A Multidisciplinary Data Synthesis for Environmentally-Relevant Beta-Emitting Radionuclides in the Back-End Nuclear Fuel Cycle

[1] Loralei Cook and [2] Haruko Wainwright

[1] loraleicook@college.harvard.edu

[2] hmwainw@mit.edu

[1] Department of Physics, Harvard College

[2] Department of Nuclear Science and Engineering, Massachusetts Institute of Technology

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Abstract

This study synthesizes multidisciplinary information—from nuclear physics and environmental science—related to environmentally relevant beta-emitting radionuclides in the back-end nuclear fuel cycle: H-3, C-14, Cl-36, Se-79, Sr-90, Tc-99, and I-129. First, our synthesis shows that these radionuclides are the key contaminants and dominant dose contributors, both in the high-level radioactive waste disposal assessments and in the measured soil and groundwater contamination at the former US nuclear-weapon production sites. In addition, these nuclides are released from commercial reprocessing facilities in significant quantities. Their importance is primarily associated with their relatively long half-lives and high environmental mobility, so that they migrate farther and faster than other radionuclides. Nuclear weak-force interactions—the reasons for their long half-lives—result in beta decays without or with little gamma radiation, which, while it limits external exposure, makes it challenging to measure their concentrations *in situ*. Their high mobility is associated with limited sorption to minerals, although we found that many transport simulations assume zero sorption due to the lack of sorption data. Our results suggest that: (1) more research is needed on these radionuclides, including *in situ* measurement capabilities, their geochemistry, and their health impacts, and (2) different regulations (from other radionuclides) might be more effective for these nuclides, given that their risk is associated with internal exposure in a similar manner as chemical substances.

1. Introduction

Nuclear energy utilization is expected to expand in the near future, with several initiatives announced recently such as the Declaration to Triple Nuclear Energy (Kimball, 2023). While nuclear energy is considered an efficient and clean alternative to fossil energy sources, its potential environmental and public health risk remains a significant concern (Sadekin et al., 2018). In particular, the back-end nuclear fuel cycle—spent fuel storage/disposal and recycling—draws considerable attention (e.g., Schneider and Marignac, 2008; Ojovan and Steinmetz, 2022; Krall et al., 2022). In the United States (US), six states have restrictions on constructing new nuclear reactors until a spent nuclear fuel disposal plan is developed (Wainwright et al., 2023). The concerns are mainly attributed to the potential release of radionuclides and subsequent contamination within the environment (Ewing, 2008).

At the same time, the past several decades of research activities have provided significant insights into radionuclide mobility and migration behaviors in the environment. There have been extensive modeling and simulation-based studies, coupled with lab experimental studies, to quantify the long-term fate and transport of radionuclides, supporting the performance assessments (PAs) of high-level radioactive waste repositories (Apted and Ahn, 2017). Three countries (Sweden, Finland, France) have completed site-specific PAs (Rechard, 2013; Svensk Kärnbränslehantering AB, 2011), while generic PAs or radionuclide transport models have been developed for multiple repositories (e.g., Finsterle et al., 2021; Jones et al., 2003). In addition,

multiple underground laboratories worldwide have provided unique opportunities to observe radionuclide migration within the geosphere in a relatively small and controlled environment (Birkholzer et al., 2024; Graupner et al., 2025; Sugita et al., 2025).

In parallel, there have been extensive soil and groundwater remediation activities at the former nuclear weapon production sites in the US (often called *legacy sites*) for the last thirty years (National Research Council et al., 2000). They include reprocessing plants to extract plutonium and uranium and other waste-related facilities, which have the same functionality as commercial back-end facilities, although the ultimate purpose is different. Most contamination is associated with low-level liquid waste discharge into the subsurface and leakages from the facilities, including high-level waste storage tanks. Some radionuclides have migrated and created a groundwater plume of more than several hundred meters, the observation of which has provided a significant understanding of radionuclides' environmental behaviors.

In these assessments and observations, a common set of radionuclides are often identified as major risk contributors in the PAs and contaminated sites. I-129 and Tc-99 have been identified as the key remediation targets for groundwater contamination at the legacy sites (Truex et al., 2015; Neeway et al., 2019), which are also the major dose contributors in the HLW PAs. Carbon-14 (C-14) is also a key radionuclide in the HLW PAs and is discharged in significant quantities from reprocessing facilities (Castrillejo et al., 2020). In addition, Chlorine-36 (Cl-36) and Selenium-79 (Se-79) are identified as the key dose contributors in the HLW PAs (Kim et al., 1993). Tritium (H-3) is a key contaminant at the legacy sites and is released from reprocessing plants in both gaseous and liquid pathways (Fievet et al., 2010; Corcoran et al., 1992).

These radionuclides share common characteristics with respect to nuclear physics and environmental chemistry. They are primarily beta emitters with long half-lives and are known to be mobile in the environment compared to actinides and other radionuclides. Although there are several review and synthesis papers on individual elements (Hou et al., 2009; Neeway et al., 2019; Wainwright et al., 2024), there is currently not a single paper, to the authors' knowledge, that provides a broad and collective overview of these beta-emitting radionuclides. Their common characteristics have been presented before in the context of *in situ* measurements (Kang et al., 2020), but they have not been explicitly linked in observations of their environmental behaviors.

In this study, we synthesize the multidisciplinary facts and information of these beta-emitting nuclides from nuclear physics and environmental science, including their production pathways in reactors, environmental mobility, and biological impacts. We first systematically identify key radionuclides through meta-analysis of (1) soil and groundwater contamination at the legacy sites, (2) effluents from reprocessing plants, and (3) HLW PAs. Subsequently, we synthesize their physical and geochemical properties, such as their fission yields and activation cross sections,

their decay characteristics, their mobility in the geosphere, and their ability to be detected and measured, as well as radionuclide exposure pathways for elucidating their key common characteristics. A multidisciplinary understanding of key radionuclides would be essential in informing environmental protection efforts at back-end facilities, particularly at the time of expanding both existing and future/advanced reactor technologies (Wainwright et al., 2024).

2. Key Radionuclides in the Back-End Nuclear Fuel Cycle

2.1. Soil and Groundwater Contamination in US Legacy Sites

Nuclear weapon production and testing during World War II and the Cold War has resulted in soil and groundwater contamination at 107 US sites (legacy sites). As of 2024, the US Department of Energy (DOE) has completed remediation at 97 sites, while the 15 sites with large and complex contamination remain under active remediation (GAO, 2024). Since these sites have different climatic, geological, and other conditions, ranging from the dry, desert-like conditions of the western US (such as the Hanford Site) to humid and moist conditions in the southeastern US (such as at the Savannah River Site), the observations at these sites provide unique insights on radionuclide migration in the natural environment. All the sites make monitoring data publicly available through databases (Gorton et al., 2016) and annual reports (e.g., *SRS—Annual Environmental Reports*).

The Tracking Restoration And Closure (TRAC) database (trac.pnnl.gov) has compiled the types and footprints of the large plumes at the remaining 15 DOE legacy sites, tracking the progress in environmental restoration and the site's closure status (Johnson et al., 2024). Specifically, it tracks the spatial contamination extent of key contaminants, including radionuclides, heavy metals, and organic contaminants. Among these sites, seven sites have significant large-scale contamination by anthropogenic radionuclides (Table 1): including the Hanford Site, the West Valley Demonstration Project, the Paducah Gaseous Diffusion Plant, the Portsmouth Gaseous Diffusion Plant, and the Savannah River Site. The Hanford Site and SRS were plutonium (Pu) production sites, which included multiple reactors and reprocessing plants (Ramsey, 2013). The West Valley site was the first commercial reprocessing plant in the US without reactors and irradiation facilities. The Portsmouth and Paducah sites were uranium-enrichment facilities to increase the U-235 concentration using the gaseous diffusion process.

Although fission reactions produce a large number of radionuclides, large-scale plumes are created by five radionuclides, all of which are beta emitters: H-3, C-14, Sr-90, I-129, and Tc-99. Hanford is the largest among these sites, with groundwater plumes of all five radionuclides. The I-129 and tritium plumes are particularly large—larger than 10,000 acres. SRS is the second largest with Sr-90 and I-129 plumes of approximately 360-390 acres. Even though Paducah was a front-end facility (involved in enrichment), the fission product Tc-99 exists in groundwater,

because reprocessed uranium was transported from Hanford and SRS, and re-integrated into new fuel production.

Table 1. Acres of Radionuclide Plumes at the DOE Legacy Sites. Data collected from the 2021 Tracking Restoration and Closure (TRAC)-based map shows the acreage size of radionuclide plumes at the legacy sites. We only included anthropogenic radionuclides. The database includes other elements: Uranium, aluminum, ammonia, benzene, beryllium, cadmium, chromium, cobalt, dichloroethane, dichloromethane, dioxane, lead, lindane, manganese, mercury, naphthalene, nitrate, nickel, PFAS, sulfate, tetrachloroethene (PCE), trichloroethene (TCE), and vinyl chloride.

Site (m^2)	H-3	C-14	Sr-90	Tc-99	I-129
Hanford	57,600,000	60,700	1,570,000	1,830,000	57,500,000
Idaho			785,000		
West Valley				130,000	
Paducah				85,000	
SRS			1,570,000		1,460,000

2.2. Effluent Releases from Reprocessing Facilities

In addition to the three locations in the US (Hanford, SRS, West Valley), there have been three existing and past commercial reprocessing facilities from which data are openly available, including the La Hague Plant in France, Sellafield in England (closed in 2022), and Tokai in Japan (closed in 2011), all of which used the PUREX (Plutonium-Uranium Extraction) process. At the reprocessing plants, uranium and plutonium are chemically extracted from spent nuclear fuel, which is beneficial for conserving nuclear fuel resources, reducing HLW, and standardizing waste forms in a vitrified form (Silverio et al., 2010).

In this process, significant quantities of radionuclides are released into the environment, mainly into the air and ocean with the intentional use of the isotopic dilution strategy. As part of this waste management strategy, the gaseous and liquid effluent discharge of these radionuclides, including H-3, C-14, Sr-90, Tc-99, and I-129, has been monitored and reported annually. We have compiled the liquid effluent release quantities (the dominant release mode compared to the gaseous release) from the three reprocessing plants for their representative years (i.e., operating near the capacity) in Table 2. We would note that the data from the Tokai plant were gathered 20 years earlier than the other two, which could have resulted in nonreported (N.R.) or non-detected (N.D.) values.

Tritium is the main radionuclide with respect to radioactivity across the three plants, followed by C-14. Among other mobile beta emitters (Sr-90, Tc-99, I-129), La Hague has a higher discharge of I-129 compared to the others, while Sellafield has a higher one of Sr-90 and Tc-99. There are other radionuclides discharged in significant quantities, including Ru-106, Sb-125, and Cs-137 (gamma emitters), and Pu-241 (primarily a beta emitter).

The release quantity is significantly different even after it is normalized by the amount of SNF processed. We would note that the amount of radionuclides processed in SNF is similar to the SNF mass given similar reactor type, neutron spectrum, and discharge burnup (i.e., the amount of energy generated per U mass), since fission-product generation depends on the number of fission, and activation-nuclide generation depends on the length of time the fuel stayed in reactors. The release quantity is variable, possibly due to different processes used in these three plants. Mizutani et al. (2009), for example, reported that the Tokai plant had multistage evaporators in the liquid-waste-treatment process, which reduced the radioactive discharge significantly, except for tritium.

Table 2. Average annual liquid radionuclide effluent discharge from nuclear reprocessing sites, and the processed spent fuel mass (MT of heavy metal). We chose a representative year in which the operational and release data are available, and the plants are operating near capacity. The La Hague data is from Orano (2021; 2025), the Sellafield data is from Sellafield (2016; 2027), and the Tokai data is from JAEA (1996) and Mizutani et al. (2009). Radionuclides with the release of larger than 1 TBq in any of these plants have been selected. "N.D." refers to a nondetectable quantity, while "N.R." refers to a nonreported one.

			Release quantity, TBq								
	Year	SNF, t	Tritium	C-14	Sr-90	Tc-99	I-129	Cs-137	Ru-106	Sb-125	Pu-241
La Hague	2021	1021	10000	6.97	0.17	0.03	1.23	1.23	2.37	0.40	0.19
Sellafield	2016	899	2000	4.80	2.00	1.90	0.52	3.70	1.10	1.20	3.00
Tokai	1994	210	490	N.R.	N.D.	N.R.	7.00E-05	5.30E-07	N.D.	N.R.	N.R.

2.3. High-Level Radioactive Waste (HLW) Performance Assessments

High-level waste (HLW) is defined as highly radioactive material, including irradiated reactor fuel, liquid wastes from reprocessing reactor fuel, and solids derived from converting such liquid wastes (NRC, 1981). We have synthesized the key radionuclides from the site-specific and generic performance assessments across multiple countries (Rechard, 2013; Finsterle, 2021; Svensk Kärnbränslehantering AB, 2011; Jones et al., 2003; Nair et al., 1999; Andra, 2013). While the conditions of these proposed HLW repositories vary depending on the waste form, geology, and compliance periods, it is valuable to compare these assessments in evaluating the key radionuclides for assessing doses and health risks.

Our meta-analysis (Table 3) has identified the dominant dose contributors common across the assessments, including I-129, Se-79, Cl-36, Tc-99, and C-14. Specifically, I-129 is the highest-ranking, or most dominant dose contributor across all the performance assessments. Tc-99 is often the second highest dose contributor. Although differing in geological-condition parameters, such as groundwater/soil, release conditions, or time scale, these assessments provide a fairly similar ranking of contribution from each radionuclide.

Our finding is consistent with the previous meta-analysis done by the Organisation for Economic Co-operation and Development Nuclear Energy Agency (OECD-NEA) in 1997 and Croff et al. (2015), which documented the dominant radionuclide contributors to exposure dose rates calculated in the safety assessments performed by ten nuclear safety organizations (OECD-NEA, 1997). In this 1997 analysis, beta emitters were the dominant dose contributors from the repositories with direct disposal. We compiled additional and/or updated assessments, including the updated PA results for the U.S. Yucca Mountain Repository. In addition, generic repository simulations (Finsterle, 2021 and Jones et al., 2003) yield similar results, with I-129 as the primary dose contributor, which parallels the results of other European-based assessments (Svensk Kärnbränslehantering AB, 2011, Nair et al., 1999; Andra, 2013).

Table 3. Ranked Dose Contributors in High-Level Waste Performance Assessments. Ranking was based on the annual dose rate reported in each report. The reference is included in the table for each site.

Site	C-14	Cl-36	Se-79	Tc-99	I-129	Simulation Timescale
Yucca Mountain Repository (United States) (Rechard, 2013)	3rd	4th	-	2nd	1st	Simulated for release between 100 and 1,000,000 years (groundwater)
Vertical Borehole Repository Simulation (Finsterle, 2021)	-	3rd	2nd	-	1st	Simulated for release between 10 and 100,000,000 years (groundwater)
Forsmark Repository (Sweden) (Svensk Kärnbränslehantering AB, 2011)	3rd	-	2nd	-	1st	Simulated release between 1,000 and 1,000,000 years (soil)

Generic deep repository (United Kingdom) (Jones et al., 2003)	3rd	4th	5th	2nd	1st	Simulated for release between 10,000 and 100,000,000 years (soil)
The Cigéo Project (France) (Andra, 2013)	-	2nd	-	-	1st	Estimate release after 100,000 years

3. Physical and Chemical Properties

3.1. Radionuclide Generation and Their Physical Properties

Most of the radionuclides identified in Section 2 are direct fission products with fission yields higher than 0.04 (from U-235 thermal fissions; Jefferson Lab, 1997). The fission yields vary among these radionuclides, such that the yield is relatively high for Sr-90, Tc-99, and I-129, which is why these nuclides are dominant dose contributors at existing contaminated sites. On the other hand, Se-79 has a lower fission yield than the other radionuclides, which is the reason—along with a long half-life—why Se-79 is not recognized in the contaminated sites, although it comes up in the HLW assessment.

In addition, H-3, Cl-36, and C-14 are created by neutron activation and transmutation. This is a major production pathway for H-3 in pressurized water reactors, particularly from boron and lithium for reactivity and corrosion control. In addition, C-14 and Cl-36 are produced by neutron activation of nitrogen and chloride that exists as impurities in fuel and other structural material (Grambow et al., 2013).

We note that some of these radionuclides are naturally occurring as well. C-14 is a naturally occurring element in the atmosphere due to the n-p reaction of N-14 with cosmic ray-induced neutrons (Lingenfelter, 1963). C-14 is incorporated into plants and animals, which has been the basis for carbon dating. Recently, the annual global production of C-14 is estimated to be 2.2 x 10^{26} atoms/yr (Kanu et al., 2016). Similarly, a relatively small amount of H-3 is also produced in the atmosphere by galactic cosmic rays (Poluianov et al., 2020). While the amount of tritium produced in the atmosphere was considered to be insignificant compared to the amount of tritium emitted into the atmosphere by anthropogenic sources, naturally occurring tritium is still used as a tracer for stratosphere dynamics (Fourré et al., 2018). I-129 can be produced by the cosmic ray interactions in the atmosphere as well as the spontaneous fission of U-238 in geological formations (Wainwright et al., 2024).

Table 4. Half-life, fission yield, and activation cross section of environmentally-relevant beta-emitting radionuclides. Note that the fission yield and cross section are for the thermal neutron spectrum for U-235.

Radionuclide	Half-Life (yrs)	Pathway	Fission Yield (%)	Cross Section (b)
H-3	12	Fission Li-6 (n, H-3) He-4 B-10 (n, H-3) Be-8 H-2 (n, gamma) H-3	0.011	N/A 940 3839 0.001
C-14	5,730	Fission C-13 (n, gamma) C-14 N-14 (n, p) C-14 O-17 (n, alpha) C-14	<0.001	N/A 0.002 1.931 0.001
Cl-36	300,000	Cl-35 (n, gamma) Cl-36	< 0.001	43.62
Se-79	295,000	Fission	0.049	N/A
Sr-90	29	Fission	5.729	N/A
Tc-99	210,000	Fission	5.400	N/A
I-129	1,570,000	Fission	0.706	N/A

3.2 Decay Characteristics

These beta emitters have relatively long half-lives with low-energy decays, having little or no gamma radiation. This is because beta decay is dictated by weak-force interactions in nuclear physics. During the beta decay, a down-quark in a neutron (n) is transformed into an up-quark, converting the neutron into a proton (p) (Randall, 2005). By this process, mediated by the W-boson, an electron (e^-) and antineutrino (ν^-_e) are emitted, creating a release of energy.

$$n \rightarrow p + e^- + \nu^-_e \quad (3.1)$$

$$A\,[Z] \rightarrow A\,[Z + 1] + e^- + \nu^-_e \quad (3.2)$$

where Z is the atomic number and and A is the atomic mass. Because the W- boson has a relatively large mass, especially compared to photons, a slower reaction takes place than those seen in gamma decay.

The radionuclides of interest in this study have beta energies from 18 keV (H-3) to 709 keV (Cl -36; Table 5). The majority of the decay daughters are at the ground state, which results in no subsequent gamma rays. The range of energies suggest different penetrating powers: for

example, H-3 has low energy beta radiation with a total beta energy spectrum of 18.6 keV without gamma radiation. Some decay daughters emit gamma rays; such as Xe-129 from I-129 and Ar-36 from Cl-36. I-129 has a beta energy spectrum of 189 keV, but emits a low-energy (38.6 keV) gamma radiation as it decays to Xe-129.

Table 5. Energy Decay Properties of Relevant Radionuclides. Daughter, beta decay energies (Q_{β}-), and gamma transition energies (E_{γ}) were collected from the JAEA nuclide database and originally sourced from the 2012 Atomic Mass Evaluation (AME2012). The energies of gamma transitions were evaluated by various sources; however, default values were selected based on their correspondence to relative intensities (I_{γ}) of 100. This represents the most intense gamma at a given initial level. N/A represents there is no gamma radiation or less than 0.1% of decay. *The gamma radiation associated with the electron capture has a branching ratio of 2% (ANL, 2001).

Radionuclide	Daughter	Beta decay energy (Q_{β}-) [keV]	Gamma transition energy (E_{γ}) [keV]
H-3	He-3	18.592	N/A
C-14	N-14	156.476	N/A
Cl-36	Ar-36	709.53	788.4236*
Se-79	Br-79	150.6	N/A
Sr-90	Y-90	546.0	N/A
Tc-99	Ru-99	297.5	N/A
I-129	Xe-129	189	39.578

The total kinetic energy released during the beta-minus decay process is the loss of mass during the decay process (Jaffe, 2018). The energy spectrum is divided by the emission of the beta particle and the emission of the antineutrino, so the beta energy at a specific moment in time is difficult to calculate. Additionally, antineutrinos have weak interactions with matter, so the exact overall energy of the system can be hard to determine. Theoretical models and calculations, such as those based on Fermi's Golden Rule, attempt to provide an exact amount of energy generated by each beta decay (Strachan, 1969), although they do not account for all interfering factors. This is the primary reason why radionuclides cannot be identified by measuring beta rays, which are different from gamma emitters (Rathbun et. al, 1984).

3.3. Radiation Detection

Traditional beta-ray detection technologies include Geiger-Mueller (GM) counters and liquid scintillation counters. GM counters are not capable of measuring beta rays from low-energy radionuclides, such as tritium and carbon-14, as these counters are less sensitive to detection

against background radiation (Morishita et al., 2020). In contrast, liquid scintillation counters can accurately report the beta spectra of these particles, although treatment and sampling processes are lengthy and require large off-site equipment.

Kang et al. (2020) have reviewed the recent advances in beta-ray detection technologies, such as the use of inorganic scintillators and variations of plastic scintillators. Nanomaterials have been implemented within inorganic scintillators, such as those containing perovskite, and specialized scintillators to detect beta emissions within soil and water with more sensitivity. In 2013, the PoRTAS (Portable Rapid Tritium Analysis System) was developed by the Savannah River National Laboratory as a specialized liquid scintillator to measure tritium concentrations, followed by the 2019 development of a plastic scintillator by Southwestern European Instruments. In 2013, a mobile instrument, based on a traditional GM counter, was designed in Japan to measure the beta radiation from Sr-90 underwater, but this tool still requires a few days of analysis to produce a result (Kang et al., 2020).

However, the detection limits of portable mobile detectors are still relatively high compared to the environmental standards (Bq/L). For example, the aforementioned Japanese Sr-90 counter has a detection limit of 2 Bq/L (roughly 54 pCi/L), which is higher than the 8 pCi/L MCL for Sr-90. Only large liquid scintillators can measure as low as several mBq/L. In addition, the energy of beta particles is distributed, so identifying a particular nuclide within a mixture is a challenge. In most cases, monitoring is done by mass-based measurements such as the inductively coupled plasma mass spectrometry (ICS-MS) method in a laboratory setting (Kang et al., 2020).

3.4. Environmental Mobility

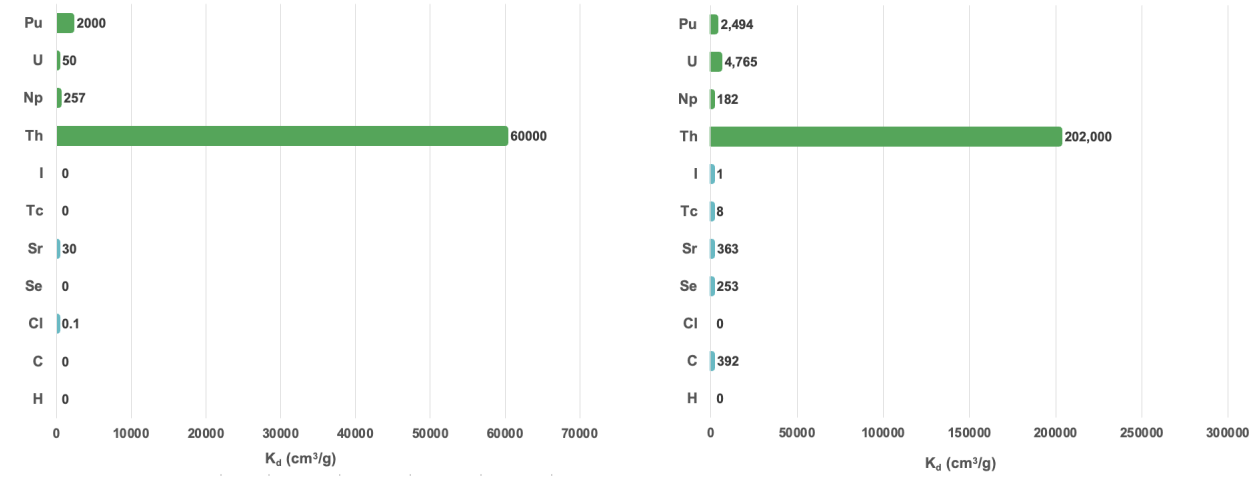
When radionuclides are released into the environment (particularly in soil and groundwater), sorption is the dominant process to retard the migration of radionuclides (Wainwright et al., 2018; Zavarin et al., 2022). Dissolved radionuclide ions can be adsorbed on mineral surfaces of rocks or sediments by electrostatic forces during their transport in surface water or groundwater. The distribution coefficient, K_d, refers to the ratio between the concentration of an adsorbed radionuclide on a solid surface and the one in an aqueous solution at equilibrium (Page et al., 1999). A high K_d value is associated with a nuclide with high retardation and slow migration, while a low K_d value is associated with a higher mobility in groundwater and other aqueous environments. K_d values vary significantly, depending on the type of soil or rock as well as the radionuclide chemical properties.

To highlight the difference between the elements, we have compiled the default K_d values for individual elements (Figure 2) used in one of the commonly used radiological and environmental assessment code RESRAD-BUILD Code (Yu et al., 2022). In addition, we have extracted the sorption experimental datasets from the L-SCIE database, which compiled sorption experimental

datasets as well as calculated K_d values for more than 10,000 data points of element-mineral pairs (Zavarin et al., 2023).

The model default K_d values (Figure 2a) are near zero for most of these beta emitters except for Cl-36 and Sr-90. We have reviewed other radionuclide transport modeling studies that have similar values (Mariner et al., 2015; Finsterle et al., 2021). The K_d values for these two nuclides are still significantly lower than those of actinides and other radionuclides. Their low K_d values suggest that they are highly mobile in the geosphere. However, the average K_d values reported by the L-SCIE database (Figure 2b) are non-zero values for Se-79, Sr-90, and I-129, which are larger than the ones in the modeling studies. This difference could be attributed to the fact that the modeling studies tend to assume lower K_d values as a conservative assumption, making radionuclides more mobile and predicted concentrations higher. Regardless, in both datasets, the same trend is observed: actinides have generally high K_d values, while the beta emitters of interest have generally low K_d values.

Figure 1 - K_d Values Among Beta-Emitters and Actinides. (a) RESRAD-ONSITE V7.2 and RESRAD-OFFSITE V4.0 default values and (b) L-SCIE database results. The L-SCIE values are representative of the median K_d values of each element for all mineral types (clay, silicates, oxides, etc.) with a pH range of 6 to 8 and an ionic strength of 0.0001 to 0.1 mol/L. The RESRAD values are estimated from plant/soil ratios and assume a logonormal distribution source.



a) **(b)**

3.5. Health Impacts and Implications

The Environmental Protection Agency (EPA) sets the drinking water standards for specific radionuclides in the US (Sondrup, 2024). Quantified by the Maximum Contamination Level (MCL), this value is derived based on 0.04 millisieverts (mSv) per year, assuming that individuals consume 2 L of water directly from the groundwater well. MCLs for the

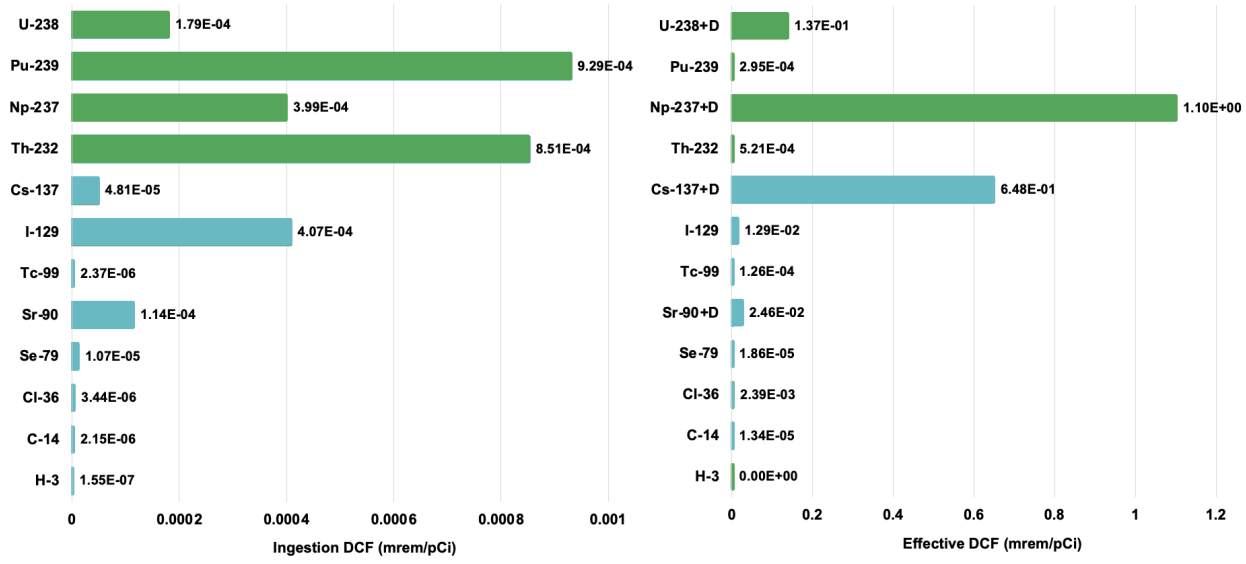
beta-emitting radionuclides of interest vary greatly, as it is generally associated with their biological risk and bioaccumulation. In addition, MCL can be converted to the mass-based standards, which are similar to other carcinogens. The mass-based standards are affected by decay half-life, such that short-lived radionuclides tend to have low concentrations.

I-129 has the lowest MCL, 1 pCi/L, which is attributed to the fact that exposure by ingestion of radioactive iodine can accumulate in the thyroid glands and lead to impaired thyroid function or cancers (Wainwright et al., 2024). Likewise, Sr-90 has a low MCL compared to others, since long-term exposure to Sr-90 in drinking water can accumulate within bone-marrow stromal cells of humans and cause cellular damage and cancer within bones and teeth (Musilli et. al, 2017). Tritium, which has not been documented to bioaccumulate in the body, has the highest MCL by pCi based on previous experiments and studies.

Table 6. Maximum Contamination Levels of Beta-Emitting Radionuclides, given in pCi/L and ppt equivalent. MCLs are based on a 2008 EPA evaluation.

Nuclide	H-3	C-14	Cl-36	Se-79	Sr-90	Tc-99	I-129
MCL (pCi/L)	20,000	2,000	700	7.55	8	900	1
MCL (ppt)	2.08E-3	0.45	21.2	0.63	5.8E-5	52.31	5.57

The International Commission on Radiological Protection (ICRP) publishes the internal and external dose coefficients of radionuclides. These dose coefficients can be used to convert given environmental concentrations to incurred radiation doses by each organ via ingestion, inhalation, immersion, and other forms of external exposure (Melo et al., 2022). These coefficients are based on the datasets and models that predict the risk and concentrations associated with radiation doses (Eckerman et al., 2012; Shubayar, 2017).



(a) **(b)**

Figure 2 - Dose Conversion Factors (DCFs) of Beta-Emitters and Actinides for: (a) Internal doses via ingestion and (b) External/effective doses from contaminated soil: Internal ingestion DCF values were populated from the ICRP Publication 72 (ICRP, 1995) and Federal Guidance Report 13 (Eckerman et al., 1999) (sourced from Table A-5 of RESRAD-BUILD Code Version 4). External DCF values were sourced from Federal Guidance Report 12 (Eckerman and Ryman, 1993). Selected actinides are represented in green and selected beta-emitters are shown in blue. Isotopes denoted with "+D" reflect aggregated values of DCFs for the principle radionuclide and their decay progenies or daughter products.

When ingested, the radiation dose received from beta-emitting radionuclides is generally smaller than that of actinides (Figure 2a), primarily because most actinides emit alpha radiations that have larger radiation weighting factors. However, I-129 and Sr-90 have high ingestion DCF, because of the accumulation in the thyroid and bones, respectively. For the external DCF (Figure 2b), the beta-emitters of our interest generally have smaller DCFs than actinides. I-129 and Sr-90 have slightly higher values, since I-129's daughter releases low-energy gamma rays when it decays and Sr-90's daughter releases high-energy beta radiation when it decays. Since low-energy beta radiations do not have enough energy to penetrate skin, external dose coefficients are low compared to Cs-137 and some actinides such as Np-237. This suggests that the radiological risk of these beta emitters is limited, except for the high ingestion risk associated with I-129 and Sr-90.

4. Discussion

Our analysis has shown that a set of beta-emitting radionuclides—H-3, Cl-36, Se-79, Sr-90, Tc-99, and I-129—are often identified as key radionuclides across the three domains associated with the back-end fuel-cycle facilities: (1) HLW disposal, (2) reprocessing effluents, and (3) soil and groundwater contamination. I-129 is the primary dose contributor in the HLW PAs, one of the significant radionuclides released from reprocessing plants, and one of the largest plumes at the legacy sites. Although the importance in each domain has been reported by other studies (Neeway et al., 2021; Kaplan et al. 2017; Mariner et al., 2015; Hou et al; Wainwright et al., 2024), our study highlights the importance of I-129 across the back-end fuel cycle. In addition, Tc-99 is also an important radionuclide in all three domains, particularly as the second highest dose contributor in many of the HLW PAs. The presence of Tc-99 at the enrichment facilities suggests that Tc-99 can appear in the front-end fuel cycle facilities when reprocessing is employed.

The other radionuclides are domain-specific. C-14 is identified as a significant dose contributor in multiple HLW disposal PAs and is also a major constituent in the effluent from reprocessing plants. Note that C-14 is recognized as an increasingly important radionuclide, since many advanced reactors plan to use graphite moderators/reflectors, the disposal of which has become a challenge during decommissioning due to the C-14 concentrations (Forsberg, 2024). H-3 and Sr-90 are more associated with existing contamination and reprocessing effluents due to their shorter half-lives. They are also major contaminants in soil and groundwater during the

decommissioning of nuclear power plant sites (Turkington et al., 2018). On the other hand, Cl-36 and Sr-79 appear only in the HLW PAs, since their extremely long half-life results in low activity in the 10-100-year time frame.

Fundamental physical properties dictate the importance of these beta-emitting radionuclides to their environmental impacts. They have relatively high fission yields or activation cross sections, so that they are generated in nuclear reactors in larger quantities than other radionuclides. In addition, these nuclides are all characterized by relatively long half-lives, from 12 years for tritium to 1.5 million years for I-129, which is associated with weak nuclear interactions. These beta emitters generally do not emit intense gamma radiation, although some of their daughter nuclides emit some low-energy gamma radiation.

Owing to these physical properties, the beta-emitting radionuclides discussed in this paper pose a significant challenge for *in situ* detection and measurements. Although technological advancements in measuring instrumentation have been made in recent years, it is still challenging to measure the energy of beta rays emitted from radionuclides *in situ* (Kang et al., 2020). The detection limit of portable detectors (10Bq/L) is often higher than the relevant environmental concentrations, 0.1 - 2 Bq/L for tritium (Sakuma et al, 2022) or the drinking water limit of 0.037 Bq/L for I-129. Since the energy of beta radiation from these radionuclides tends to be low, only scintillators can measure them, which can be done only in high masses or over several days of analysis. This can pose a challenge in detecting leakages and characterizing contaminations of these radionuclides (Kang et al., 2020). More research is needed to develop devices for the accurate and rapid detection and quantification of beta emitters.

In parallel, the chemical properties dictate their mobility in the environment. These beta emitters are often present in the environment as anionic species, so that they have limited sorption to clay minerals in soil and rock (Zavarin et al., 2022). This low sorption leads these radionuclides to migrate faster and farther compared to actinides and other radionuclides. Although sorption theory and related models have been developed mainly based on laboratory-scale experiments and mechanistic models, the real-world observations at contaminated sites confirm this theory and our understanding (He et al., 2024; Friday et al., 1996). In addition, their mobility, mainly represented by K_d, has a significant impact on future HLW disposal assessments in the million-year time frame.

The importance of environmental mobility has been recognized before, but frequently ignored. Waste management is, for example, still evaluated only considering the mass of waste, volume, and/or radiotoxicity, representing the source term without considering transport (e.g., Kim et al., 2022). It has been known that radiotoxicity does not directly correspond to the potential environmental impact, since it overlooks the fact that many radionuclides (e.g., actinides) are not mobile. Apted et al. (2012) thus suggested a “modified-RI” that accounts for the long-lived and

mobile nature of fission products. Our result re-emphasizes the importance of considering radionuclides' mobility in the environmental impact assessment.

At the same time, many models, including RESRAD, assume zero K_d for many beta-emitting radionuclides (e.g., Mariner et al., 2016; Wainwright et al. 2024), considering that lower Kd values are more conservative. However, recent studies have often shown that K_d is actually higher than zero for many species. In particular, non-negligible sorption has been observed in field studies. For example, Kaplan et al. (2010) investigated the variations in K_d values of I-129 accumulated in the Savannah River Site F-Area; the organically rich soil had nearly two orders of magnitude higher Kd values than the other soil samples. Such accumulation has been observed in deep marine deposits as well (Ohta et al., 2024). In general, the sorption experimental data for these species are limited compared to uranium and actinides. Given their importance in the fuel cycle, more research is needed to measure the K_d values of these radionuclides for various soil and rock conditions.

In addition, we have synthesized the datasets associated with the biological and health impacts of these radionuclides, including the drinking water standards and dose conversion factors. We have found that these long-lived beta emitters are primarily internal dose contributors (via inhalation, consumption, or absorption), similar to chemical carcinogens. The drinking water standards (MCL) exist for individual radionuclides—accounting for bioaccumulation and other factors—similar to how chemical carcinogens are regulated (Ashfold and Caldart, 2008). However, this fact is often lost in the assessments in which the health impacts are discussed similarly to gamma emitters.

Radiation protection is based on the assumption that the cancer probability follows a linear no-threshold (LNT) risk model (Richardson et al., 2023). LNT has also been used for chemical carcinogens, like aflatoxins, nitrosamines, and benzo(a)pyrene (Neumann, 2009). However, the current standards are developed primarily based on external exposure data. Although there are studies on internal exposure, they are often focused on short-lived nuclides for assessing the impacts of the Chernobyl and Fukushima incidents or medical treatments (Brenner et al., 2011; Matsuda et al., 2013). Given that the long-lived beta-emitting nuclides are important in the fuel cycle and nuclear waste management, and that there are only six of these radionuclides, it would be critical to design more focused experiments and simulation studies to quantify the biological impacts of these radionuclides

We acknowledge the limitations of our study, such as not covering all the nuclear facilities in the world. For representing environmental mobility of radionuclides, Kd is a simplified approach; more sophisticated surface complication models are available (Zavarin et al., 2022). However, this paper aims to highlight the importance of specific beta emitters, through the lens of nuclear physics and environmental science, for evaluating the environmental and health impacts

associated with the back-end fuel cycle. With the projected expansion of the nuclear energy industry in the coming years, scientific research, environmental regulations, and public knowledge should reflect an awareness of the potential effects of these beta-emitting radionuclides.

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