Risks and Uncertainties Associated With High-Level Waste Tank Closure

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ABSTRACT

Closure of tanks containing high-level radioactive waste (HLW) is a challenging problem involving potentially competing influences from economic, societal, and technological considerations. The U.S. Department of Energy (DOE) is faced with protecting public health and the environment while making economically responsible decisions. Risk (i.e., annual dose) is becoming more prominent as DOE's metric to evaluate the economic consequences of its decisions. Risks are assessed through modeling and calculations commonly known as performance assessment (PA). In the process of tank closure, the U.S. Nuclear Regulatory Commission (NRC) is typically consulted to perform an independent review of DOE's PAs.

The NRC staff developed a generic PA model, applicable to HLW tank closure, which NRC utilizes to complete its independent review. The model was developed using the generic simulation software, GoldSim, because of its probabilistic capabilities and its adaptability to different problems [1]. The NRC staff uses the resultant risk from the generic models to evaluate the reasonableness of performance assessment models submitted by DOE. Large differences in the estimates of risk between the generic PA model and the DOE PA would likely indicate a need for stronger technical basis for processes significantly contributing to annual dose (risk) reduction.

BACKGROUND

Liquid HLW results from the reprocessing of spent nuclear reactor fuel. Reprocessing involves the dissolution of spent nuclear fuel followed by chemical separation to selectively remove fissile material for reuse. Various waste streams have been produced during reprocessing operations, depending on the technology and waste management practices utilized at DOE sites. From the Nuclear Waste Policy Act of 1982, as amended, both the supernate and sludge resulting from the reprocessing of spent nuclear fuel are classified as HLW. In other words, wastes from reprocessing are classified by their source, and not by risk to human health or the environment. In most cases, it is not technically or economically practical to remove all reprocessing wastes prior to tank closure; therefore, DOE may choose to leave some HLW in the tanks and close the tank in place. In the past, NRC has provided criteria, as guidance to DOE, for which the residual materials in the tank can be determined to be incidental, and therefore would not need to be disposed of as HLW. The criterion evaluated in this paper is: "…wastes are to be managed, pursuant to the Atomic Energy Act, so that safety requirements comparable to the performance objectives set out in 10 CFR Part 61, Subpart C are satisfied." The performance objectives of 10 CFR Part 61 include protection of the general population, protection of an inadvertent intruder, protection of workers, and stability of the disposal site.

However, the emphasis of this paper is on the models and analyses developed to evaluate long-term protection of the general population. An annual dose limit of 250 μSv/yr (25 mrem/yr) is specified for the general population in 10 CFR Part 61. A detailed summary of incidental waste from a regulatory perspective can be found in [2].

NRC acts in an advisory capacity and does not provide regulatory approval for incidental waste determinations. It is DOE's responsibility to determine whether or not waste is "incidental." In essence, NRC is an independent technical reviewer of determinations made by DOE. To evaluate the long-term protection of the general population, DOE generates site-specific performance assessment calculations. In order to more efficiently identify the key assumptions, the important uncertainties, and to probe the impact of alternative conceptual models, NRC developed a flexible model applicable to assessing HLW tank closure at various DOE sites. The results of the NRC model are not used to demonstrate compliance; rather, they are used to identify key aspects of the problem and focus the review on those aspects.

MODEL DETAILS

PA models are typically probabilistic simulations of multiple (process) submodels. They should be able to represent lack-of-knowledge (epistemic) uncertainty, as well as natural variability (aleatoric uncertainty), if the uncertainties can influence the conclusions. Because long-term projections of impacts are the goal of the PA, temporal evolution of the system should be represented. The complexity of the models should be influenced by the amount of information available to support the models and the risks of the problem. The process of performance assessment is usually iterative in nature (i.e., results of the initial model are used to improve the model further and indicate where additional data collection is needed).

The generic PA model for HLW tank closure developed by NRC utilized the software platform GoldSim [1]. GoldSim was ideal for this application because it is graphically-oriented and very flexible. To allow for different features and characteristics of different sites, the platform for building and editing the model had to be inherently flexible. Although GoldSim is a general purpose simulator, it has basic building blocks in the environmental module developed specifically for mass transport problems. Most of the NRC model made use of the basic GoldSim elements. The flexibility of the software was especially helpful when developing the PA model by facilitating review, modification, and analyses.

The problem that was evaluated was a simulation of the long-term risks associated with leaving residual materials (originally classified as HLW) in tanks. The purpose of the NRC model was to focus the NRC review on those assumptions, uncertainties, and conceptual models that most influence the risks. Tank closure involves removal of HLW to the maximum extent technically and economically practical, which may require tank cleaning. After cleaning, the below-grade tanks are filled with various forms of grout to stabilize the residual materials, provide environmental conditions conducive to radionuclide retention, limit water contact with the waste, provide long-term stability, and limit inadvertent intrusion into the waste. An engineered cap may be included above the tank to reduce infiltration of water. The PA model must be capable of representing the infiltration of water, the degradation of the tanks and engineered features (such as grout), the mobilization and release of radionuclides from the waste, the transport of the radionuclides through the environment, and the exposure to public receptors. The final model had approximately 150 elements, of which about 60 were stochastic elements to represent parameter uncertainty. In GoldSim, stochastic elements can be assigned a variety of probability distributions. It is impractical to summarize all submodels and parameters of the PA in this paper, so this paper only focuses on several key areas. The model was developed with the desire to include all processes essential to the calculation of risk, while maintaining a manageable degree of complexity.

The main submodels represented in the PA are: infiltration, engineered barrier degradation, inventory, source-term release, transport through the unsaturated zone, transport through the saturated zone, and conversion of radionuclide concentrations to annual dose. Select aspects are discussed. Engineered barrier degradation is represented using a GoldSim Source element. Both the tank and grout failure have initiation times and rates of degradation, which are uncertain. Because it is unlikely the grout will degrade prior to tank degradation, the grout degradation cannot initiate until after tank failure. The grout may degrade via a number of mechanisms, including sulfate and magnesium attack, calcium hydroxide leaching, alkali-aggregate reaction, carbonation, acid attack, and reinforcement corrosion [3]. Some of the grout degradation mechanisms depend on water flow rates and the ingress/egress of deleterious species. Therefore, the amount of infiltration contacting the grout was proportional to the extent of tank failure up to a user defined number (e.g., 20%), at which point the failed tank is considered to no longer be a hydrologic barrier.

The inventory model allows the user to specify the physical form of the waste and its radiological composition. Upon release from the waste, the radionuclides can partition between the grout solid phase and cement-modified liquid phase. Flexibility was added to the model to represent the distribution coefficients for the grout, based on reducing or oxidizing conditions and the aging of the grout [4]. Solubility limits are applied to the liquid phase before radionuclides are released from the tanks. Decay chains can be represented, and radiological decay is included in all of the mass transport calculations. Transport through the unsaturated zone (UZ) is assumed to be vertical with no lateral dispersion. The UZ transport model utilizes Pipe elements with a partitioning factor so that the user can specify the proportions and lengths of the flowpaths for the fractured and porous media. For fractured flow, the Pipe elements can represent matrix diffusion, but that capability is not used in the base case model. Transport through the saturated zone (SZ) is assumed to be lateral in one-dimension with longitudinal dispersion and sorption. Distribution coefficients, dispersivity, transport length, and groundwater flow rates are all uncertain parameters. Finally, the radionuclide concentrations at the receptor location are converted to a radiological exposure with stochastic biosphere dose conversion factors.

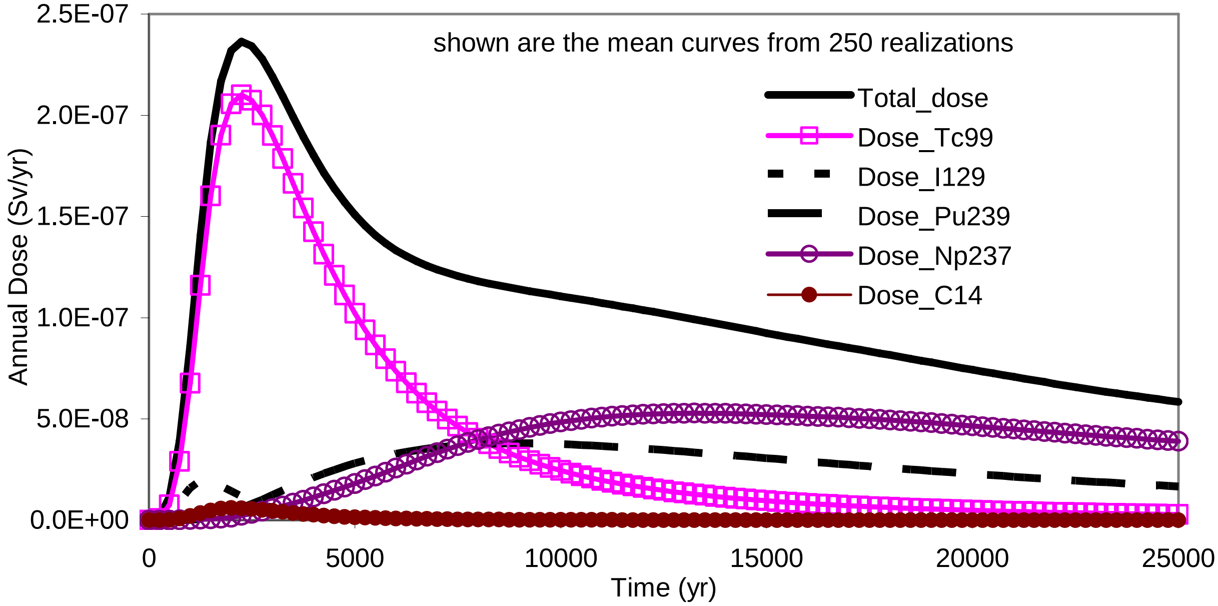
DISCUSSION

The generic PA model was applied during the review of recent documentation from the Idaho National Engineering and Environmental Laboratory (INEEL) for HLW tank closure, in order to risk-inform the review [5]. The NRC staff used the generic PA model to identify the key assumptions and parameters of the DOE performance assessment. The analyses that follow are designed to convey some important aspects of assessing the risks from HLW tank closure, but are not necessarily applicable to any specific site.

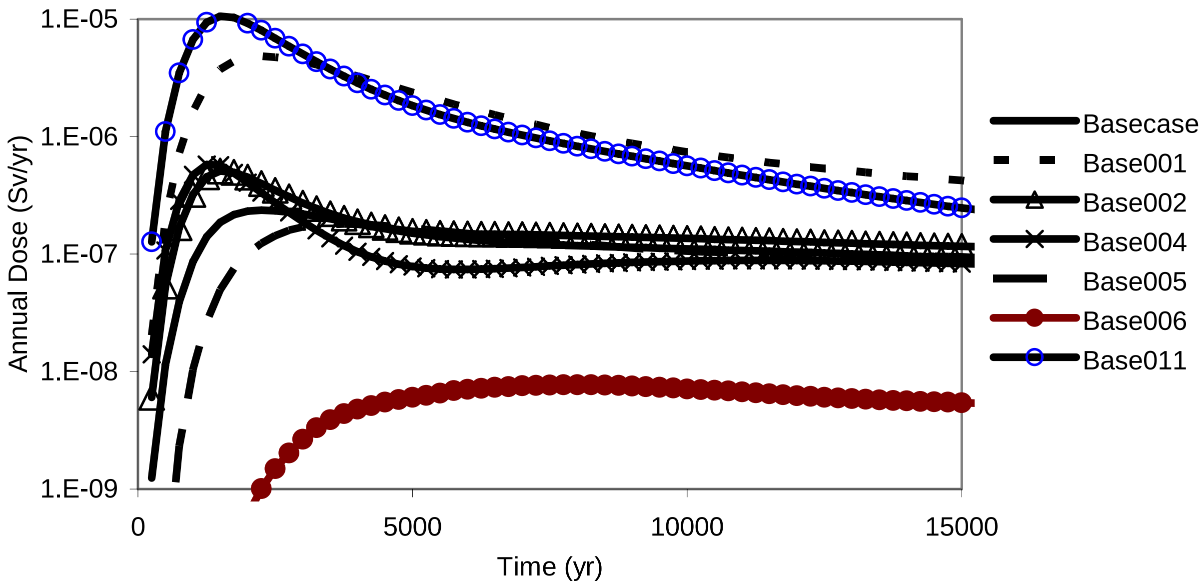
The base case model results and select sensitivity results are summarized in Figure 1 (a). The base case model was run with 250 realizations for 25,000 years. The mean annual dose is the average of the individual realizations of annual dose at a point in time. The performance metric is then the peak of this mean curve, hereafter referred to as peak of the mean. The compliance period is not specified in 10 CFR Part 61; therefore, the analysis period was designed to capture the peak response, as well as pertinent system behavior. The inventory and many of the other parameters in the model were defined to be consistent with the INEEL site [5].

Numerical stability of the results was examined by performing multiple simulations with different seeds for the Latin Hypercube Sampling algorithm, and the peak of the means were within 10% of each other. Also, there was little change in the results, using either 500 or 1000 realizations. The peak mean annual dose for the base case is 0.24 μSv/yr (0.024 mrem/yr) at 2250 years, primarily from Tc-99 and I-129 with small contributions from Pu-239 and Np-237. At greater than 10,000 years, Pu-239 and Np-237 are the dominant contributors. Stepwise linear regression was performed on both the raw variables and the ranks of the variables. The most sensitive parameters were identified as: the biosphere dose conversion factor for Tc-99, infiltration rate, groundwater flow rate in the SZ, the distribution coefficient (Kd) for Grout Tc-99, the likelihood of only fractured flow, gradual failure rate of the grout, and the length of porous media in the UZ. The most sensitive parameters are consistent with the authors' understanding of the problem. Tc-99 dominates the annual dose and thus, parameters that directly influence the Tc-99 release rate (infiltration rate, Kd Grout Tc-99, gradual failure rate of the grout) should be sensitive parameters. Infiltration rate and groundwater flow rate in the SZ contribute to sensitivity because their uncertainties influence the amount of dilution to the contaminants as they exit the unsaturated zone and enter the saturated zone (which have substantially different water flow rates in the model). The presence (or absence) and length of porous media parameters were used to represent conceptual model uncertainty for the amount of transport through porous media in the unsaturated zone.

A number of additional analyses completed to address various model or scenario uncertainties are presented in Figure 1 (b). The legend of Figure 1 (b) provides a number of identifiers for sensitivity cases. The base case curve is simply the total annual dose curve shown in Figure 1 (a). Base001 is an analysis with a more pessimistic inventory. The amount of Tc-99 is roughly twenty times larger, which results in a peak mean annual dose that is about twenty times larger. When a highly-soluble species is the largest contributor to annual dose, the risk is directly proportional to the inventory. This result highlights the importance of reducing uncertainty in the inventory and removing the tank inventory to the maximum extent that is technically and economically practical. Base002 is an analysis using distribution coefficients for sorption of radionuclides to grout, representative of chemically reducing conditions for environment III [4]. Environment III is defined in the literature as the environmental conditions associated with severely weathered cement [4]. The time at which the grout may be considered "severely weathered" would depend on many factors, including degradation mechanisms, and in many cases, the infiltration rate. The annual dose for Base002 is larger by roughly a factor of 2 compared to the base case, largely due to the higher release rates from the tanks resulting from lower sorption. Base004 utilized distribution coefficients for Tc representative of oxidizing conditions, and the annual dose was a factor of 3 larger than the base case. Base005 and Base006 show the influence of adding an engineered cap to reduce the infiltration rate. In the former case, the engineered cap is a flow barrier that degrades over time. In the latter case, the engineered barrier is conceptualized as more of a capillary barrier type system, which may result in a permanent long-term reduction of infiltration. For a flow barrier type system that degrades over time, the reduction in annual dose is relatively small (~30%). For a capillary barrier type



(a)



(b)

Figure 1 (a) Annual dose response showing individual radionuclides for the tank closure PA model. (b) Sensitivity analyses evaluating alternative scenarios and models: 1) Base001 was a 20x increase in key radionuclide inventory, 2) Base002 used distribution coefficients representative of an severely weathered grout (environment III) [4], 3) Base004 used a distribution coefficient for Tc-99 with grout representative of oxidizing conditions, 4) Base005 had an infiltration-reducing engineered cap that lost its functionality over time, 5) Base006 had an engineered cap that permanently reduced infiltration by a factor of 10, 6) Base011 had a combined effect of pessimistic inventory and environment III grout Kds [4].

system that results in a long-term reduction in infiltration by a factor of 10, the peak annual dose is roughly a factor of 30 less and is shifted later in time. The results for the influence of the engineered cap are conditional on many factors (e.g. the release rates, the conceptual models for release, the hydrogeology, and the exposure pathways) and should not be generalized. The analysis presented with Base011 used the pessimistic inventory and grout sorption coefficients consistent with environment III. The results show a non-linear response, such that the peak is larger than simply multiplying the changes in the peak from Base001 and Base002. The magnitude of the non-linear response was larger for Pu-239 than it was for Tc-99. Probabilistic PAs with many stochastic variables appropriately capture the non-linearity of the submodel- and integrated submodel-responses.

CONCLUSIONS

The PA model has proven to be an effective tool in risk-informing reviews of HLW tank closure impacts. For the tank closure problem studied, the parameters and models determining radionuclide release rates, the inventory remaining in the tanks, and the geochemical environment for radionuclide release, were important. The amount of dilution expected prior to receptor exposure, whether via the fundamental hydrological system or via wellbore dilution, can linearly influence the risks. Therefore it is important to have adequate technical basis for the amount of dilution represented in the PA model. The presence of porous media capable of preventing the transport of strongly sorbing species was a key barrier to the minimization of risks from many radionuclides.

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