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Original Article

Radiotoxicity flux and concentration as complementary safety indicators for the safety assessment of a rock-cavern type LILW repository

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A B S T R A C T

This study presents a practical application of complementary safety indicators, which can be applied in a safety assessment of a radioactive waste repository by excluding a biosphere simulation and comparing the artificial radiation originating from the repository with the background natural radiation. Complementary safety indicators (radiotoxicity flux from geosphere and radiotoxicity concentration in seawater) were applied in the safety assessment of a rock-cavern type low and intermediate level radioactive waste (LILW) repository in the Republic of Korea. The natural radionuclide (^{40}K, ^{226,228}Ra, ^{232}Th, and ^{234,235,238}U) concentrations in the groundwater and seawater at the Gyeongju LILW repository site were measured. Based on the analyzed concentrations of natural radionuclides, the levels of natural radiation were determined to be $8.6 \times 10^{-5} - 8.0 \times 10^{-4}$ Sv/m^2/yr and 6.95×10^{-5} Sv/m^3 for radiotoxicity flux from the geosphere and radiotoxicity concentration in seawater, respectively. From simulation results obtained using a Goldsim-based safety assessment model, it was determined that the radiotoxicity of radionuclides released from the repository is lower than that of the natural radionuclides inherently present in the natural waters. The applicability of the complementary safety indicators to the safety case was discussed with regard to reduction of the uncertainty associated with biosphere simulations, and communication with the public.

1. Introduction

The first priority in designing a repository for radioactive waste is to ensure that the disposal system does not pose any radiological hazard to human health or the environment. Most approaches to safety assessments of repositories have involved primary safety indicators (i.e., dose and risk). However, the evaluation of repository safety only in relation to these primary indicators has been regarded as insufficient because dose and risk are calculated based on many assumptions [1]. In particular, a biosphere system simulation, which is the final modeling component in the calculation of dose and risk, has the clear disadvantage of large uncertainties associated with its time evolution, as ecological and biological characteristics change over time and are thus rarely predictable [2]. For instance, ingesting seafood containing radionuclides, which have released from the Gyeongju repository in Korea, may be considered one of the main pathways for human exposure to radiation, because the repository is located adjacent to the ocean. The Korean sea surface temperature increased by approximately 1.5 °C from 1881 to 1990 [3], and the marine ecosystem has changed in response to this climate change. In a case study conducted over thirty years (1975–2004), annual catches of warm water species such as squid, mackerel, and anchovy increased, but those of cold water species such as pollock, saury, and sardine deceased [4]. In addition, the dietary habits and agricultural behavior of humans have changed markedly over time. For example, Korean intake of fish and shellfish has increased by a factor of five over the last fifty years [5]. Therefore, owing to its dynamic characteristics, the reliability of biosphere system modeling for the future is weak, and the relatively high uncertainty of ecosystem simulations with regard to long-term safety are cause for concern compared to those of engineered barriers and the geosphere. In light of this, safety assessments in terms of dose and risk alone should be supported by

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multiple evidences and other indicators to address uncertainty concerns [2,6].

The safety case can be reinforced with a variety of complementary evidence. To this end, the use of complementary indicators based on comparison with the background radiation in the environment has been proposed as a method for strengthening the robustness of the safety case for repositories [6]. Indicators such as the flux and concentration of radionuclides have been recommended for safety assessments of repositories because their comparison with naturally occurring radionuclides is accessible and no biosphere simulation is required [7]. Inter-European projects such as SPIN (Testing of Safety and Performance Indicators) [8] and PAMINA (Performance Assessment Methodologies in Application to Guide the Development of the Safety Case) [9] have intensively explored the use of such complementary indicators for the safety case of repositories. In these joint research projects, different types of complementary indicators were evaluated according to various criteria, e.g., their comparability with natural radiation reference values and predictability during modeling. The radiotoxicity concentration in biosphere water, radiotoxicity flux from the geosphere, and power density were selected as complementary safety indicators in the PAMINA project [9]. Safety assessments using complementary safety indicators can exclude biological pathways, reducing the problematic uncertainties related to time evolution in biosphere simulation.

A reference value for each safety indicator is a prerequisite for evaluating the modeling results at the endpoint of the safety assessment. Typically, repository safety is assessed by comparing measured and/or calculated quantities with appropriate safety standards. For example, a LILW repository in Korea may not exceed an annual dose for individuals of 0.1 mSv. While the determination of safety standards for primary indicators has no clear yardstick, complementary safety indicators have the advantage that their reference values can be derived based on observation of the natural system. The concentrations of naturally occurring radionuclides in groundwater may be used to set the reference values for complementary safety indicators. Moreover, in communication with the public, the ability to directly compare modeling results with values obtained from local sites (e.g., a well, river, or groundwater) is very useful.

The main objective of this study is a practical application of complementary safety indicators for the safety assessment of a rock-cavern type LILW disposal facility in Korea. The radiotoxicity flux from geosphere and radiotoxicity concentration in seawater were selected as complementary safety indicators in this study (Fig. 1). To apply these indicators to a safety assessment, reference values were derived from measurement of the concentration of natural radionuclides (^{40}K, ^{226,228}Ra, ^{232}Th, and ^{234,235,238}U) in the groundwater and seawater at the Gyeongju repository site. The radiotoxicity flux from the geosphere and the radiotoxicity concentration in seawater originating from the repository were calculated using a total system performance assessment model. The safety of the repository was assessed by comparing the modeling results with the established reference values.

2. Methods

2.1. Analysis of natural radionuclide concentrations in groundwater

Groundwater samples for analysis were obtained from boreholes at the Gyeongju repository site in cooperation with the Korea Radioactive Waste Agency (KORAD). To obtain the background radiation of the repository site, a multi-packer system of boreholes allows the collection of approximately 15 L of groundwater at depths of −120 to −190 m at each sampling point, which is a similar to that of the silo (−80 to −130 m). Three sampling points were located in a coastal area in the groundwater pathway (A), a coastal area outside the groundwater pathway (B), and a groundwater recharge area (C). The aquifer and groundwater recharge area sites are based on previously reported data in numerical simulations of groundwater [10,11]. The A − C distance was sufficiently far to avoid interference between the sampling positions.

The concentrations of ^{40}K, ^{226,228}Ra, and ^{232}Th in the groundwater samples were analyzed with a high-purity germanium (HPGe) gamma-ray detector. The ^{232}Th content was obtained from the ^{228}Ac signal based on the assumption of secular equilibrium. The uranium isotopes, ^{234,235,238}U, in the groundwater were assayed using liquid scintillation counting. Seawater at the Gyeongju repository site was collected near the shoreline. Only the concentration of ^{40}K was determined in seawater samples owing to the difficulty in measuring trace amounts of other nuclides. Samples for analysis of ^{40}K were stored in 1 L Marinelli beakers, and the gamma peak (1460.7 keV) of ^{4} K was measured for 12 h using a detector calibrated with IAEA-RGK-1. The detector calibration and sample measurement for radium [12], actinium [13], and uranium isotopes [14] followed standard analytical procedures. The confidence level of all measurement is approximately 95%.

2.2. Modeling

The radiotoxicity flux from the geosphere and the radiotoxicity concentration in seawater originating from the rock-cavern type

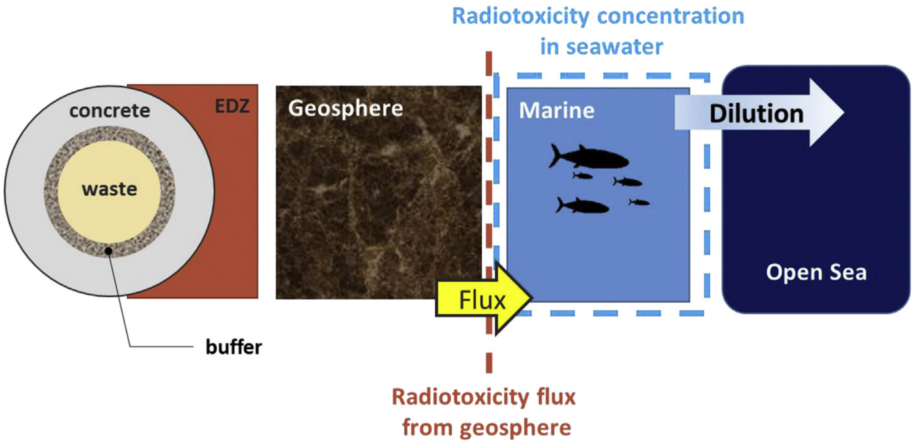

Fig. 1. Concept of the radiotoxicity flux from geosphere and radiotoxicity concentration in seawater.

LILW repository were calculated on the basis of the safety assessment model developed in a previous study [15]. The model consists of three sub-modules (engineered barrier system (EBS), geosphere, and biosphere), and was implemented using the commercially available Goldsim [16] program based on the design of six silo-type repositories for LILW in Gyeongju (Fig. 2). In the EBS module, steel waste drums are disposed of in concrete packages, and these concrete packages are stacked in silos. Crushed granitic rock is used to fill the gaps between the waste packages and the silo walls. It is assumed that the silos become completely saturated with groundwater immediately after being closed. In addition, it is assumed that diffusion is the only migration process for the radionuclides in the silos. It is also postulated that an excavation damage zone (EDZ) surrounds the concrete wall. In the EDZ, it is assumed that groundwater flows vertically upwards, and then discharges into the geosphere through the top part of the silo, which is connected to an operational tunnel. The geosphere submodule represents an aquifer in fractured rock, which is assumed to be a porous medium. Radionuclides from the geosphere are transported to the sea and then distributed homogeneously within coastal seawater. An ocean current towards the open ocean dilutes the radionuclide concentration in seawater. Details on the methods and input data for the safety assessment model have been provided elsewhere [15].

3. Results and discussion

3.1. Reference values for the radiotoxicity flux from geosphere and radiotoxicity concentration in seawater

Table 1
Radionuclide concentrations and detection limits in groundwater samples (A – C).

Natural radionuclide	Concentration (Bq/m^3)			
	A	B	C	MDA
^{40}K	n.d.	7470 ± 910	n.d.	<3000
^{226}Ra	329 ± 7	15 ± 4	48 ± 4	<11
^{228}Ra	244 ± 19	n.d.	152 ± 7	<22
^{232}Th	n.d.	4070 ± 460	n.d.	<270
^{234}U	n.d.	224 ± 10	n.d.	<2
^{235}U	n.d.	5 ± 1	n.d.	<2
^{238}U	n.d.	156 ± 8	n.d.	<2

n.d.: below the minimum detectable activity (MDA) value.

Reference values for the radiotoxicity flux and concentration indicators were determined based on the concentrations of radionuclides present in natural groundwater. The measured radionuclide concentrations in the groundwater samples (A – C) are summarized in Table 1. The radionuclide concentrations varied considerably across the sampling sites. This heterogeneous distribution of radionuclides is typically observed in natural samples. For example, the concentration of ^{40}K in Chinese rivers varies between 8 and 7149 Bq/m^3 [2], while the concentrations of ^{235}U and ^{238}U in a crystalline rock type in a Spanish aquifer were found to be in the range of 0.06–188 Bq/m^3 and 1.19–3890 Bq/m^3, respectively [9].

The radionuclide concentration can be converted to radiotoxicity (in Sv) by multiplying by the dose conversion factor for each radionuclide. The effective dose coefficients for ingestion of radionuclides for adults presented in publication 119 of the International Commission on Radiological Protection (ICRP) [17] were used in this study. Table 2 summarizes the natural radiotoxicity in groundwater determined in this study. The total radiotoxicity of the Gyeongju groundwater varied between 1.18×10^{-4} Sv/m^3 (Site C) and 1.00×10^{-3} Sv/m^3 (Site B). This degree of radiotoxicity at the Gyeongju repository site is comparable to reported values from other sites [9,18,19] (Table 3).

The reference value for the radiotoxicity flux from geosphere has been determined from the total radiotoxicity in the groundwater, as given by Equation (1) [9]:

$$J = \sum_n s_n \times D_n = \sum_n C_n \times Q \times D_n \tag{1}$$

where J is the reference value for the radiotoxicity flux (Sv/yr), s_n is the activity flux of nuclide n (Bq/yr), D_n is the ingestion dose

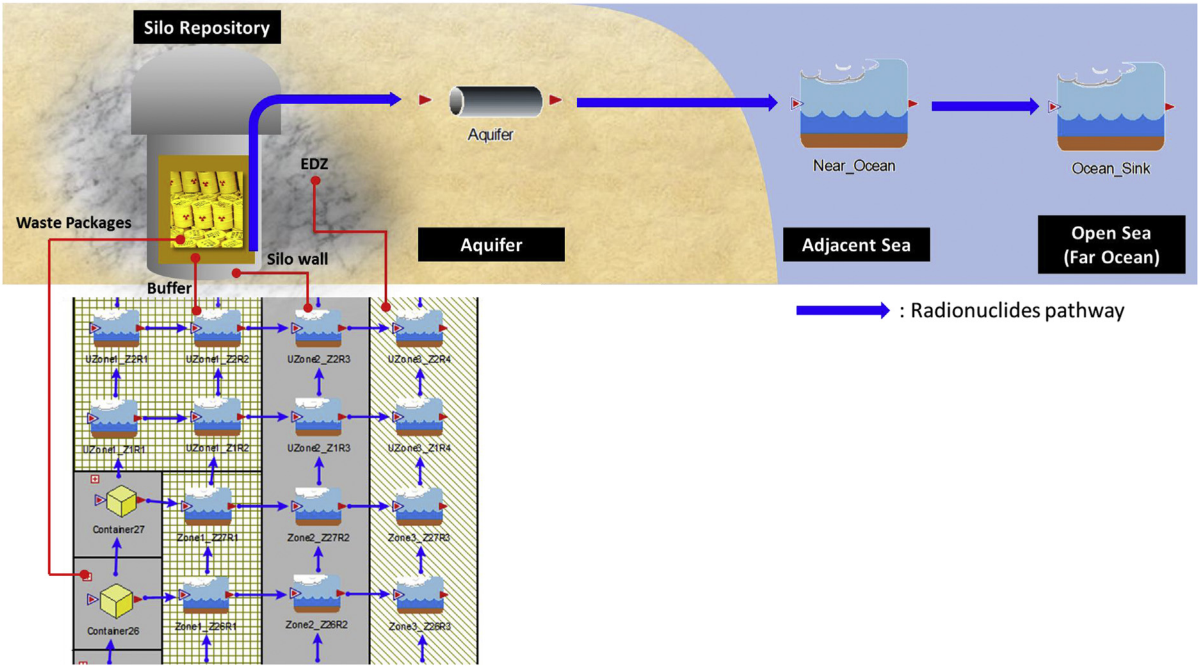

Fig. 2. Goldsim modeling scheme.

Table 2
Radiotoxicity concentrations of radionuclides in groundwater samples (A − C).

Natural radionuclide	Radiotoxicity concentration (Sv/m^3)		
	A	B	C
^{40}K	n.d.	4.63×10^{-5}	n.d.
^{226}Ra	9.22×10^{-5}	4.14×10^{-6}	1.35×10^{-5}
^{228}Ra	1.68×10^{-4}	n.d.	1.05×10^{-4}
^{232}Th	n.d.	9.36×10^{-4}	n.d.
^{234}U	n.d.	1.10×10^{-5}	n.d.
^{235}U	n.d.	2.36×10^{-7}	n.d.
^{238}U	n.d.	7.01×10^{-6}	n.d.
Total	2.60×10^{-4}	1.00×10^{-3}	1.18×10^{-4}

n.d.: below the minimum detectable activity (MDA) value.

conversion factor of nuclide n (Sv/Bq). In general, a flux is defined as a magnitude that flows through a unit area per unit time. However, in SPIN and PAMINA projects [8,9], the radioactivity flux (Bq/yr) represented a magnitude that flows via an integral area of a specific surface (i.e. geosphere). According to the conventional definition of flux used in previous studies of complementary indicator [8,9], the radioactivity flux from geosphere (s_n) is determined by the multiplication of radioactivity concentration in groundwater (C_n) and volumetric groundwater flow rate (Q) (the units of C_n and Q in equation (1) are Bq/m^3 and m^3/yr, respectively). However, since the concept of using integral area in equation (1) is different from that of using a unit area, which has been generally used in various fields, the use of equation (1) can cause a misleadingness. For example, the unit of radiotoxicity flux (Sv/yr) defined by previous works [8,9] is not a general unit of flux (magnitude/area/time) whereas the unit of radiotoxicity is identical to that of effective dose rate, instead. Therefore, in order to avoid a confusion and improve a clarity, the flux defined by a unit area was adopted in this work. Accordingly, the radiotoxicity flux from geosphere (Sv/m^2/yr) is given in equation (2):

$$F = \sum_n C_n \times D_n \times v_{GW} \tag{2}$$

where F is the reference value for the radiotoxicity flux from geosphere (Sv/m^2/yr), C_n is the radioactivity concentration of nuclide n (Bq/m^3), D_n is the effective dose coefficient for ingestion of nuclide n (Sv/Bq) recommended by ICRP [17], and v_{GW} is groundwater flux (m/yr). From the simulation of groundwater flow at the Gyeongju repository site [11], the groundwater flux was calculated to be 0.73–0.80 m/yr. This value was applied in Equation (2) and the reference value for radiotoxicity flux from the geosphere was determined to be $8.6 \times 10^{-5} - 8.0 \times 10^{-4}$ Sv/m^2/yr. Note that the proposed reference value of radiotoxicity flux from geosphere in this work is not a single value but a range of values derived from natural radiation. Complementary safety indicator can provide evidence of the disposal safety by comparing artificial radiation from a repository and natural background radiation in geosphere. Complementary safety indicator is distinct from the primary indicator that is used to assess the disposal safety in comparison with regulatory criteria. For instance, the annual effective dose is a measure which can be utilized to quantitatively assess the radiological hazard to human. On the other hand, the safety assessment using complementary safety indicators (flux and concentration) does not make a deterministic conclusion whether the repository is safe or unsafe, but provides information about how large the radiological interference would be induced by a repository based on the comparion of artificial radiation with natural background radiation. In this regard, the use of the representative natural radiation amount with its variation as the reference value is more proper than the use of a single value such as an average or a minimum value. Therefore, the range of natural radiation ($8.6 \times 10^{-5} - 8.0 \times 10^{-4}$ Sv/m^2/yr) as reference level is proposed for the safety assessment by radiotoxicity from geosphere in this work.

To determine the reference value for the radiotoxicity concentration in seawater, the concentration of ^{40}K in a seawater sample from the Gyeongju site was used. ^{40}K is predominant in the radiotoxicity of seawater owing to the abundant concentration of potassium in the marine system. The ^{40}K concentration was determined to be 11,200 ± 940 Bq/m^3, which corresponds to 6.95×10^{-5} Sv/m^3 after multiplying by the dose conversion factor. Therefore, a radiotoxicity concentration of 6.95×10^{-5} Sv/m^3 is used as a yardstick for the safety assessment based on the radiotoxicity concentration in seawater. The values for the complementary safety indicators used in this and other studies are summarized in Table 4.

3.2. Modeling results and safety assessment using complementary safety indicators

A safety assessment using the complementary safety indicators was carried out by comparing the reference values obtained from the natural radionuclides present in the groundwater and seawater with the modeling results. Figs. 3 and 4 show the calculated radiotoxicity flux from the geosphere (Sv/m^2/yr) and the radiotoxicity concentration in seawater (Sv/m^3), respectively, with the proposed reference values. In order to determine the radiotoxicity flux from the geosphere released from the repository, the total radiotoxicity flux through the geosphere/biosphere interface was calculated by Goldsim modeling and the calculated flux was divided by the size of repository. To estimate the size of the repository, previous European studies [9,18,20] used a footprint of the repository area, varying from about 2 to 10 km^2 (Table 4). However, these postulated areas appear arbitrary because their estimations are based on hypothetical repositories. Only the Dutch study provided detailed information for estimating the repository size [18]. Based on the concept of flux indicator from geosphere, the size of the repository represents the boundary area between geosphere and biosphere. According to the simulation of groundwater flow at the Gyeongju site [11], the groundwater from the recharge area would pass

Table 3
Comparison of radiotoxicity concentrations in groundwater reported in the literature [9,18,19,20].

	Considered Isotopes	Radiotoxicity concentration (Sv/m^3)
This study (Korea)	^{40}K, ^{226}Ra, ^{228}Ra, ^{232}Th*, ^{234}U, ^{235}U, ^{238}U	$1.18 \times 10^{-4} - 1.00 \times 10^{-3}$
Amphos21 (Spain)	^{40}K, ^{87}Rb, ^{223}Ra*, ^{224}Ra*, ^{226}Ra, ^{228}Ra*, ^{222}Rn, ^{228}Th*, ^{230}Th*, ^{232}Th, ^{234}Th*, ^{234}U*, ^{235}U, ^{238}U	$4.19 \times 10^{-7} - 1.25 \times 10^{-3}$
NRI (Czech Republic)	^{40}K, ^{226}Ra, Gross alpha, Gross beta, Uranium	$4.80 \times 10^{-5} - 1.74 \times 10^{-2}$
GRS (Germany)	^{232}Th, ^{235}U, ^{238}U, all daughter nuclides* of ^{232}Th, ^{235}U, and ^{238}U	6.88×10^{-6}
NRG (Nederland)	^{235}U, ^{238}U, all daughter nuclides* of ^{235}U and ^{238}U	8.00×10^{-6}
JAEA[a] (Japan)	^{210}Po, ^{210}Pb, ^{228}Th, ^{226}Ra, ^{228}Ra, ^{234}U, ^{238}U	$2.34 \times 10^{-5} - 3.29 \times 10^{-4}$

* Concentration was derived assuming equilibrium between mother and daughter nuclides.

[a] Radiotoxicity was calculated in this work using the effective dose coefficients and concentrations of natural radionuclides given in [19].

Table 4
Comparison of the groundwater flux, repository size, radiotoxicity flux from geosphere, and radiotoxicity concentration in seawater.

	Groundwater flux (m/yr)	Repository size (m^2)	Radiotoxicity flux from geosphere ($Sv/m^2/yr$)	Radiotoxicity flux from geosphere × Size of repository (Sv/yr)	Radiotoxicity concentration in biosphere water (Sv/m^3)	Ref.
This study (Korea)	0.73–0.80	3×10^4	$8.6 \times 10^{-5} - 8.0 \times 10^{-4}$	2.4–24.0	6.95×10^{-5}	p.w.
Amphos21 (Spain)	10^5 m^3/yr[a]		-[a]	0.001–100	1.00×10^{-5}	[9]
NRI (Czech Republic)	0.02–0.32	1×10^7	$8.0 \times 10^{-7} - 5.5 \times 10^{-3}$	$8.0–5.5 \times 10^4$	2×10^{-5}	[9]
GRS (Germany)	4.8×10^4 m^3/yr[a]		-[a]	0.27–14.78	2×10^{-6}	[20]
NRG (Nederland)	$5.28 \times 10^{-4} - 1.47 \times 10^{-2}$[b]	1.86×10^6	$5.4 \times 10^{-9} - 1.2 \times 10^{-7}$	0.01–0.23	8×10^{-6}	[18]

In this table, radiotoxicity flux from geosphere ($Sv/m^2/yr$) represents a radiotoxicity flux flowing through a unit area of geosphere. Radiotoxicity flux from geosphere × size of repository (Sv/yr) in this work corresponds to the reference value of radiotoxicity flux from geosphere in previous studies [8,9].

[a] Only the value of groundwater flux × area of repository was given.

[b] Only the vertical portion of groundwater flux was considered.

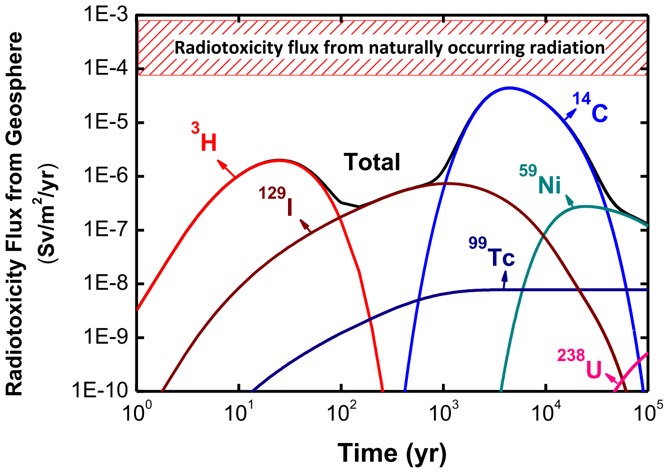

Fig. 3. Radiotoxicity flux from the geosphere calculated from the safety assessment model with the proposed reference value.

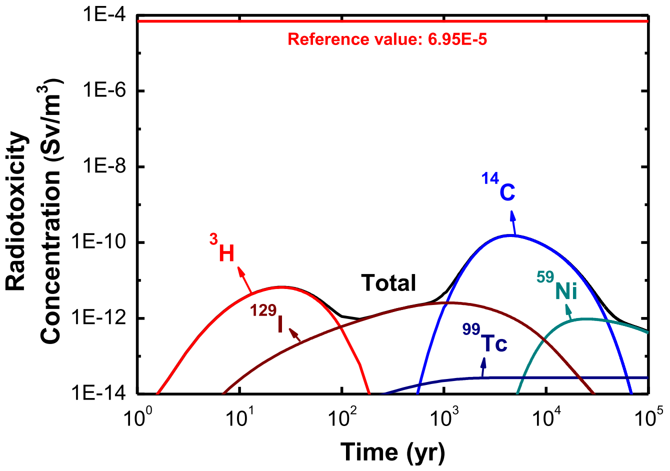

Fig. 4. Radiotoxicity concentration in seawater calculated from the safety assessment model with the proposed reference value.

through the silos and then travel to the adjacent coast. Thus, the size of the Gyeongju repository used in this study was estimated as the longitudinal section of shoreline (300 m × 100 m) through which the groundwater flows, as in the previous study [21]. This approach based on groundwater modeling seems more reasonable than that adopted by the previous European studies as the area was estimated from simulation results for the actual repository site. Moreover, the average groundwater flow for the six silos at the Gyeongju repository after closure was estimated to be 15,500–21,300 m^3/yr (converted from the reported value of 7.08–9.72 m^3/day per silo [11]), which is similar to the approximation in this study (21,900–24,000 m^3).

In Fig. 3, the calculated radiotoxicity flux from the geosphere reaches a maximum of 4.4×10^{-5} $Sv/m^2/yr$ at approximately 4450 years after closure of the repository. ^3H and ^{14}C were identified as the main contributors to the radiotoxicity flux. Note that all ^3H and ^{14}C were assumed to be soluble species although they may exist as a gas phase in LILW repository system. It is very difficult to evaluate how much these radionuclides are formed as gas phases due to the lack of information on waste forms and the complicated physicochemical reactions, such as degradation of waste and redox reactions of radionuclides. In addition, it is expected that gas phases of ^3H and ^{14}C are rarely dissolved in groundwater and cause no significant effect on the radiotoxicity in groundwater.

The radiotoxicity flux from the geosphere contributed by the repository was determined to be less than the natural radiation level ($8.6 \times 10^{-5} - 8.0 \times 10^{-4}$ $Sv/m^2/yr$). This indicates that the radiological effect due to the repository is much lower than that of naturally occurring radionuclides. Similarly, the radiotoxicity concentration in seawater released from the LILW repository reaches a peak of 1.5×10^{-10} Sv/m^3 at approximately 4450 years after closure (Fig. 4). Based on these simulation results, the amount of radionuclides released from the rock-cavern type LILW repository is negligible compared to the intrinsic ^{40}K in seawater of Gyeongju repository site with respect to their radiological effects.

4. Conclusions and implications

This study is focused on the practical application of complementary safety indicators for the safety assessment of a LILW disposal system. The radiotoxicity flux from geosphere and the radiotoxicity concentration in seawater were used as complementary safety indicators, and the reference levels were determined to be $8.6 \times 10^{-5} - 8.0 \times 10^{-4}$ $Sv/m^2/yr$ and 6.95×10^{-5} Sv/m^3, based on the concentrations of naturally occurring radionuclides (^{40}K, $^{226,228}Ra$, ^{232}Th, and $^{234,235,238}U$) in the groundwater and seawater near the Gyeongju repository site, respectively. According to the modeling results, it is expected that the radiological effects from the rock-cavern type repository will not exceed the naturally occurring radiation in the local groundwater and seawater. To the best of our knowledge, site-specific yardsticks for the radiotoxicity flux and the radiotoxicity concentration in Korean rock-cavern type LILW repositories are proposed for the first time in this study.

The factors and assumptions used to determine the reference value for the radiotoxicity flux indicator in each study varied, although the assessment principle appears to be identical. As can be seen in Tables 3 and 4, there is a large discrepancy of parameters in each calculation in previous studies. Therefore, it is important to develop a logical method that is appropriate for the local circumstances and to measure the site-specific radiotoxicity concentration at an actual disposal site to ensure precise application of the complementary safety indicator to the safety assessment.

The main concept in the use of complementary safety indicators is that naturally existing radiotoxicity can provide evidence for the safety case and multiple lines of reasoning [22]. Regarding the long-term safety of the repository, safety assessments can be strengthened through introduction of complementary safety indicators that exclude biosphere simulations, which have high uncertainty in the prediction of long-term safety. In addition, use of complementary safety indicators may facilitate communication with the public, such as the local residents near a repository. For instance, the radiotoxicity concentration indicator can answer the question “Will my great-grandchildren be able to swim or fish at the beach?” [22]. Complementary safety indicators can provide intuitive information about the safety of repository to the public by allowing for direct comparison with background radiation from regional natural waters.

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