

A SIMPLE IMPROVED MEASURE OF RISK FROM A GEOLOGIC REPOSITORY

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Calculation of the expected long-term doses from spent nuclear fuel or high-level waste stored in a geologic repository via a performance assessment is a time-consuming and expensive undertaking because large amounts of data are required for engineered and geological systems, and there are many features, events, and processes that need to be taken into account. While necessary for repository licensing, it is impractical to do so for the purpose of providing input to comparisons of nuclear fuel and fuel cycle alternatives. Therefore, in lieu of detailed performance assessments, analysts typically rely on surrogate measures such as radiotoxicity. This paper identifies commonly used surrogate measures of long-term repository risk, assesses the strengths and weaknesses of these measures, and examines the consequences of the limitations in such existing measures. An improved measure of long-term repository risk that is risk based, yet still simple to calculate (because it is based on the results of existing performance assessments), is then proposed. The implications of the improved measure are discussed, and potential next steps are suggested.

I. INTRODUCTION

Estimates of long-term risks from repository waste¹ disposal are used for two broad purposes. The first purpose is to provide one part of the technical basis for licensing facilities that will dispose of the repository wastes. This technical basis is very detailed and is specific to the facility being licensed. Typical steps to provide this technical basis – called a “performance assessment” – include:

- Characterizing the radionuclide concentrations and inventory;
- Establishing the waste package design including the waste form and engineered barriers such as spent nuclear fuel (SNF) cladding, SNF or high-level waste (HLW) canister, an overpack made of corrosion-resistant metals such as high-nickel or copper alloys, and material to restrict the

¹ For the purposes of this paper, “repository waste” includes spent nuclear fuel, high-level waste, and non-high-level wastes containing most of radionuclides such as the transuranic elements, I-129, and C-14, and a fraction of the major fissile and fertile radionuclides.

- access and egress of water from the waste package (e.g., bentonite clay);
- Obtaining a thorough understanding of the geological, chemical, thermal, and hydrological conditions at the proposed repository site at present and into the future (as perturbed by repository wastes) through an extensive site characterization program;
- Predicting degradation rates for waste package components in the post-closure repository environment for times ranging from millennia to possibly thousands of millennia;
- Predicting the rate at which radionuclides will be released from the repository waste, dissolved in ground water, and transported to a point in the environment where they enter the biosphere;
- Estimating the transport of radionuclides through complex pathways in the biosphere and ingested by humans in water and food;
- Calculating the risks from ingesting the radionuclides; and
- Estimating uncertainties in the risks due to conceptual and data variability.

As a consequence of the foregoing, a repository performance assessment is site-specific, time-consuming, complex, costly, and often controversial. Preparation of a performance assessment is not undertaken lightly and is not likely to be done except in anticipation of operating a repository.

The second purpose of estimating the long-term risks from a repository is to provide one basis for evaluating preferences for entire nuclear fuel cycle options (FCOs) and/or technology options such as reprocessing or actinide partitioning and transmutation. Such evaluations typically involve quantifying and weighting multiple criteria such as operational safety, cost, proliferation resistance, and repository risk [1, 2]. Preparing a repository performance assessment for each of the FCOs and technology options is impractical because of the many waste forms and differing source terms, and frequently the absence of a specific repository site at the time the evaluations are being done. Instead, the repository risk criterion is typically quantified by using surrogate measures that are relatively simple to calculate such as: repository waste volume, radioactivity, or radiotoxicity; such surrogate measures are, at best, imperfect measures of repository risk.

The objectives of this paper are to identify commonly used surrogate measures of long-term repository risk, assess the strengths and weaknesses of these measures, and examine the consequences of the limitations in existing measures. We then propose an improved measure of long-term repository risk *for use in comparing nuclear FCOs* that is still simple to calculate, examine its implications, describe some of the unknowns in its calculation, and suggest potential next steps to address those limitations.

II. COMMONLY USED SURROGATE MEASURES OF LONG-TERM REPOSITORY RISK

II.A. Rudimentary Surrogate Measures: Volume and Radioactivity

One simple measure of repository risk that has been used from the beginning of the nuclear power era [3] [4] to the present [5] is the volume² of waste to be emplaced in a repository. Volume is typically calculated by assuming standard waste package designs for various wastes [6, 7]; these designs specify the amount of waste in terms of the number of fuel assemblies or HLW canisters, etc. per unit of throughput (e.g., metric tons of heavy metal) for each FCO and the size of the package. Taken together, this allows calculation of the volume of waste to be emplaced in a repository per unit throughput (e.g., liters/MTHM). This value can be used directly as a measure to compare various FCOs operating at steady-state or as the basis for projecting repository waste volumes in a dynamic nuclear energy scenario. The strengths of this measure are that it is relatively simple to calculate and it can be related to operational risks such as transportation, interim storage, and emplacement. The weakness of this measure is that the total volume of repository waste – which might be comprised of wastes types having a wide range of radioactive material concentrations – has little relationship to long-term repository risk. If the measure were used to compare the volume of waste of a specific waste type (e.g., SNF) from FCO A to that from FCO B, there would be a distant relationship to repository risk, but waste volume still does not account for the amount of radioactivity in the waste (e.g., differing fuel burnup) and other factors listed in Section I (above) that are taken into account in a performance assessment.

An incremental improvement over volume as a measure of repository risk can be achieved by adding the radionuclide concentration (e.g., MBq/L) in the waste to

² Waste mass has the same attributes because volume and mass are proportional, with density being the conversion constant.

the measure. When multiplied by the volume of waste per unit throughput the result is the inventory of radionuclides in the repository waste. The strength of using radioactivity is that it has an increased correlation with risk because – under the tenets of the linear non-threshold hypothesis of radiological impacts – the risks from such wastes are related to the radioactivity. The weaknesses of using radioactivity as a measure of long-term risk are that (1) it does not account for each radionuclide emitting different types of radiation, having different energy spectra and biological behavior once the receptor is exposed as well as other factors listed in Section I (above), and (2) it is incrementally more complex to calculate than volume.

II.B. The Popular Surrogate Measure: Radiotoxicity

A further incremental improvement in developing a surrogate measure for repository risk had its beginnings early in the 20th century when radiation protection focused on limiting total dose to humans, especially to workers handling specific radionuclides such as Ra-226 [8]. Since the middle of the 20th century, increased availability of information on the radiological impacts of individual radionuclides, and the biological behavior of each radioelement in various chemical forms in the human body, allowed limits to be established for each radionuclide [9]. These limits have been expressed in various units such as intake limits or a maximum allowable concentration in water, corresponding to a specific dose limit (e.g., 5 mSv/yr) for a member of the public; the units can be readily interconverted. The limits have a range of names (e.g., annual limiting intake (ALI), maximum permissible concentration (MPC), radionuclide concentration guide (RGC)) and units (e.g., radioactivity per unit volume of water, radioactivity intake per year, or radiation dose per unit of radioactivity). These limits are recommended by scientific advisory bodies such as the International Commission on Radiation Protection (ICRP) [10] and established by national regulatory authorities like the Nuclear Regulatory Commission (NRC) [11] in the US.

Radionuclide ingestion limits can be used to calculate a measure of the radiotoxicity³ of a radioactive waste. This is typically done by dividing the radioactivity (MBq) of each radionuclide by its concentration limit corresponding to a dose limit (MBq/L) to yield the volume of water (L) required to dilute the radionuclide so that the dose limit is met. Alternatively, the radioactivity (MBq) of each radionuclide can be multiplied by the dose resulting from ingesting a unit amount of radioactivity of each radionuclide (Sv/MBq) to yield the dose (Sv) that would result from ingesting that amount of the

³ In some of the literature this is called “hazard” instead of “toxicity”.

radionuclide. The water volumes or doses for each radionuclide are summed over all radionuclides in a waste to yield a radiotoxicity value for the waste which can be compared with the radiotoxicity of other wastes. The radiotoxicity of a waste can also be normalized by dividing it by the radiotoxicity of a reference radioactive material – for example, the uranium required to produce the waste – to yield a toxicity index.

Using radiotoxicity measures as a surrogate for repository risk addresses two important deficiencies in surrogate measures such as waste volume or radioactivity: (1) it takes the nature of the radiation from each radionuclide into account, and (2) it addresses the unique biological effects of each ingested radionuclide. Additionally, with the advent of digital computers and storage devices, the additional difficulty in calculating radiotoxicity, as compared to just radioactivity, is insignificant because radiotoxicity is simply the sum of the product of the radioactivity of each radionuclide multiplied by a fixed value defining the toxicity of the radionuclide.

One artifact of the capability to readily calculate the radiotoxicity of a mixture of radionuclides – and the relatively small incremental effort to calculate it as compared to waste volume or radioactivity – has been its longstanding and widespread use as a surrogate for repository risk when evaluating preferences for FCO options or technologies [12-15] and compendia of radiotoxicities have been prepared for various radioactive materials [16, 17]. However, despite its popularity, radiotoxicity still suffers from some of the same weaknesses as do the more rudimentary measures. In particular, it does not account for many of the factors listed in Section I (above) as being important to determining the radiological risk from repository waste, such as waste form degradation rates, radionuclide solubility, radionuclide mobility in ground water, radionuclide decay with time, and radionuclide movement in the biosphere before intake. In essence, using radiotoxicity as a surrogate for risk assumes that a radioactive material, such as SNF, is directly ingested which seems implausible – at best.

III. CONSEQUENCES OF LIMITATIONS IN CURRENT MEASURES OF LONG-TERM REPOSITORY RISK

This paper will use radiotoxicity – the best of the existing, simple measures of long-term repository risk – to highlight the consequences of limitations in these measures by comparing the radiotoxicity of typical⁴ PWR SNF (Fig. 1) with the results of a performance assessment

⁴ The “typical” nature of the repository risk results from a performance assessment will be further supported below.

for a repository containing PWR SNF (Fig. 2). One difference between the two results is that the performance assessment does not predict significant doses to occur for several thousand years, with the peak dose occurring even later, whereas the radiotoxicity is at a maximum at short decay times and monotonically declines thereafter. Perhaps more importantly, the main contributors to radiotoxicity are actinides (plutonium, Am-241) at all decay times beyond a few centuries, while fission and activation products are inconsequential at all times. In contrast, the main contributors to repository risk based on the performance assessment are an assortment of long-lived fission and activation products, with the decay products of the uranium (which constitutes the bulk of the SNF) only beginning to make an appearance after several million years.

The very different suite of key contributors to repository risk when using these two different measures has significant potential consequences for selecting preferred FCOs and technologies. Specifically, the actinides – and plutonium and the minor actinides in particular – are essentially irrelevant to the calculated long-term repository risk. This conclusion is driven by the actinide elements having very low solubility in typical ground water at a potential repository site, in combination with the fact that actinides migrate through the geosphere much more slowly than the bulk ground water because of sorption effects. An important consequence is that the removing actinides from SNF for alternative disposition such as recycle and fission (i.e., partitioning and transmutation) does not lead to a proportional⁵ reduction in calculated repository risk. This result calls into question the use of radiotoxicity to justify FCOs and technologies designed to recover and recycle the minor actinides to convert them to fission products.

The fact that fission products dominate long-term repository risk, based on performance assessment results, also has implications. One implication is that the calculated long-term repository risk of FCOs producing the same amount of energy will not differ greatly because the fission product yields of various fissile species are not very different (more on this below). Another implication is that the potential to reduce long-term repository risk by recovering the dominant fission products and transmuting them to shorter-lived species exists.

It is also noteworthy that in this assessment two activation products – C-14 and Cl-36 – are contributors to long-term repository risk, albeit at a low level. These species are produced by neutron captures in stable nitrogen and chlorine contaminants, respectively, in many fuel matrix and fuel assembly structural materials. This raises the issue of the amount and variability of these

⁵ Removing actinides such as Pu and Am-241 does significantly reduce decay heat which may have indirect benefits.

contaminants that are in such materials, which largely depends on methods of manufacture. Additionally, it means that calculated long-term repository risk could be lowered by reducing the amount of these contaminants in nuclear fuels.

In short, available estimates of long-term repository risk, based on performance assessment calculations, turn the implications of using radiotoxicity ‘on its head’ by de-emphasizing the importance of actinides⁶ (to the point of being largely inconsequential) and bringing long-lived fission and activation products into prominence. While the limitations of radiotoxicity as a measure of long-term repository risk that lead to this conclusion are not new [18], radiotoxicity continues to be widely used. However, use of radiotoxicity as a measure of long-term repository risk lends support to FCOs and technologies such as actinide partitioning and transmutation that seem to offer modest reduction of long-term repository risk.

IV. AN IMPROVED SIMPLE MEASURE OF LONG-TERM REPOSITORY RISK

The foregoing leads to the dilemma alluded to in Section I: radiotoxicity is easy to calculate but yields results that are not representative of the best current estimates of long-term risks to the public from a geologic repository. Thus, using radiotoxicity as a surrogate for repository risk can be misleading, while a performance assessment yielding results that are more representative of reality are much more difficult to obtain and are usually site-specific (or at least geology-specific). This dilemma can be resolved by recognizing a fortuitous commonality among performance assessments: the list of radionuclides that dominate calculated risk is short and mostly contains the same radionuclides, irrespective of the site or geology being assessed. To illustrate this point results from 15 repository performance assessments for a variety of geologies [19-25] were analyzed to identify the radionuclides that dominate risk and the time at which the peak exposure occurs. Results⁷ of the analysis are given in Figure 3 which identifies the radionuclides that contribute at least 0.0001% of the sum of the peak dose rates out to a 1 million year decay time to a member of the public from an undisturbed repository containing spent LWR fuel. The observations based on this figure and the performance assessments underlying it are as follows:

- I-129 dominates long-term repository dose by at least a factor of five in all but one of

⁶ There may be reasons other than radiotoxicity (e.g., decay heat reduction, proliferation concerns) to remove actinides.

⁷ The data underlying the Figure 3 were read from graphs and are likely accurate to only one significant figure.

the 15 studies examined and even there it remained a significant contributor to risk.

- Other frequently important contributors to long-term repository dose are C-14, Cl-36, and Se-79.

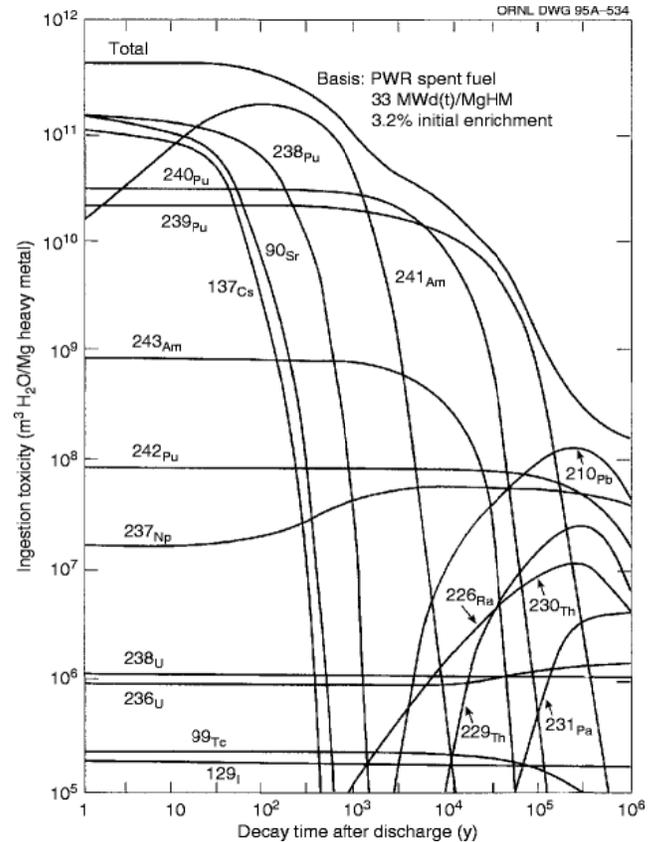


Fig. 1. Ingestion radiotoxicity of PWR SNF as a function of decay time [26]

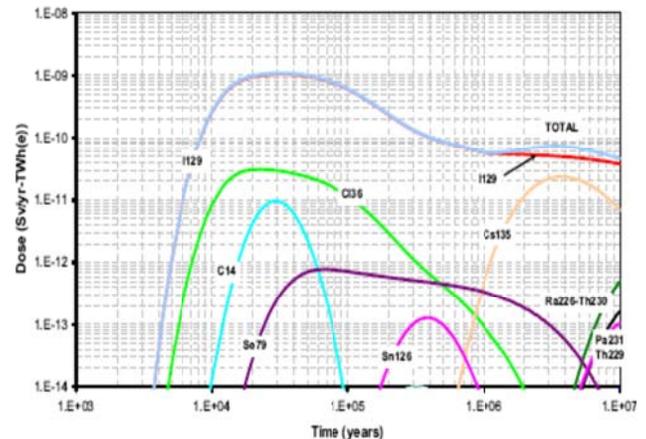


Fig. 2. Dose from SNF disposal in granite calculated by a performance assessment [19]

- Radionuclides that are sometimes important contributors to long-term repository dose are Tc-99, Sn-126, and Cs-135.

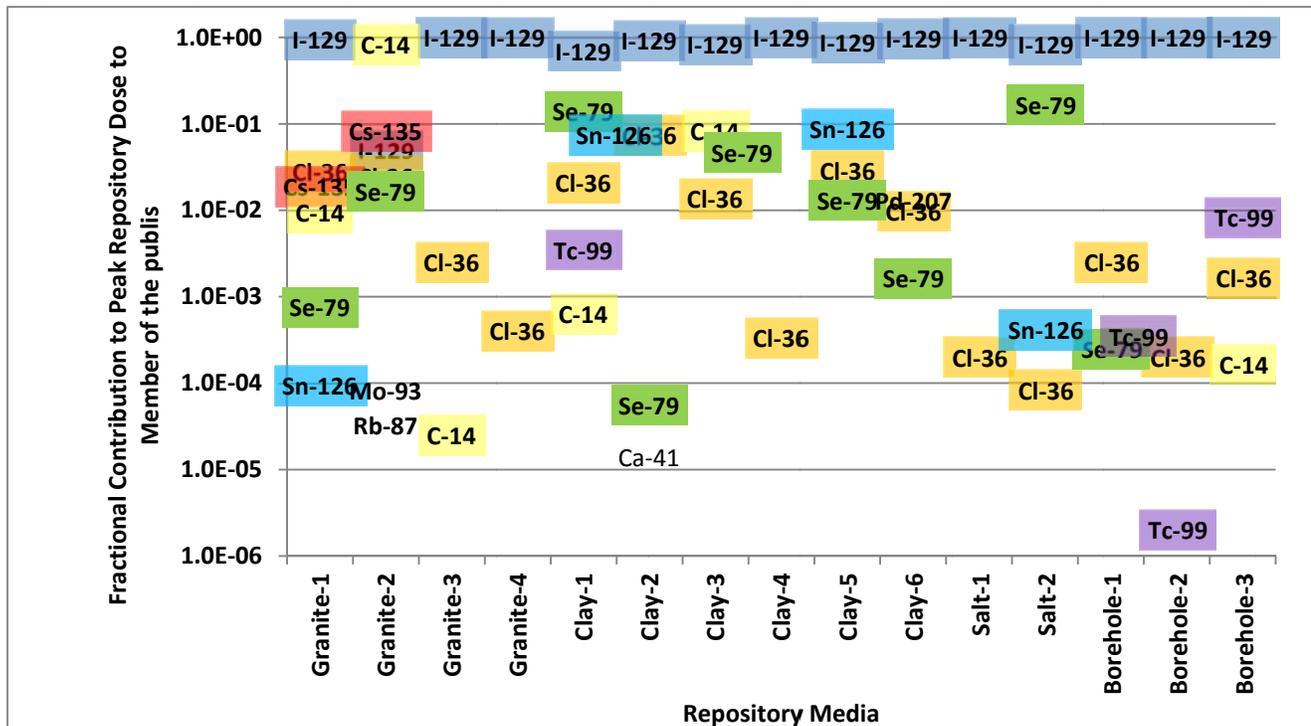


Fig. 3. Fractional contribution of the most important radionuclides contributing to the absolute calculated dose rate to a member of the public from undisturbed performance of a repository containing spent LWR fuel

- Radionuclides that infrequently become important contributors to long-term repository dose are Mo-93, Rb-87, and Ca-41.
- Actinides and their decay products only begin to contribute to long-term repository dose, at levels that are orders-of-magnitude below the smallest values in Figure 3, at times close to or beyond 1 million years. Assessments that went beyond 1 million years (about half) typically showed increasing doses from actinides (usually from the U-238 decay chain that constitutes the bulk of the fuel matrix, but in a couple of instances from the Np-237 decay chain) primarily because essentially everything else has decayed to innocuous levels or is decaying away rapidly.
- Fission products such as Sr-90 that dominate SNF radiotoxicity for the first century or so do not contribute at all because they decay to unimportant levels before they can be released and migrate from the repository.

Thus, the list of radionuclides that accounts for >99% of long-term repository dose to the public out to 1 million

years contains seven fission or activation products: C-14, Cl-36, Se-79, Tc-99, Sn-126, I-129, and Cs-135.

On this basis, the proposed simplified measure of repository risk is the ratio of the amount of each listed radionuclide in one nuclear material or set of nuclear materials from a FCO to the amount in another nuclear material or set of nuclear materials for a second FCO. The following section will discuss some implications of using this measure for the purpose of comparing FCO alternatives and technologies, and limitations of the measure.

V. IMPROVED SIMPLE MEASURE OF REPOSITORY RISK: IMPLICATIONS AND UNKNOWNNS

This section discusses some implications of using the list of seven radionuclides as a measure of long-term repository risk and some of the unknownns that would need to be faced to confidently use the measure.

V.A. Fission Products

Five of the seven radionuclides that dominate long-term repository risk are fission products. As such, the concentration of these fission products in various SNFs

and nuclear materials recovered from the SNFs can be calculated relatively accurately. These concentrations are related to the long-term risk from the disposal of the subject material in a repository. Parameters used to illustrate application of the measure are given in Table I. The capture cross sections (and by extension, removal

rates) of these five fission products are small compared to their production rates so this process can be neglected for the purposes of FCO comparison and the concentration of the radionuclides in SNF is proportional to the fission yield of their mass chain.

Table I. Cross section and fission product yields of Se-79, Tc-99, Sn-126, I-129, and Cs-135 for a PWR [27, 28]

Radionuclide	Cross Section, barns	Fission Product Yield							
		U-235		Pu-239		Pu-241		U-233	
		%	Ratio to U-235	%	Ratio to U-235	%	Ratio to U-235	%	Ratio to U-235
Se-79	0.31	.045	1	.044	.98	.015	.33	.14	3.1
Tc-99	9.3	6.11	1	6.21	1.0	5.96	.98	4.91	.80
Sn-126	0.0329	.059	1	.20	3.4	.082	1.4	.23	3.9
I-129	3.1	.54	1	1.37	2.5	.82	1.5	1.6	3.0
Cs-135	2.5	6.5	1	7.6	1.2	7.2	1.1	6.3	.97

The ratio of the yields of the five listed radionuclides from fission of Pu-239, Pu-241, and U-233 to that of U-235 is given in Table I. Ratios greater than 1.0 indicate that SNF from fresh fuel containing more of a particular fissile species would pose a higher repository risk than a fuel with less of that fissile species as compared to U-235, and conversely for a ratio less than 1.0. To take a specific example, the U-233/U-235 fission product ratio for the dominant I-129 is significantly greater than 1.0 so one would expect PWR SNF that had been enriched with U-233 to have a higher repository risk than one enriched with U-235, and similarly for plutonium.

V.B. Activation Products

Two activation products were shown to be consistently important to long-term repository dose: C-14 and Cl-36. Their concentration in SNF and nuclear materials derived from it depend on the initial concentration of stable precursor radionuclides: C-13 and N-14 in the case of C-14, and Cl-35 in the case of Cl-36. Carbon, nitrogen, and chlorine are present in fresh fuel matrices, fresh fuel structural material (cladding, spacer grids), and moderators to widely varying degrees. In some fuel cycle options these materials can be present in substantial quantities, e.g., reactors using graphite fuels and moderators (C-13 leading to C-14), nitride fuels (N-14 leading to C-14), and chloride-based molten salt reactors (Cl-35 leading to Cl-36). However, for current fuel cycles involving LWRs, CANDU reactors, etc. all are present in trace amounts (up to tens of ppm by weight). C-13 is not an important precursor of C-14 compared to N-14 production routes in current reactors because the amount of C-13 – which is a small fraction of the total carbon – is very small. However, even tens-of-ppm of nitrogen and chlorine lead to amounts of C-14 and Cl-36 that have been shown to be important contributors to

long-term repository risk. The challenge in using these activation products as measures of long-term repository risk is that their concentrations current fuel cycle materials is poorly known because: (1) at these low levels the contaminants do not have any adverse impacts on material properties and (2) they are difficult to measure at these concentrations. Information for different materials in alternative fuel cycles is essentially non-existent except in special cases like carbon in reactors such as those using hexagonal block fuel. Further, information on carbon and nitrogen concentrations that were assumed in the nuclear fuel depletion calculations underpinning repository performance assessments is not readily available. Thus, while it is conceptually possible to use these activation products to compare alternative FCO materials or technologies, data does not presently exist to support such comparisons.

V.C. High-Level Waste

Essentially all of the foregoing has focused on SNF. In the case of HLW the dominance of fission and activation products as contributors to long-term repository risk would be increased. At times less than one million years essentially all of the fission and activation products remain while actinides such as plutonium *per se*, the Am-241 produced from plutonium decay, or U-233 have been reduced by reprocessing to even lower concentrations than in the parent SNF. For times beyond one million years, similar logic prevails except that it is the reduction in the concentration of actinides that constitute the bulk of the fuel matrix (U-238 or Th-232) that contributes to the increased dominance of the fission and activation products.

VI. IMPROVEMENTS AND NEXT STEPS

The following sections identify a range of potential improvements to the measure of long-term repository risk proposed in this report and steps that could be taken to accomplish the improvements.

VI.A. Reconcile differences in existing repository performance assessments

As is evident from Fig. 3, I-129 is the dominant contributor to calculated long-term repository dose in most performance assessments. However, while there is substantial variability in which radionuclides are the next-most-important contributors, the cause of the variability has not been identified. It would be helpful to perform a more detailed review of existing performance assessments to determine the cause(s) of difference in radionuclide importance to long-term dose. In particular, it would be useful to know whether inconsistent generic assumptions are the main driver of variation, as opposed to the attributes of the rock type or site, and refine list and importance accordingly.

VI.B. Further investigation of the importance of risks from oxidizing repository geologic settings

The dominance of the seven long-lived activation and fission products is a result of their being significantly-to-highly soluble and relatively little retarded by sorption on subsurface geology during transport in ground water for the repository geologies being evaluated. For the most part, this results from the subsurface redox conditions in these geologies being at least neutral but usually reducing so that actinides and other species having multiple valence states are relatively insoluble and highly retarded by sorption mechanisms. For example, the solubility of neptunium under reducing conditions is about 10,000 times less than under oxidizing conditions in ground water typical of the site at Yucca Mountain, NV [29]. The situation concerning retardation by sorption effects is more variable depending on the element but under reducing conditions retardation is larger by at least 70x for technetium, 10x for neptunium, and 80x for uranium [30].

One exception to the presence of reducing conditions is the proposed repository site at Yucca Mountain in the U.S. which was projected to have oxidizing conditions under which some actinides – especially Np-237 – would have been a significant contributor to long-term repository dose. Two Yucca Mountain performance assessments were reviewed to identify dominant contributors to long-term dose. For the performance assessment underpinning the DOE license application for Yucca Mountain [31] the most important radionuclides, constituting 94% of the calculated dose at 1 million years,

were I-129, Tc-99, Pu-242, Cs-135, and the Np-237 chain in decreasing order of importance. The contribution of the actinides was about 18% of the total at 1 million years. In the EPRI performance assessment [32] the dominant contributors to dose were the U-238 decay chain, I-129, and the Np-237 decay chain in decreasing order of importance with the actinides accounting for about 70% of the dose at 1 million years.

These considerations lead to the need to assess the importance of accounting for risks from repositories in oxidizing geologic settings. Such an assessment would provide the basis for deciding whether the additional radionuclides that are important in oxidizing settings should be included in the simple repository risk measure and, if so, how to weight them. Questions to be addressed in support of addressing this need include: (1) explaining the differences in the list of dominant contributors in the EPRI and DOE performance assessments described in the previous paragraph, (2) examining the technical basis for selecting the Yucca Mountain site and locating the disposal facility above the water table, and (3) conducting a survey to determine the extent to which deep geologic waste disposal is being considered in any other oxidizing geologic environment.

VI.C. Data acquisition needs

As discussed in Section V.B, the amount and variability of stable precursor (i.e., carbon, nitrogen, chlorine) concentrations of activation products that can be significant contributors to long-term repository risk from nuclear fuels, structural material, and moderators are poorly known. Improving knowledge in this regard should be a priority target of further investigations.

In addition, none of the performance assessments used in preparing this paper considered thorium-based or graphite-based fuels using any fissile material and there do not appear to be any in existence. Such fuels are being considered in FCO comparisons [2], and both graphite and thorium are projected to be resistant to degradation in both reducing and oxidizing geochemical environments [33]. A useful first step would be a more thorough literature search emphasizing difficult-to-access information sources, such as those not in English. However, new performance assessment calculations may be needed to underpin generalization of the simple measure of repository risk for thorium- and graphite-based fuels. The simple improved measure of risk will need to be updated once performance assessments are available for these fuels to determine if the list of radionuclides important to dose needs to be revised based on the physical and chemical properties of these fuels.

VI.D. Conceptual refinement

The examples given in Section V were based on ‘eyeball’ assessment of simple ratios of fission product yields or gross differences in the concentration of activation product precursors. The simple improved measure of risk would be greatly enhanced if sufficient consistency could be found among performance assessment results to support derivation of defensible weighting factors for each radionuclide. The weighting factors could then be used to calculate a single value for the measure, hopefully with enough information to allow uncertainty analysis to be conducted. Conceptual refinements necessary to determine generic weighting factors are as follows:

- Reconcile radionuclide ‘outliers’: Delve into the assumptions that lead to large differences in the importance of various radionuclides as discussed Section VI.A, e.g., the dominance of C-14 in one case in Fig. 3. This includes the need to deal with oxidizing geologic settings as discussed in Section VI.B.
- Accounting for variability in dose curve shapes: As is evident from Fig. 2, the times over which particular radionuclides lead to doses calculated to be close to their associated peak dose varies substantially. For example, C-14 has a sharply defined peak that persists over a relatively short time whereas near-peak doses from I-129 persist for long times. The implication of this variation is that the collective dose from a radionuclide having near-peak doses over a long time would be larger than for a relatively short-lived peak even if the peak doses were the same. Further work is needed on how to account for this difference in the simple improved measure of repository risk. One possibility is to use time-integrated dose instead of the peak dose.
- Determine the time horizon to be considered: Most national authorities limit consideration of post-closure repository risks (doses) to timeframes of one million years or less. Doses at longer times are not considered, or are addressed by comparison to naturally occurring ore bodies [34-36]. The rationale for this is that geological predictions have little scientific basis at longer times because unpredictable large scale changes can take place, e.g., mountain building, continental drift, and massive erosion [37]. The potential lack of scientific basis notwithstanding, performance assessments that have been extended to times longer than one million years generally show risks are driven by naturally occurring radioactive radionuclides such as U-238 and Th-232 – often the major constituents of the nuclear fuel – as they achieve secular equilibrium with their decay products. As noted above, in performance assessments for geologies having oxidizing

geochemical conditions, Pu-242 and the Np-237 decay chain may also be calculated to be significant contributors in the few-million-year timeframe. Further evaluation is needed to select and justify the time horizon that will limit the radionuclide peaks to be considered in establishing the list of key radionuclides and to fix the upper time boundary for calculating integrated dose as described in the preceding bullet.

- The simple improved measure of long-term repository risk developed in this paper is based on assessments of undisturbed repository performance. Regulators often consider scenarios that disturb the repository, typically involving human intrusion by drilling. The key feature of these scenarios is the beneficial impact of solubility limits and retardation by sorption in reducing the rate of radionuclide transport to the biosphere are largely bypassed. The result is that the importance of radionuclides in these scenarios is more closely represented by radiotoxicity than by the results of performance assessments for undisturbed repositories, i.e., the actinides are much more important contributors to risk in the disturbed repository scenario. The purpose of analyzing disturbed repository performance “...is to examine the site- and design-related aspects of repository performance under an assumed intrusion scenario to inform a qualitative judgment” [38] and they are not comparable to assessments of undisturbed repository performance because event probabilities are not taken into account. However, it may be possible to develop a separate simple measure of risk for a disturbed repository based on existing performance assessment results for such scenarios using the approach described in this paper.

VI.E. Facilitate Use of the Simple Improved Measure of Repository Risk

Even though simple, calculation of the improved repository risk measure could still be time consuming if one were comparing a large number of FCOs or performing sensitivity studies. Calculating the measure becomes even more time-consuming if additional performance assessments must be performed due to the absence of an existing relevant assessment. Thus, it would be beneficial to facilitate calculation of the improved risk measure and providing flexibility in the range of scenarios that could be addressed. A first step is to obtain existing performance assessment results in numerical (not graphical) form to allow better estimation of peak and integrated doses and, thus, weighting factors as described above.

Obtaining new performance assessment results that are in a numerical form and which encompass a range of geologic settings and waste types would be a major step

in facilitating use of the measure. By definition, this would involve doing performance assessments for multiple combinations of geologic settings and waste types which, as discussed in Section I, can be costly and time-consuming. However, simplified repository performance assessment methodologies have been developed for some commonly considered geologies [25, 39]. Results from simplified performance assessment methodologies appear to compare well with the results of more complex methodologies [23]. It may be possible to use the simplified performance assessment methodologies as part of a software package that calculates repository doses for FCOs as a basis for a simplified measure for comparing the repository risks of the FCOs.

The ultimate goal should be to make calculation of the simple measure of long-term repository risk both rapid and flexible enough to allow efficient comparisons of the repository risks from various FCOs or technologies. The existence of codes such as ORIGEN2 [40] can provide a platform for this by calculating the generation and removal of radionuclides as a function of time. The simple repository risk measure can then be calculated as the amount of each radionuclide important to repository risk, as the weighted amount of each radionuclide with input weighting factors, or by combining the results of simplified repository performance assessment methodologies with the calculated radionuclide concentrations.

VII. SUMMARY AND CONCLUSIONS

Surrogate measures of long-term repository risk such as radiotoxicity are easy to calculate. However, comparison of ingestion radiotoxicity results with the results of existing performance assessments for a variety of repository media reveals that there is little correspondence between the radionuclides that dominate radiotoxicity and those that dominate dose to the public as calculated by repository performance assessments. In particular, radiotoxicity of spent nuclear fuel is dominated by transuranic actinides such as plutonium and Am-241 whereas repository risk in terms of dose to a member of the public is dominated by several long-lived fission products and activation products. These products are relatively soluble in ground water and have relatively low retardation rates during transport to and through the biosphere. The discrepancy between the radiotoxicity measure and the results of repository performance assessments has the potential to result in dysfunctional outcomes such as emphasis on removing actinides from SNF as a means to reduce repository risk.

An alternative measure is proposed for use in evaluating nuclear fuel cycle options (but not for repository siting or licensing) that reflects the results of existing performance assessments but which is still simple

to calculate. Existing performance assessments of repositories in a variety of geologic media (granite, clay, salt, deep boreholes) were analyzed to identify radionuclides that contribute greater than 0.0001% of the peak dose to the public during a one million year time horizon. This yielded the following list of radionuclides that are significant contributors to long-term repository dose to the public:

- I-129 dominates long-term repository dose by at least a factor of five in all but one of the 15 studies examined and even there it was a significant contributor to risk.
- Other frequently important contributors to long-term repository dose are C-14, Cl-36, and Se-79.
- Radionuclides that are sometimes important contributors to long-term repository dose are Tc-99, Sn-126, and Cs-135.

A simple example used this list of significant contributors to long-term repository risk by comparing the fission yield of the most significant fission product – I-129 – from U-235 fissions to the yield from fissions in other fissile materials. The comparison showed that using fissile materials other than U-235 would lead to higher amounts of I-129 in repository wastes than using U-235. Fuels that use mixtures of fissile materials would lead to intermediate results.

The situation concerning the two activation products in the list of significant contributors to repository risk (C-14, Cl-36) is less clear because the concentrations of their precursors in materials comprising nuclear fuels (N and Cl, respectively) are poorly known.

The proposed simple improved measure is by no means fully developed. Beneficial improvements to the measure are identified as follows

- Investigating how to address radionuclide risks from oxidizing repository media
- Obtaining data on the concentration of activation product precursors in present and future nuclear fuels, fuel assembly structural materials, and solid moderators.
- Obtaining data on the performance of thorium- and graphite-based fuels in repositories and repository performance assessment results.
- Determining how to address the differing profiles of dose curves from various radionuclides (sharp, short-duration peak vs broad, long-duration peak), and whether to address repository risks beyond one million years.
- Developing an approach for calculating weighting factors related to the importance of each radionuclide to repository risk.
- Facilitating calculation of the simple improved measure of long-term repository risk by

integrating the time-dependent calculation of radionuclide inventories and radionuclide weighting factors to enable parametric study of the dose implications of various fuel cycle options and technologies.

Implementing these improvements will involve a combination of conceptual development, evaluation of details in existing repository performance assessments, obtaining new data, and integrating modifications of existing software.

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