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Bioaccumulation Factor for Radium in Rio Grande Biota

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1 Purpose

The Los Alamos National Laboratory (LANL or Laboratory) Environmental Protection and Compliance Division's Environmental Stewardship Group (EPC-ES) performs the radiological analysis portion of biota dose assessments using the computer program RESRAD-Biota (DOE 2004, DOE 2020). RESRAD-Biota uses a default bioaccumulation factor, B_{iv}, for radium in aquatic animals. This bioaccumulation factor, also listed in Department of Energy (DOE) Standard, DOE-STD-1153-2019, is the ratio of a radionuclide concentration in fresh weight biota to the environment in which it resides, which for aquatic animals is water. DOE-STD-1153-2019 offers conservative values for these ratios that are specific to the radiological contaminant, the biota, and the environment. The B_{iv} in RESRAD-Biota can be used for general screening (Level 1 screening). If the factor proves to be too conservative, then a site-specific B_{iv} may be used. The biota dose assessment of fish from the northern New Mexico region of the Rio Grande fail the general screen, which indicates that further analysis is necessary. Level 2 analysis is site-specific but still has conservative limitations. Level 3 analysis is a more realistic and representative, site-specific analysis.

The purpose of this paper is to use data specific to the Rio Grande to calculate a new aquatic animal radium B_{iv}. This B_{iv} can then be used for Level 3 analysis of aquatic animals specifically in the northern New Mexico region of the Rio Grande.

The calculated radium B_{iv} is a useful value for performing detailed, accurate biota dose assessments and calculating dose to fish using RESRAD-Biota when only sediment data are available.

2 Context

2.1 Biota Dose Assessments

Few aquatic locations exist near the Laboratory besides the Rio Grande. Every 3 years, the Rio Grande is a focus of investigations by the Soil, Foodstuffs, and Biota (SFB) Program; for example, it was the focus of the investigations during 2014, 2017, 2020, and 2023. The SFB Program collects samples for analysis of various radionuclide and chemical concentrations in soil, food, and biota. SFB data are then used to conduct biota dose assessments.

2.2 DOE Regulations

Biota dose assessments are mandated by U.S. Department of Energy (DOE) Order (O) 458.1. At LANL, these assessments are published in the Annual Site Environmental Reports (ASERs), (e.g., LANL 2014; LANL 2017; LANL 2020; LANL 2023), and they are supported by the technical documents referred to in the ASERs. DOE O 458.1 states explicitly that biota dose assessments can use "the RESRAD family of codes," which includes RESRAD-Biota (DOE 2004, DOE 2020).

The DOE technical standard, 1 rad/d for aquatic animals, is listed in DOE-STD-1153-2002 and -2019 and is used as the dose limit value in RESRAD (DOE 2002; DOE 2019).

2.3 RESRAD Default Value Calculations

This assessment uses RESRAD-Biota version 1.8, which is the most recent release at the time of writing. The following example in Table 1 and Table 2 uses RESRAD-Biota to clarify the calculations and to show the difficulty with radium. This example is equivalent to a Level 1 (screening level) analysis of Ra-226 for aquatic animals; however, the example requires selecting RESRAD's Level 3 analysis so that riparian animals can be removed from the model, as they will not be considered in this paper.

Table 1. Inputs to RESRAD-Biota Level 1 (Screening Level) Aquatic Animal Assessment

Step	Inputs
1.	Select Ecosystem "Aquatic," Level 3, Units "Traditional."
2.	De-select "Riparian Animal" under "Organism."
3.	Add Ra-226 and remove Am-241; enter the sediment value = 1 pCi/g, which is a typical value for natural radium and for each radionuclide in the uranium decay chain.
4.	Un-check the box next to "Water"; you should see "Kd: 70 mL/g." The calculated water concentration is 1/70 pCi/mL or 1000/70 pCi/L, which is 14.2857142 pCi/L.
5.	Click "Run" near the top-right corner. A new box will appear. The numbers in the top row should be 1.40 with a red background, which means that the assessment has failed by a factor of 1.40.
6.	To see more details, select the "Dose Rate" tab at the top. Note that the aquatic-animal dose rate is 1.4 rad/d and is mostly "Internal Dose." Select the "Tissue Report" button to view the calculated radium concentration in fish tissue, which is 45.7 pCi/g.
7.	To see even more details, click "Close" on this box and then click "Edit" on the original box. Note the values of the dose conversion factor, "DCF." Also note that only water is selected as a contributor to ingestion dose. Click the "Input" tab and the "BIV" tab and note the values of the bioaccumulation factor, "BIV."

Table 2. RESRAD-Biota Example Screening Level Calculations for Aquatic Animals

Sediment	1	pCi/g	Typical value for natural radium
Radium K_d	70	mL/g	From Table 4.3 page M3-52 of DOE-STD-1153-2002
Water	14.2857142	pCi/L	RESRAD-Biota shows the result with units pCi/L
Radium B_{iv}	3200	mL/g	From Table F-1, page F-4 of DOE-STD-1153-2019. Organism-to-water ratio for aquatic animals
Tissue	45.7	pCi/g wet	Aquatic animal (= water · B_{iv})
Radium DCF (per year)	11.2	(rad/y)/(pCi/g)	Divide by 365d/y to get rad/d in next row
Radium DCF (per day)	0.031	(rad/d)/(pCi/g)	Same as Table E-2 page E-9 of DOE-STD-1153-2009
Dose rate	1.4	rad/d	Aquatic animal total dose rate (= tissue · DCF)

K_d: sediment-to-water partition coefficient
DCF: dose conversion factor

The calculated dose rate in Table 2 agrees with the RESRAD-Biota dose rate. The extra information in this table (not calculated in RESRAD-Biota) is the tissue concentration for aquatic animals (fish).

DOE-STD-1153-2019 and RESRAD-Biota use conservative default B_{iv} values (DOE 2019). The default Ra-226 and Ra-228 bioaccumulation factor was chosen from bioaccumulation factors in the literature for different aquatic organisms; B_{iv} = 3200 mL/g came from measurements of freshwater Gammarus, a crustacean, reported by Till and Meyer (DOE 2019; Till and Meyer 1983). The calculated wet-weight tissue concentration for aquatic animals in Table 2 is 46 pCi/g, which is much larger than the measured tissue data for fish in the Rio Grande. It is too large because the bioaccumulation factor, B_{iv}, is too large. The default B_{iv} value must be replaced by a site-representative value. RESRAD-Biota's model for aquatic animals assumes that radionuclide uptake comes only from water, not soil or sediment, as shown in Step 7 of Table 1; therefore, only a new organism-to-water bioaccumulation factor needs to be derived.

In addition to the B_{iv} value, two other variables should be changed for running a Level 3 RESRAD-Biota analysis. First, the default sediment/water distribution coefficient, K_d, value for radium is too large because RESRAD-Biota uses the DOE-STD-1153-2002 K_d value. In the current standard, DOE-STD-1153-2019, the K_d for Ra-226 and Ra-228 was updated to 1,000 mL/g for deposited sediment. Second, the radiation weighting factor (treated as the relative biological effectiveness [RBE] in RESRAD-Biota) should be changed to 10 for alpha particles, as advised by DOE-STD-1153-2019 (page B-1).

3 Procedure for Calculating and Using B_{iv}

3.1 Data

Environmental samples are collected by LANL staff, sent to an outside analytical laboratory for measurement, and then the results are uploaded into the Laboratory's publicly accessible environmental database, Intellus (https://intellusnm.com/). For this paper, a subset of the Ra-226 and Ra-228 measurements in Intellus was used.

3.1.1 *Procedure for Using Intellus*

To pull data from Intellus, navigate to https://intellusnm.com/ and use the "quick search" under the "search data" dropdown. Users must answer seven questions to get the type of data they want. For the current study, questions were answered as shown in Table 3. The following sections discuss what data were used from Intellus search results and why.

Table 3. Intellus Quick Search Inputs

Question	Input
Data provider	Los Alamos National Laboratory
Type of data	Analytical results
Sample types	For fish data: select "biota," then when asked "what type of biota?" choose "this type includes all fish types" and "animal." Note: When the sample type is recorded as "animal," the "field sample comments" data column will usually describe what kind of animal, allowing for later removal of non-fish samples.
	For soil and sediment data: Select "solid" without specifying anything in the "what type of solid?" dropdown.
	For water data: Select "water" without specifying anything in the "what type of water?" dropdown.
Dates	1942–present to include all sample collection years
Locations	Everywhere in Los Alamos area
Analytical parameters	Choose "select parameter(s) from a list." For "select parameters by," make sure "individual parameter" is chosen. Include Ra-226 and Ra-228.
Data columns	Add "report uncertainty" and "% moisture" to the selected parameters.

3.1.2 *Fish Samples*

Fish have been collected by LANL since the 1970s for analysis of various radionuclides. Starting in 2013, the SFB team at LANL included Ra-226 and Ra-228 in the list of radionuclides requested for analysis in fish samples. Fish were collected in 2013 and then triennially starting in 2014, during late spring, summer, and early fall. Samples were collected in Abiquiu Reservoir, in Cochiti Reservoir, and in the Rio Grande between these two reservoirs; see Figure 1.

When deciding which data to use for this evaluation, the *report basis* and the *% moisture* variables were considered. The report basis is the amount of analyte per gram of either wet, dried, or ashed sample, where “wet” refers to analyzing the sample as is. Knowing the basis is necessary in order to convert analyte concentrations to wet-weight concentrations, using conversion ratios reported in the literature. Alternately, the % moisture variable, which represents the amount of material removed if drying or ashing were performed before determining analyte concentrations, provides a way to convert radium concentrations to wet-weight concentrations. If “basis” is selected as a data column in Step 7 from Table 3, this designation is present in all fish samples since 2013; however, review of detailed data package and discussions with Stas Marczak from LANL’s Sample Management Office and the analytical laboratory staff indicate that the basis is usually incorrect. The % moisture variable, present only in 2023 samples, was therefore considered necessary in order to use fish results. For this evaluation, samples from only 2023 are used.

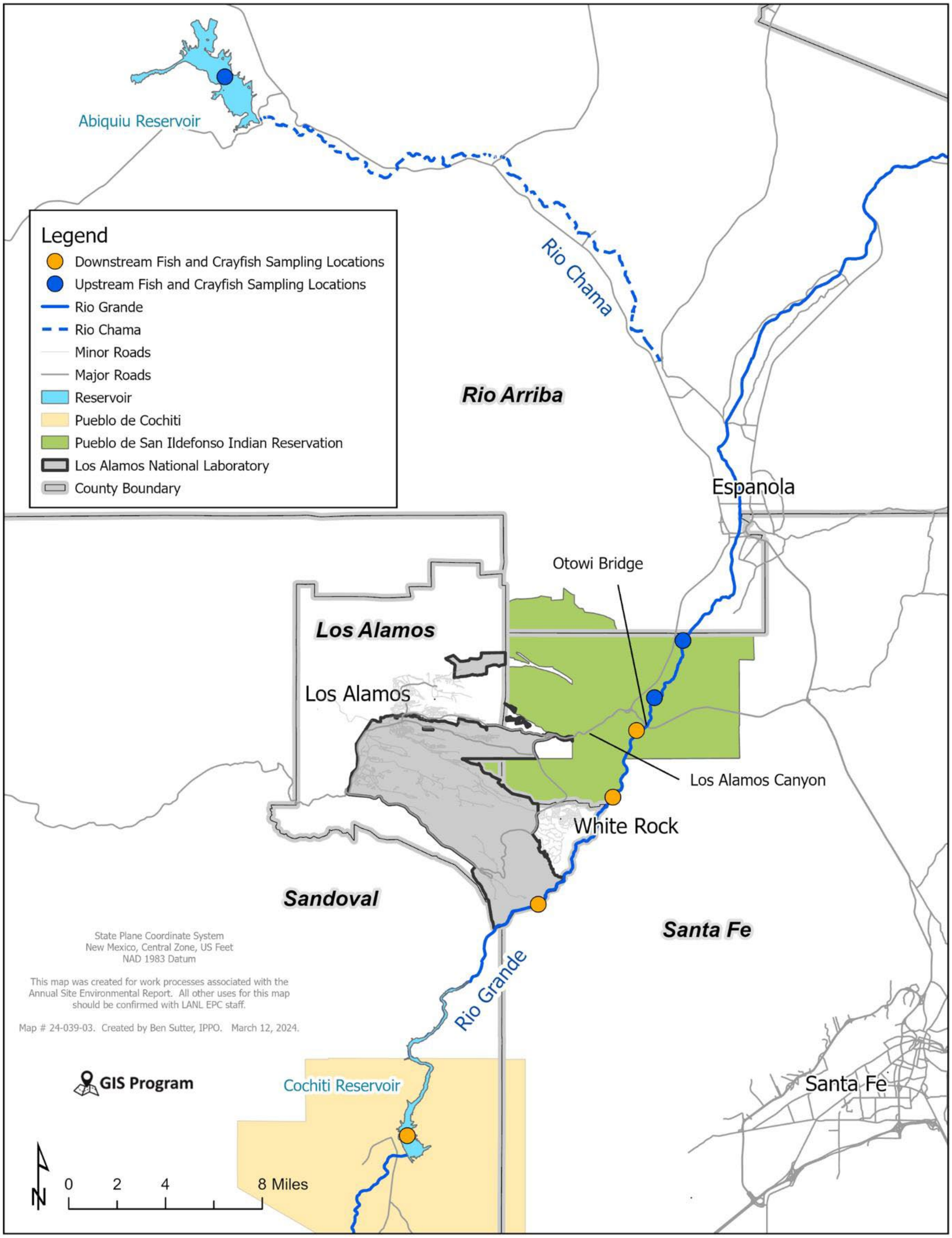

Figure 1: Fish and crayfish collection locations upstream and downstream of the Laboratory in 2023. (LANL 2023)

In 2023, 10 fish samples were analyzed from Cochiti Reservoir. The analytical laboratory initially reported Ra-226 concentrations for seven of the fish samples that were an order of magnitude smaller than the other three fish samples from Cochiti. The analytical laboratory dilutes samples in liquid before analysis; the original analysis for Ra-226 data inadvertently used diluted sample masses, which made the concentrations appear to be much smaller than they actually were. The Ra-226 concentrations for these seven fish samples were revised by the analytical laboratory using correct sample masses and updated in Intellus in November 2024. The corrected concentrations were used in this paper.

The final sample size for the fish dataset was n = 40, where each sample was analyzed for both Ra-226 and Ra-228. Table 4 lists characteristics of the sample set, such as number of each fish species; Figure 1 shows where fish were collected.

Table 4. Fish Dataset Characteristics for Ra-226 and Ra-228

Number of Fish in Each General Collection Area		Number of Fish in Each Feeding Category		Number of Each Fish Species	
Abiquiu	10	Predators	7	Crappie	1
				Small mouth bass	3
				Walleye	3
		Bottom feeders	3	Carp	1
				Catfish	1
				White sucker	1
Cochiti	10	Predators	4	Pike	3
				Walleye	1
		Bottom feeders	6	Carp	3
				Catfish	3
Upstream of Los Alamos Canyon	10	Bottom feeders	10	Carp	2
				Channel Catfish	7
				White Sucker	1
Downstream of Los Alamos Canyon	10	Bottom feeders	10	Blue catfish	1
				Channel catfish	9

LAC = Los Alamos Canyon

3.1.3 *Sediment and Soil Samples*

A search of Intellus for soil and sediment samples in and around the Rio Grande yielded Ra-226 and Ra-228 measurements as far back as 1993.

Many of the Ra-226 measurements were done using gamma spectroscopy; these samples were excluded from the dataset. Gamma spectroscopy measurements should be examined to see what gamma-ray energy was used. As discussed by McNaughton and Patel, if the 186.2 keV peak of Ra-226 is used, there will be spectral interference between that peak and the 185.7 keV gamma emission of any U-235 present. Measuring the 609.3 keV gamma peak of Bi-214 gives the activity of Ra-226 but only if samples are sealed long enough so that daughter products, including Bi-214, come into secular equilibrium with Ra-226 (McNaughton and Patel 2010). The Ra-226 soil and sediment dataset size of 79 samples was considered to be large enough despite excluding measurements done using gamma spectroscopy. Therefore, all measurements included in the dataset used Environmental Protection Agency method 903.1 for counting Ra-226.

Only samples taken by the SFB team from the bottom of Abiquiu and Cochiti and within the active channel of the Rio Grande were used; sediment collection descriptions appear in Chapter 7 of the relevant ASERs (LANL 2014; LANL 2017; LANL 2020; LANL 2023). The reason both soil and sediment samples were included is that, regardless of the soil or sediment designation, deposited material collected from the bottom of the Rio Grande, Abiquiu Reservoir, and Cochiti Reservoir was deemed relevant to uptake into fish.

The final soil and sediment sample size was n = 79, where each sample was analyzed for both Ra-226 and Ra-228. Table 5 lists characteristics of the sample set, including collection years. Figure 2 shows sample locations. Radium concentrations in soil and sediment were reported on a dry-weight basis.

Table 5. Number of Soil and Sediment Samples Collected by General Area and Year

Collection Year	Abiquiu	Cochiti	Upstream of LAC	Downstream of LAC	All Locations
2014	5	5	7	8	25
2017	–	–	7	8	15
2020	6	6	7	8	27
2023	6	6	–	–	12

LAC = Los Alamos Canyon

ASERs published by the Laboratory refer to sample locations along the Rio Grande as being either upstream or downstream of the point where Los Alamos Canyon (LAC) drains into the Rio Grande. This distinction is useful because comparing upstream and downstream locations allows for determining concentrations of chemicals or radionuclides contributed by LANL. For the same reason, it is useful to compare samples from Abiquiu Reservoir and Cochiti Reservoir. One soil and sediment collection location used in this paper was incorrectly labeled as a downstream Rio Grande location in 2014 and 2017 samples (sample IDs began with BMI-RGDS) and labeled as an upstream location in 2020 samples (sample IDs began with BMI-RGUS). All samples at this location will be treated as upstream samples here.

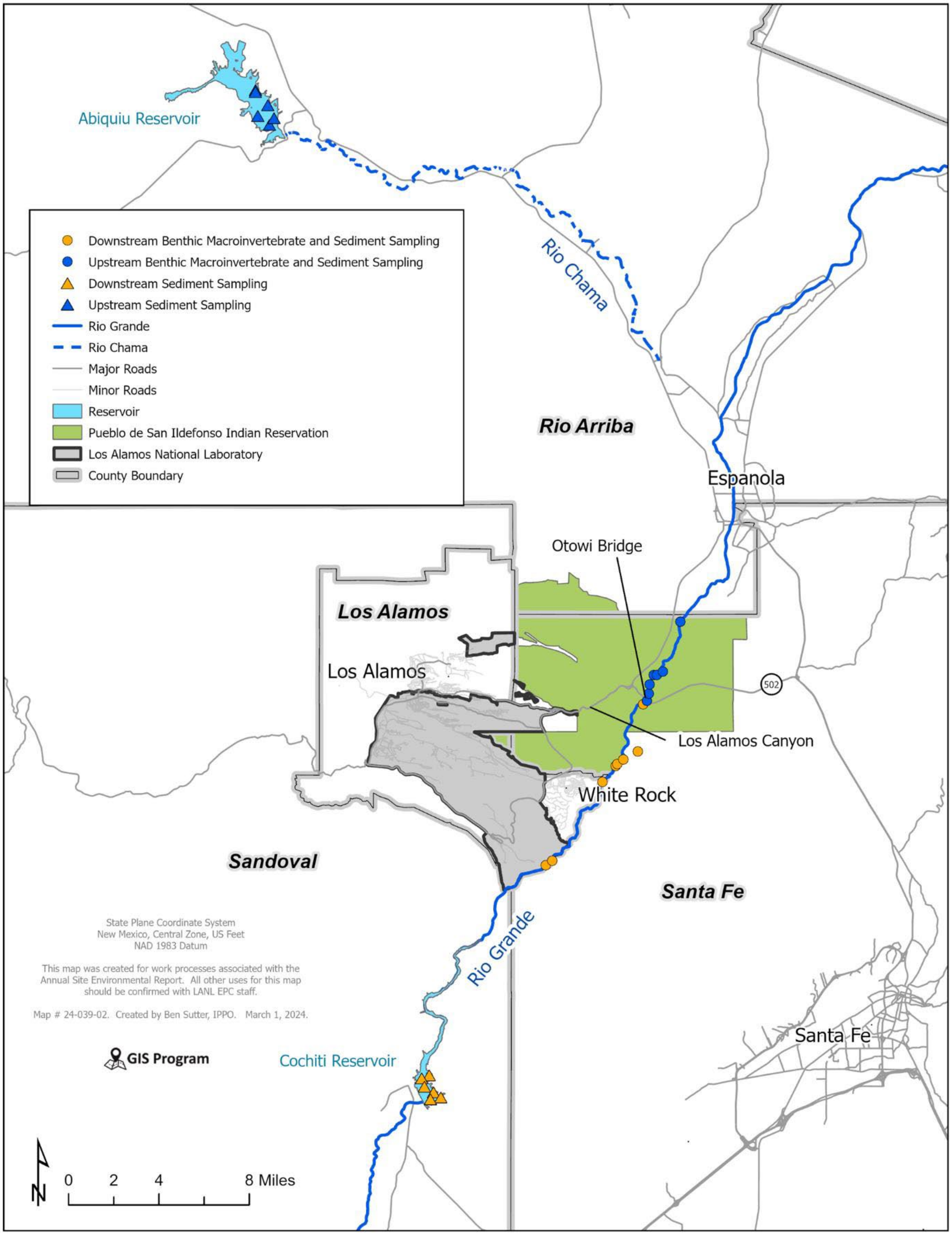

Figure 2: Sediment and benthic macroinvertebrate sample collection locations upstream and downstream of the Laboratory in 2023. (LANL 2023)

Ra-226 is part of the U-238 decay chain; Ra-228 is part of the Th-232 decay chain. Both of these decay chains are naturally occurring in the environment. Soil and sediment samples taken by the SFB team from the bottom of Abiquiu and Cochiti and within the active channel of the Rio Grande were measured for many radionuclides, including several from the U-238 and Th-232 decay chains. Measurements of radionuclides in the U-238 and Th-232 decay chains, including Ra-226 and Ra-228, were investigated for indication that the Ra-226 and Ra-228 in the samples were naturally occurring (discussed in Section 3.6, Assessing Soil and Sediment Samples for Secular Equilibrium). Most of these measurements were performed on the same soil and sediment samples as those in the radium dataset. Collection locations were the same as those in the radium dataset, and the range of collection years was the same. If the radionuclides within each decay chain are in secular equilibrium in soil and sediment, it indicates that the radium in the area is natural (DOE-STD-1153-2019, page F-12). The reason no Ra-226 and Ra-228 data were downloaded and assessed for secular equilibrium in fish is that bioaccumulation varies by element (see Appendix F, Table F-2, of DOE-STD-1153-2019 for default bioaccumulation values of different radionuclides in aquatic animals). This variation leads to different relative amounts of each radionuclide in a decay chain. Similarly, Ra-226 and Ra-228 will not be assessed for secular equilibrium in water samples because solubility varies for each radionuclide in their decay chains. (See Appendix D of DOE-STD-1153-2019 for K_d values of radionuclides.)

3.1.4 *Water Samples*

A search of Intellus for water samples in and around the Rio Grande yielded Ra-226 and Ra-228 measurements going back to 1965.

This analysis does not include storm water—which often has high amounts of suspended sediment—spring, well, or ground water. For reasons discussed in Section 3.1.3, Sediment and Soil Samples, gamma spectroscopy measurements of Ra-226 were not included. Finally, only samples taken within the Rio Grande, Abiquiu, and Cochiti were used.

The final water sample size was n = 28, where each sample was analyzed for both Ra-226 and Ra-228. These samples were designated as base flow from two Rio Grande locations, one upstream and one downstream of LAC. The upstream water collection location is at the point marked “Otowi Bridge” in Figure 1. The downstream water collection location is about 0.1 miles downstream of the fish sample location above the words “White Rock” in Figure 1. Table 6 lists characteristics of the sample set, including collection years.

Table 6. Water Dataset Characteristics for Ra-226 and Ra-228

Number of Samples per Year		Number of Samples per Location		Number of Filtered and Unfiltered Samples	
2008	11	Upstream Rio Grande	4	Filtered	2
				Unfiltered	2
		Downstream Rio Grande	7	Filtered	3
				Unfiltered	4
2009	11	Upstream Rio Grande	7	Filtered	2
				Unfiltered	5
		Downstream Rio Grande	4	Filtered	2
				Unfiltered	2
2010	6	Upstream Rio Grande	4	Filtered	2
				Unfiltered	2
		Downstream Rio Grande	2	Filtered	1
				Unfiltered	1

3.2 Analysis

Table 7 lists descriptive statistics for Ra-226 and Ra-228 concentrations in fish samples. These concentrations are not based on whole-body measurements. Fish were filleted, and the non-fillet parts of the fish—including viscera for the Abiquiu and Cochiti samples—were sent to the analytical laboratory for radionuclide analysis, since the muscle was used for measurements of other analytes (LANL 2023, page 7-29). Some skin was present on samples analyzed for radionuclides (Gaukler 2024). The liver was removed from Abiquiu and Cochiti fish viscera in all but two Cochiti samples, and the head was usually not included in radionuclide measurements, except for a few Rio Grande fish, as described in chain-of-custody documents (Gaukler 2024) and the "Field Sample Comments" data column. Radium concentrations are expected to be higher in fish bones and viscera (IAEA 1990), so the concentrations in Table 7 will be used here as conservative measures of whole-body radium.

Table 7. Radium Concentrations in 2023 Fish Samples (pCi/g)

	Abiquiu	Cochiti	Upstream of LAC	Downstream of LAC	All Locations
Ra-226					
Average ash-weight concentration	0.67	0.49	1.2	1.0	0.85
1 Std Dev	0.26	0.17	0.92	0.98	0.72
Average wet-weight concentration	0.030	0.018	0.048	0.035	0.033
1 Std Dev	0.020	0.008	0.039	0.043	0.032
Ra-228					
Average ash-weight concentration	1.4	1.6	0.81	0.52	1.1
1 Std Dev	1.1	1.8	0.97	0.91	1.2
Average wet-weight concentration	0.054	0.062	0.039	0.013	0.042
1 Std Dev	0.043	0.070	0.056	0.029	0.053

Table 8 lists descriptive statistics for Ra-226 and Ra-228 concentrations found in water, soil, and sediment samples. The average radium concentration was the same for filtered and unfiltered water measurements within 1 standard deviation; therefore, filtered and unfiltered water measurements were averaged together without a correction for the contribution of radium activity from suspended sediment in unfiltered water samples.

Table 8. Radium Concentrations in Samples of Water (pCi/L), Soil, and Sediment (pCi/g)

Radionuclide	Sample Matrix	Average	1 Std Dev
Ra-226	Water: both filtered and unfiltered	0.32	0.26
	Soil and Sediment	1.1	0.55
Ra-228	Water: both filtered and unfiltered	0.42	0.26
	Soil and Sediment	0.88	0.48

3.2.1 *B_{iv} Calculation*

The wet-weight bioaccumulation factor in fish is calculated as

$$bioaccumulation\ factor = \frac{concentration\ of\ analyte\ in\ wet\ fish}{concentration\ of\ analyte\ in\ water} \quad .$$

Table 9 shows inputs for an example B*iv* calculation; it is for a Ra-226 measurement of a catfish collected in Cochiti in 2023.

Table 9. Data for Example Ra-226 B_{iv} Calculation

% Moisture	K_d	Ra-226 Concentration in Cochiti Ash-Weight Catfish in 2023 (pCi/g)	Average Ra-226 Concentration of Soil and Sediment Dataset* (pCi/g)
96.1	$\left(1000 \frac{\frac{pCi}{g\ sediment}}{\frac{pCi}{mL\ water}}\right) = 1000 \frac{mL\ water}{g\ sediment}$.	0.504	1.1

* The average of all soil and sediment data was used to calculate individual B_{iv} values in this paper; however, when Rio Grande sediment data are collected, the average value at a specific location can be used along with the final B_{iv} value in this paper to perform calculations at that particular Rio Grande site. (See Section 3.2.2, Using New B_{iv} in RESRAD, for an example of this method.)

Calculation of a bioaccumulation factor for the catfish in Table 9 is as follows. Ash-weight concentration of radium is converted to wet-weight using % moisture:

$$\left(0.504 \frac{pCi}{g\ ash\ fish}\right)\left(\frac{(1-0.961)\ g\ ash\ fish}{g\ wet\ fish}\right) = 0.0197 \frac{pCi}{g\ wet\ fish} \quad . \tag{1}$$

Average Ra-226 concentration in all soil and sediment samples is converted to concentration in water using K_d = 1000 mL/g (refer to Appendix D of DOE-STD-1153-2019):

$$\frac{\left(1.1 \frac{pCi}{g\ sediment}\right)}{\left(1000 \frac{mL\ water}{g\ sediment}\right)} = 1.1 \cdot 10^{-3} \frac{pCi}{mL\ water} \quad . \tag{2}$$

The outputs of equations (1) and (2) are used to get the bioaccumulation factor:

$$B_{iv} = \frac{\left(0.0197 \frac{pCi}{g\ wet\ fish}\right)}{\left(1.1 \cdot 10^{-3} \frac{pCi}{mL\ water}\right)} = 18 \frac{mL\ water}{g\ wet\ fish} = 18 \frac{L\ water}{kg\ wet\ fish} \quad . \tag{3}$$

Table 10 uses the same calculation method as above to find the Ra-226 and Ra-228 B_{iv} for individual fish samples before averaging by general fish collection area.

Table 10. Average Fish B_{iv} for Ra-226 and Ra-228 by Collection Area (L water)/(kg wet fish)

	Abiquiu	Cochiti	Upstream of LAC	Downstream of LAC	All Locations
Ra-226					
Average	27	16	44	32	30
1 Std Dev	18	7.7	35	39	29
Ra-228					
Average	61	70	44	15	48
1 Std Dev	49	79	64	33	60

The Ra-226 and Ra-228 B_{iv} for all locations is the same within 1 standard deviation of uncertainty; therefore, the B_{iv} values were averaged, giving a single radium B_{iv} of 39 L/kg, with 1 standard deviation of 66 L/kg. To derive a B_{iv} that leads to conservative calculations of radium concentrations in fish, use the mean plus 2 standard deviations, or 170 L/kg.

Table 11 compares measured versus predicted wet-weight radium concentrations in fish. Measured values come from converting ash-weight concentrations to wet-weight concentrations as shown in Equation (1). Predicted values come from multiplying B_{iv} = 170 L/kg by the average radium water concentration at a given location, where the water concentration was calculated using Equation (2). Note that predicted radium concentrations in fish tissue were 2 to 10 times larger than the measured values. In contrast, the default RESRAD B_{iv} leads to calculated tissue concentrations that are 45 to 250 times the measured tissue concentrations in Table 11. Both the default B_{iv} and the B_{iv} calculated here are too large and give fish radium concentration predictions that are too large; however, the B_{iv} calculated here is conservative while still giving a tissue concentration that is within about a factor of 10 of our data average. Therefore, the new B_{iv} is an improvement for the stretch of the Rio Grande in northern New Mexico because it is more accurate while still being conservative.

Table 11. Measured and Predicted Radium Concentrations in Wet-Weight Fish (pCi/g)

	Abiquiu	Cochiti	Upstream of LAC	Downstream of LAC	All Locations
			Ra-226		
Measured concentration	0.030	0.018	0.048	0.035	0.033
Predicted concentration	0.21	0.24	0.16	0.14	0.18
			Ra-228		
Measured concentration	0.054	0.062	0.039	0.013	0.042
Predicted concentration	0.20	0.15	0.12	0.14	0.15

3.2.2 Using New B_{iv} in RESRAD

As in the RESRAD default value calculations, the example in Table 12 and Table 13 uses RESRAD-Biota version 1.8. To clarify the calculations for a site-specific analysis, this example uses the scenario of Ra-226 at Cochiti in 2023.

Table 12. RESRAD-Biota Example Inputs for Cochiti Fish in 2023

Step	Inputs
1.	Select Ecosystem "Aquatic," Level 3, Units "Traditional."
2.	De-select "Riparian Animal" under "Organism."
3.	Add Ra-226 and remove Am-241; enter the sediment value = 1.83 pCi/g, which was the average value in Cochiti sediment in 2023.
4.	Un-check the box next to "Water"; you should see "Kd: 70 mL/g." Change the K_d value to 1,000 mL/g.
5.	In the bottom right corner, change the alpha RBE to 10.
6.	Under "Organism," click "Edit," "Input," "BIV"; change the water BIV to the value found for the Rio Grande: 170 mL/g (in the "Water" column).
7.	Click "Close" on this box and "Run" near the top-right corner. A new box will appear. "Total" in the top row should be $4.92 \cdot 10^{-3}$ with a white background, which means the assessment has succeeded. The "Total" indicates that the Ra-226 concentration is only 0.492% of the limit.
8.	To see more details, select the "Dose Rate" tab at the top. Note that the aquatic animal dose rate is $4.92 \cdot 10^{-3}$ rad/d and is mostly "Internal Dose."

Table 13. RESRAD Example Calculations for Ra-226 in Cochiti in 2023

Input	Value	Unit	Description
Sediment	1.83	pCi/g	Average value in Cochiti sediment in 2023
Radium K_d	1,000	mL/g	From Table D-1 page D-2 of DOE-STD-1153-2009
Water	1.83	pCi/L	Water value for K_d equal to 1,000 mL/g
Radium B_{iv}	170	mL/g	Calculated organism-to-water ratio for aquatic animals
Tissue	0.311	pCi/g wet	Aquatic animal (= water · B_{iv})
Radium DCF (per year)	5.62	(rad/y)/(pCi/g)	RESRAD internal DCF for Ra-226 when alpha RBE = 10
Dose rate	$4.92 \cdot 10^{-3}$	rad/d	Aquatic animal total dose rate (RESRAD calculated)

Note that the dose rate in Table 13 ($4.92 \cdot 10^{-3}$ rad/d total dose as calculated by RESRAD) is slightly larger than the internal dose from multiplying (tissue Ra concentration) × (internal DCF), which gives $4.79 \cdot 10^{-3}$ rad/d. The internal dose is small enough in comparison with the external dose calculated by RESRAD-Biota that there is a non-negligible contribution from external dose of about 3%.

3.3 Discussion

The calculated tissue concentration for aquatic animals in Table 13 is 0.311 pCi/g, which is larger than the average of 2023 Cochiti wet-weight fish data (0.018 pCi/g) by about a factor of 17. In contrast, the default RESRAD B_{iv} leads to a calculated tissue concentration of 46 pCi/g, which is about 2,600 times the average of 2023 Cochiti fish data.

The 2014 ASER explains why B_{iv} is much smaller than the RESRAD default, although it disagrees with the bioaccumulation factor calculated here. It was noted that,

> The radium data show that the bioaccumulation factor is less than 1.0. Radium is chemically similar to calcium, and fish take up radium when they cannot get enough calcium (Eisenbud and Gesell 1997). However, calcium is abundant in the Rio Grande, so this relatively small bioaccumulation factor is reasonable (LANL 2014).

The B_{iv} estimate in the 2014 ASER, < 1 mL/g, disagrees with the B_{iv} calculated here in large part due to K_d. If this paper had used the older K_d = 70 mL/g from DOE-STD-1153-2002, the calculated B_{iv} before adding 2 standard deviations would have been 3 mL/g, which agrees more closely with the 2014 estimate. Another variable that contributes greatly to the disagreement in bioaccumulation factors is the radium concentration reported in fish. Comparing original data from Intellus, the fish data analyzed in the 2014 ASER were around two orders of magnitude smaller than the 2023 ash-weight fish data. Given the uncertainty in report basis before 2023, smaller radium concentrations in fish could have been due in part to samples being less thoroughly ashed or even reported on a dry- or wet-weight basis.

3.4 Sources of Uncertainty

Radium in Los Alamos can be divided into two categories: background and LANL-made. These two categories each have causes for fluctuations and uncertainties that should be considered. However, as discussed below in Section 3.6, Assessing Soil and Sediment Samples for Secular Equilibrium, radium in the samples used for this paper is likely to be from background, not LANL operations. The LANL radium contribution—because it is indistinguishable from background—is therefore not a variable of concern when analyzing data fluctuations.

The second category, background radium, has many possible causes for variability of measured concentrations in fish and water. We do not know the source of fish, water, or sediment data fluctuations within the Rio Grande (chemical processes, geology, meteorological conditions, type of fish, radium source depending on the season, etc.), so the mean is not well known. Taking 2 standard deviations is therefore the appropriate uncertainty calculation (as opposed to the standard error of the mean, which is used when error sources are understood and the mean can be more accurately calculated). Our final B_{iv} is therefore the average plus 2 standard deviations. Adding 2 standard deviations ensures that our value is conservative (95% upper confidence limit). This decision agrees well with the uncertainty estimations discussed in Section 3.5, Estimating Uncertainty.

3.5 Estimating Uncertainty

3.5.1 *% Moisture*

Fish samples analyzed in 2023—and fish samples in general—are likely dryer than live fish by varying amounts because samples can dry out and lose fluids between the time of collection and analysis, which includes transit time.

Fresquez reported average ash/dry and dry/wet weight conversion ratios for muscle and bones and separately for viscera of fish collected in the Rio Grande. The dataset comprised 251 fish samples; species included bass, carp, carp sucker, catfish, crappie, pike, sucker, trout, and walleye (Fresquez et al. 2007). These species, excluding trout and carp sucker, were all present in the 2023 dataset used in this paper.

Combining all fish species analyzed by Fresquez, the average ash-weight-to-dry-weight ratio for muscle, bone, and viscera was 0.11; the average dry-weight-to-wet-weight ratio was 0.27. This result would indicate a % moisture of around 73% for dried samples and around 97% for ashed samples. The 2023 fish dataset had % moisture values ranging from 92 to 99%, with an average of 96%. These values are most consistent with the Fresquez ash-weight percentage. Comparing the average % moisture in 2023 fish data with Fresquez indicates better than a factor of 2 uncertainty when converting ash-weight analyte concentrations to wet-weight analyte concentrations.

Toppe et al. found average ash weights from ashing bones of several saltwater fish species; values ranged between 264 and 565 (grams ash)/(kg bone) (Toppe et al. 2007), which gives % moisture values between 74 and 44%, respectively. The IAEA referenced measurements of 2.54% ash from ashing muscle and 12% from ashing bone, giving 97.5% and 88% moisture, respectively (qtd. in IAEA 1990). The U.S. Department of Agriculture has measured average amounts of ash from ashing raw fillets of many fish species, ranging from 0.93 to 2.54% ash, which gives % moisture values between 99 and 97% (USDA 2024). These different measurements of the amounts of ash from fish, including the Fresquez measurements stated above, show that % moisture varies greatly in different parts of fish, with much lower percentages in bone. Because the 2023 fish data did not include muscle, there is a higher relative amount of bone than is present in whole-body fish. Therefore, it can be expected that the ash-to-dry conversion using 2023 % moisture values leads to an overestimate of the radium concentration in wet-weight fish, increasing the conservatism in calculated B_{iv} values.

3.5.2 *Radium Concentration in Fish*

With 1 standard deviation of uncertainty, the concentrations of Ra-226 and Ra-228 in fish were 0.85 ± 0.72 and 1.0 ± 1.2 pCi/g, respectively, indicating about a factor of 2 uncertainty. There is additional uncertainty between the whole-body concentration of radium in fish and the 2023 measurements in Intellus, the latter of which are expected to be higher because they pertain to only fish bones and sometimes head and viscera. However, using measurements of only parts of fish with more radium accumulation increases the conservatism of uptake calculations. In addition, the same parts of fish tend to be measured for radium in each triennial SFB sampling event at LANL, so the current B_{iv} is readily applicable to all fish sampled by standard SFB methods.

3.5.3 *Radium Concentration in Water*

The concentration of radium in soil and sediment, within 1 standard deviation, was 1.1 ± 0.6 pCi/g for Ra-226 and 0.88 ± 0.48 pCi/g for Ra-228, indicating less than a factor of 2 uncertainty. The concentration of radium in water, calculated from average sediment concentration of radium and K_d = 1,000 mL/g, was 1.06 for Ra-226 and 0.88 for Ra-228. In comparison, the measurements of radium in water, including 1 standard deviation, were 0.32 ± 0.26 pCi/g for Ra-226 and 0.42 ± 0.26 pCi/g of Ra-228. These uncertainties and differences between measured and calculated water concentrations indicate around a factor of 3 uncertainty in the concentration of radium in water.

3.5.4 *Uncertainty in B_{iv}*

Taken together, the uncertainties in % moisture and radium concentrations in fish and water indicate a factor of 3 uncertainty in the calculated B_{iv}. Using water data instead of sediment data and K_d would have given an average B_{iv} of around 100 mL/g, also indicating a factor of 3 uncertainty when comparing to the average B_{iv} using sediment data and K_d, which was 39 mL/g before adding 2 standard deviations. Since the calculated B_{iv} was originally 39 mL/g, adding 2 standard deviations such that the final B_{iv} = 170 mL/g accounts for more than a factor of 3 uncertainty, providing a conservative estimation. The discussion of Table 11 showed the conservative nature of using B_{iv} = 170 mL/g for Rio Grande fish because it consistently overestimated the concentration of radium in fish samples by 2–10 times.

3.6 Assessing Soil and Sediment Samples for Secular Equilibrium

Soil and sediment samples taken by the SFB team from the bottom of Abiquiu and Cochiti and within the active channel of the Rio Grande were measured for several radionuclides in the U-238 and Th-232 decay chains in addition to Ra-226 and Ra-228. Descriptive statistics for these data are listed in Table 14.

Table 14. Soil and Sediment Data for Radionuclides in the U-238 and Th-232 Decay Chains

Radionuclide	n	Average (pCi/g)	1 Std Dev (pCi/g)
U-238 decay chain			
U-238	94	0.90	0.33
Th-234	16	1.7	2.4
U-234	94	0.88	0.31
Ra-226	79	1.1	0.6
Pb-214	16	1.2	0.3
Bi-214	16	0.96	0.26
Th-232 decay chain			
Ra-228	79	0.88	0.48
Ac-228	16	1.2	0.6
Pb-212	16	1.3	0.5
Bi-212	16	1.5	0.9
Tl-208	16	0.36	0.17

Two Th-234 results (each 7 pCi/g) were much higher than the other Th-234 data and had associated larger lab uncertainties (3 pCi/g). On average, the lab uncertainty for Th-234 was 1.5 pCi/g. Both the lab uncertainties and the standard deviations between samples were large compared with the reported Th-234 concentrations. The Th-234 measurements were inaccurate because they were done with gamma spectroscopy. Th-234 has three gamma-ray peaks that should be used together to measure Th-234 activity; these peaks are at 63.3, 92.4, and 92.8 keV (McNaughton and Patel 2010). The wrong peak may be selected for the 63.3 keV emission because Am-241 has a nearby peak at 60 keV; additionally, below 110 keV, many X-ray emissions from various radionuclides cause spectral interference with the desired gamma-ray peaks (McNaughton and Patel 2010); however, within 1 standard deviation, the radionuclides in the U-238 decay chain (including Th-234) have an activity of about 1 pCi/g.

Secular equilibrium between the radionuclides in the decay chain in which radium belongs indicates that the radium is natural (DOE-STD-1153-2019). Therefore, the U-238 decay chain data in Table 14 indicate that the Ra-226 present in Rio Grande, Abiquiu, and Cochiti samples was likely natural because the radionuclides have equal activities within 1 standard deviation of uncertainty.

Within 1 standard deviation, all radionuclides in the Th-232 decay chain have an activity of about 1 pCi/g except for Tl-208. Bi-212 decays to Tl-208 with a 36% probability (McNaughton and Patel 2010), so if the activity of Bi-212 is roughly 1 pCi/g, it can be expected that the secular equilibrium activity of Tl-208 is roughly 0.4 pCi/g. The data in Table 14 show that the Ra-228 in Rio Grande, Abiquiu, and Cochiti samples was likely natural because the Th-232 decay chain radionuclides were in secular equilibrium.

3.7 Future Study

There are perennial water systems near Los Alamos other than the Rio Grande—including Water Canyon and Sandia Canyon (LANL 2017, chapter 6)—that could be analyzed for dose to aquatic animals. In these locations, B_{iv} is likely to be similar to the one calculated in this paper. For accuracy, B_{iv} values specific to these locations should be calculated, but they are likely much lower than the RESRAD-Biota default value.

In fish, bioaccumulation of radium is expected to vary by species, fish age, and metabolism, and by the concentrations of calcium, strontium, and barium in the environment, duration of exposure and accumulation differences in various parts of the fish body (IAEA 1990). Only the last of the aforementioned variables was addressed in this paper: Basing our calculated B_{iv} mostly on parts of the fish body that accumulate more radium leads to a less accurate but conservative estimate of the B_{iv}. This paper also did not look for differences in bioaccumulation depending on water pH, time of year, possible trends over time, or feeding category (bottom feeding or predatory fish).

Other aquatic animals in the Rio Grande, such as crayfish, would be valuable to study. DOE-STD-1153-2019 mentions that mollusks and crustaceans often have the highest B_{iv} values of aquatic animals (see page F-3), so it might be expected that the B_{iv} of radium in Rio Grande crayfish is higher than the B_{iv} calculated here for fish. No trout were collected in 2023; their bioaccumulation of radium is likely similar to the fish in this paper, but it would be useful to include them in bioaccumulation studies due to public interest in consumption of this species.

Riparian animals have not been included in biota dose assessments at LANL (LANL 2014; LANL 2017; LANL 2020; LANL 2023), and no data are available in Intellus for riparian animals. RESRAD-Biota has a default bioaccumulation factor for riparian animals (B_{iv} = 800 mL/g) that causes them to fail Level 1 analysis for the Rio Grande, so deriving a site-representative B_{iv} value is appropriate. In RESRAD-Biota, putting the B_{iv} calculated here for fish into "Food Source Characteristics" for riparian animals predicts that the dose to riparian animals should pass Level 3 analysis; however, this prediction should be confirmed with measurements.

4 Conclusion

Fish in the northern New Mexico region of the Rio Grande fail RESRAD-Biota Level 1 analysis, mostly because of radium; its default bioaccumulation factor (3,200 mL/g) is extremely conservative. The existing data were used to calculate a wet-weight aquatic animal B_{iv} for Ra-226 and Ra-228 that was more accurate for the region: 170 mL/g.

The value of finding a site-specific B_{iv} in this paper is that (1) it provides another way to perform biota dose assessments of radium in the Rio Grande, and (2) fish dose rates can be calculated using sediment data alone. With the new B_{iv} calculated in this paper, Rio Grande fish pass Level 3 RESRAD-Biota radium dose assessment.

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7 Acronyms and Abbreviations

Acronym	Definition
ASER	annual site environmental report
BIV	bioaccumulation factor
DCF	dose conversion factor
DOE	(U.S.) Department of Energy
EPC-ES	Environmental Protection and Compliance Division-Environmental Stewardship Group
IAEA	International Atomic Energy Agency
ID	identification number
LAC	Los Alamos Canyon
LANL or Laboratory	Los Alamos National Laboratory
O	Order
RBE	relative biological effectiveness
SFB	Soil, Foodstuffs, and Biota