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Leachate/Contaminant Reduction Time Study (Title I)

ABSTRACT

Concentrations of selected design inventory constituents in INEEL CERCLA Disposal Facility landfill leachate were simulated over the 15-year operations period. The purpose of the study was to examine the change in leachate concentration over time, as it is directed toward the evaporation pond. The results may be applied toward performance assessment modeling in the future. Two groups of screened design inventory constituents were applied to the leachate/contaminant reduction time study. One group was screened on the basis of partition coefficients and the other on the basis of concentration in the design inventory. The latter group was entered into a geochemical model to simulate solubility constraints on the resulting leachate concentrations. Concentrations of each group were entered into a spreadsheet program that simulated partitioning to the solid phase via adsorption, radioactive decay, and leachate removal from the landfill. The leachate will be a brackish to saline water dominated by sodium and sulfate and buffered by carbonates to a pH of around 8.2. The results indicate less than 10% of the inventory masses of the most mobile constituents (iodine and technitium) are expected to be removed from the landfill during the operation period.

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ACRONYMS

CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
cm	centimeter
g/kg	grams per kilogram
ICDF	INEEL CERCLA Disposal Facility
INEEL	Idaho Engineering and Environmental Laboratory
kg	kilogram
kg/L	kilograms per liter
mg/kg	milligrams per kilogram

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1. INTRODUCTION

The purpose of the Leachate/Contaminant Reduction Time Study is to document the composition of both the leachate and the landfill mass over the period of operation (15 years) at the Idaho National Engineering and Environmental Laboratory's (INEEL) proposed INEEL CERCLA Disposal Facility (ICDF) landfill. During this period, the leachate will flow through the leachate collection system and will be discharged to the evaporation pond. Results of this study will be used to estimate pond water composition over time and may also be used in performance assessment modeling.

The design inventory elements were screened based on a process provided by BBWI. This process produced two sets of elements (described below). Each set was converted from a solid concentration or radionuclide activity to an aqueous concentration. Geochemical modeling of solubility was used in this determination for one of the two sets. The resulting aqueous concentrations were sent to a spreadsheet where leaching rates were applied along with partition coefficients (which are used to estimate the degree of adsorption to soil mineral surfaces) and radioactive decay rates (where applicable) to determine the mass removal from the landfill over time. Plots were constructed of landfill mass concentrations and landfill leachate concentrations over time for each element set.

Details of the procedure described above are presented in the following sections: Screening Process, Geochemical Modeling, Model Leachate Generation, and Conclusions.

2. SCREENING PROCESS

Quantities of inorganic (non-radioactive) and organic compounds were reported as total kilograms (kg), whereas concentrations of radionuclides were provided as total curies in the "INEEL CERCLA Disposal Facility Design Inventory," (DOE-ID 2001a). These were converted to kg using published half-lives and atomic mass for each isotope. For elements with reported concentrations of radioactive and non-radioactive forms, the isotope masses were summed, although in these cases the radioactive concentrations were far less than those of non-radioactive isotopes. The source of radioactive constituents was Table D-3 from the design inventory (DOE-ID 2001a).

The screening process is illustrated in Figure 2-1. Two screening exercises were completed. The first was based solely on partition coefficient (K_d) values. As shown on the portion of Figure 2-1 marked "1," all elements with K_d values less than 20 were considered significantly leachable during the operations period and were therefore included in the leachate generation calculations presented in Section 4. A K_d of 20 corresponds to 1% of the waste mass leaching into solution. These constituents were assumed to have no solubility controls, and were assumed to partition to the aqueous phase according to their K_d values. The resulting list of elements, including their milligrams per kilogram (mg/kg) concentrations, K_d values, and corresponding aqueous concentrations, is provided in Appendix A as Table A-1. The constituents with K_d values greater than 20 are displayed in Table A-2.

The second screening exercise (marked "2" on Figure 2-1) was based on the nature and concentration of inventory constituents. All constituents existing in solution as anions were included, regardless of their concentrations in the inventory. Plutonium is predominantly in anionic form above pH 8, and uranium carbonate species are anionic, so both of these elements were included, even though they are assumed to be cations in many studies. Organic compounds were assumed to be neutral species. All inventory organics were eliminated because none was over 1% of the total mass fraction, calculated by dividing the total kg for a given compound by the assumed total waste stream mass of 5.85×10^8 kg. Inorganic constituents were screened to include only those that constituted over 0.01% of the total mass fraction. Since there is a minimal amount of organic constituents in the design inventory, 1% was selected as the criteria to identify the major chemical species contributing to the leachate chemistry. Similar screening was used for the inorganic species with the intent to identify the species that contribute 99.99% of the constituents in the leachate. The final list of constituents for this screening exercise is shown in Appendix A as Table A-3. Those constituents that were screened out are provided in Table A-4.

The second list was included in a geochemical model to correct for solubility in the leachate before being passed to the leachate generation calculations. The geochemical modeling is described in Section 3.

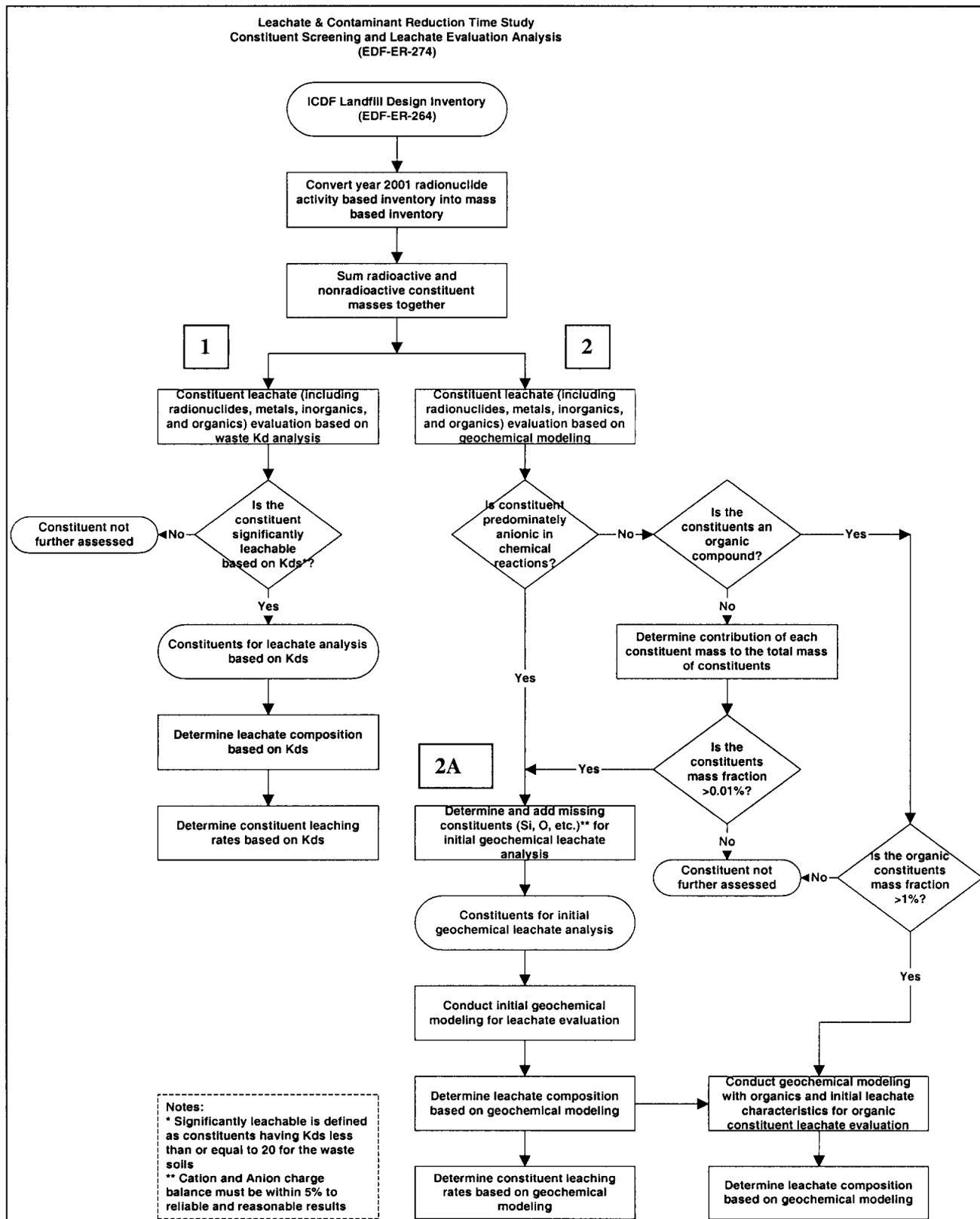


Figure 2-1. Screening process flowchart for leachate/contaminant reduction time study.

3. GEOCHEMICAL MODELING

The screened inventory constituents at the box labeled “2A” in Figure 2-1 were input into a geochemical model to estimate their solubility-controlled concentrations in leachate.

3.1 General Principles

The geochemical model PHREEQC, v. 2.3 (Parkhurst and Appelo 1999), was used for this study, using a recently released database, LLNL.DAT, which contains species and mineral phase data for all of the screened elements. PHREEQC receives as input all aqueous concentrations of constituents in the infiltrating water, along with quantities of all mineral phases believed to be present in the landfill material. Geochemical conditions are also specified, namely the pH, redox potential, and the presence of atmospheric gases (O₂, CO₂, etc.). These conditions may either be set or allowed to change as the system equilibrates.

The model takes into account all elements present and, using the database, calculates concentrations and activities (concentrations corrected for ionic strength) of all aqueous species present. For example, calcium exists in various aqueous forms including Ca²⁺, CaHCO₃⁺, CaCl⁺, etc. Saturation with respect to applicable mineral phases is also calculated by comparing the ion activity product with the solubility product constant (K_{sp}) for a given mineral. For example, the mineral calcite (CaCO₃) has a K_{sp} of approximately 10^{-8.4}. This means that under equilibrium conditions, the product of the aqueous activities of the ions Ca²⁺ and CO₃²⁻ is 10^{-8.4}. If this product is greater than the K_{sp}, the solution is said to be supersaturated with respect to calcite, and undersaturated if the opposite is true. This demonstrates the importance of calculating the free concentration of mineral constituent ions such as Ca²⁺. Other ions in solution may complex Ca²⁺ or reduce its activity by contributing to higher ionic strength. PHREEQC calculates these effects for all input elements and minerals.

One of many other features of the PHREEQC model is the ability of the user to assign equilibrium conditions to selected mineral phases. In this way, if minerals are known or suspected to exist at a site, the user may instruct PHREEQC to dissolve the necessary amounts of those minerals to achieve equilibrium with the surrounding solution. This feature was used in the present study.

3.2 Site-Specific Methodology

The first step in the geochemical model setup was to input the infiltrating water chemistry. The average annual rainfall at the site was assumed to be approximately 8 in./yr (NOAA 1989). In addition, site water supplies will be used for dust suppression and compaction during the operating period at an estimated rate of 10,000 gallons per week (DOE-ID 2001b). Given the completed landfill will cover an area 810 ft by 720 ft, this corresponds to an equivalent of 1.43 in./yr in applied water. The infiltrating water will therefore have a proportion of 85% rainwater and 15% applied water. Major chemical constituents for each water source were input to PHREEQC, using an average rainwater analysis (Brownlow 1996) and site records for Well CPP-1 (Marty Doornbos personal communication 2001). The two waters were mixed in the proportions described above and the resulting solution was saved for further modeling. For the 90% submittal, a sensitivity analysis will be performed examining the effect of increasing the amount of supplied water to the mixture.

As a conservative approach, the waste mass was assumed to be water-saturated. For each kilogram of waste mass, a void volume was calculated using reported values of dry density (1.86 kilograms per liter [kg/L] of soil volume) and porosity (0.3 L of void space per L of soil volume) from the Geotechnical Report (DOE-ID 2000). This void volume (0.162 L/kg) corresponds to 6.18 kg of landfill soil exposed to

each liter of leachate. In the geochemical model, 1 liter of the water mixture described above was equilibrated with specified molar quantities of minerals present in 6.18 kg of soil. In reality, the waste mass may not be completely saturated. For minerals with low solubility, the concentration in leachate is limited by solubility, so that a smaller amount of leachate will show the same concentration. The amount of mass removed will be less with a smaller volume of water. For soluble compounds such as iodide salts, the leachate would be more concentrated. However, the same amount of mass would be extracted during the operations period as the mass calculated assuming complete water saturation. A sensitivity analysis demonstrating these concepts will be provided in the 90% submittal.

The second step was to assign mineral phases to site soil and/or controls on constituent solubility. Mineralogy (with average percent abundance) reported in soils on the Chemical Processing Plant consisted of quartz, plagioclase feldspar, potassium feldspar, calcite, pyroxene, and detrital mica (USGS 1989). Simplifying assumptions were made to assign specific minerals to plagioclase (albite), potassium feldspar (orthoclase), pyroxene (enstatite), and mica (illite). The average percent abundance of each mineral was converted to grams per kilogram (g/kg) with the assumption that the minerals have approximately equal density. The g/kg concentrations were converted to total grams by multiplying by 6.18, the total kg exposed to each liter of leachate (see above). Finally, the grams of each mineral were converted to moles by dividing by the molecular weight of each mineral. The calculations are summarized in Table 3-1. There was a significant difference between the calculated major cation masses (sodium, potassium, calcium, and magnesium) from this mineral assemblage and those reported in the inventory. For example, if all sodium reported in the inventory is assigned to albite, there is far less mass of this mineral present than that suggested by this assemblage. If a more calcic plagioclase were used, there would be an excess of both sodium and calcium. It was concluded that inventory masses of major elements were measured by partial dissolution of samples by acid extraction, rather than a complete digestion of waste sample soils, and therefore the calculated masses in Table 3-1 are most representative.

Table 3-1. Calculation of mineral abundance in site soils.

	Quartz	Albite (plagioclase feldspar)	Orthoclase (potassium feldspar)	Calcite	Enstatite (pyroxene)	Illite (mica)
Average Abundance (%) ^a	38.3	22.7	13	3.7	13	9.3
g/kg soil	383	227	130	37	130	93
mineral molar mass (g/mol)	60	262	278	100	100	383.5
mol/kg soil	6.39	0.87	0.47	0.37	1.30	0.24
mol exposed to 1 L leachate ^b	39.5	5.3	2.9	2.3	8.0	1.5

a. Average of BLRB-7, BLRB-8, and BLRB-9 (USGS 1989).

b. Calculated by multiplying mol/kg by 6.18 kg soil per L void space (see text).

Some of the soil minerals listed above are typically not in direct equilibrium with pore water in natural environments. Feldspars will have a microscopic layer of clay mineral on the weathering surface. Though minute in overall mass concentration, this clay mineral (kaolinite was assigned for modeling purposes) controls the aqueous concentrations of aluminum and silicon. As magnesium-rich minerals such as enstatite dissolve, the aqueous concentration of magnesium is typically controlled by dolomite in

this environment. Kaolinite and dolomite were input to PHREEQC only as solubility controls, not as quantified masses.

Inventory elements not accounted for by the calculations in Table 3-1 were assigned to realistic mineral phases based on the soil environment. Metal cations were commonly assigned oxides or hydroxides that form in near-surface regimes. Whenever possible, the most common near-surface mineral phase was assigned as a source for a given element. Although not expected in oxidizing environments, significant sulfide was reported in the inventory. To account for this mass, the common sulfide minerals pyrite (FeS_2) and sphalerite (ZnS) were included. In a soil environment open to the atmosphere such as this one, equilibrium calculations predict that all sulfides will be oxidized to sulfate and dissolve. Zincite (ZnCO_3) was used as a control on zinc solubility, and ferric hydroxide ($\text{Fe}(\text{OH})_3$) was used both as a source of iron and a control on its solubility. A list of the screened elements and their mineral source(s) and control(s) is provided in Appendix A as Table A-5.

Two very soluble anions, cyanide (CN^-) and iodide (I^-), were not represented by realistic minerals in the LLNL database. To account for these constituents, they were added as sodium salts under the reasonable assumption of complete dissolution.

One liter of the infiltrating water mixture was equilibrated with the molar quantities of the minerals listed in Table A-5. Because the system is open to the atmosphere during the operating period, the system was also equilibrated with atmospheric oxygen ($P_{\text{O}_2} = 0.2 \text{ atm}$) and carbon dioxide ($P_{\text{CO}_2} = 10^{-3.5} \text{ atm}$). Redox potential was calculated on the basis of oxygen fugacity and pH was calculated as part of the equilibration process.

3.3 Results of Geochemical Modeling

The modeled concentrations of the screened elements are reported in Appendix A as Table A-6. The complete PHREEQC input and output files are provided in Appendix B. The modeled leachate is a brackish to saline water (total dissolved solids between 20,000 and 30,000 mg/L) dominated by sodium and sulfate with a pH of 8.2. The water chemistry is most influenced by the oxidation of sulfide minerals, equilibration of carbonates, and dissolution of the more plentiful of the soluble components of the design inventory (such as boron, phosphorus, terbium, vanadium, and ytterbium, along with the major elements).

Elements that were only slightly soluble included barium, zinc, plutonium, and uranium. Salts of most anionic constituents were completely dissolved before equilibration could be reached. These constituents included chloride, iodide, technitium, selenium, and arsenic.

These results represent an approximation of chemical conditions. The mineral phases were chosen on the basis of the best available data and what were considered reasonable assumptions regarding the geochemical environment. As discussed above, the model predicted complete oxidation of sulfide due to the equilibration condition with atmospheric oxygen. This process may be limited during the operation period by chemical kinetics, but insufficient data were available for quantification.

Partition coefficients (K_d values) were not applied during the geochemical modeling stage. The leachate generation calculations, described in Section 4, involved application of K_d values and radioactive decay.

4. LEACHATE GENERATION RATE AND CONTAMINANT REDUCTION OVER TIME

Application of partition coefficients (K_d values) and radioactive decay were combined with assumed annual leachate losses from the proposed ICDF landfill to calculate changes in leachate and landfill composition over the operating period. As discussed in Section 2, two groups of screened constituents were examined: one screened on the basis of K_d values and one on the basis of mass concentrations and solubility. The methodology and results for each group are presented below.

4.1 Leachate Generation with K_d -Screened Group

4.1.1 Introduction

An analytical solution was applied to estimate the loss of substantially soluble contaminants from the emplaced waste in the ICDF during the period of time that the landfill is in operation and prior to placement of the final cover. This model is intended to support decisions regarding waste placement and to facilitate understanding of the mobility of contaminants within the facility. The approach and methodology used in this evaluation are described in the following sections.

4.1.2 Approach

An analytical model utilizing Microsoft Excel™ spreadsheet tools was prepared. The spreadsheet format allows rapid development of the model and ease of modification of specific input parameters to support sensitivity analyses. Input to the model includes the ICDF design inventory for contaminants, estimated waste mass and volume, and project-specific distribution/partition coefficients (K_d) for site contaminants. The model utilizes simplified assumptions regarding moisture content of the waste soil within the landfill and water recharge through the emplaced waste. The details of the model methodology are described in Section 4.1.3.

4.1.3 Methodology

The inventory and release model estimated the concentration of each contaminant in the leachate exiting the ICDF yearly, and the amount of contaminant mass remaining within ICDF. These parameters were modeled over a 15-year period during which the landfill is assumed to be open to ambient precipitation. The model assumptions and computational details are described in the following subsections.

4.1.3.1 Assumptions. The following assumptions were applied to support development of the analytical model:

1. The landfill was assumed to be filled to capacity at the start of the computations (i.e., the design volume of waste soil is in place at time zero [T_0], or the start of the simulation).
2. The waste soil is assumed to be saturated at the beginning of the simulation.
3. The initial leachate concentration was assumed to be the contaminant mass present in the design inventory, distributed between the solid and solution phases according to the site-specific distribution coefficients (K_d).
4. Only contaminants considered to be significantly leachable (i.e., contaminant $K_d < 20$) were included in the simulation.

5. Contaminant mass leaves the landfill only by leachate transport and/or by radioactive decay, as applicable.

4.1.3.2 Computations. Contaminant mass exited the ICDF only by means of leachate transport or radioactive decay. The model essentially consisted of determining the equilibrium concentration of each contaminant in solution and on solids contained within the ICDF, and then calculating the amount of leachate exiting the ICDF. Thus, the amount of contaminant remaining in the ICDF equaled the initial mass minus the mass transported by the leachate and the mass lost to radioactive decay.

Beginning with the initial inventory for each contaminant, the total mass (m_T) equaled the mass of contaminant on the solids (m_s) and the mass of contaminant in solution (m_{aq}):

$$m_T = m_s + m_{aq} \quad \text{or} \quad m_T = c_s M + c_{aq} V$$

where the contaminant concentration on the solids (c_s) equals the mass of the contaminant on the solids divided by the total mass content of the ICDF (M), and the contaminant concentration in solution (c_{aq}) equals the mass of contaminant in solution divided by the total water content of the ICDF (V). The ICDF was assumed to be saturated at the start of the simulation time with no change occurring in the water content. At equilibrium, the distribution coefficient (k_d) equals the ratio the contaminant concentration on the solids to the contaminant concentration in solution:

$$k_d = \frac{c_s}{c_{aq}}$$

Combining the two equations results in the following relationships:

$$c_{aq} = \frac{m_T}{(k_d M + V)} \quad \text{and} \quad c_s = k_d c_{aq}$$

The mass of contaminant exiting the ICDF each year equals the leachate concentration multiplied by the leachate volume (v_i), and the mass remaining (m_{Ti+1}) is the initial mass minus the leachate mass, less the mass lost to radioactive decay:

$$m_{Ti+1} = m_{Ti} * e^{-\ln(2)/T_{1/2}} - c_{aq} v_i$$

where $T_{1/2}$ is the half-life of the radioactive isotope. For non-decaying contaminants ($T_{1/2} = 0$), the exponential term equals 1. The contaminant concentrations for the following year are then calculated using the remaining total mass, and the preceding contaminant concentration equations.

4.1.3.3 Variable Input Parameters. Estimated water recharge through the landfill waste mass was the only input parameter that was varied during this simulation. Two simulations were performed using 1.0 centimeter (cm) and 10.0 cm annual recharge, respectively. The naturally occurring background recharge for the INEEL site is estimated to be 1.0 cm per year.

4.1.4 Simulation Output and Discussion

Concentrations of all screened constituents from Table A-1 were calculated for each year of the operation period, and the results are presented in Appendix C of this report. The leachate reduction simulation indicates that contaminant mass reduction due to leaching of selected leachable contaminants

(i.e., iodine-129, technetium-99, and neptunium-237) is typically less than 10% of the design inventory mass over the 15-year simulation period at the 1.0 cm/yr recharge rate. Iodine-129 was assigned a K_d of zero, and therefore represents the most conservative conditions. Technitium-99 and neptunium-237 have K_d values of 0.2 and 8.0, respectively. The leachate concentration and residual contaminant mass for the selected iodine, technetium, and neptunium isotopes for the 15-year simulation period at the 1.0 cm/yr and 10 cm/yr recharge rates are shown in Figures 4-1 and 4-2, respectively.

The estimated volume of landfill leachate generated annually under this simulation is approximately 550,000 L at the 1.0 cm/yr recharge rate and 5,500,000 L at the 10 cm/yr recharge rate.

Under the landfill leaching scenario developed for this simulation, an annual recharge rate of approximately 0.3 m/yr would be required to remove 90% of the design inventory masses of iodine-129, technetium-99, and neptunium-237.

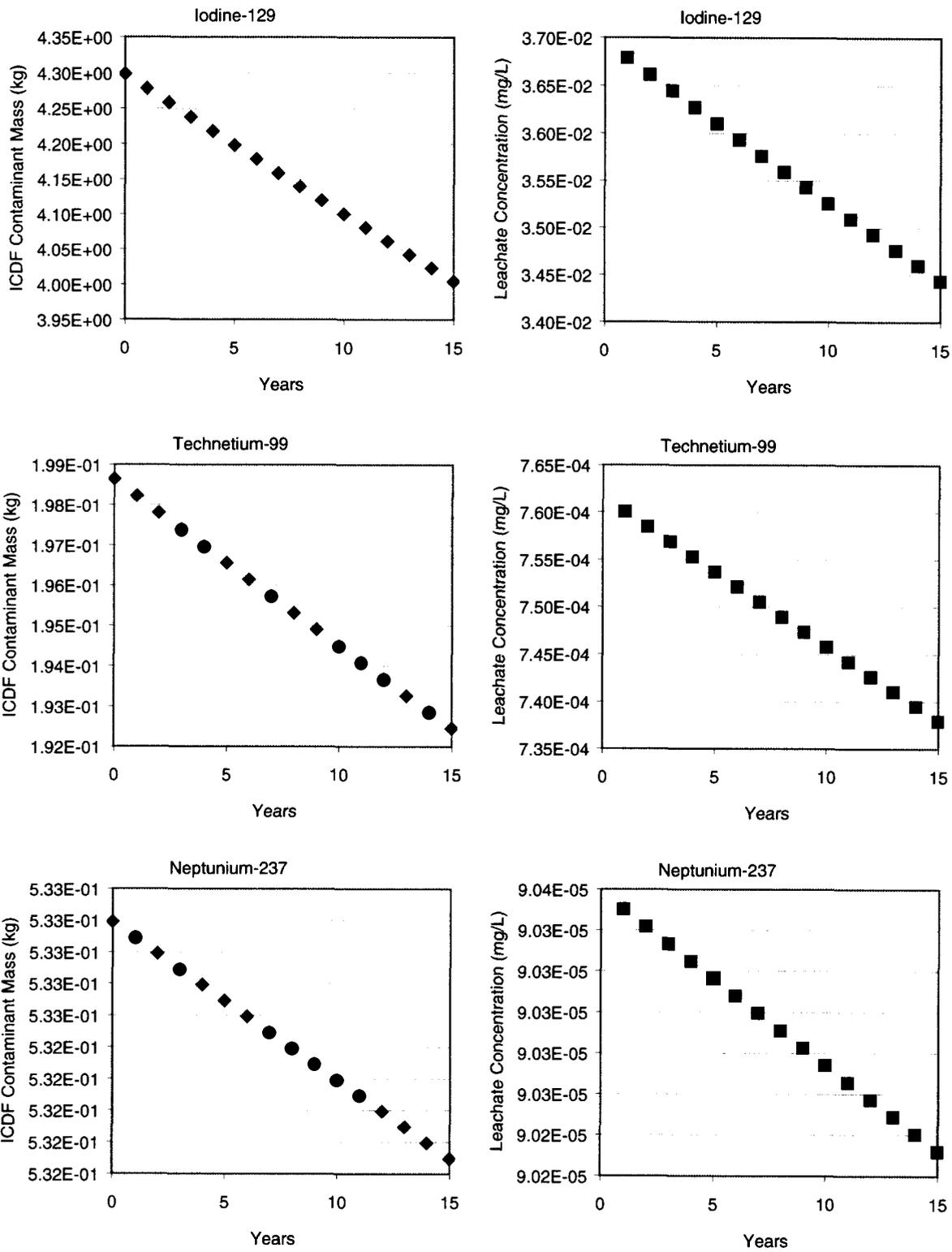


Figure 4-1. Evaluation of leachate concentration and residual mass in the ICDF at 1.0 cm/yr recharge.

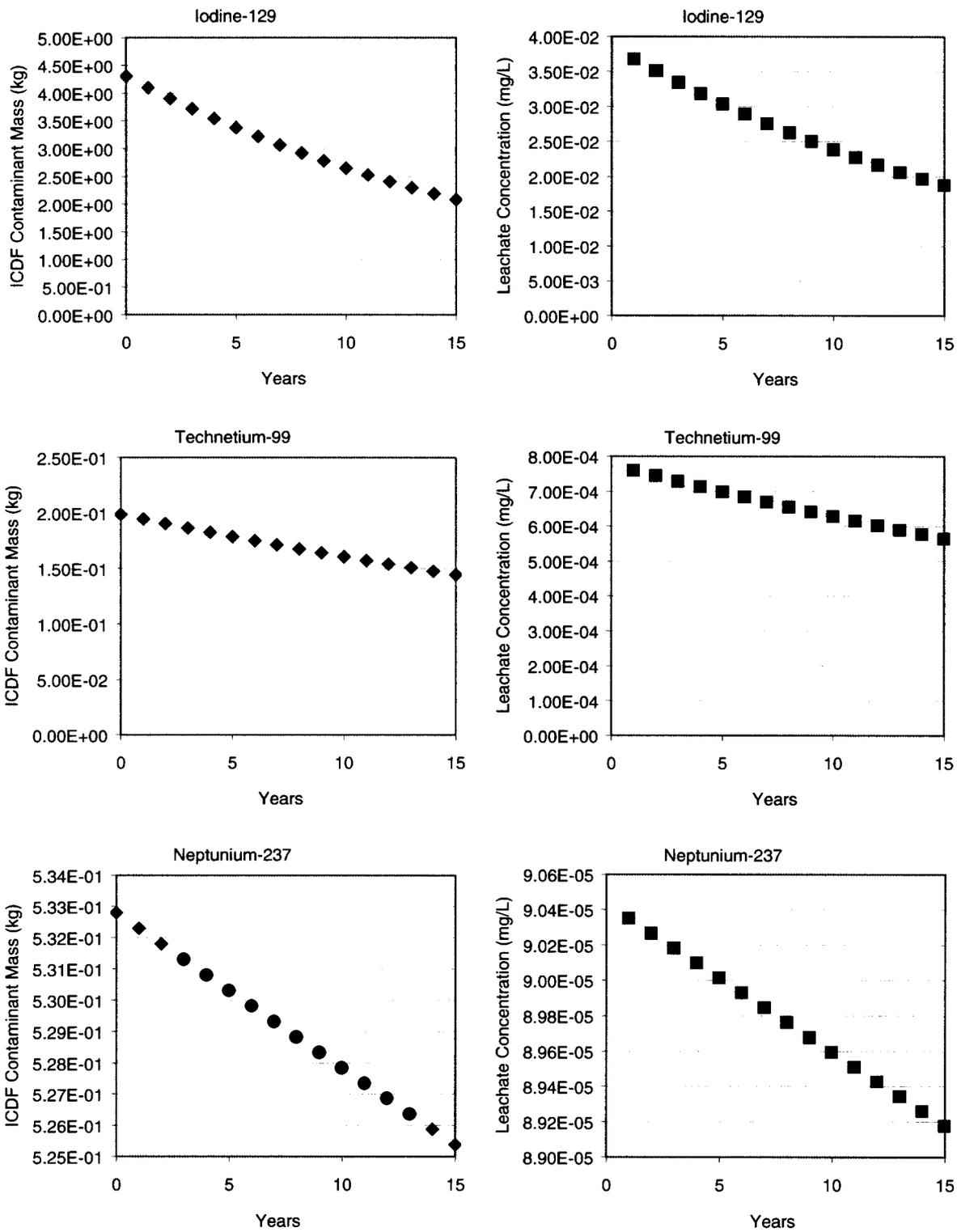


Figure 4-2. Evaluation of leachate concentration and residual mass in the ICDF at 10.0 cm/yr recharge.

4.2 Leachate Generation with Concentration/Solubility-Screened Group

The second group of constituents was screened on the basis of concentration (see Section 2) for cations and organics. No anionic constituents were screened. The group was subjected to solubility modeling using PHREEQC (Section 3). The resulting aqueous concentrations from the PHREEQC output were then corrected for adsorption partitioning using the retardation factor, $r_f = 1 + (B_d/n)K_d$, where B_d is bulk density (1.855 kg/L) and n is porosity (0.3). Because the PHREEQC simulations only accounted for solubility constraints, the output concentrations were divided by the r_f to correct for adsorption.

The corrected aqueous concentrations were input to the spreadsheet, and the same methodology was applied as that described in Section 4.1. For iodine and technitium, the same plots of leachate concentration with time were generated as shown in Figures 4-1 and 4-2 above, because both these elements are completely soluble. The complete model output for this group is provided in Appendix D.

5. CONCLUSIONS

Concentrations of selected design inventory constituents in ICDF landfill leachate were simulated over the 15-year operations period. The purpose of the study was to examine the change in leachate concentration over time, as it is directed toward the evaporation pond. The results may be applied toward performance assessment modeling in the future.

Two groups of screened design inventory constituents were applied to the leachate/contaminant reduction time study. One group was screened on the basis of partition coefficients and the other on the basis of concentration in the design inventory. The latter group was entered into a geochemical model to simulate solubility constraints on the resulting leachate concentrations. Concentrations of each group were entered into a spreadsheet program that simulated partitioning to the solid phase via adsorption, radioactive decay, and leachate removal from the landfill.

The leachate will be a brackish to saline water dominated by sodium and sulfate and buffered by carbonates to a pH of around 8.2. The results indicate less than 10% of the inventory masses of the most mobile constituents (iodine and technitium) are expected to be removed from the landfill during the operation period of 15 years, mainly due to the modest amount of leaching that is expected to take place in this time frame.

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