CALCULATION OF DOSE RATES FROM CRITICAL RADIONUCLIDES IN LOW-LEVEL RADIOACTIVE WASTE DISPOSAL

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ABSTRACT

Dose rates to persons using a well near a low-level radioactive waste disposal site for drinking and irrigation are calculated. Major contributors to those dose rates are shown to be tritium, carbon-14, and iodine-129. Suggestions for reducing potentially high dose rates from tritium and carbon-14 are provided. However, the volume of the waste containing iodine-129 and the long life of that isotope make the methods used for tritium and carbon-14 either technically impractical or costly. Suggestions are made for further research into the assumptions underlying the predictions of serious dose rates from iodine-129.

INTRODUCTION

Many pathways for migration of radionuclides from a low-level waste disposal facility have been postulated and analyzed. One pathway--movement downward from the waste into an aquifer and thence to a well used by humans for drinking or irrigation--is almost always revealed by analyses to be one of the critical paths of nuclide migration whenever significant percolating water is present at the disposal site. Any well close to the disposal site and downstream (in the sense of the aquifer flow) has the potential of intercepting radionuclides in concentrations essentially identical to those leaving the bottom of the waste at the disposal site.

This paper describes analyses of potential dose rates to users of water drawn from a well 50 meters from the nearest point of a low-level waste disposal site. While the analysis employs parameters typical of a shallow land disposal facility for low-level waste, some of the conclusions drawn are also valid for other disposal methods being considered.

The analysis described below uses the average U.S. nuclide concentrations for low-level waste as prescribed by the NRC in Ref. 1 and 2. The RAEPATH code used is described in Ref. 3, and its output was compared to the results given in Ref. 1 for concentrations and dose rates from wells near the disposal site. The critical radinuclides in the water withdrawn from the well are determined and separate methods of dealing with each nuclide to reduce its potential impacts on users of the well water are discussed.

TECHNICAL APPROACH

The RAEPATH computer code used in the analyses reported represents migration of radionuclides from a shallow land burial facility to a nearby well in the manner illustrated in Fig. 1. Water percolating through waste is assumed to contain a certain concentration of each nuclide of interest as it moves vertically downward from the bottom of the waste volume to the aquifer below. The concentration of each nuclide in the water depends on its concentration in the waste and its release rate from the waste. Because there is little opportunity for mixing with the aquifer water in the short horizontal distance (50 meters) of travel from under the waste volume to the well, it is assumed that the contaminated water forms a distinct layer in the aquifer, as shown. This is essentially valid for the more mobile nuclides, which have little opportunity for lateral diffusion as they move toward the well.

The well is conservatively assumed to withdraw water from the largest possible thickness of the contaminated layer. This thickness is limited only by the assumed perforated length of the well casing or the thickness of the contaminated layer, whichever is smaller. If the contaminated layer is smaller than

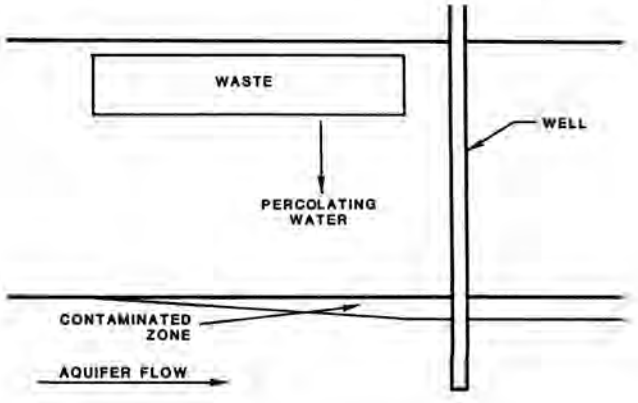

Fig. 1. Relationship of the well to the disposal facility.

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the perforated length of the well casing, then the contaminated water is assumed to be mixed with pure water drawn in through the rest of the perforated casing.

The analysis using RAEPATH assumed the disposal facility and site parameters shown in Table I. The number of mesh points listed in the table indicates the number of line sources perpendicular to the aquifer flow direction that were used to represent the release of nuclides from the waste. Two aquifer velocities were analyzed--1 meter per year and 10 meters per year. The parameters in Table I were chosen to be representative of a site in the northeastern U.S. The nuclide-specific retardation coefficients used in the analysis are those in set 3 in Table R.15 of Ref. 1 which correspond to a soil that has retardation characteristics between those of clay and sand.

Two sets of release rates were used in the calculation of nuclide concentrations in water that passes through the waste. One set assumed that the water leaves the trench bottom chemically saturated (i.e., the nuclides are completely equilibrated between the waste and the percolating water). In the terminology of Ref. 1 and 4, the "contact time" for this case is unity. The second set of release rates assume equilibrium is not obtained and represents a contact time less than unity. These release fractions were calculated using the alogorithm of the GRWATER code described in Appendix H in Ref. 1. The two sets of nuclide-specific release rates are given in Table II.

Dose conversion factors for ingestion were taken from ICRP Publication 30.[5] Annual uptakes of well water and food grown using well water for irrigation, as well as bioaccumulation factors, were calculated in the manner specified in Ref. 5.

CALCULATED DOSE RATES

Dose rates to an individual using water from a well 50 meters from the edge of the trenches are indicated for the two aquifer velocities in Figs. 2 and 3. The safety factor plotted in these figures is the ratio of the maximum permissible organ dose prescribed in 10CFR61 to the organ dose calculated. The plots show the critical nuclide at each low point in the safety factor. It can be seen that the critical nuclides are the highly mobile tritium, carbon-14 and iodine-129. With the higher aquifer velocity, the tritium has a greater impact because there is less opportunity to decay before reaching the well.

TABLE I

Site Parameters

Length of burial area parallel to aquifer flow	750 m
Width of burial area	267 m
Thickness of waste	5 m
Total waste volume	5×10^5 m^3
Waste density	1400 kg/m^3
Horizontal aquifer velocity	1, 10 m/yr
Aquifer porosity	0.30
Vertial distance from aquifer to waste	1.0 m
Average vertical water velocity	0.3 m/yr
Percolation rate (meters of water per year)	0.10 m/yr
Number of mesh points (sectors)	20
Distance between waste volume and well	50 m
Length of perforated well casing	10 m

TABLE II

Release Fractions

Radionuclide	Release Fraction (unit contact time) (yr^{-1})	Release Fraction Calculated Using GRWATER Algorithm (yr^{-1})
H-3	2.30×10^{-2}	4.70×10^{-4}
C-14	1.15×10^{-4}	2.36×10^{-6}
Fe-55	2.96×10^{-4}	6.06×10^{-6}
Co-60	2.96×10^{-4}	6.06×10^{-6}
Ni-59	2.96×10^{-4}	6.06×10^{-6}
Ni-63	2.96×10^{-4}	6.06×10^{-6}
Sr-90	1.97×10^{-4}	4.03×10^{-6}
Nb-94	2.22×10^{-4}	4.54×10^{-6}
Tc-99	2.30×10^{-3}	4.70×10^{-5}
I-129	2.30×10^{-3}	6.63×10^{-8}
Cs-135	3.24×10^{-6}	6.63×10^{-8}
Cs-137	3.24×10^{-6}	2.00×10^{-4}
Ra-226	2.00×10^{-4}	2.00×10^{-4}
Th-230	2.00×10^{-5}	5.11×10^{-8}
U-235	2.50×10^{-6}	5.11×10^{-8}
U-238	2.50×10^{-6}	1.91×10^{-7}
Np-237	9.34×10^{-6}	1.91×10^{-7}
Pu-238	9.34×10^{-6}	1.91×10^{-7}
Pu-239	9.34×10^{-6}	1.91×10^{-7}
Pu-241	9.34×10^{-6}	1.91×10^{-7}
Pu-242	9.34×10^{-6}	1.91×10^{-7}
Am-241	8.22×10^{-5}	1.68×10^{-6}
Am-243	8.22×10^{-5}	1.68×10^{-6}
Cm-243	9.34×10^{-6}	1.91×10^{-7}
Cm-244	9.34×10^{-6}	1.91×10^{-7}

Carbon-14 also has a slightly greater impact with the higher aquifer velocity. Peak dose rates (minimum safety factors) caused by iodine-129 are independent of aquifer velocity because of I-129's long (17 million years) half-life.

Figures 2 and 3 also illustrate the effects of the two contact times used in the analyses. Peak dose rates (minimum safety factors) are more pronounced when the conservative unity contact times are used in the analyses. However, even with contact times calculated according to the GRWATER code (NRC Contact Times in Figures 2 and 3) are used, unsatisfactory safety factors are predicted due to the presence of tritium and iodine-129. It is also worth noting that the release fractions calculated using the NRC contact times, shown in Table II, are extremely small--generally smaller than those required by the NRC for high-level waste after container failure. They may be unrealistically small and are at least open to question.

Briefly, the analysis has shown that using a published inventory of nuclide concentrations in average U.S. low-level waste, a computer code that compares well with at least one other published code and typical shallow land burial parameters the mobile nuclides tritium, carbon-14 and iodine-129 were calculated to cause peak dose rates that approach or exceed the limits specified in 10CFR61. The remainder of this paper will address methods for resolving this difficulty.

DISCUSSION

Reducing the potential threat from tritium is relatively easy because of the short half-life (12.3 years) of this nuclide and the fact that most of the tritium in the U.S. inventory comes from a few sources. Burying the majority of the tritium in long-

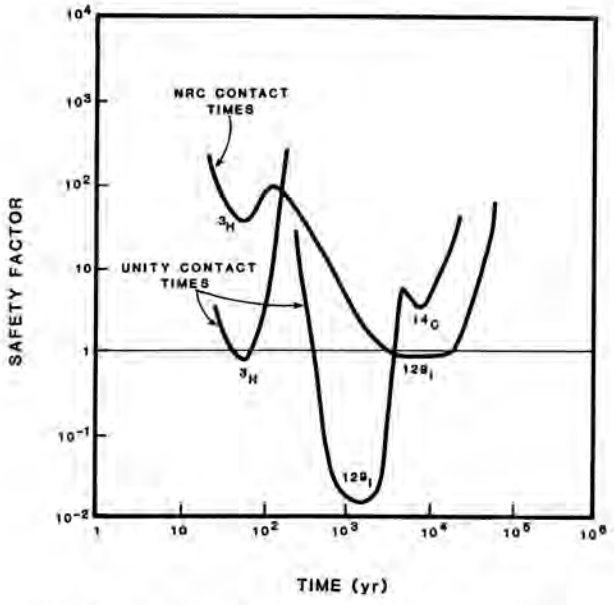

Fig. 2. Well pathway safety factor - 1 m/yr aquifer velocity.

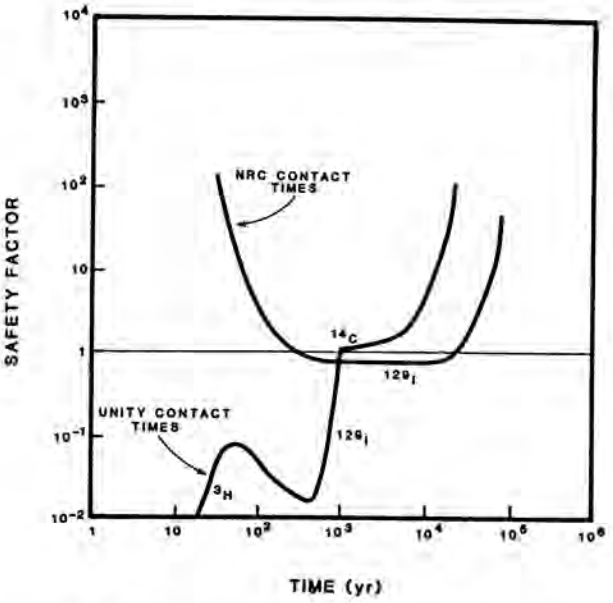

Fig. 3. Well pathway safety factor - 10 m/yr aquifer velocity.

lived containers can greatly reduce potential peak dose rates from this nuclide. Figure 4 shows the normalized potential peak dose rate from tritium as a function of container life if 90 percent of the tritium activity is placed in long-lived containers. (The remaining 10 percent of the tritium is assumed to be available for release at some specified rate immediately after burial.) It can be seen that a container life of more than 40 years is of no value in reducing the peak dose rate. That is, while using a long-lived container can reduce the peak potential dose, there is no benefit to using containers that can last beyond 40 years. Of course this limit is a function of the 90 percent/10 percent split assumed. If 99 percent of the tritium was assumed to be in long-lived containers the cutoff would be slightly more than 40 years.

The impacts of carbon-14 and iodine-129 on doses to maximally exposed individuals cannot be reduced by placing them in long-lived containers because of their long half-lives--5,700 years and 17 million years respectively. Assuring the life of containers over periods of these magnitudes, and longer, is difficult or impossible. However, most (more than 98 percent according to the source terms in Ref. 1 and 4 of the carbon-14 in U.S. low-level wastes comes from industrial source manufacturing. Those wastes exceed the allowable concentrations for Class C wastes and represent only a small fraction (around 0.01 percent) of U.S. low-level waste volumes. For these reasons, disposal in facilities that are more secure than shallow land burial or most of its alternatives is likely to be required anyway and will not impose an excessive cost burden on the country in general.

The most difficult problem to address is how to deal with the calculated dose rates from iodine-129. Most alternatives to shallow land burial offer no potential relief because of this isotope's long half-life. The alternatives usually suggested are not likely to provide significantly better protection from release long enough to distinguish them from shallow land burial where iodine-129 is concerned. Also, the waste streams indicated in Ref. 1 and 4 as contributing about 90 percent of all I-129 constitute over 12 percent of U.S. low-level waste volume; therefore the total cost of using expensive methods for their treatment and disposal will be significant.

The thesis of this paper is that many elements of calculations of iodine-129 doses from low-level radioactive wastes should be reexamined in the light of Figs. 2 and 3. Simply stated, predictions of doses to maximally exposed individuals from iodine-129 in average U.S. low-level wastes, based on generally accepted waste inventories, dose conversion factors, and release rate calculations, and based on a computer code that produces similar results to the NRC suite of codes--for the problem under investigation--exceed dose rate limits given in 10CFR61. There are a number of reasons why these high predicted dose rates may have nothing to do with actual dose rates that may occur. These reasons are discussed briefly below. The intent of this paper is to prompt a reexamination of elements of the dose rate calculation to see if there is reason to believe they are overly conservative.

There are at least three areas in the prediction of iodine-129 dose rates that are worthy of close scrutiny. They are:

- The waste streams that References 1 and 2 indicate to be the major sources of iodine-129 in U.S. low-level wastes.
- The possibility of dilution of I-129 released from low-level wastes by stable iodine in the soil.
- The underlying assumptions behind the iodine-129 ingestion dose conversion factor given in Reference 6.

References 1 and 2 indicate that the major source of I-129 in U.S. low-level wastes are ion exchange resins, non-compactible trash and filter sludges from boiling water reactors. Together these three waste streams account for about 90 percent of all I-129 in U.S. low-level wastes and over 12 percent of the volume of low-level waste. The concentrations of iodine-129 given for these waste streams is about that of the threshold level for most commonly used methods

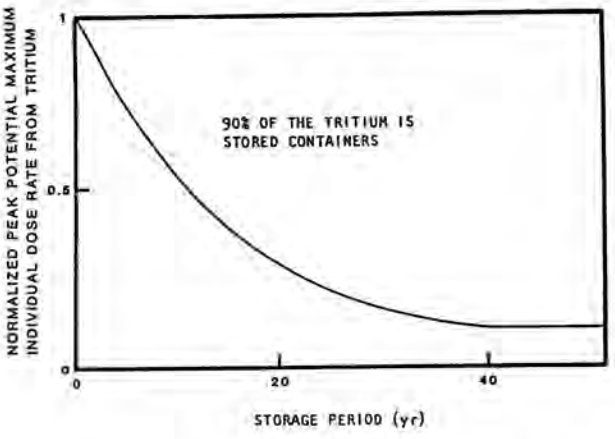

Fig. 4. Reduction in potential dose from tritium by storage.

for making measurements of this isotope in the wastes. Therefore, it is difficult to prove by direct measurement that average I-129 concentrations in these waste streams are substantially lower than those given in the references. There are claims to that effect derived from basic calculations. A thorough program to determine if it is possible to measure or infer substantially lower I-129 concentrations in these and perhaps in other reactor waste streams, if successful, would permit a lowering of the peak predicted dose rates from this isotope that are evident in Figs. 2 and 3.

Dilution of the I-129 by stable iodine (I-127) present in the soil has been proposed as a means of reducing the impacts of I-129 from waste disposal sites. This phenomenon was not modeled in the calculations described above. From the information in Soldat [7] it can be calculated that a thousandfold dilution is needed to keep thyroid dose rates at or below the 75 mrem/yr limit set in 10CFR61. That is, if the I-129/I-127 ratio in the thyroid is assumed to be the same as is in food and drink ingested, the food and drink must not contain more than 1 atom of I-129 for every thousand atoms of I-127. Oztunali [8] provided a good discussion of the prospects for such dilution and the current inability to state with any certainty whether it will take place.

Key difficulties in predicting the role of dilution include questions about the chemical similarities between iodine in the waste and in the soil and the observation that if the iodine in the soil was soluble enough to act as a dilutant for that in the waste it would have all washed out of the soil a long time ago. While dilution with natural iodine is suggested by some as a viable mechanism for lowering the dose rate from iodine-129, Oztunali indicates there are many open questions regarding this phenomenon.

The assumptions underlying the ingestion dose conversion factor for I-129 given in Ref. 6, or any similar dose conversion factors, may need to be reexamined. The dose conversion factor gives a ratio of mrem/yr dose to the thyroid to pCi/yr of I-129 ingested. If the model [6] that indicates dose rate to the thyroid is proportional to the ratio of I-129 to stable iodine in the food and drink ingested is used, then any dose conversion factor must be based on an assumed ratio. Since the thyroid will only retain a narrow range of iodine atoms a statement of dose rate per total ingested iodine-129 activity must be based on several assumptions about total annual iodine intake and the dilution ratio. These assumptions should be reexamined in the light of the questions concerning natural dilution discussed by Oztunali and of the potentially critical role played by iodine-129 in dose rates from low-level waste disposal that is illustrated in Figs. 2 and 3.

One additional area of interest is the dose rate limit (75 mrem/yr) given for the thyroid in 10CFR61. Measures of relative cancer risk from radiation doses to different organs [9] indicate that the risk from a given dose to the thyroid is as much as eight to ten times lower than the risk from the same dose to some other organs. 10CFR61 gives a 25 mrem/yr dose rate limit to all organs other than the thyroid. If 25 mrem/yr is appropriate for other organs then a limit between 200 and 300 mrem/yr seems more appropriate for the thyroid, based on relative cancer risks.

SUMMARY

This paper has presented calculations which predict potential individual dose rates from iodine-129 in low-level wastes. The calculations are based on published inventories and release rates and typical disposal site parameters and the predicted dose rates approach or exceed the limits stated in 10CFR61. Several key considerations in predicting dose rates from iodine-129 along the critical pathway are recommended for close scrutiny. If any one of them is found to be overly conservative the concern over iodine-129 ingestion doses raised here can be considerably lessened.

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