Po-216		Po-212
Bismuth					↓	Bi-212/ [^]	↓
Lead					Pb-212/ [^]	↓	Pb-208 (stable)
Thallium						Tl-208/ [^]	

See table notes for Table 5. Sources of thorium-232 include natural thorium-232 in rock and soil. Plutonium-240 and uranium-236 which results from neutron capture in a reactor also decay to thorium-232. Depleted uranium can include uranium-236. The higher actinides that decay to plutonium-240 are not shown but include californium-252 and -248, curium-248 and -244, plutonium-244, and neptunium-240.

Table 7. Uranium-235 decay series.

Californium	Cf-251						
Berkelium	↓	Bk-247					
Curium	Cm-247	↓	Cm-243				
Americium	↓	Am-243	↓				
Plutonium	Pu-243 / ^	↓	Pu-239				
Neptunium		Np-239 / ^	↓				
Uranium			U-235				
Protactinium			↓	Pa-231			
Thorium			Th-231 / ^	↓	Th-227		
Actinium				Ac-227 / ^	↓		
Radium				↓	Ra-223		
Francium				Fr-223 / ^	↓		
Radon					Rn-219		
Polonium					Po-215		
Bismuth					↓	Bi-211 / ^	
Lead					Pb-211 / ^	↓	Pb-207 (stable)
Thallium						Tl-207 / ^	

See table notes for Table 5. Sources of uranium-235 include natural uranium in rock and soil. It should not be ignored where enriched uranium is released to the environment. Plutonium-239 also decays to uranium-235 and higher actinides (californium, curium, americium and neptunium) are shown. Dispersion of reactor fuel from reactor accidents and spent fuel reprocessing can spread uranium-235 in the environment.

Table 8. Uranium-233 decay series.

Californium	Cf-241						
Curium	Cm-245	↓					
Americium	↓	Am-241					
Plutonium	Pu-241 / ^	↓					
Neptunium		Np-237					
Uranium		↓	U-233				
Protactinium		Pa-233 / ^	↓				
Thorium			Th-229				
Actinium			↓	Ac-225			
Radium			Ra-225 / ^	↓			
Francium				Fr-221			
Radon				↓			
Astatine				At-217			
Polonium				↓	Po-213		
Bismuth				Bi-213 / ^	↓	Bi-209	
Lead				↓	Pb-209 / ^	↓	
Thallium				Tl-209 / ^			Tl-205

See table notes for Table 5. Uranium-233 is not naturally occurring. This weapons fissile material can only be produced in a reactor or by the higher actinide decays shown including plutonium-241 and americium-241 decay. Higher actinides (californium, curium, americium and neptunium) are shown. Uranium-233 can and has been used in nuclear weapons testing. Its dispersion can also result from various weapons production and separations processes. Disposal of americium-241 following plutonium purification may be a significant source. It can also result from spent fuel reprocessing particularly of high enriched uranium fuel because of the high buildup of neptunium-237 in HEU reactor operations.

Sources of uranium-235 include natural uranium in rock and soil but are typically considered to be of small enough abundance to be ignored. But this decay series should not be ignored where enriched uranium is released to the environment. Sources of the U-235 decay series also include plutonium-239 which decays to uranium-235. Dispersion of reactor fuel from reactor accidents and spent fuel reprocessing can spread uranium-235 in the environment. Waste water disposal from HEU spent fuel reprocessing has put uranium-236 in the Snake River Plain Aquifer. Fuel reprocessing and calcining and reactor fuel melt tests or accidents spread various radionuclides present in nuclear fuels to air and soil.

Depleted uranium is uranium that is left over after extraction of uranium-235. Enriched uranium includes more than 0.72 percent up to 93.5 percent U-235 enrichment. Commercial nuclear power reactors typically use 3 to 5 percent enrichment. Enriched uranium also includes increased amounts of uranium-234 which cannot be separated from the uranium-235. Most depleted uranium includes between 0.2 and 0.4 percent uranium-235. Depleted uranium composition can vary and can include uranium-236 if it resulted from reactor fuel reprocessing. The health harm caused by inhalation or ingestion of depleted uranium includes illness and increased risk of birth defects.^{13 14}

Uranium-233 is not naturally occurring. This weapons fissile material can only be produced in a reactor or by the higher actinide decays shown including plutonium-241 and americium-241 decay. Uranium-233 has been dispersed by its production, separation and limited use in nuclear weapons testing. Disposal of americium-241 following plutonium purification may be a significant source. It can also result from spent fuel reprocessing particularly of high enriched uranium fuel because of the high buildup of neptunium-237 in HEU reactor operations.

Higher actinides such as californium, curium, americium and neptunium may be produced using target material in nuclear reactors in order to produce weapons related materials or to produce a heat source for radiothermal generators such as plutonium-238 which is used as a power supply in spacecraft.¹⁵ These materials have been disposed of routinely to an open-air evaporation pond at the INL's ATR Complex. These materials have not necessarily been included in required federal reporting under the National Emissions Standards (NESHAPs) because they are not monitored but only estimated. Therefore, whenever unplanned releases are occurring via escaping resin beads, for example, the emissions would be underestimated. Frankly, the NESHAPs reporting by the INL appears to lack validation and may substantially understate INL's airborne emissions of transuranics and other radionuclides.

¹³ Rosalie Bertell, International Journal of Health Services, "Depleted Uranium: All the Questions About DU and Gulf War Syndrome Are Not Yet Answered," 2006. p. 514
<https://ntp.niehs.nih.gov/ntp/roc/nominations/2012/publiccomm/bertellattachmentohw.pdf>

¹⁴ Depleted Uranium Education Project, *Depleted Uranium Metal of Dishonor How the Pentagon Radiates Soldiers & Civilians with DU Weapons*, 1997. ISBN:0-9656916-0-8

¹⁵ Transuranics are radionuclides often having extremely long half-lives. Many decay progeny may be created before reaching a stable, non-radioactive state. See our factsheet at <http://www.environmental-defense-institute.org/publications/decayfact.pdf>. See also an ANL factsheet at <https://www.remm.nlm.gov/ANL-ContaminationFactSheets-All-070418.pdf>

The environmental monitoring of airborne radioactivity that is conducted tends to ignore peaks and appears to be missing weeks of data in graphs charting alpha and gamma airborne radiation levels. This can be observed for various years, but is particularly obvious in 2006.¹⁶ Particulate matter in filters for 2006 provide instances of elevated levels of radionuclides such as plutonium-239, plutonium-238 and americium-241 in the filters along with cesium-137 and strontium-90. A high statistical bar allows denial that a “detection” of the radionuclide occurred.

Numerous “detections” were admitted in assessing filter particulate in 2006, see first quarter 2006 air monitoring at www.idahoesser.com.¹⁷

The coincidence of elevated levels of airborne radioactivity seem to correspond to elevated gross alpha and gross beta levels in drinking water monitoring.

Weapons material that is fissile include uranium-235 which is concentrated by enrichment while plutonium-239 is created from uranium-238 by neutron capture in a nuclear reactor. Fission products such as cesium-137 and strontium-90 (and many others) are created in a nuclear reactor by the splitting apart of uranium atoms. Actinides are created by neutron capture and these include the actinides neptunium, plutonium, americium, curium and californium.

As shown in the decay series tables, man-made actinides can decay to “natural” decay series. But natural does not mean healthy especially when the levels of decay progeny are elevated. And the experts that pretend that the decay progeny are from “natural” background are not admitting that the reason the levels of decay progeny are elevated is due to the release of radionuclides from the INL and other nuclear operations.

Many of these decay progeny are harmful to health but are not monitored because of the techniques used to perform sampling or due to a mistaken belief that since uranium is natural it does not need to be monitored. Uranium health effects depend on the solubility and the concentration and health studies of miners are not necessarily exposed to comparable chemical forms of uranium.

Uranium, including depleted uranium, persists in the environment essentially forever and causes illness, cancer and increased risk of birth defects. Gulf war veterans found this out as their babies were born with missing fingers and arms. See our 2017 EDI report about radiological and chemical exposures at the INL.¹⁸

¹⁶ Annual and quarterly environmental monitoring reports of the Idaho National Laboratory and surrounding communities is available at http://www.idahoesser.com/Publications_surveillance.htm as the Department of Energy funded and overseen Idaho National Laboratory Site Environmental Surveillance, Education, and Research Program. Some charts are edited to reduce clarity but charts using raw data show significant gaps in monitoring airborne gross alpha and gross beta the graphs available by community.

¹⁷ Annual and quarterly environmental monitoring reports of the Idaho National Laboratory and surrounding communities http://www.idahoesser.com/Publications_surveillance.htm

¹⁸ Tami Thatcher, Environmental Defense Institute, *Radiological and Chemical Exposures at the INL That Workers May Not Have Known About*, April 2017. <http://www.environmental-defense-institute.org/publications/Radchemreport.pdf>

I have to think that the nice folks who make such efforts to cover up the extent of and the source of the radionuclide contamination in Idaho just don't understand enough about the health effects. But it makes it all the more imperative that citizens do learn about the health effects and press state officials to better monitor the releases and seek to reduce future releases. Citizens also need to press for treatment of drinking water for themselves and their children.

Gamma Spectrometry – Deconvolution of Multiple Decay Series Behind the Scenes

In a 1981 document describing gamma spectrometry, the necessary process of deconvolution of one decay series from another is described. And as with sausage, sometimes you will find it more palatable if you don't know what goes into the sausage. Perhaps that is why we are not allowed to know what is in the libraries actually used during the lung count process at the Idaho National Laboratory. The public might be interested in the “expected” and ever changing elevated contaminated background levels of various radionuclides that are subtracted from lung count results.

In addition to ever changing background levels, it seems that for as long as lung counts have been conducted, lung count operators have been selecting gain values via the software — to address the “noise” as one Rocky Flats lung count operator described it. The tweaking of gain settings is typically not available on lung count results but could be manipulating the results.

Table 9 below presents a very limited set of radionuclides and corresponding prominent energy peaks. Let's say you are told that your lung count results don't show any contamination above expected background levels of radionuclides. Yet your lung count report shows a hand written explanation that lead-214 (Pb-214) must be the reason for the program not being able to identify the radionuclides present using the program used to conduct your lung count.

Why would normal expected levels of the uranium decay series raise flags in the gamma spectrometry lung counting report if the levels were due to the normal expected levels of background that they frequently calibrate to? And you know that you were exposed to very high levels of the uranium decay series along with various man-made transuranics.

Figure 1 depicts a very limited range of energy levels from 200 to about 400 keV on the horizontal axis to show how uranium and thorium spectra overlap. The vertical axis is count rate. The width of the peaks as well as peak shape and the center of the peaks are typically within roughly plus or minus 1 keV on either side of the center of the peak.

Table 9. Average background count rates at selected energies.¹⁹

Radionuclide	Energy (keV)	Area (counts)	Error (%) Area	Count Rate (cpm)	Count Rate (cps)
Uranium-238 series					
Th-234	63.3	468	31.20	1.11E-2	1.85E-4
Th-234	92.8	1395	8.85	3.32E-2	5.5E-4
Pa-234m	1001.04	?			
Ra-226	185.7	997	11.85	2.37E-1	3.95E-3
Pb-214	295.16	?			
Pb-214	351.8	670	14.25	1.60E-1	2.7E-3
Bi-214	609.4	378	11.95	9.06E-2	1.5E-3
Bi-214	1238.2	52	26.0	1.24E-2	2.1E-4
Pb-210	46.5	250	26.90	5.9E-2	9.8E-4
Thorium-232 Decay Series					
Bi-212/ Ac-228	727.3	100	34.0	2.38E-2	3.97E-4
Ac-228	911.1	306	12.25	7.29E-2	1.2E-3
Ac-228	988.1	179	13.90	4.26E-2	7.1E-4
Pb-212	238.6	1368	7.50	3.26E-1	5.4E-3
Tl-208	510.7	5692	7.95	1.36	2.27E-2
Ra-224	240.98	?			
Uranium-235 Decay Series					
U-235	185.7	997	11.85	2.37E-1	3.95E-3
Th-231	163.16	?			
Pa-231	302.64	?			
Th-227	235.9	?			
Fr-223	49.8	?			
Ra-223	269.41	?			
Ra-219	271.20	?			
Pb-211	404.89	?			
Bi-211	351.8	670	14.25	1.60E-1	2.7E-3
Uranium-233 Decay Series (keV from ANL Contaminant Fact Sheets)					
Am-241	59.5	?			
Am-243	56.0	?			
Pa-233	200	?			
Np-237	35	?			
Cm-243	130	?			
Cm-245	96	?			
Cm-247	320	?			

Table notes: Counting time of 70 hours. Gamma ray energy levels in kiloelectron Volts in the table are not the newest or most refined keV values. Each nuclide has other peaks but the table includes only those highest in abundance and detector efficiency generally above 50 keV. Note that many gamma energies overlap which requires deconvolution to estimate the prominent nuclide contributors to the counts at that energy level. Reference: ANL/ES-118, September 1981.

¹⁹ Gamma ray energy levels in kiloelectron Volts in the table are not the newest or most refined keV values. Michael H. Momeni, "Analysis of the Gamma Spectra of the Uranium, Actinium, and Thorium Decay Series," ANL/ES-118, September 1981. <http://www.iaea.org/inis/collection/NCLCollectionStore/Public/13/663/13663724.pdf>

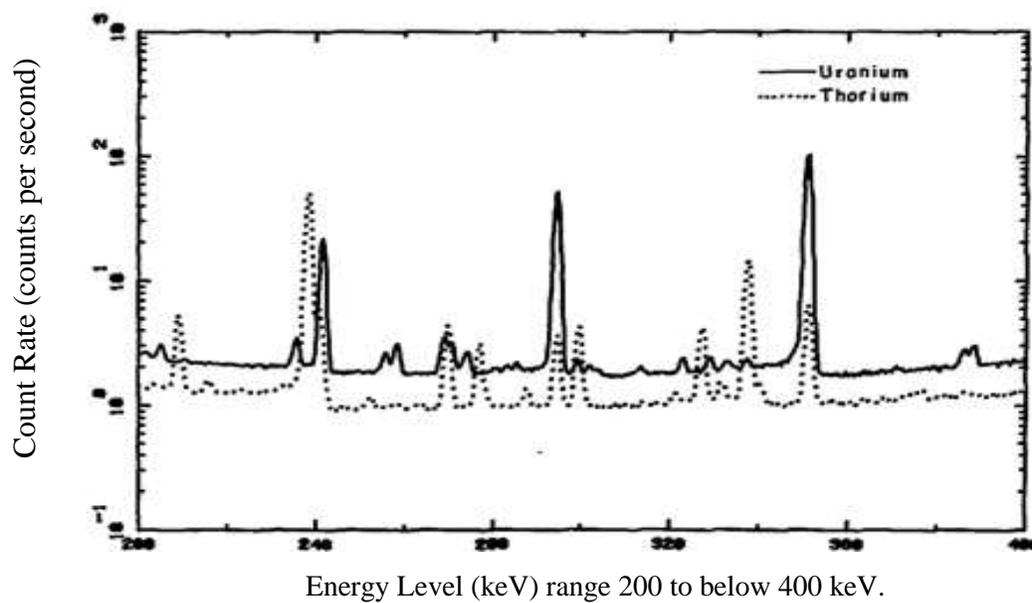


Figure 1. An example of gamma spectra of uranium and thorium ores between 200 and 400 keV. See this and other energy ranges from 40 to 1400 keV in ANL/ES-118 or other documents.

The various prominent peaks result from the elevated count rate (in counts per second or counts per minute) at specific energy levels (in keV). This allows the known energy levels for various nuclides to be recognized by known tendency for gamma emission at or very near that energy level. The typical range evaluated by gamma spectrometry ranges from 40 keV to 1400 keV.

See our January 2018 EDI newsletter for more information about lung counting.²⁰

Articles by Tami Thatcher for February 2018.

²⁰ Environmental Defense Institute newsletter for January 2018. <http://www.environmental-defense-institute.org/publications/News.18.Jan.pdf>