Journal of the Korean Nuclear Society
Volume 33, Number 2, pp.156~165, April, 2001

Important Radionuclides and Their Sensitivity for Groundwater Pathway of a Hypothetical Near-Surface Disposal Facility

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(Received May 26, 2000)

Abstract

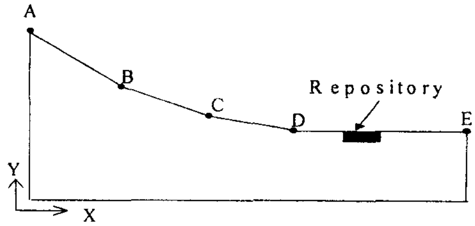
A radiological safety assessment was performed for a hypothetical near-surface radioactive waste repository as a simple screening calculation to identify important nuclides and to provide insights on the data needs for a successful demonstration of compliance. Individual effective doses were calculated for a conservative groundwater pathway scenario considering well drilling near the site boundary. Sensitivity of resulting ingestion dose to input parameter values was also analyzed using Monte Carlo sampling. Considering peak dose rate and assessment timescale, C-14 and I-129 were identified as important nuclides and U-235 and U-238 as potentially important nuclides. For C-14, the dose was most sensitive to Darcy velocity in aquifer. The distribution coefficient showed high degree of sensitivity for I-129 release.

Key Words : radioactive waste disposal facility, groundwater scenario, sensitivity

1. Introduction

Upon release from the waste form, the radionuclides will migrate out of the radioactive waste disposal facility, mainly through the groundwater pathway to geosphere. The geosphere has the function of retarding the movement of the radionuclides by various physical and chemical interactions such as dispersion and sorption. Radionuclides are delayed by these retardation processes in the geosphere and these will finally be transported with dilution to the biosphere, where these radionuclides can enter the human exposure pathways, such as food chains. Therefore the goal of the radiological safety assessment is to determine whether radiological exposure doses or risks to members of the public will meet the appropriate regulatory requirements of quantitative performance objectives. The safety assessment calculations can be required iteratively at various repository development phases such as site selection and characterization, facility construction, disposal operations and final site closure. A safety assessment at early stage aims at identifying the most important nuclides that influence the performance of the disposal system, optimizing disposal facility design, and eliminating unnecessary effort and resource needs in site characterization and other activities. U.S. NRC recommended a tiered approach, which starts with the highly conservative inventories and release mechanisms and processes to a less conservative approach be used. This approach recommends to screen out radionuclides with half-lives less than 5 years, which are not present in significant activity levels and do not have long-lived daughter products. And it also recommends that a dose calculation should be performed under the assumption that the engineered barriers are completely ineffective in delaying or retaining radionuclides within the disposal facility. Important nuclides will be determined by calculating the transport of the radionuclides in soil and groundwater under the conservative assumptions[1].

In this study, the influence of radiological impacts on a potential member of the critical group was evaluated on the basis of a modular approach. It was mainly composed of source term analysis and transport analysis of radionuclides through groundwater pathway, and was coupled each other in such a way that the radionuclide mass release flux from the bottom of the disposal vault was read as input to the groundwater transport model. A highly conservative groundwater pathway scenario considering well drilling near the site boundary was assumed. Sensitivity of resulting ingestion dose to input parameter values was also analyzed using Monte Carlo sampling.



Topography Coordinate	A	B	C	D	E
X(m)	0	550	790	1230	1700
Y(m) (Unit: Sea level)	360	150	100	50	50

Fig. 1. Local Topography of a Hypothetical Disposal Repository

2. Near-surface Disposal System Descriptions

The local topography of the arbitrary site is illustrated in Figure 1. Granite is well developed in the site. The disposal facility was assumed to be constructed on a hillside that runs from southwest to northeast. The site map shows relatively steep gradient in hilltop region and slow gradient in the rice field region adjacent to the main road. The native soils at the site are assumed to be relatively permeable. The climatic condition was set to be humid environment with annual average rainfall of 1.5 meters[2].

The capacity of the disposal facility in the initial phase is around 100,000 drums (based on 200*l* drum). It consists of three types of disposal vault depending on the durability and/or size of waste packages. The typical disposal vaults are as follows:

- Vault Ⅰ : waste packages with long durability (such as concrete containers) backfilled with gravel,
- Vault Ⅱ : standard size waste packages with short durability (such as 200*l* steel drums) grouted with concrete,
- Vault Ⅲ : large size waste packages with short durability(such as 350*l* repackaged steel drums) grouted with concrete.

Twenty vaults (including 17 grouted vaults (7 for vault II and 10 for vault III) and 3 backfilled vaults for vault I) will be constructed and laid out in two rows. The facility is oriented such that the longitudinal direction of the facility is perpendicular to the main direction of aquifer

Table 1. Facility Dimensions(100,000 Drums Equivalent)

Description	Dimensions (m)
Length	266
Width	70
Height	8.7
Thickness of barrier layer (bentonite-sand mixture)	2
Distance above water table	3

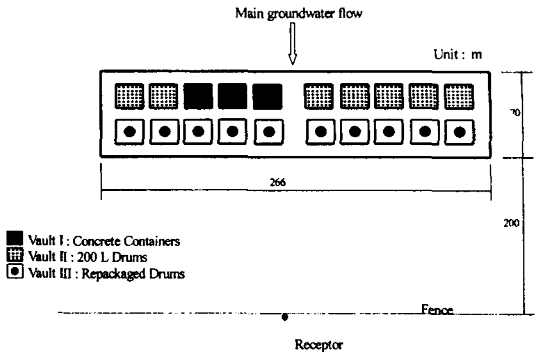


Fig. 2. General Arrangement of the Disposal Facility

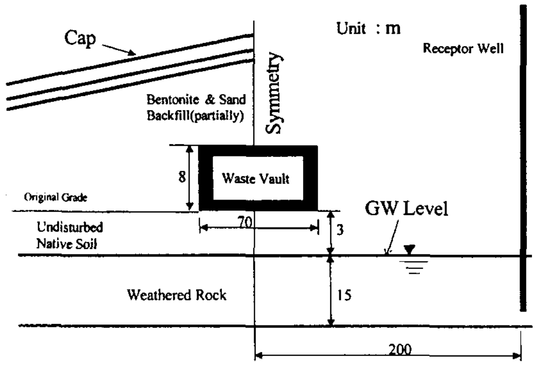


Fig. 3. Cross-sectional View of the Disposal Facility

flow. In the conceptual design, the interval of each vault is 2 m in the longitudinal direction and 10 m in transverse direction in consideration of subsidence stability, waste package handling, and vault maintenance and constructional aspects.

The final disposal cover will be constructed after the disposal vaults in a disposal area are completely filled. The final cover adopts multi-layer system to ensure low percolation, water drain, and intrusion resistance. Top soil surface layer with native soil of the site and lowermost barrier layer with composite of 20%-bentonite and 80%-sand was suggested in the conceptual design. The facility dimension for the initial phase of disposal is given in Table 1. Two kinds of layers were assumed, one for unsaturated native soil layer with 3 m thickness directly below the base of the concrete vault, the other for saturated relatively unconsolidated weathered rock layer. Saturated zone is assumed as a horizontal layer with constant thickness of 15 m, and it is also assumed that Darcy velocity of 10 m/yr along the perpendicular direction to the longest dimension of the facility. A well is assumed to be located at 200 m away from the disposal facility. A well-water drinking scenario was also used for biosphere modeling. Figures 2 and 3 show the layout and the cross-sections of the disposal facility, respectively.

3. Assessment Modeling

As a conservative analysis of the radiological impact via groundwater pathway a well drilling scenario near the site boundary is considered. During the institutional control period, waste forms in non-degraded disposal containers would not contact with water, thus leaching could occur only in degraded containers. The artificial barrier layers such as asphalt and geomembrane among the multi-layer cover are assumed to be degraded after 100 years of closure. At the end of institutional control period, assumed in this study as 300 years after closure of the disposal facility, all engineered barriers including disposal containers and waste matrices are also assumed to be completely deteriorated so that water infiltration rate is almost

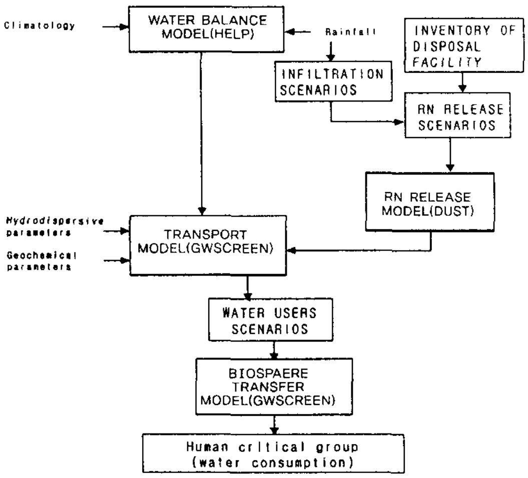


Fig. 4. Groundwater Pathway Assessment Scheme

the same as that of native soils in the site. The radionuclides leached from waste forms begin to migrate to soil and aquifer, and finally reach to wells located near the site boundary. The nearest well is assumed to be 200 meters away from the centerline of the waste vaults.

In this study, the radiological safety assessment was mainly divided into two parts, source term modeling and radionuclide transport modeling. Figure 4 represents the groundwater pathway assessment scheme and computer tools used.

Table 2 lists the radionuclide inventory of the facility considered in the safety assessment. Radionuclides with half-lives less than 5 years are eliminated. However, long-lived radionuclides with a highly potential mobility (e.g., C-14, Tc-99, I-129 and alpha-emitting transuranics such as Pu-239) are included. In addition, the radionuclides with a relatively high dose conversion factor and/or significant ingrowth of daughter radionuclides (e.g., U-238) are also included in the inventory.

Conceptual model for the source term evaluation is illustrated in Figure 5. Radionuclide release rates through the bottom of concrete vault were calculated with the DUST-MS code[3]. Homogenization by repository-averaging process

Table 2. Radionuclides, Half Lives, and Inventories Considered in the Assessment

Radionuclide	Half Life(years)	Inventory(Bq)
H-3	1.24E+1	2.60E+13
C-14	5.73E+3	1.68E+13
Co-60	5.27E+0	1.73E+14
Ni-59	7.50E+4	3.65E+12
Ni-63	9.60E+1	9.51E+13
Sr-90	2.91E+1	1.39E+12
Nb-94	2.03E+4	1.00E+11
Tc-99	2.13E+5	4.07E+10
I-129	1.57E+7	1.25E+10
Cs-137	3.00E+1	6.11E+13
U-235	7.04E+8	1.91E+08
U-238	4.47E+9	4.74E+10
Pu-238	8.77E+1	1.28E+11
Pu-239	2.41E+4	5.88E+10
Total		3.77E+14

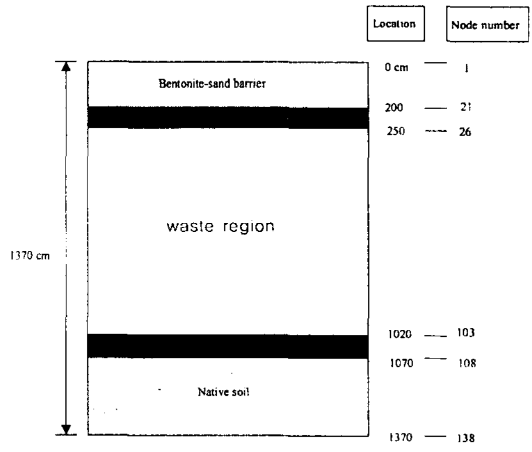


Fig. 5. DUST-MS Model for Source Term Evaluation

was made in the source-term modeling. The appropriate volume for averaging radionuclide concentration was assumed the entire volume of the disposal facility, including any uncontaminated regions between individual disposal vaults. Three time intervals - namely, the first 100 years, 100 to 300 years, and from 300 years on - were considered to take into account the degradation of

Table 3. Distribution Coefficient and Solubility

Nuclides	Distribution coefficient (m^3/kg)							Solubility, (M)
	Bentonite -sand barrier	Concrete layer	Waste region			Native Soil	Aquifer (weathered rock)	
			Vault I	Vault II	Vault III			
H-3	0	0	0	0	0	0	0	-
C-14	5.0E-3	2.5E+0	2.5E+0	2.5E+0	2.0E-3	5.0E-3	1.0E-2	6.4E-2
Co-60	2.0E-1	2.0E-2	2.0E-2	2.0E-2	1.0E-2	1.5E-2	1.0E+0	1.2E-3
Ni-59, Ni-63	1.0E-1	2.0E-2	2.0E-2	2.0E-2	9.0E-3	4.0E-2	1.0E+0	-
Sr-90	1.0E-1	2.5E-3	2.5E-3	2.5E-3	8.0E-3	1.5E-2	2.0E-2	2.5E-1
Nb-94	2.0E+0	5.0E-1	5.0E-1	5.0E-1	0	0	1.0E+0	5.6E-8
Tc-99	5.0E-1	6.0E-1	5.0E-1	6.0E-1	5.0E-4	1.0E-4	1.0E+2	3.8E-8
I-129	1.0E-2	6.0E-4	6.0E-4	6.0E-4	5.0E-3	1.0E-3	5.0E-3	-
Cs-137	2.0E-1	2.5E-4	2.5E-4	2.5E-4	1.0E-3	3.0E-1	1.0E-1	-
U-235, U-238	1.0E+1	2.0E+0	2.0E+0	2.0E+0	-	-	1.0E+2	3.3E-7
Pu-238, Pu-239	2.0E+0	4.0E+1	4.0E+0	4.0E+1	-	-	5.0E+0	1.2E-9

engineered barrier. To estimate the water influx into the vault for the source term analysis, the long-term performance of the final cover was simulated by considering the degradation of artificial barrier materials under various precipitation conditions[4]. These infiltration rates were used as the input data for the initial condition in the DUST-MS code to estimate the mass release rate for each radionuclide.

Finally, the GWSCREEN code[5] was used to analyze the radionuclides transport in unsaturated soil and the aquifer, human uptake, and doses to the individual in a critical group. The code was developed to perform initial screening calculations for the groundwater pathway impacts resulting from the leaching of radioactive contaminants from surface or buried sources. The results from the codes have been shown to provide bounding estimates of groundwater concentrations when compared to those from other numerical codes[6]. The modeling approach used in GWSCREEN is based on a mass balance concept, which has its roots in the methodology presented by the Nuclear Regulatory Commission. Three primary models are employed in GWSCREEN: the source model, the unsaturated zone mass transport model, and the saturated zone mass transport model. These models are coupled together by the contaminant fluxes across the interface between model boundaries. In this assessment the source mass fluxes at the base of the concrete vault, calculated from DUST-MS as a function of time, were replaced with the source model. The unsaturated and saturated zone was modeled as an isotropic, homogeneous porous medium.

Input parameters as shown in Tables 3 to 5, such as material properties, distribution coefficient, solubility and dose conversion factor of

Table 4. Flow and Transport Material Properties

Materials			Effective diffusion coefficient(m^2/sec)		Dispersion coefficient(m)	Bulk density (kg/m^3)	
Bentonite-sand barrier			1.0E-10		0.10	2,000	
Concrete layer			1.6E-12		0.05	2,500	
Waste region	Vault I		1.7E-12		0.05	2,500	
	Vault II		7.9E-12		0.05	2,000	
	Vault III		1.3E-11		0.10	1,800	
Materials	Water Content		Van Genuchten Parameters		Saturated hydraulic conductivity (m/yr)	Dispersion coefficient (m)	Bulk density (kg/m^3)
	Residual	Saturated	$\alpha(m^{-1})$	n			
Native soil	0.21	0.3	3.5	3.0	4.42E-02	0.10	1,820
Aquifer (Weathered rock)	-	0.25	-	-	1.0E+1	5~10	2,522

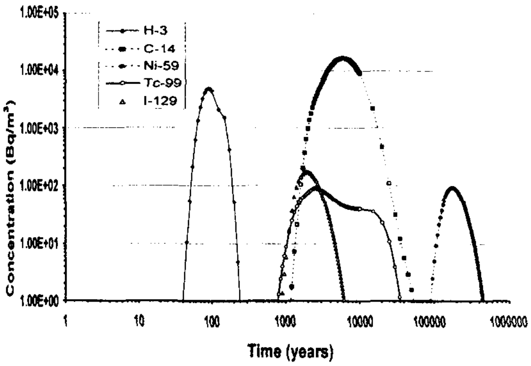


Fig. 6. Concentration in Well Water

each radionuclide were determined from the related previous studies[7,8].

4. Results and Discussion

4.1. Radionuclide Screening Results

The time evolution of the concentration of radionuclides(Bq/m^3) extracted from the well is shown in Figure 6. Figure 7 shows the peak ingestion dose and the time of the peak for each radionuclide release, calculated by GWSCREEN. Calculations have been undertaken until it can be demonstrated that the total peak dose has been found. The dose is calculated by assuming that a person consumes drinking water of 0.73 m^3/yr from the well[7]. This generally yields the highest dose in the absence of irrigation. Considering the peak dose and assessment timescale of 1,000 years stipulated in the regulation, the contribution of C-14 and I-129 to the final dose is significantly higher than that of other nuclides. Therefore, these nuclides could be identified as important nuclides. The time evolution of the total doses shows two peaks in Figure 8. These peaks correspond to the contribution of each nuclide. The first and second peaks reach at about 2,000 and 6,000 years after closure of the disposal facility, and these are mainly due to C-14 and I-129, respectively. Total effective dose received by the critical individual in the case of the water consumption from the well shows around 0.01 mSv/yr. This value is below the performance objectives, the predicted radiological risk of 10^{-6}/yr or its dose equivalent 0.02 mSv/yr to any individual in a critical group, set up in the national regulation applicable to the LILW disposal facility

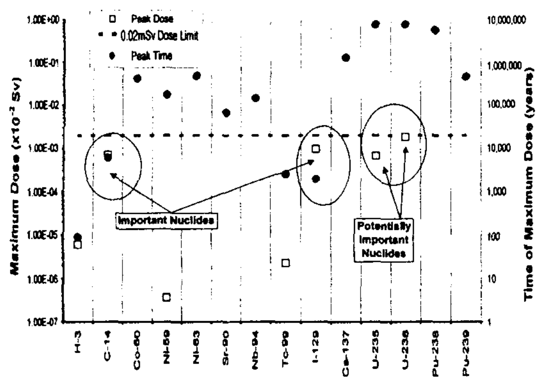


Fig. 7. Peak Dose and the Time of the Peak for Each Radionuclide

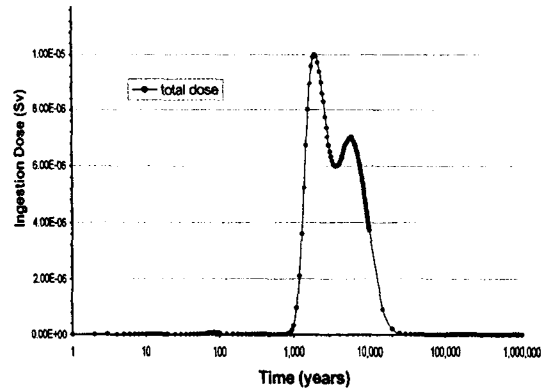


Fig. 8. Effective Dose Due to Ingestion of Contaminated Well Water

Table 5. Ingestion Dose Conversion Factors

Nuclides	Half-life	Molecular weight	Dose Conversion Factor
	(years)	(g/mol)	(Sv/Bq)
H-3	1.24E+01	3	1.70E-11
C-14	5.73E+03	14	5.68E-10
Co-60	5.27E+00	60	7.03E-09
Ni-59	7.50E+04	59	5.41E-11
Ni-63	9.60E+01	63	1.46E-10
Sr-90	2.91E+01	90	8.11E-10
Nb-94	2.03E+04	94	1.38E-09
Tc-99	2.13E+05	99	3.51E-10
I-129	1.57E+07	129	7.57E-08
Cs-137	3.00E+01	137	1.35E-08
U-235	7.04E+08	235	6.78E-08
Pa-231	3.28E+04	231	2.97E-06
Ac-227	2.18E+01	227	3.95E-06
U-238	4.47E+09	238	6.62E-08
U-234	2.45E+05	234	7.03E-08
Th-230	7.54E+04	230	1.43E-07
Ra-226	1.60E+03	226	2.97E-07
Pb-210	2.23E+01	210	1.81E-06
Pu-238	8.77E+01	238	1.03E-06
U-234	2.45E+05	234	7.03E-08
Th-230	7.54E+04	230	1.43E-07
Ra-226	1.60E+03	226	2.97E-07
Pb-210	2.23E+01	210	1.81E-06
Pu-239	2.41E+04	239	1.16E-06
U-235	7.04E+08	235	6.78E-08
Pa-231	3.28E+04	231	2.97E-06
Ac-227	2.18E+01	227	3.95E-06

(Note: Pu-238 modelled as U-234)

in the post-closure period[9].

In the meanwhile, some actinides included in the inventory such as U-235 and U-238 show that these can be potentially important nuclides even though the peak time is too long to be considered in the LILW repository. Decay of radionuclides to produce other radionuclides was considered to account for production of progeny in the radiological dose assessment of uranium containing wastes. To evaluate the movement of radioactive progeny, radioactive progeny were assumed to travel at the same rate as their parent in the GWSCREEN. The assumption is also made that no progeny exist at the time of waste emplacement. Figures 9 and 10 show the effective dose from U-235, U-238 and their progeny, respectively.

4.2. Sensitivity Analysis

Sensitivity analysis was carried out with the variation of different input parameters such as percolation rate, unsaturated zone thickness, transverse dispersivity, Darcy velocity in aquifer, distribution coefficients in unsaturated zone and aquifer, and water intake rate. The objective of the sensitivity analysis was focused to identify the important parameters by determining the relative

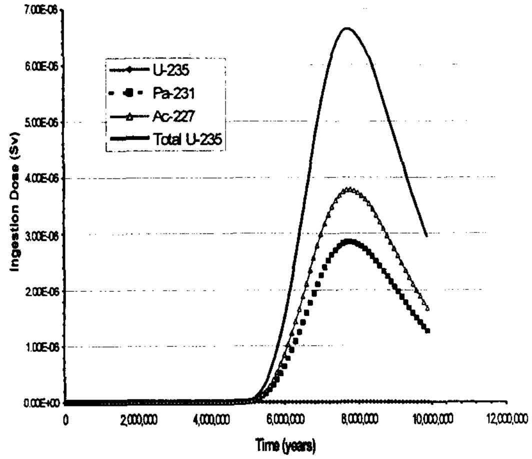


Fig. 9. Effective Dose from U-235 and its Daughter Nuclides

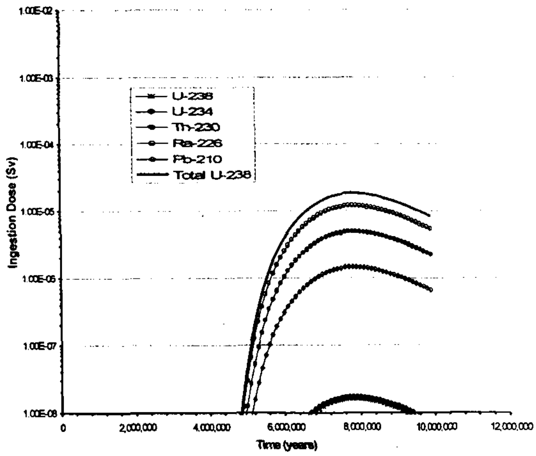


Fig. 10. Effective Dose from U-238 and its Daughter Nuclides

contributions to the resulting dose. Monte Carlo sampling technique was applied to find the sensitivities of ingestion dose of C-14 and I-129 exposure to input parameter values. The simulation was performed for five hundred Monte Carlo trials. Parameter distribution type of each variable was assumed among the normal, lognormal, uniform, and triangular distributions as given in Table 6. The ranges of values for these parameters were also selected to represent a wide range of field condition. After site characterization

Table 6. Distribution and Range of Input Parameters Used in the Sensitivity Analysis

Parameters	Distribution type	Unit	Range of values
PERC *	Lognormal	m/yr	0.005 ~ 10
DEPTH	Triangular	m	1.0 ~ 5.0
AY	Normal	m	0.01 ~ 0.5
U	Triangular	m/yr	3.15 ~ 20.0
KDU	Lognormal	m^3/kg	0.00004 ~ 0.03
KDA	Lognormal	m^3/kg	0.00005 ~ 0.1
WI	Uniform	m^3/yr	0.18 ~ 0.73

* PERC : percolation rate, DEPTH : unsaturated zone thickness, AY : transverse dispersity, U : Darcy velocity in aquifer, WI : water intake rate, KDU, KDA : distribution coefficients in unsaturated zone and aquifer, respectively.

has been completed, the range of input parameter values will be further refined, and the distributions should also be available. Sensitivities were calculated by computing the rank correlation coefficient between each stochastic variable and the resulting dose. Percent contribution to the variance in dose is approximated by squaring the rank correlation coefficients and normalizing them to 100%. Sensitivity of ingestion dose to input parameter values is given in Table 7. It is found that the dose is most sensitive to Darcy velocity in aquifer followed by distribution coefficient in unsaturated zone for the case of C-14. The release of I-129 is highly sensitive to the distribution coefficient.

5. Conclusions

A radiological safety assessment was performed for a hypothetical near-surface radioactive waste repository as a simple screening calculation to identify important nuclides and to provide insights on the data needs for a successful demonstration of compliance. Individual effective doses were calculated under a highly conservative groundwater pathway scenario considering well

Table 7. Sensitivity Analysis Results for C-14 and I-129 Releases

Parameters*		PERC	DEPTH	AY	U	KDU	KDA	WI
C-14	Correlation coefficient(r)	0.224	-0.073	0.047	-0.626	-0.307	-0.202	-
	Contribution to variance(%)	8.61	0.90	0.39	66.94	16.16	7.01	-
I-129	Correlation coefficient(r)	0.309	-0.145	0.029	-0.221	-0.532	-0.035	0.234
	Contribution to variance(%)	18.89	4.14	0.17	9.68	56.05	0.24	10.84

* Abbreviations are the same as Table 6.

drilling near the site boundary. The results of preliminary safety assessment showed that the effective doses for groundwater pathways were below the current regulatory limit in Korea. That means the conceptual design of disposal vaults could be properly implemented from the radiological safety point of view. C-14 and I-129 were identified as important nuclides and U-235 and U-238 as potentially important nuclides, considering peak dose rate and assessment timescale. Sensitivity of resulting ingestion dose to input parameter values was also analyzed using Monte Carlo sampling. For C-14, the dose was most sensitive to Darcy velocity in aquifer. The distribution coefficient, which is usually a very sensitive parameter, showed high degree of sensitivity for I-129 release. This sensitivity analysis provided insights on the data needs for a complete demonstration of compliance. A refined and comprehensive safety assessment approach along with site-specific data would be required for the next stage of disposal project implementation.

Acknowledgment

This work has been performed as a part of the Nuclear R&D Program supported by Ministry of Science and Technology(MOST).

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