

# A QUANTITATIVE RISK ASSESSMENT DESCRIPTION OF A RADIONUCLIDE SOURCE TERM

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The purpose of this paper is to provide a numerical illustration of the details of the calculation of the release rate of a radionuclide from nuclear waste in a geologic repository using the quantitative-risk-assessment (QRA) approach described by Garrick.<sup>1</sup> This illustration is based on a hypothetical set of three events that can result in the release of radionuclides to the near-field environment which is defined as immediately outside a waste package. Two of these events are based on reality, one is strictly hypothetical. The reason for providing the numerical detail is to aid in illustrating how results are obtained, what the form of the results are, and the mathematical and numerical information required to obtain the results. The QRA result is in the form of a "probability of frequency" (POF) radionuclide release rate from the waste. This release rate is sometimes called the "source term."

## I. INTRODUCTION

The use of the Quantitative-Risk-Assessment approach for deriving a standard for a repository was recently described by Moeller.<sup>2</sup> Moeller brings to attention what the National Academy of Sciences and the National Research Council stated: "We recommend the use of a standard that sets a limit on the risk to individuals of adverse health effects from releases for (a) the repository." A primary reason for this statement is that if the limits were expressed in terms of dose, it would not be possible to estimate accompanying risk at times in the future because the relationship between dose and risk will change with time. The reason risk will change with time is because the baseline rates for consequences, such as cancers, will change due to medical progress in decreasing the current baseline consequences rate. The preference is that the performance of a repository be expressed in terms of the risk of receiving a given radiation dose.

"Risk" is not a number, but a collection of numbers, or more precisely a collection of curves that display scenarios, likelihoods, and consequences. The so-called "risk parameter" is usually expressed as the frequency with which an undesired event occurs in a probability-of-

frequency format. In the case of the disposal of nuclear waste the undesired event is the release of radionuclides to a water flow that can ultimately affect the environment and human health. Another way to describe "risk" is that risk can be thought of as a structured set of scenarios, their likelihoods and consequences. This structured set is generally depicted as an event tree or a number of event trees.

This QRA illustration is composed of two parts: the QRA technical approach, and a quantitative description of the chemistry for a radioisotope of interest. The chemistry chosen for this illustration is that of Murphy and Grambow<sup>3</sup> who describe interpretation and modeling of recent experimental data that yield thermodynamic constants for the distribution of trace Np(V) between aqueous solutions and uranophane for the sequestration of neptunium in uranophane. By using the results from Murphy and Grambow a more realistic source term description of the release of neptunium is obtained compared to using a neptunium solubility based on a neptunium-oxide controlling phase.

The QRA technical approach is cast in terms of a "set of triplets" written as:<sup>1</sup>

$$R = \left\{ \left\langle S_i, L_i, X_i \right\rangle_c \right\} \quad (1)$$

The brackets denote "the set of," R denotes the risk attendant to the system or activity of interest, S<sub>i</sub> denotes the i-th risk scenario (a description of something that can go wrong), L<sub>i</sub> denotes the likelihood of that scenario happening, X<sub>i</sub> denotes the consequence of that scenario, and the subscript c denotes "complete," meaning all, or at least all of the important scenarios, must be included in the set. Likelihood for each scenario can be quantified using appropriate probability distributions. For the purpose of illustrating the release rate of a radionuclide from nuclear waste, a nominal scenario is illustrated which is only one of many scenarios that can result in the release of a radionuclide. The use of the word "nominal" pertains to the nominal flow of percolation water across a waste package with subsequent corrosion and waste form

dissolution. Other scenarios include earthquakes and volcanism that can damage waste packages resulting in immediate release or damage that can affect the rate of corrosion or timing for failure due to corrosion.

A discussion of a QRA perspective on the source term for the proposed repository at Yucca Mountain was given by Murphy and coworkers<sup>4</sup> where a methodology was outlined for quantifying the radionuclide source term for a nuclear waste repository. The illustration that follows here is a numerical example based on that discussion. The release rate calculated by the QRA approach is a quantity of radionuclide “i” per unit time (a mass rate), and the quantity can be mass, curie, or mole. This illustrative example considers a nominal scenario comprising (only) three *events*: a one-dimension percolation flux that uniformly enters and flows through a drift (no flow focusing or defocusing), loss of a waste package by corrosion that allows percolation water to enter and leave the waste package as a function of time in proportion to the area of the waste package area corroded, and dissolution of the waste form and release of a radionuclide.

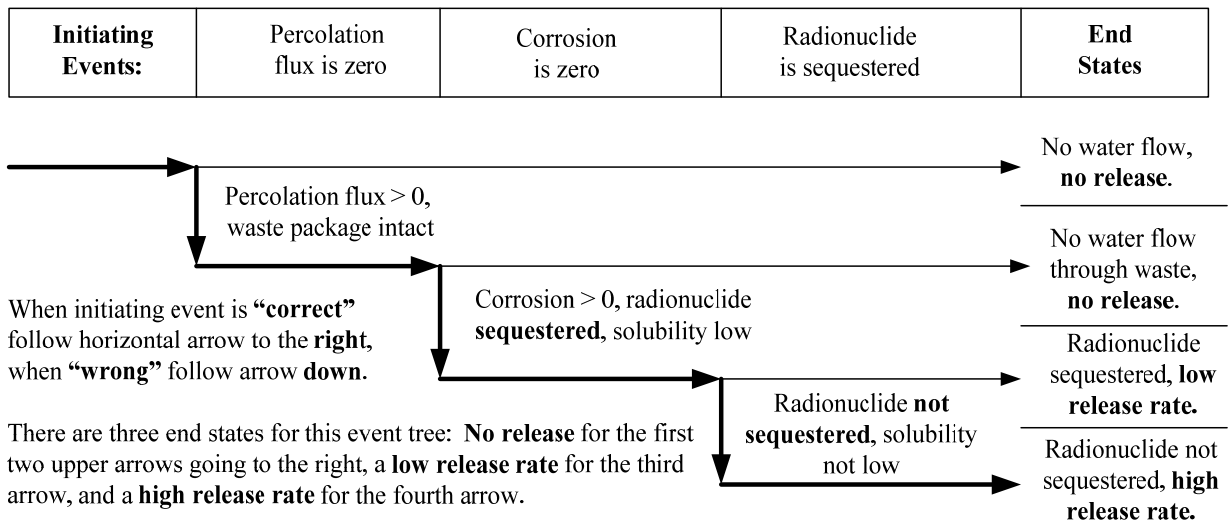
A realistic description of the dissolution of a radionuclide and its subsequent interaction in the local chemical environment should consider more than just the pure solid component solubility of a radionuclide which has sometimes been used by others.<sup>5</sup> Descriptions of dissolution and local chemical environment interactions that are more realistic are provided by Cui and coworkers<sup>6</sup> for a deoxygenated environment where significant amounts of radionuclides (U, Np, Tc, Sr) were found to be immobilized on the corrosion layer of iron canister material. Ferriss and coworkers<sup>7</sup> describe a corroding-iron environment that may sorb uranyl ions and may also immobilize other radionuclide species such as  $\text{TcO}_4^-$ ,  $\text{I}^-$ , and  $\text{NpO}_2^+$ . There is also the case of manufactured waste forms for actinides where the waste form dissolution rate is extremely low.<sup>8,9</sup> For the illustration that follows here the results by Murphy and Grambow<sup>3</sup> are used primarily because quantitative information is available, and thus this illustration is for the release of neptunium.

## II. SCENARIO DESCRIPTION

For the events noted in the preceding paragraphs consider the interpretation of the percolation flux in terms of risk assessment. Percolation flux,  $S$ , or water flow, across the waste is clearly something that can go wrong because it is desirable to have no flow. A disposal or storage site may have been chosen for exactly this no-water-flow characteristic. If the chosen site is dry, and remained so, there would be no need to assign a likelihood,  $L$ , of the percolation flux. But consider that

flow does occur and an assignment of the likelihood of the flow rate has to be developed. The consequences of percolation flux,  $X$ , is the flow of a quantity of water across the waste package. The corrosion of the waste package and radionuclide dissolution can be described in terms of risk assessment in the same manner as the percolation flux.

The three events comprising this scenario, percolation flux, corrosion, and radionuclide release, are illustrated in the event tree diagram in Figure 1. The interpretation of the logic of this event tree diagram is as follows: Enter this diagram on the upper horizontal line that begins on the left under the initiating events block and proceed to the right to the “percolation flux is zero” block. If the percolation flux is indeed zero then proceed on this horizontal line to the right and exit at the end state where “no water flow” appears, and subsequently there is no release of a radionuclide. However, in the event that the percolation flux is not zero, and this is a “what went wrong” event, proceed on the down arrow at the percolation event block to the second horizontal line. Given that the percolation flux is not zero there will be a probability distribution for the likelihood of occurrence of a percolation flux. Upon entering the second horizontal line proceed to the right to the corrosion event block. If corrosion is zero proceed to the right to the end state where “no water flow through waste” appears, and subsequently there is no release of a radionuclide. However, in the event corrosion is not zero proceed on the down arrow to the third horizontal line and then to the right to the “radionuclide is sequestered event.” Given that the corrosion rate is not zero there will be a probability distribution that describes likelihood of the rate of corrosion which is used to calculate how much of the waste-package surface area has been breached allowing percolation water to flow across the waste form. Clearly this waste-package surface area breach due to corrosion is a function of time and this event tree diagram is thus a “snapshot” in time. If the “radionuclide is sequestered” is true, or the radionuclide has a low solubility, then proceed on this third horizontal line to the end state “low release rate” and a radionuclide release rate will occur. However, in the event the radionuclide is not sequestered proceed on the down arrow at the “radionuclide is sequestered” event to the fourth horizontal line where the radionuclide has a high solubility. Proceed from here to the end state “high release rate.” There will be probability distributions that describe the likelihood for both the “sequestered” and “not sequestered” states. This event tree Boolean logic is that of an “and” series of events for the end state of a radionuclide release to occur.



**Figure 1. Event tree for the scenario comprised of three events (percolation, corrosion, dissolution) used in this illustrative calculation for the source-term frequency of release rate of a radionuclide. The dark-arrow path is that for which all events are “wrong” and results in the worse possible end state.**

In the illustration that follows the events that comprise the scenario are quantitatively described, and then the calculation steps for the QRA release rate of a radionuclide (call this radionuclide “i”) are described.

### III. EVENT DISTRIBUTIONS

The percolation flux likelihood is described by a lognormal probability distribution, chosen for illustration only, which yields the water flow rate across the horizontal plane of the waste package. It is this water flow rate that can enter, dissolve radionuclides, and leave the waste package and drift. For this illustration assume that the percolation flux immediately above the drift(s) is described by a glacial transition climate period which is assumed to occur at times greater than 2,000 years after emplacement of waste.<sup>10</sup> This percolation flux is assigned a mean of approximately 28.3 cm/year with a standard deviation of 5.1 cm/year (for the purpose of illustration only; the proposed Yucca Mountain repository reported 16.9 to 37.9 cm/yr, with a standard deviation of 5.1 cm/yr.<sup>11</sup>).

The likelihood of breach of the waste package by corrosion is described as the fractional area breached, or fraction of the area that is gone, as a function of time. This breach of the waste package description is not the same as used by others where when a waste package fails the water flux entering is assumed to be 5% of the wet percolation flux rate times the horizontal cross-sectional area of the package and the fraction of the container area

available to flow is thereafter constant.<sup>12</sup> In order to describe the likelihood of breach of a waste package fractional area as a function of time, which simply means the waste package continues to “open up” over time, there are two corrosion measures considered for this illustration. The first is the time at which half of the wall area is breached (or gone), and the second is the fractional area of the waste package that is breached as a function of time. The use of these two corrosion measures is described and illustrated in the following paragraphs.

The time at which half the waste package area is breached is based on the time it takes for a laboratory-measured corrosion rate to corrode through the waste-package wall thickness. This laboratory-measured corrosion rate is based on data described by a distribution with a mean and standard deviation. The waste-package wall thickness divided by the laboratory-measured corrosion rate yields the time at which the waste package would be instantly and completely “gone” if the corrosion rate were uniform over the waste package surface. The entire waste package is not expected to disappear instantly. Some portions of the waste package area will corrode slower and some faster. Slower and faster corrosion rates result in the waste package area being *fractionally* breached rather than the entire area instantly disappearing as a function of time.

The fractional breach of the waste-package area as a function of time is described by another distribution with

a mean time of occurrence and a standard deviation; the mean time of occurrence is the time for half of the waste-package surface to be breached (or disappear) as described in the preceding paragraph. Again, a standard deviation for the fractional area breached about the time of occurrence of the mean must be determined. How to make this determination is not known, but can possibly be accomplished through analysis of laboratory corrosion data

The calculation of the waste-package fractional-area-breached proceeds by generating a random number between 0 and 1. Call this random number  $n_r$ . From a cumulative lognormal distribution with a mean  $\mu$  (for this illustration 70,000 years) and sigma  $\sigma$  (for this illustration 10,000 years) for the time when half the waste package area is gone, determine the time given this random number  $n_r$ . Thus for each random number generated (or roll of the dice) a time when half the waste package area is gone will be generated. Call this time when half, also 50%, of the waste package area is gone  $\theta_{50}$ . Use this  $\theta_{50}$  as the mean along with another sigma (for example 20,000 years) to define a distribution (for example lognormal) for the fractional area breached as a function of time. The time for the calculation of the radionuclide release rate is specified, so use this specified time to determine the fractional area breached from the cumulative distribution for the fractional area breached. Each time a random number is generated a  $\theta_{50}$  time is generated. Use this  $\theta_{50}$  mean time for when half the waste package area is breached along with a sigma of 20,000 years to define a distribution for the waste-package fraction area breached. Use the specified time to calculate the fraction area breached by integrating this distribution up to the specified time.

Consider now qualitatively what the waste-package fraction area breached will look like when the mean time at which half the area is breached is 70,000 years. Considering a specified time of 10,000 years, much less than 70,000 years, the fraction area breached for numerous random numbers  $n_r$  is expected to be close to zero with a cluster of very small fractions just above zero, or the waste package is largely intact. At the other extreme a specified time of 100,000 years would yield a cluster of fractions very close to unity, or the waste package is mostly gone, very little intact area left. For a specified time around 70,000 years a cluster of fractions centered close to 0.5 is expected.

The solubility likelihood, which is also referred to as a distribution of concentration limits, has been discussed in a general perspective by Murphy and coworkers.<sup>13</sup> They note that both theoretical and empirical bases fail to provide definitive technical bases for concentration distribution functions and illustrate this with natural

nickel, lead and calcium concentration data. However for the purpose of this illustration the solubility likelihood of a radionuclide, sequestered or not, is described by a lognormal distribution, and the units for the solubility, or concentration, are whatever units are convenient. For the aqueous neptunium concentration controlled by sequestration in secondary uranophane, use the result of Murphy and Grambow<sup>3</sup> who report a total neptunium concentration of  $1 \times 10^{-9}$  m (0.00024 ppm) for a solution at equilibrium with uranophane containing this nuclide in solid solution at a Np/U ratio of 1/1,000. Assign a standard deviation of 0.0005 ppm (for this illustration only). For the not-sequestered radionuclide solubility use the result from Bernot<sup>4</sup> for the solubility (aqueous concentration) of neptunium of 10 ppm with an assigned standard deviation of 2 ppm (for this illustration only). The probability distribution parameters for each of the above events are summarized in Table 1

**Table I. Probability distribution parameters for illustrative release rate simulations.**

Parameter	$\mu$	$\sigma$
Percolation flux	28.3 cm/yr	5.1 cm/year
Time of half waste-package area failure	70 kiloyear	10 kiloyear
Fraction area failed	Obtained from random pick of time of half-area failed; kiloyear	20 kiloyear
Sequestered neptunium concentration	0.00024 ppm	0.00005 ppm
Not-sequestered neptunium concentration	10 ppm	2 ppm

For this illustration a lognormal distribution was used as described in Hahn and Shaprio<sup>14</sup> to determine all the likelihoods. However, if discrete data are available for these phenomena, then discrete probability arithmetic should be used because using "fitted" distributions introduces additional uncertainty.<sup>15</sup>

#### IV. FREQUENCY-OF-RELEASE-RATE SOURCE TERM

The frequency of release rate of radionuclide "i" is calculated using the Monte Carlo technique<sup>14</sup> to calculate the release rate for many simulations at a specified time. The release rates from these many simulations are then cast into the form of a histogram

from which a distribution can then be derived (such as a lognormal distribution). The results will be in the form of a probability-of-frequency (POF) release rate distribution for a *specified time*. A POF release rate distribution is derived for many specified times. The calculation procedure for a specified time is: (1) Generate a random number between 0 and 1 to pick (select) a percolation flux from the cumulative distribution. (2) Generate another random number between 0 and 1 to pick (select) the time of occurrence when half the waste package outer barrier area is breached. (3) Use the time of occurrence when half the waste package outer barrier is breached as the mean time for the fraction of the package area breached along with a specified standard deviation for the fraction of the package area breached. The resulting lognormal distribution from these two parameters yields the fraction of the package area breached as a function of time. Use the *specified time* to calculate the fraction of the package area breached from the cumulative distribution function. (4) Generate another random number between 0 and 1 to pick (select) a radionuclide solubility (concentration) depending on which branch is taken in Figure 1. Then calculate the rate of radionuclide release per package from:

$$release\ rate = flux \times (area\ fraction) \times (package\ area) \times concentration \quad (2)$$

Perform steps 1 through 4 and evaluate the release rate many times, N, for a *specified time*, and then plot the N results in the form of a histogram. Note that for each of the N evaluations the percolation flux, corrosion mean time, area breached, and radionuclide concentration are randomly picked. The resulting histogram is a plot of the number of occurrences of a release rate range as a function of release rate “bins.” These “bins” are release rate ranges chosen by the user. From this histogram a lognormal distribution (or whatever distribution is convenient) can then be derived.

## V. FREQUENCY-OF-RELEASE RESULTS

The probability-of-frequency release rate for this illustration is calculated for a set of specified times; these are 50, 70 and 120 kiloyears. These specified times are chosen to span the mean time for half the waste package to have corroded, 70 kiloyears, and the simulation results will show how the probability-of-frequency release rate increases with respect to time as more of the package is corroded away. The probability-of-frequency release rate for neptunium with a mean solubility of 10 ppm is presented in Figure 2, and in Figure 3 for the aqueous neptunium concentration of 0.00024 ppm controlled by

sequestration in secondary uranophane for a solution at equilibrium with uranophane containing this nuclide in solid solution at a Np/U ratio of 1/1,000; both of these figures are at a specified time of 70 kiloyears and derived from 1,000 Monte Carlo simulations. These figures present the histogram data, and the scaled lognormal distribution functions to fit the histogram data are also plotted. These probability-of-frequency release rate figures are typical in appearance for all the times specified.

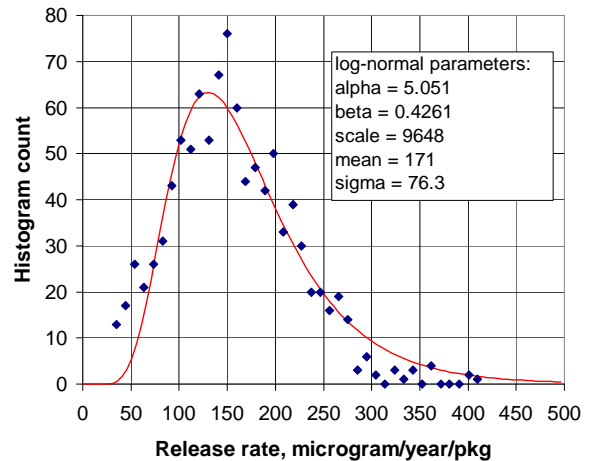


Figure 2. Probability of frequency release rate for neptunium at 70 kiloyears derived from a mean pure solid solubility of 10 ppm and 1,000 Monte Carlo simulations.

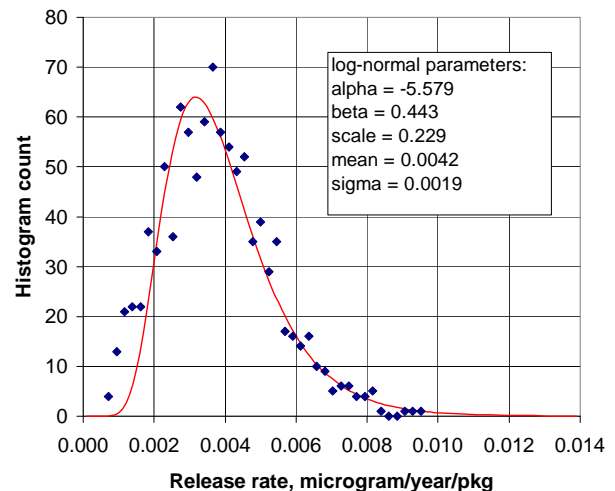
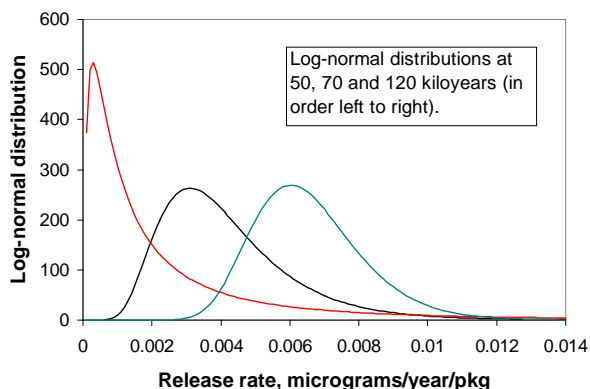


Figure 3. Probability of frequency release rate for neptunium at 70 kiloyears derived from a neptunium concentration of 0.00024 ppm controlled by sequestration in secondary uranophane and 1,000 Monte Carlo simulations.

From these figures it is apparent by visual inspection that the lognormal functions are approximately the same shape, and note that the  $\beta$  parameter for each are close; 0.426 for Figure 2 and 0.443 for Figure 3. The mean for the distribution in Figure 2 is 171  $\mu\text{grams/year/package}$  while for Figure 3 it is 0.0042  $\mu\text{grams/year/package}$ . These means clearly show the effect of the different concentrations; that due to a pure component solid and that of a sequestered component in a solid. The probability-of-frequency release rate changes with respect to time because the waste package corrodes with respect to time. This change is illustrated in Figure 4 for neptunium sequestered in uranophane at specified times of 50, 70 and 120 kiloyears.

The lognormal distributions in Figure 4 for the probability-of-frequency release rate for sequestered neptunium illustrate the progress of corrosion of the waste package. At 50 kiloyears, the fraction of the waste package corroded is relatively small compared to the mean time for half the package to corrode, which is 70 kiloyears. As such most of the release rates are “bunched” at small values. At 70 kiloyears the frequency of release has moved to higher values with a peak (not the mean) around 0.003  $\mu\text{gram/year}$  and the distribution still looks like a lognormal distribution (skewed to the left). But at 120 kiloyears the distribution peak has moved to 0.006  $\mu\text{gram/year}$  and the distribution has taken on a symmetrical shape; at this time most of the waste package is gone.



**Figure 4. Lognormal distributions for the probability of frequency release rate of neptunium concentrations controlled by sequestration in secondary uranophane for specified times of 50, 70 and 120 kiloyears and 1,000 Monte Carlo simulations for each time.**

## VI. CONCLUSIONS AND OBSERVATIONS

At this point in this illustrative exercise the probability-of-release rate results would be applied to a

repository containing many waste packages (possibly thousands). The transport of radionuclides to the accessible environment would be described by additional events in the event tree in Figure 1. These additional events would be based on how water flow would transport the radionuclide to the accessible environment and include any interactions and chemical reactions with the media through which the flow would occur. The consequences of the release of this, or any radionuclide, are the impacts of the dose received on human health as the result of drinking water and consuming food that contains this radionuclide at the accessible environment. The consequences would be illustrated by the frequency of occurrence of low doses being relatively high while the frequency of occurrence of high doses would be relatively low. The operative word here is “relatively” because to receive a high dose the radionuclide solubility has to be relatively high and the water flow rate relatively high (greater source strength) at any particular time. An illustration of a high frequency of occurrence for low doses and a low frequency of occurrence for high doses is given by Garrick and coworkers<sup>16</sup> where 31 scenarios were considered for a QRA for the New York State operated West Valley radioactive waste disposal area.

Clearly, quantitative information describing the likelihood of these events is required; this is especially true for the “chemistry.” Qualitative information is insufficient. It has become apparent that attention has been and is being given to the “chemistry,” but it is also clear that more effort is required to fully take into account the interactions and chemical reactions of the myriad of radioisotopes in spent nuclear fuel. A discussion is given by Bernot<sup>4</sup> on what additional information is required to predict the dissolved neptunium concentrations likely to be controlled by the solubilities of Np-bearing corrosion products. This particular discussion by Bernot is exactly what is discussed by Garrick and Kaplan<sup>17</sup> regarding an “information gathering” option in a decision-theory perspective on the disposal of high-level radioactive waste; go get some more information (on the chemistry). There is also an engineering component that has to occur when the chemistry of interest requires the addition of a component that does not occur in sufficient quantity naturally, such as iron. The addition of iron to the system can be intentional by adding steel shot to the interior of the waste package along with the waste, or the addition of a thick steel overpack on the waste package can be installed. In each case corroding iron would be immediately available in and outside the waste package through which the radioisotopes would flow.

The QRA technical approach can be derived from and reviewed in many sources, a few of which are cited

here. However, it is clear that quantitative information has to be developed to derive a realistic source term. This quantitative information includes, and is only part of the larger conceptualization, the breaching of the waste package as a fractional area lost as a function of time, and the chemistry of the radionuclides of interest in the local environment. To that end experimental programs will have to be designed and carried out to obtain quantitative information so that the risk assessment will be “evidence based.”

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