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December 13, 1999
Contract No. NRC-02-97-009
Account No. 20-1402-871

U.S. Nuclear Regulatory Commission
ATTN: Dr. John W. Bradbury
Division of Waste Management
Two White Flint North
Mail Stop TD-13
Washington, DC 20555

Subject: Transmittal of the deliverable "Effect of Reducing Chemistry on Technetium Transport—Materials Research Society Manuscript Paper"

Dear Dr. Bradbury:

This letter transmits "Effect of Reducing Chemistry on Technetium Transport—Materials Research Society Manuscript Paper" (Intermediate Milestone 20.01402.871.000). The final title for this deliverable is "Technetium-99 Chemistry in Reduced Groundwaters: Implications for the Performance of a Proposed High-level Nuclear Waste Repository at Yucca Mountain, Nevada." It will be submitted for publication in the proceedings volume for the Materials Research Society Symposium on the Scientific Basis for Nuclear Waste Management XXIII.

This work was conducted as an activity under the Radionuclide Transport (RT) Key Technical Issue (KTI). In the current U.S. Department of Energy (DOE) Total System Performance Assessment (TSPA) for the proposed Yucca Mountain high-level nuclear waste repository, Tc-99 has consistently represented a significant proportion of the dose to the critical group. This is due to the high solubility and low sorption of technetium under oxidizing conditions. As part of its site characterization activities in preparation for a license application, the DOE has obtained Eh-pH measurements at various depths from the saturated zone in a single well in fractured tuff, indicating localized zones of reducing conditions. Under reducing conditions, technetium has low solubility and is strongly sorbed. The work presented in this manuscript used the NRC TPA Version 3.2 code to explore the potential effects of reducing conditions on Tc-99 solubility and transport and the resulting impact on repository performance. This manuscript documents ongoing issue resolution activities in the RT KTI.

The sensitivity analyses indicate that localized reducing conditions have the potential to significantly reduce the dose from Tc-99 to the critical group. Although there is some effect in the transport of technetium through the fractured tuff aquifer in TPA Version 3.2, the major effect is due to increased retardation in the alluvium. This is due to treating the alluvial aquifer as a homogeneous porous medium with readily accessible sorption sites. The suggested importance of the alluvium in this analysis underscores the importance of alluvium characterization, either as part of site characterization, through the Nye County Early Warning Drilling Program, or as part of performance confirmation.

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The results of these sensitivity analyses will be used to investigate alternative conceptual models, provide added realism to TPA, and to develop the flexibility to evaluate changes in future DOE work as new data come out on redox conditions in the SZ and the alluvium. These analyses will also be used to develop the technical bases for acceptance criteria and review methods in the Yucca Mountain Review Plan.

If you have any questions about this deliverable, please call me (210) 522-5540 or Dr. David Turner (210) 522-2139.

Sincerely,

E. C. Pe

English C. Pearcy, Manager
Geohydrology and Geochemistry

ECP:ar

cc: J. Linehan, B. Stiltenpole, B. Meehan, J. Greeves, J. Holonich, W. Reamer, K. Stablein, D. Brooks, S. Wastler, J. Ciocco, B. Leslie, D. Esh, W. Dam, N. Coleman, T. McCartin, C. Lui, W. Patrick, CNWRA Directors, CNWRA Managers, CNWRA ISI Leads, T. Nagy (SwRI Contracts)

TECHNETIUM-99 CHEMISTRY IN REDUCING GROUNDWATERS: IMPLICATIONS FOR THE PERFORMANCE OF A PROPOSED HIGH-LEVEL NUCLEAR WASTE REPOSITORY AT YUCCA MOUNTAIN, NEVADA

Roberto T. Pabalan, David R. Turner, and Michael P. Miklas, Jr.
Center for Nuclear Waste Regulatory Analyses, Southwest Research Institute
6220 Culebra Road, San Antonio, TX 78238-5166 (rpabalan@swri.edu; dturner@swri.edu)

ABSTRACT

Performance assessment calculations by the U.S. Department of Energy and the Nuclear Regulatory Commission indicate that Tc-99 is a major contributor to dose to a hypothetical receptor group 20 km downgradient of a proposed high-level nuclear waste repository at Yucca Mountain, Nevada, within the first 10,000 yr after permanent closure. This result is due in large part to the high solubility and low retardation of Tc under oxidizing conditions in the Yucca Mountain environment. Recent site characterization data on the chemistry of saturated zone groundwater at Yucca Mountain and vicinity indicate the presence of locally reducing geochemical conditions, which could decrease the solubility and enhance the sorption and retardation of Tc-99. In this study, a preliminary assessment of the potential effects of reducing conditions on the transport and release of Tc-99 was conducted. Sensitivity analyses using the NRC/CNWRA Total-system Performance Assessment code (TPA Version 3.2) indicate that decreased Tc solubility and increased Tc sorption due to reduction of Tc(7+) to Tc(4+) can significantly delay the arrival of Tc-99 at the receptor group location. Decreased Tc solubility can decrease the Tc-99 dose by three orders of magnitude relative to the TPA 3.2 base case. Enhanced Tc retardation in the tuff aquifer only does not greatly decrease the calculated Tc-99 peak dose, whereas increased Tc retardation in the alluvial aquifer alone prevents Tc-99 from reaching the receptor group in 50,000 yr. The release and transport of other redox-sensitive radioelements could be affected in a manner similar to Tc. Thus, reduced groundwater conditions could significantly enhance the performance of the geologic barrier system and reduce the dose to the receptor group.

INTRODUCTION

Technetium-99 is a radionuclide of concern in safety assessments of high-level nuclear waste (HLW) repositories because of its long half-life (2.13×10^5 yr) and its relative abundance in HLW (fission yield ~6 percent). A redox-sensitive element, Tc exists in a heptavalent (7+) state under nonreducing conditions as the pertechnetate (TcO_4^-) anion (Fig. 1). The TcO_4^- anion does not sorb significantly onto mineral surfaces, and compounds of Tc(7+) generally have high solubility so that the concentration of Tc in groundwater is not expected to be limited to low values by solubility constraints. Conversely, under reducing conditions Tc is present in a tetravalent (4+) state. The solubility of Tc(4+) solids, such as $TcO_2 \cdot 1.6H_2O$ (Fig. 1), is much lower compared to Tc(7+) solids, and Tc(4+) aqueous species sorb more strongly onto mineral surfaces compared to Tc(7+) species. For example, sorption experiments by Lieser and Bauscher[1] using sediments, predominantly quartz, and groundwaters from Gorleben, Germany, indicate that at pH = 7 and Eh > 0.17 V, in the stability field of TcO_4^-, Tc K_d is low and relatively constant at ~ 0.2 mL/g (Fig. 2). At lower Eh, where the Tc(4+) species $TcO(OH)_2$(aq) is the dominant aqueous form, K_d is high, with a value of ~ 1,000 mL/g. Although Lieser and Bauscher[1] indicated that precipitation of low solubility Tc(4+) solids may have contributed to Tc removal in their experiments, the effect of low Eh on sorption is significant.

As part of the United States HLW program, the Department of Energy (DOE) and the Nuclear Regulatory Commission (NRC) conducted total-system performance assessment (PA) simulations of the proposed geologic repository at Yucca Mountain, Nevada (YM).[2,3,4] The DOE and the NRC use PA to systematically analyze what can happen at the repository after permanent closure, and to calculate as a function of time the total dose affecting a hypothetical human receptor group located some distance downgradient from YM in the event of failure of waste packages (WPs) to isolate the radionuclides. The

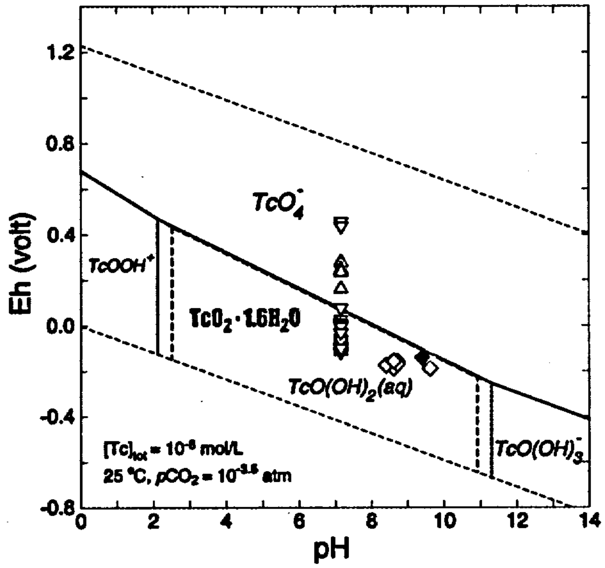

Figure 1. Eh-pH diagram for aqueous Tc species calculated using the NEA thermodynamic data.[9] Also shown are Eh measured in well UE-25 WT#17 (Δ,∇)[5], and on samples from wells USW H-3 (♦) and USW H-4 (◇).[6]

NRC PA activity, part of an ongoing process at the NRC to prepare for the review of a potential DOE license application for a HLW repository at YM, is designed to provide information on the potentially important isolation characteristics and capabilities of the proposed repository system based on the available information regarding the geologic setting and the natural and engineered barrier systems. An important aspect of the NRC PA is quantifying the sensitivity of the results to, and the uncertainty associated with, the input parameters.

In the DOE and NRC PA analyses, oxidizing conditions were assumed over the entire transport path from YM through both the unsaturated and saturated zones. Under these conditions, there is no solubility limiting phase for Tc-99, and the PA source term models for Tc assumed a high solubility limit such that Tc release is essentially constrained by the dissolution rate of the waste form. After mobilization from the waste form, the dominant aqueous species is assumed to be TcO_4^-, and Tc sorption or retardation, modeled using a K_d or a retardation coefficient (R_f), is typically assumed to be either very low[3,4] or zero[2] over the entire transport path length. Because of the assumed high solubility and low retardation, both the DOE and the NRC PA results indicate that Tc-99 is a dominant contributor to the dose rate during the first 10,000 yr after closure of the repository and remains as a significant contributor to the total dose at longer times.[2,3]

Some site characterization data suggest that localized reducing conditions may exist in the groundwaters at Yucca Mountain. These data include downhole measurements of Eh in well UE-25 WT#17 (Fig. 3),[5] Eh measurements on a water sample from well USW H-3 and on "thief" samples from well USW H-4,[6] and indirect evidence in the form of reduced sulfide minerals observed in cuttings from NC-EWDP-1D,[7] a well located in the alluvial zone. In this study, a preliminary assessment of the potential effects of reducing conditions on Tc-99 transport and release and on the performance of the proposed repository system was conducted using the NRC/CNWRA PA code (TPA Version 3.2).

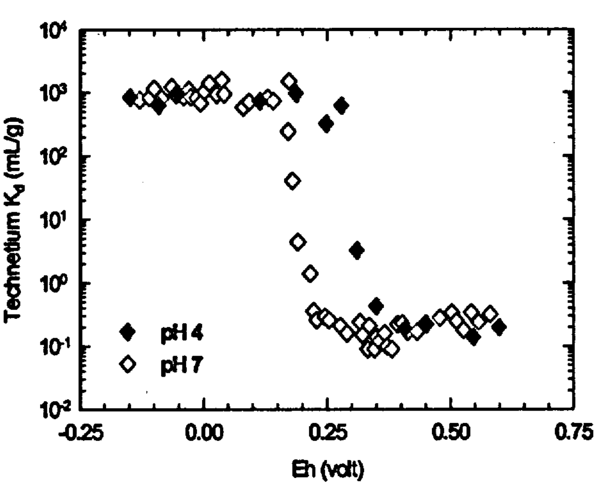

Figure 2. Technetium sorption coefficient, K_d, as a function of Eh. Values taken from Lieser and Bauscher[1]

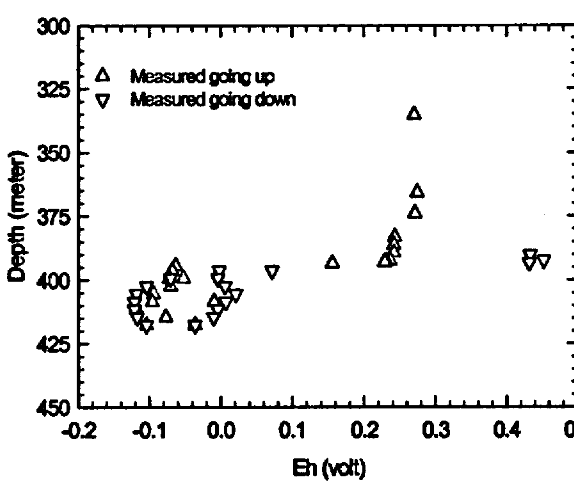

Figure 3. Eh measured downhole in well UE-25 WT#17. Data taken from DOE (1999; data tracking number LAAM831311AQ98.003)[5]

TECHNICAL APPROACH

The NRC has selected a base case (defined as a particular set of models and model parameters describing the YM repository system) to perform sensitivity and uncertainty analyses and to study the general behavior of the system as simulated by the TPA Version 3.2 code.[4,8] Because oxidizing conditions were assumed throughout the geologic barrier system at YM, previous NRC PA calculations assumed zero retardation of Tc in the fractures and matrix of the volcanic tuff in the saturated zone, and a log uniform probability distribution function, with minimum and maximum values of 1 and 30, respectively, was assumed for Tc R_f in the alluvium.[8] In addition, the solubility limit of Tc in the NRC PA calculations was set to a high value (1 mol/L). To investigate the effects of saturated zone reducing conditions on Tc-99 transport and repository performance, the Tc R_f parameters for the saturated zone and the Tc solubility limit were modified from the TPA Version 3.2 base case (oxidizing) values.

Downhole measurements of Eh and pH in well UE-25 WT#17[5] indicate an average pH of 7.14 ± 0.09 and Eh values less than 0.17 V below a depth of about 390 m (Fig. 3). In the absence of data on Tc sorption on Yucca Mountain rocks under reducing conditions, the Tc K_d of 1,000 mL/g derived by Lieser and Bauscher[1] for pH 7 and low Eh was used. For Tc-99 sorption on the matrix of volcanic tuff in the saturated zone, this K_d corresponds to an R_f of 2.6×10^4, based on porosity (ϕ) and bulk density (ρ) base case values of 1 percent and 2.65 g/cm^3, respectively.[8] Zero retardation ($R_f = 1$) of Tc in the fractures of volcanic tuff was maintained in the calculations due to uncertainty about the type and extent of fracture coatings. In TPA Version 3.2, transport through alluvium is modeled using a porous media approach, and there is no component of fracture flow and transport. A Tc R_f value of 1.7×10^5, based on a K_d of 1,000 mL/g, ϕ of 12.5 percent, and ρ of 2.47 g/cm^3, for transport through the alluvium was used in the sensitivity analyses. The TPA Version 3.2 does not have the capability to handle localized groundwater conditions, although one can specify different transport properties for the tuff and the alluvial aquifers. In this study, higher retardation factors are used (i) over the entire length of the saturated zone flow path from the repository to the receptor group location 20 km away, (ii) for the tuff aquifer only, or (iii) for the alluvial aquifer only.

The Tc solubility limit used in the sensitivity analyses, calculated using the NEA Tc thermodynamic data[9] and the chemistry of groundwater reported for UE-25 WT#17[5,10] (data tracking numbers LAAM831311AQ98-001, -002, and -003) and summarized in Table 1, is 4×10^{-9} mol/L. The solubility-limiting phase was assumed to be $TcO_2 \cdot 1.6H_2O$. Using an anhydrous phase, TcO_2, results in a much lower calculated solubility (4×10^{-13} mol/L), but $TcO_2 \cdot 1.6H_2O$ is more relevant to the formation of Tc oxides in reducing groundwaters.[11]

Table 1. Cation and anion concentrations in water samples taken from well UE-25 WT#17[5,10]

Component	Concentration (mg/L)
Sodium	21.1 ± 1.4
Silicon	20.0 ± 3.7
Calcium	11.2 ± 1.8
Potassium	3.24 ± 1.24
Magnesium	1.13 ± 0.38
Iron	0.78 ± 0.78
Manganese	0.41 ± 0.13
Sulfate	15.2 ± 7.0
Chloride	9.20 ± 4.30
Nitrate	3.80 ± 2.17
Fluoride	3.34 ± 0.56
Bromide	1.11 ± 1.22
Phosphate	<0.1
pH	7.14 ± 0.09

It is important to note that radionuclide release begins only after WP failure, thus WP lifetime significantly affects repository performance and the Tc dose to the receptor group. The WP, based on the DOE Viability Assessment design,[2] is robust. It has an outer corrosion allowance material composed of carbon steel and an inner material of Alloy C-22. The latter is highly resistant to localized corrosion, with a passive dissolution rate independent of environmental factors, such as pH and chloride concentration, and only slightly dependent on temperature.[12] Thus, for the base case, WP failure by corrosion does not occur until greater than 18,000 yr on average. To investigate the effect of container life on the sensitivity analysis results, additional TPA runs were done using the properties of Alloy 625, a less corrosion resistant material, instead of Alloy C-22 in the input file to induce earlier WP failure and focus on the performance of the geologic barrier.

Only dose from Tc-99 is discussed here, but the potential effects of reducing groundwater conditions on dose from other redox-sensitive radioelements (Pu, Np, U) may be evaluated using the same approach. All TPA runs were conducted for 50,000 yr using 250 realizations for each run. All parameters, except those for Tc and the WP material discussed previously, were maintained at the base case values listed in appendix A of the TPA Version 3.2 user manual.[8] The base case data set has 838 parameters, with 592 defined as constants and 246 sampled parameters specified with probability distribution functions.

RESULTS

The sensitivity analyses used peak dose as the measure of repository performance to eliminate the time dependency of the performance measure and simplify the interpretation. The results in Figs. 4 and 5 are presented in terms complementary cumulative distribution functions (CCDFs), which are estimates of the probability that a given consequence (in this case, peak dose) will be exceeded. For the TPA Version 3.2 base case with an Alloy C-22 corrosion resistant material, using solubility and retardation parameters for Tc based on oxidizing conditions results in peak doses to the receptor group from Tc-99 at 50,000 yr (closed circles in Fig. 4) that are less than the 25 mrem/yr peak mean total effective dose equivalent (TEDE) limit proposed by the NRC in Draft 10 Code of Federal Regulations Part 63. Using a lower solubility limit of 4×10^{-9} mol/L to simulate the potential effect of reducing conditions on Tc mobilization (but with low Tc retardation), the calculated dose from Tc-99 was decreased by about three orders of magnitude (open circles in Fig. 4). Using enhanced Tc retardation in the tuff aquifer to reflect stronger sorption of Tc(4+), but maintaining a high (oxidizing condition) solubility limit, the calculated Tc-99 dose (open diamonds in Fig. 4) decreased by less than an order of magnitude compared to the base case. On the other hand, with enhanced Tc retardation in the alluvial aquifer only or along the full length of the saturated zone, Tc-99 did not reach the receptor group in 50,000 yr, even with a high solubility limit.

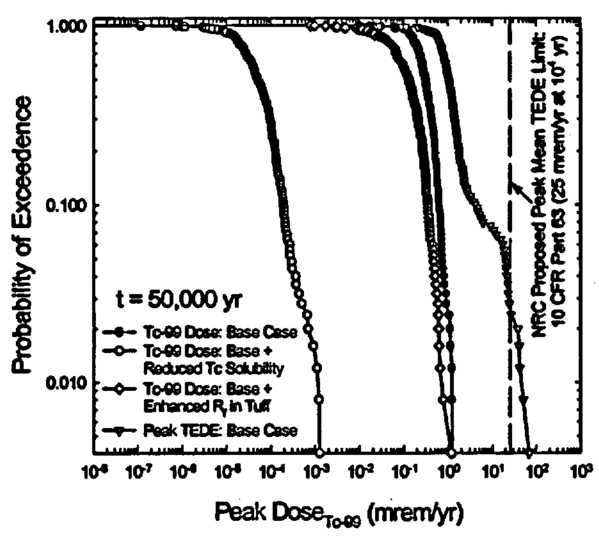

Figure 4. CCDF for Tc-99 dose to a receptor group 20 km from the repository calculated using the base case parameter set,[8] the base case parameters with decreased Tc solubility limit, or the base case parameters with enhanced Tc retardation in the tuff aquifer. Calculations with enhanced retardation along the full length of the saturated zone or in the alluvial aquifer only resulted in zero Tc-99 dose and are not plotted. Shown for reference are the peak total dose from all radionuclides and the proposed NRC 10 CFR Part 63 TEDE limit at 10,000 yr.

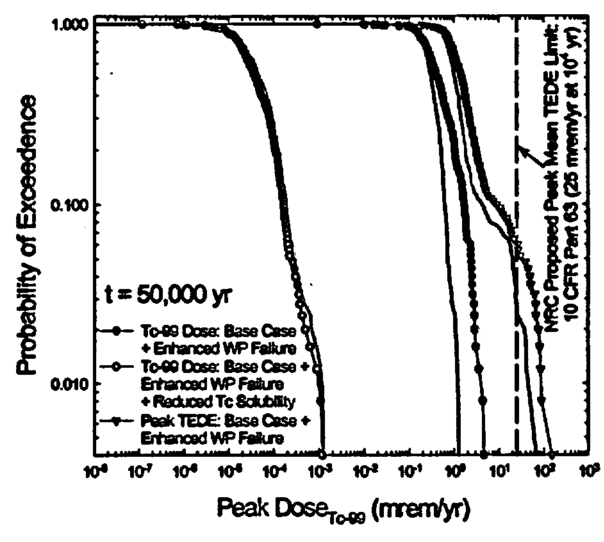

Figure 5. CCDF for Tc-99 dose calculated using Alloy 625, instead of Alloy C-22, properties to simulate earlier WP failure, and the base case parameter set[8] or the base case parameters with decreased Tc solubility limit. Calculations with enhanced retardation along the full length of the saturated zone, which resulted in zero Tc-99 dose, are not plotted. The heavy lines are the CCDFs for Tc dose and TEDE for the base case with Alloy C-22 taken from Fig. 4.

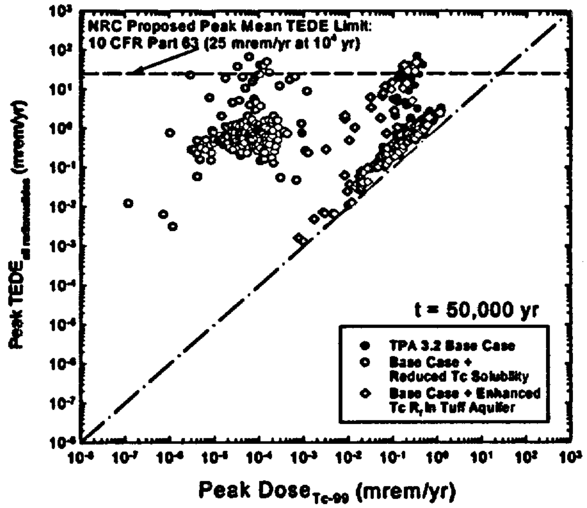

Figure 6. Calculated peak TEDE and Tc-99 dose. Values for each of 250 realizations are plotted. Symbols as in Fig. 4. Calculations with enhanced retardation along the full length of the saturated zone or in the alluvial aquifer only resulted in zero Tc-99 dose and are not plotted. The dashed-dotted line represents a 1:1 relationship between log (Peak TEDE) and log ($Dose_{Tc-99}$). Points along the line indicate that the peak dose is all due to Tc-99.

The resulting CCDFs for the cases in which the effect of decreased container life on Tc release was evaluated are shown in Fig. 5. The simplified approach of using Alloy 625 properties, instead of Alloy C-22, for the corrosion resistant material in the WP reduced the average time to WP failure to about 10,200 yr. The CCDFs indicate that, under oxidizing conditions (high Tc solubility and low Tc retardation), the Tc-99 peak dose to the receptor group at 50,000 yr is slightly higher with Alloy 625 than with Alloy C-22 due to the earlier WP failure. Lowering the Tc solubility limit decreased the Tc-99 dose by more than three orders of magnitude relative to the oxidizing condition case, although the results for Alloy 625 and Alloy C-22 are about the same. Enhancing Tc retardation in the saturated zone delayed Tc transport sufficiently such that no Tc-99 dose was received by the receptor group at 50,000 yr, even with the enhanced canister failure.

The calculated values of Tc-99 peak dose and peak TEDE are plotted in Fig. 6. Under TPA Version 3.2 base case conditions, the doses from Tc-99 calculated for 250 realizations lie close to the diagonal line, i.e., Tc-99 makes up a significant proportion of the peak TEDE at 50,000 yr. Other radionuclides, predominantly I-129, also contribute to the peak TEDE at 50,000 yr. A reduction in Tc solubility lowers the calculated dose from Tc-99 and also minimizes the contribution of Tc-99 to the peak TEDE (indicated by the greater vertical distance of the open circles from the diagonal line). Enhanced Tc retardation in the tuff aquifer alone does not greatly decrease the calculated Tc-99 peak dose, whereas increased Tc retardation in the alluvial aquifer only or along the full length of the saturated zone prevents Tc-99 from reaching the receptor group in 50,000 yr.

CONCLUSIONS

A preliminary assessment of the potential effect of reducing groundwater on the performance of a proposed HLW repository at YM was conducted using the TPA Version 3.2 code. Perhaps not surprisingly, the sensitivity analyses indicate that decreased Tc solubility and increased Tc sorption due to reduction of Tc(7+) to Tc(4+) can significantly delay the arrival of Tc-99 at the receptor group location. In general, the reduced form of redox-sensitive radioelements, such as Np, Pu, and U, are less soluble and sorb more strongly than oxidized species.[13] The release and transport of other redox-sensitive radioelements will be affected in a manner similar to Tc. Thus, reduced groundwater conditions could significantly enhance the performance of the geologic barrier system and reduce the calculated dose to the receptor group.

In performance assessment calculations for a multiple barrier geologic repository, dose to the receptor group is extremely dependent on the robustness of the engineered barrier. If it can be demonstrated with certainty that container life exceeds a significant proportion of the compliance period, then using conservative assumptions to model radionuclide release and transport through the geologic system may be sufficient to provide confidence in the total system performance of the repository. If, however, uncertainty remains in the long-term performance of the engineered materials, then additional realism in PA simulations of the potential isolation capability of the geologic barrier at YM is necessary to demonstrate whether the proposed repository can meet its performance objectives.

Under the current structure of TPA Version 3.2, it is not possible to specify localized reducing zones. Thus, the dependence of Tc release and repository performance on the relative extent of reduced conditions was not studied in detail. In these preliminary sensitivity analyses, conditions sufficient to enhance Tc sorption were assumed for the tuff aquifer only, for the alluvial aquifer only, or for the entire saturated zone transport path. The results indicate that, under the assumptions used in the TPA 3.2 calculations, the tuff aquifer plays a relatively minor role in retarding Tc-99 transport.

Only little information is available on the redox state of groundwaters in the YM area. Given that retardation in the tuff aquifer does not significantly affect the calculated dose to the receptor group, any characterization of the extent of reducing conditions in the saturated zone at YM should focus on the alluvial aquifer. Also, a revision of the NRC/CNWRA TPA code to permit simulation of localized effects is necessary to more realistically evaluate the effect of hydrochemistry on the solubility and transport of redox-sensitive radionuclides.

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