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APPLYING INEEL RESEARCH AND TECHNOLOGIES TO IMPROVE LLW DISPOSAL

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ABSTRACT

The low-level radioactive waste (LLW) disposal facility at the Idaho National Engineering and Environmental Laboratory (INEEL) is operated in compliance with United States Department of Energy (DOE) Order 435.1, "Radioactive Waste Management." DOE Order 435.1 includes requirements to conduct a performance assessment and composite analysis (PA/CA) to project the potential doses that may be received by a member of the public as a result of releases that may occur from the facility and other surrounding sources. Although the PA/CA were deemed sufficient to justify issuing the Disposal Authorization for the disposal facility, the DOE Low-Level Waste Disposal Facilities Federal Review Group (LFRG) identified some assumptions that required additional confirmation. Thus, the Disposal Authorization was granted with some conditions that required resolution in order to maintain the capability to operate the LLW disposal facility. This paper summarizes INEEL research and technologies that contributed to successful resolution of all of the conditions imposed on the Disposal Authorization. The research and technologies described in this paper can also be applied at other LLW disposal facilities and broader applications in the United States and internationally.

INTRODUCTION

The Idaho National Engineering and Environmental Laboratory (INEEL) is a multi-purpose national laboratory located in southeastern Idaho conducting research and operations to support a variety of government, defense, and commercial customers. Many of these activities lead to the generation of low-level radioactive waste (LLW) requiring disposal. Onsite LLW disposal has been practiced for more than 50 years at the Radioactive Waste Management Complex (RWMC), located in the southwest corner of the INEEL.

The active LLW disposal facility at the RWMC is operated in compliance with United States Department of Energy (DOE) Order 435.1, "Radioactive Waste Management." Permission to operate the disposal facility is granted by the DOE Low-Level Waste Disposal Facilities Federal Review Group (LFRG) through the issuance of a Disposal Authorization. The LFRG conducts detailed reviews as part of the process of granting a Disposal Authorization. Following the reviews at the INEEL, the Disposal Authorization for the active part of the LLW disposal facility was granted in 2000 with some conditions. The conditions addressed specific areas of need, including the need for additional assurance regarding modeling assumptions, monitoring plans, and long-term stewardship and closure. The conditions must be satisfactorily addressed in order to maintain the Disposal Authorization.

Activities related to maintaining the Disposal Authorization for the LLW disposal facility at the RWMC are closely integrated with technology development and research from the INEEL. In

this regard, LLW disposal serves as an example of how research and development (R&D) can contribute directly to operational needs. The integration of LLW disposal and R&D includes use of existing research and previously developed technologies, development of new technologies, as well as initiation of new research projects funded through research programs and operations programs.

REVIEW GROUP ISSUES

A major emphasis of the LFRG review is to consider the defensibility of assumptions that are made in the performance assessment [1] and composite analysis [2] (PA/CA) related to long-term performance of a disposal facility. In the case of the RWMC LLW disposal facility, C-14 is a key contributor to the projected doses to a hypothetical receptor assumed to live near the disposal site. Thus, the LFRG wanted reassurance that assumptions that could affect the rate at which C-14 would be projected to be released and migrate in the environment were conservative. To meet this need, the LFRG requested that research be conducted to address specific issues and that additional sensitivity/uncertainty analyses be conducted to better bound the range of possible results.

The LFRG also emphasized the importance of environmental monitoring to provide objective evidence that environmental conditions remain similar to those assumed for the PA/CA and to reassure the public that migration of key radionuclides is not occurring above projected rates. A monitoring plan has been prepared to describe the program being implemented to address the requirements of DOE Order 435.1 and specific LFRG areas of interest [3].

At the request of the LFRG, an Options Analysis [4] was prepared that identified a path forward to resolve key issues. Some of the activities described below were identified in the Options Analysis. Waste management has worked together with research and development staff to help resolve all of the issues identified by the LFRG, including those mentioned above. This paper includes several examples of research and technology development that contributed to resolving the issues raised by the LFRG.

APPLIED RESEARCH

The INEEL has considerable expertise in subsurface science and corrosion research. This expertise was drawn upon to help address key issues identified by the LFRG. The research conducted to address two of the issues is discussed here. The first issue discussed is a need to provide additional justification for the release rates assumed for C-14 from beryllium, and the second issue is to provide additional justification to reduce the conservatism of the doses projected due to exposure to C-14 after closure of the disposal facility.

Corrosion Testing

A large proportion of the total inventory of C-14 buried at the RWMC is in the form of activated metals, including beryllium. Release rates of radionuclides contained in activated metals are assumed to be controlled by corrosion of the metal (i.e., as the metal corrodes, the radionuclides can be released from the metal matrix). Thus, a corrosion rate is used as the release rate for radionuclides in activated metal for the purposes of PA/CA modeling. Corrosion rates used in the

current PA/CA were determined based on a literature review. Conservative values from somewhat saline environments were used in order to ensure that corrosion rates were not underestimated. The LFRG still believed that it was necessary to conduct some additional testing to verify corrosion rates, given the importance of C-14 in the results of the PA/CA.

To address the LFRG issue, the INEEL has undertaken a research project to assess the corrosion rates of several different metals in vadose zone soils found at the RWMC [5]. The test is being conducted in a berm located adjacent to the RWMC. The berm is constructed using soils representative of fill soil used at the RWMC. Corrosion rates are being assessed using buried 7.6 cm (3 in) square by 0.3 cm (1/8 in) thick metal coupons. The coupons were buried in arrays with one array at 1.2 m (4 ft) depth and another at 3.0 m (10 ft) depth at each test location. Each coupon array comprises four test coupons of each of the following metals: 1018 carbon steel, Type 304L stainless steel, Type 316L stainless steel, welded Type 316L stainless steel, Inconel 718, Beryllium S200F, aluminum 6061, Zircaloy-4, and Ferralium 255, for a total of 36 coupons. All of the coupons in a given array are retrieved at the same time. Table I includes a summary of the installation and retrieval dates for the arrays.

Table I. Coupon installation and retrieval schedule

Coupon Array	Depth (m)	Installation Date	Retrieval Date
CA01	1.2	Oct. 22, 1997	Oct. 23, 1998
CA02	3	Oct. 21, 1997	Nov. 3-5, 1998
CA03	1.2	Oct. 22, 1997	Oct. 15, 2000
CA04	3	Oct. 21, 1997	Oct. 23, 2000
CA05	1.2	Nov. 3, 1997	Oct. 30, 2003
CA06	3	Nov. 3, 1997	Nov. 13, 2003
CA07	1.2	Oct. 22, 1997	Oct. 2008
CA08	3	Oct. 22, 1997	Oct. 2008
CA09	1.2	Nov. 11, 1998	Oct. 2013
CA10	3	Nov. 11, 1998	Oct. 2013
CA11	1.2	Oct. 26, 2000	Oct. 2018
CA12	3	Oct. 26, 2000	Oct. 2018

To date, three sets (six arrays) of coupons have been retrieved, representing one year, three years and six years of burial. This year's and future retrievals were scheduled such that new data are available in time to support revisions of the PA/CA, which must be addressed every five years. Fig. 1 is a photo from the 2003 retrieval for a typical set of coupons. The soil is sampled around the coupons to evaluate migration of corrosion products and to determine the presence of microbes.

Fig. 2 includes examples of corrosion on beryllium coupons from the 3 m level after one, three, and six years of burial, respectively. Note that the amount of corrosion on other coupons from the same depth and exposure times varies. A summary of the results obtained to date for beryllium corrosion at the 3 m level is provided in Table II. The corrosion rates observed from the test (110 – 260 yr/mm or 0.004 – 0.009 mm/yr) have all been consistently less than the rates assumed for the PA/CA (39.4 yr/mm), which provides added assurance that the assumed rates are likely to be conservative and, if necessary, could possibly be used to justify a decrease in the assumed release rate for C-14.



Fig. 1. Coupon retrieval and soil sampling.

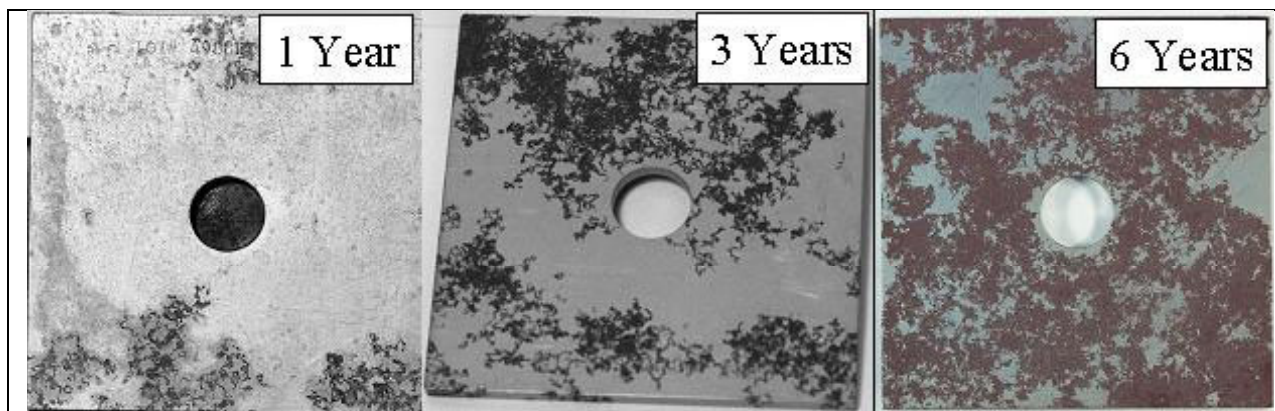


Fig. 2. Examples of cleaned beryllium coupons retrieved at different times from 3 m depth.

Table II. Observed Ranges of Be Corrosion Rates at 3 m Depth (PA/CA Assume 39.4 yr/mm).

One Year Test		Three Year Test		Six Year Test	
% Mass Loss	Corrosion Rate	% Mass Loss	Corrosion Rate	% Mass Loss	Corrosion Rate
0.26-0.35	195-260 yr/mm 0.004-0.005 mm/yr	0.98-1.68	110-195 yr/mm 0.005-0.009 mm/yr	2.03-3.28	120-195 yr/mm 0.005-0.008 mm/yr

C-14 Migration

As noted above, C-14 is the primary dose contributor in the PA/CA. However, an important conservative assumption was made in the PA/CA. For the groundwater pathway, it was assumed that all C-14 migrated downward to the aquifer contributing to the projected water pathway doses. In addition, for the atmospheric pathway, it was assumed that all C-14 migrated upward to the atmosphere contributing to the projected air pathway doses. This assumption guarantees that the migration of C-14 via both pathways will be overstated. In the case of the air pathway, the doses are projected to be well within acceptable levels and the over conservatism is not a concern. However, in the case of the water pathways, although the projected doses were within acceptable levels, the LFRG believed that they were high enough to warrant additional study to provide further assurance that the conclusions remain valid.

Monitoring data for gas phase C-14 releases over known disposal locations have confirmed that C-14 is migrating upwards in the gas phase [6]. However, it is difficult to quantify the fraction migrating upwards versus downward in the field. Thus, an experimental program was started to examine radionuclide migration in a mesoscale column using unsaturated soils representative of the RWMC [7]. A mesoscale experiment is intended to more closely approximate migration in the field than previous smaller laboratory scale column experiments.

The mesoscale soil column used for this experiment is roughly 2.6 m tall and 1 m in diameter (see Fig. 3) and set up in a laboratory in order to control the environmental conditions. The column is designed to monitor migration in the liquid and gas phase at eight different levels along the length of the column. Coring has also been conducted to assess partitioning of radionuclides on the solid phase. Water was added to the column in 2001 and allowed to reach equilibrium at moisture levels similar to those found at the RWMC. After several months of testing with SF₆ and lithium bromide; C-14, uranium, and H-3 were injected into the soil column. Migration of C-14 is the focus of the discussion that follows.

C-14 was observed to migrate via diffusion at a slower rate than SF₆, which is consistent with the fact that a fraction of the C-14 partitions into the liquid and solid phases. Model comparisons with the experimental results suggest that migration in the column is represented well using standard diffusion theory and the Millington expression to describe the reduction in diffusivity due to partial saturation and constant partitioning factors for partitioning of C-14 between solid, liquid and gas. Fig. 4 illustrates the excellent agreement between observed and modeled breakthrough curves for C-14 in the column. Conclusions from the experimental work and modeling to date suggest that, because C-14 migration is dominated by diffusion and the disposal is much closer to the surface than to the aquifer, the majority of the C-14 would migrate upward and vent to the atmosphere.

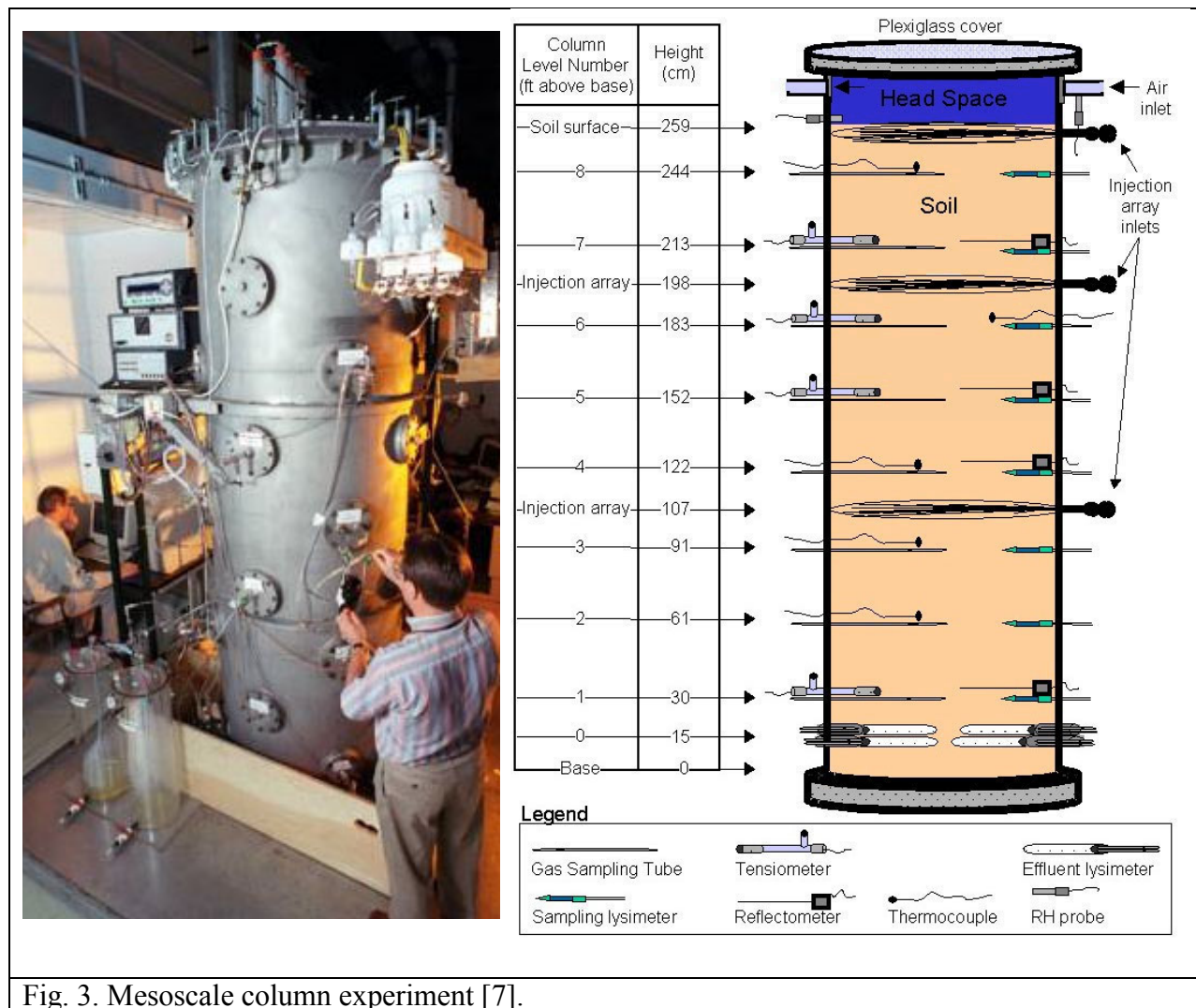


Fig. 3. Mesoscale column experiment [7].

Although all evidence from experiments and modeling to date confirms that a majority of the C-14 would vent to the atmosphere, there are complicating factors in the field that need to be addressed. For example, occasional increases in infiltration rates would temporarily increase the amount migrating down. However, it is known that these increases in infiltration occur rarely during the course of a year, and only temporarily alter the soil moisture content in the upper few meters of cover. Such variations may increase slightly the downward flux of C-14 but overall they do little to limit the diffusive process that transports C-14 more rapidly along its short path to the atmosphere. There are also factors in the field that reduce the rate at which C-14 migrates downward (e.g., higher moisture contents, as can be found beneath disposal locations, would tend to reduce the rate of diffusion in the gas phase). The modeling and experimental results provide compelling evidence that at least 50%, and likely, much more of the C-14 would migrate upwards and vent to the atmosphere. In order to maintain a substantial level of conservatism, the models used for the PA/CA will initially be updated to reflect a 50% split.

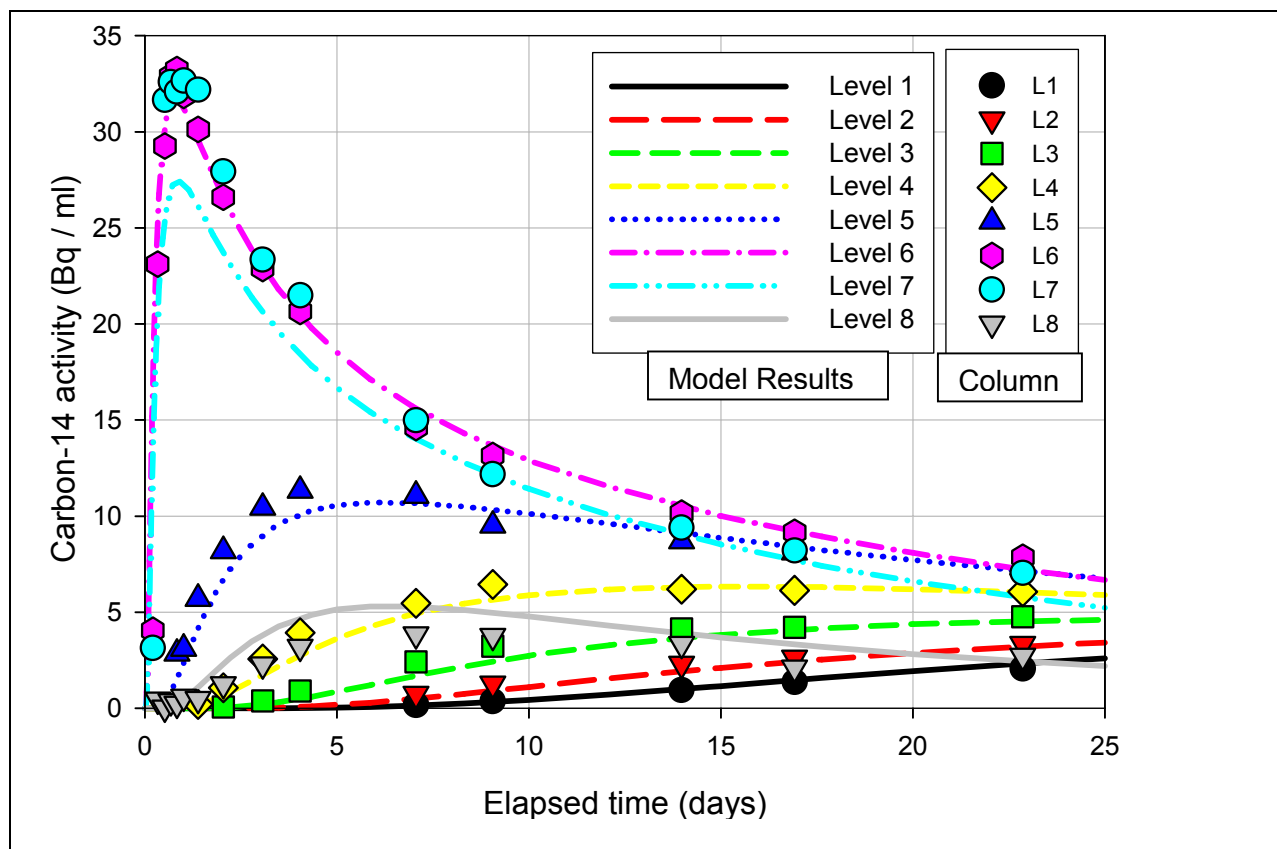


Fig. 4. Observed gas carbon-14 breakthrough curves at the eight sampling levels (symbols) and modeled breakthrough curves (curves) using a pH-dependent gas-aqueous partitioning ratio (see Fig. 3 for the locations of the sampling levels on the column).

TECHNOLOGY DEVELOPMENT

Several technologies developed at the INEEL are being used to address LFRG issues and support continued disposal of LLW at the RWMC. Three examples are briefly discussed in the following sections. The first technology is the Advanced Tensiometer, which is used to track changes in moisture content in deep vadose zone soils below the waste as part of the active pit monitoring system. The second technology is a new approach to sampling C-14 in the soil gas above existing disposal locations that makes the sampling more cost effective and reduces exposure time during sampling. The third technology is a simplified radionuclide transport model, GWSCREEN, which is used to facilitate the numerous simulations required to conduct sensitivity/uncertainty analysis for the PA/CA.

Advanced Tensiometers

Tensiometers are used to measure the soil water potential in unsaturated soils. The soil water potential provides a means to detect changes in moisture content, which is an indication of moisture migration. In the very dry soils at the RWMC, such a measurement provides valuable information about the potential for contaminant migration. Thus, tensiometers are being installed as part of the instrument clusters used for active pit monitoring.

Conventional mechanical tensiometers have been limited by the depth at which they could be used. Given that the vadose zone is nearly 200 m (600 ft) thick at the RWMC, a new tensiometer was needed that could be used at greater depths. The INEEL developed an advanced tensiometer to meet this need. These advanced tensiometers are being used as part of the active LLW disposal monitoring system being installed as LLW is disposed at the RWMC. The monitoring ports will provide data regarding moisture and radionuclide migration under the containers that have been disposed. This is an important contributor to building confidence that the PA/CA modeling is providing a conservative estimate of actual radionuclide migration.

C-14 Sampling

C-14 is primarily present in soil gas as $^{14}\text{CO}_2$, which can migrate in gas or liquid phases. The relative difficulty and expense of sampling and analysis for C-14 has been an impediment to efforts to characterize migration of C-14 from LLW and through the unsaturated soil environment. A method for sampling and analysis of C-14 and total carbon in gaseous samples was developed at the INEEL, enabling large-scale sampling programs to be conducted safely and at reasonable cost. The method is particularly well suited for soil gas sampling because the CO_2 content of soil gas typically ranges from 2000 to 20,000 ppm.

Soil gas samples are being collected in 1-liter Tedlar[®] bags. The CO_2 concentration of the sample is measured by gas chromatography, and the sample volume is determined using a large gas syringe. Approximately 12 mL of 0.5 N NaOH solution is injected into the bag to absorb the CO_2 from the gas sample. Nine to 10 mL of the solution is recovered, mixed with a compatible liquid scintillation counting (LSC) cocktail, and counted in a low-background LSC instrument. The method has been used routinely at the INEEL to measure the C-14/C-12 ratio in CO_2 near buried, activated steel and beryllium at levels ranging from 10 to 10^5 times the ratio in modern carbon. This method has the following benefits: no hazardous material is used in the field; sample collection is fast and not technically challenging; total carbon is determined without wet chemistry procedures; and there is minimal production of waste. These benefits translate into lower costs and reduced personnel exposure in the field.

Modeling Software

The current approach for PA/CA modeling is to use a fully three-dimensional, process-level model, TETRAD, to simulate moisture flow and radionuclide transport for a base set of parameters. TETRAD requires anywhere from several days to several weeks to complete a suite of simulations for one set of contaminants. Sensitivity/uncertainty analysis can require hundreds of simulations for each contaminant of interest. Thus, a more simplified approach was needed to conduct the simulations for the purpose of the sensitivity/uncertainty analyses.

GWSCREEN [8], a computer code originally developed at the INEEL as a screening tool to support environmental restoration decision-making, is used to support the PA/CA for the LLW disposal facility at the RWMC. GWSCREEN is based on a two-dimensional semi-analytical solution to the transport equation. This results in very rapid run times necessary to conduct sensitivity/uncertainty analyses, although without the level of detail provided by a model like TETRAD. In practice, GWSCREEN is calibrated to the results from the TETRAD modeling for

each radionuclide of interest. The calibrated GWSCREEN model is then used as the base case from which to conduct the numerous sensitivity/uncertainty analysis simulations. This provides the suite of modeling results needed to better understand the range of possible results that can be obtained given the uncertainties inherent in modeling the long-term performance of a LLW disposal facility.

CONCLUSION

Several issues were raised by the LFRG after reviewing the information necessary to grant a Disposal Authorization for the LLW disposal facility at the RWMC. Some of these issues were attached as conditions to address when the initial Disposal Authorization was granted. The conditions included, among other things, a need for research to provide additional justification for assumptions made in the PA/CA and enhanced monitoring around the LLW disposal facility.

This paper has outlined how INEEL research and technologies contributed to resolving some of the issues identified by the LFRG. The research discussed above addressed corrosion rates of metals and migration of C-14 in the vadose zone. The corrosion study to date has illustrated that the observed corrosion rates in the test are less than the corrosion rates assumed for the PA/CA. Likewise, the mesoscale column study has provided convincing evidence to support reducing the conservatism of assumptions regarding the fraction of C-14 that migrates down to the aquifer.

The role of three technologies developed at the INEEL in helping address issues raised by the LFRG was also discussed. Advanced tensiometers developed at the INEEL are part of a suite of instruments being used to monitor the region beneath the waste being disposed in the LLW disposal facility. A new technique developed at the INEEL for sampling C-14 in soil gas is being used in areas where previous disposals of LLW containing C-14 have taken place. The new technique allows the samples to be taken at a lower cost and with reduced exposure of personnel. The third technology discussed is a computer code, GWSCREEN, that is being used to make the numerous simulations required to conduct sensitivity and uncertainty analyses that help to better bound the range of possible results to reflect the uncertainties associated with long-term simulations of the behavior of LLW disposal facilities.

In August 2003, in recognition of the research and application of technologies discussed in this paper as well as other activities to address remaining issues raised by the LFRG, the INEEL was given formal notice that all conditions associated with the initial Disposal Authorization have been satisfactorily addressed. This paved the way for continued operation of the LLW disposal facility. The R&D described in this paper can also be applied at other LLW disposal facilities and broader applications in the United States and internationally.

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