


## Article

# Impact of Concrete Degradation on the Long-Term Safety of a Near-Surface Radioactive Waste Disposal Facility in Korea

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**Abstract:** The migration of radionuclides from radioactive waste into the environment poses a public safety concern. Thus, the long-term safety assessment for near-surface disposal sites for radioactive waste in South Korea entails providing reasonable assurance that the annual radiation dose exposure from radionuclide release from the waste repository into the biosphere will not exceed the regulatory limit of 0.1 mSv/yr. At the first near-surface disposal site in Gyeongju, concrete was a crucial component of the engineered barriers designed to contain radionuclides within the disposal site. The ability of concrete to retain radioactive waste within the disposal site is attributed to its high sorption capacity for radionuclides. However, research has shown that the degradation of concrete can affect its radionuclide retention capabilities, which are defined by sorption properties of distribution ( $K_d$ ) and diffusion ( $D_s$ ) coefficient parameters. As a result, changes in sorption properties may lead to radionuclides migrating out of the disposal vault. In light of the geochemical deterioration of engineered concrete barriers, this study assesses the long-term safety of near-surface disposal sites. To simulate the impact of concrete degradation on radionuclide migration, we employed RESRAD-OFFSITE's extended source-term features, which can model the release of radionuclides from radioactive waste shielded by concrete barriers. Using carefully screened published sorption data of four radionuclides ( $^{14}\text{C}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{99}\text{Tc}$ ) in different stages of concrete degradation, the results indicated that released radioactivity during the most degraded state of concrete will result in a maximum radiation exposure dose of  $1.4 \times 10^{-8}$  mSv/yr from  $^{99}\text{Tc}$  which is below the permissible limit of 0.1 mSv per year, thus demonstrating that concrete is a reliable component of the engineered designed barriers for near-surface disposal facilities.

**Keywords:** engineered barrier system; near-surface disposal; radionuclide sorption properties; geochemical concrete degradation; distribution coefficient; diffusion coefficient; RESRAD-OFFSITE



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## 1. Introduction

When it comes to the disposal of radioactive waste, various concerns must be addressed. One of the most critical issue is ensuring the long-term safety of the disposal site once it has been released from regulatory oversight (after 300 years) [1]. Numerous radionuclides have long half-lives, indicating that they will continue to be hazardous for longer than 300 years. Therefore, it is essential to adopt a disposal method that will minimize the danger of radiation exposure to current and future generations. Near-surface disposal is a sought-after disposal method that involves holding waste in a facility designed to isolate it from the environment by utilizing both natural and engineered barriers. This disposal approach is suitable for low- and intermediate-level radioactive waste, the majority of which consists of radionuclides with short half-lives and a few with long half-lives that decay over thousands of years beyond the operational lifetime of the disposal site [2].

As the second phase of the Gyeongju LILW radioactive waste repository, the first near-surface waste disposal facility in South Korea was developed [3]. The first stage of the LILW facility is a 100-m-deep underground silo with a capacity of 35,200 cubic meters (130,000

drums) [4]. The near-surface repository was constructed with a capacity of 32,875 cubic meters (125,000 drums) fewer than 30 m below the surface [4]. This disposal includes a subterranean gallery, three mobile cranes, and twenty concrete vaults. These man-made barriers were incorporated to supplement natural barriers in order to meet the regulatory design goals of isolating radionuclides from the environment.

Important criteria to consider when choosing barrier structures and materials are their long-term durability, compatibility with site characteristics, associated media and availability [5]. Concrete structures at the Gyeongju LILW site are made of Ordinary Portland cement, as is the case with many near-surface disposal sites [6]. Ordinary Portland cement and composite cements (with supplementary cementitious materials in the form of iron blast furnace slag, silica fume, pozzolans, and fly ash) are the commonly used types of cement due to the well-understood technology transferred from civil engineering research and many years of experience with its performance in various environments [7,8]. In addition to Portland cement, other materials are being evaluated to determine their suitability for use in the construction of engineered barriers to radioactive waste repositories [9]. For instance, geo-polymers produced by mixing a source of silicates and aluminates (such as fly ash or metakaolin) with an alkali activator have been proposed for use, since they were allegedly utilized in the construction of ancient pyramids [9]. Despite this claim, there is still uncertainty surrounding their long-term performance; and their use of caustic Na/K(OH) as an activator presents handling challenges [9].

When evaluating the long-term performance of a near-surface waste disposal facility, it is often assumed that concrete barriers will maintain their initial chemical buffering function for thousands of years [10]. Consequently, many studies on the long-term performance of concrete engineered barriers focus on the evolution of hydraulic behavior over long timescales, with the assumption that the concrete barriers will retain their initial designed hydraulic properties for a certain period of time before disintegrating into porous sand with poor hydraulic properties (rapid water infiltration) [11]. This does not account for the progressive degradation of concrete barriers resulting from geochemical changes. Concrete is a mixture of water, hydrated cement, and fine aggregates, such as sand and gravels [12]. Portland-based concrete is made of several anhydrous minerals, mainly calcium silicates (alite, belite, aluminates, and ferrites), and traces of gypsum [13]. During the hydration process, the anhydrous OPC forms aqueous and solid cement minerals such as portlandite ( $\text{Ca}(\text{OH})_2$ ), calcium-silicate-hydrate (C-S-H) and other minor phases comprising aluminum, sulfate, and iron, whose surface areas provide the large sorption capacity for dissolved radionuclides [12]. Additionally, the alkaline nature of cement phases provides a favorable environment in which radionuclides are less soluble [7].

The gradual degradation of concrete is based on the assumption that, over time, the disposal facility will deteriorate, allowing rainwater to infiltrate the concrete structures within the disposal facility, as shown in both the conceptual diagram in Figure 1 and in the assumed degradation timeframe in Figure 2. The rainwater with a lower pH will not be in equilibrium with the high alkaline hydrated cement mineral system that makes up the concrete structures [14]; consequently, chemical reactions will occur that gradually alter the aqueous and solid phases of concrete, thereby causing further disintegration of concrete [15]. The reduced pH of infiltrating water will increase radionuclide solubility, leading to the transport of radionuclide ions via an electrochemical potential gradient and an advection caused by a flow of aqueous solution [15]. As depicted in Figure 2, this study assumes that infiltration of water will occur after 300 years of regulatory oversight and that the disposal site will have experienced normal wear, with the possibility of a radionuclide-containing waste drum failing after 100 years of operation, followed by failure of a concrete vault after 300 years, a cover failure after 500 years, and further concrete degradation after 1000 years. The natural deterioration of the disposal site is projected to be caused by, among other things, general influences on construction, lengthy operation periods (>100 years), climate change (e.g., pro-longed flooding), and ground water geochemistry [7].

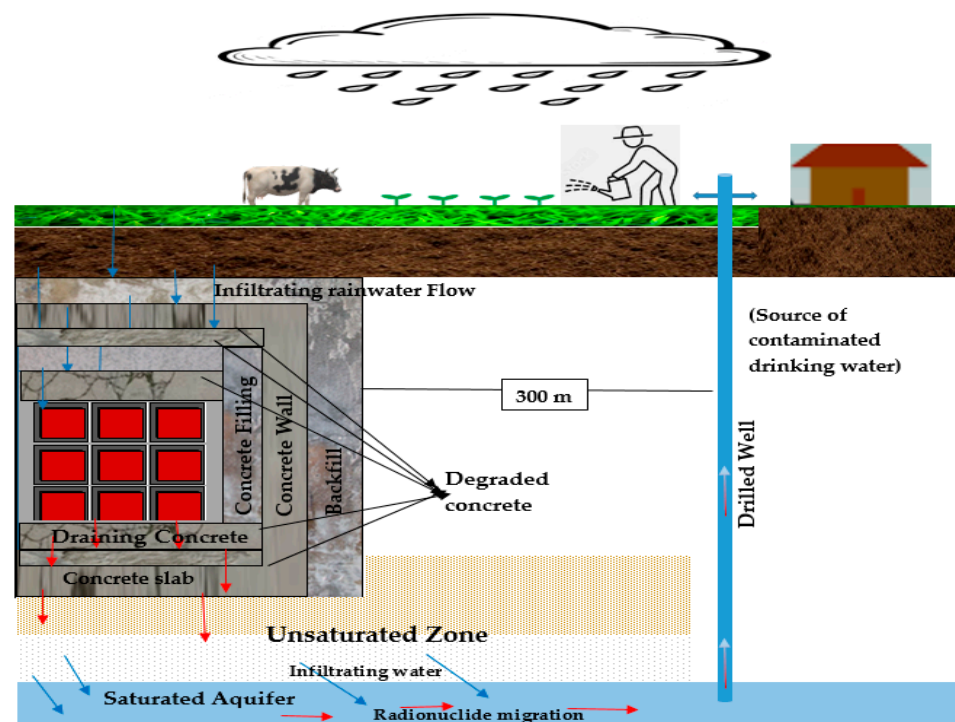


Figure 1. Conceptual Model of a near-surface disposal site.

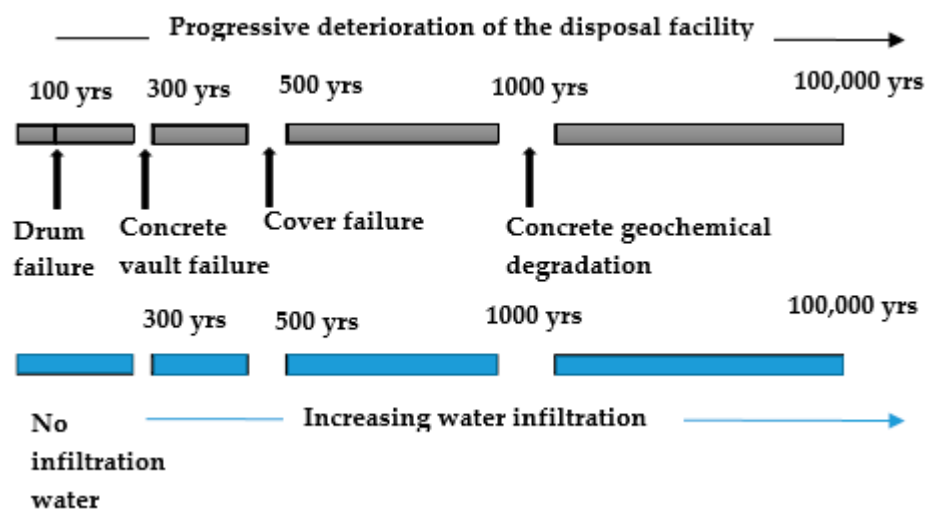


Figure 2. Time frame to reflect gradual deterioration of disposal facility.

Literature indicates that concrete deterioration goes through four phases, defined by gradual decrease in pore water pH and calcium to silica (C/S) ratio [11]. The initial state (I) is distinguished by a pH greater than 12.5. This is due to the high amounts of alkalis, including sodium and potassium. In the second state, the pH is around 12.5 and controlled by the solubility of portlandite ( $\text{Ca}(\text{OH})_2$ ) which has the ability to firmly bind metal ions, anions, and halogens by substituting its hydroxyl functional group [11,16]. The pH of the third state (III) is governed by the equilibrium between Calcium-Silica-Hydrate (C-S-H) and infiltrating water from the surrounding non-cementitious materials. The packing of the C-S-H tiny polymers into platelets provides the sorption binding surface area in this condition [16]. Lastly, in the fourth state (IV), the pH decreases to approximately 10 under the influence of calcite ( $\text{CaCO}_3$ ) which has a smaller reactive surface area, hence exhibiting lower sorption abilities than the other phases [16]. A few studies evaluating the effect of

geochemical evolution on concrete barriers have demonstrated that the sorption behaviors of radionuclides vary with the stages of concrete degradation [12].

Sorption refers to the binding of radionuclides on the surface or within the structure of a substance [7]. The radionuclide sorption process on cementitious surfaces is evaluated using the distribution coefficient ( $K_d$ ) and diffusion coefficient ( $D_s$ ) parameters, which are employed to assess radionuclide migration through the cementitious medium [17]. The distribution coefficient is the ratio of the radionuclide's mass concentration in the solid phase to its concentration in the liquid phase at equilibrium, whereas diffusion is the movement of vapor and ions through concrete [18]. The reported sorption values for a number of radionuclides can vary by more than two orders of magnitude in different environments. This study employed carefully selected sorption values from a database [10] developed by a global panel of experts to eliminate the ambiguity caused by the existing pool of varying sorption data for radionuclides. This sorption database was adopted as an integral component of the supporting documentation for the safety analysis of a near-surface disposal of LILW in Dessel, Belgium [10]. In doing so, our study seeks to contribute to the evaluation of the long-term safety of the Gyeongju LILW disposal site.

For the long-term containment of radionuclides, the effect of sorption properties on the migration of radionuclides in disposal facilities can be significantly more substantial than the hydraulic properties of the designed barriers [11]. Therefore, it is important to assess the long-term safety of a near-surface waste disposal site, based on the hypothesis that concrete degradation will lead to migration of radionuclides into the biosphere, as depicted in Figure 1. To examine this risk, this study used RESRAD-OFFSITE computer software to assess radiation exposure to the surrounding environment as a result of concrete degradation. This software was developed by the Nuclear Regulatory Commission (NRC) of the United States to assess radiation exposures of human receptors situated on top of or at a distance from radioactive contaminated site [19]. Further improvement of the software's capabilities has allowed it to model the transport of radionuclides beyond the soil medium, allowing it to simulate the release of radionuclides from radioactive waste material shielded by concrete-engineered barriers. Using the extended features of the RESRAD-OFFSITE code, the radiological impact of radioactivity released as a result of concrete degradation will be estimated as the exposure dose via the drinking water pathway, assuming that a contaminated well is 300 m from the near-surface disposal facility.

## 2. Materials and Methods

### 2.1. Geochemical Degradation of Concrete

The chemical approach to cementitious deterioration developed by Prof. F. Glasser established that the degradation of hydrated cementitious materials follows a pattern characterized by decreasing pore water pH and C/S ratios culminating in four distinct phases [8,15]. These four states of concrete degradation were used by Belgian researchers to evaluate the performance of the near-surface low-level radioactive repository at Dessel; thus, this study assumes the same four cementitious degraded phases described below for the concrete waste vault barrier in Gyeongju near-surface disposal. The published radionuclide sorption data for each phase (Table 1) will be used as the distribution coefficient in the contaminated medium for the RESRAD-OFFSITE simulation.

State I: ( $13.5 > \text{pH} > 12.5$ ) in which hydrated cement pore fluids include substantial quantities of free alkali metal ions due to the first dissolution of alkali metal sulfate and oxides. This state's pore fluid composition is dominated by Na, K, and OH. Due to the limited solubility of portlandite ( $\text{Ca}(\text{OH})_2$ ) at these pH levels, only trace amounts of Ca are liberated from the solids, resulting in a high C/S ratio. This state lasts for only about three years [12,20].

State II: ( $\text{pH} = 12.5$ ), the solubility of portlandite at 12.5 (25 °C) will influence the pH of the pore fluid after all alkali ions have been removed at the end of State I [10]. The composition of the pore fluid in this stage is essentially a 20 mM calcium hydroxide solution [10]. The duration of this condition is proportional to the amount of portlandite in

the hydrated cement. Hardened cement paste can contain up to 25 wt% portlandite. This state is said to last around 3500 years [12].

State III: ( $12.5 > \text{pH} > 10$ ) after the total disintegration of portlandite at the conclusion of State II. The incongruent dissolution of the C-S-H phases regulates pH in this state. At States I and II, CSH phases with a high C/S ratio ( $\text{C/S} > 1.5$ ) coexist with portlandite. This phase lasts approximately 36,400 years [12].

State IV: ( $\text{pH} < 10$ ) the C-S-H phases and other hydrated cement components are entirely dissolved and the pH drops below pH 10 [10]. The composition of the pore fluid is controlled by the remaining aggregate minerals and incoming water.

**Table 1.** Sorption data of radionuclides in states II and IV of concrete degradation.

Radionuclide	Half-Life (Years)	Distribution Coefficient ( $\text{cm}^3/\text{g}$ )		Diffusion Coefficient ( $\text{m}^2/\text{yr}$ )
		State II	State IV	
$^{14}\text{C}$	5700	5000	100	$1 \times 10^{-12}$
$^{90}\text{Sr}$	28.8	30	1	$5 \times 10^{-11}$
$^{99}\text{Tc}$	211,000	3000	1	$1 \times 10^{-8}$
$^{137}\text{Cs}$	30	20	0	$5 \times 10^{-10}$

The distribution coefficient ( $K_d$ ) data for all radionuclide was obtained from [12]. As for  $D_s$ ,  $^{14}\text{C}$  and  $^{137}\text{Cs}$  data was obtained from [11] while  $^{90}\text{Sr}$  and  $^{99}\text{Tc}$  data is from [20].

## 2.2. RESRAD-OFFSITE Modeling Overview

To simulate the amount of radiation released by degrading concrete, we assumed that the repository would remain intact for one hundred years following the scheduled disposal of all radioactive material. It is assumed that, after 100 years, 200 L of waste drums will collapse, allowing radionuclides to escape from the drum into the concrete waste vault. After 300 years of regulatory oversight, water will begin to infiltrate the concrete barriers; this infiltration will worsen after 500 years following the failure of the disposal site's top cover. Infiltrating water transports radionuclides that have desorbed from concrete structures from one barrier to the next until they reach the biosphere through groundwater. The concentration of each radionuclide in groundwater is predicted to be affected by both sorption abilities and decay rate.

To investigate the impact of concrete deterioration on the long-term safety of concrete engineered barriers, the RESRAD-OFFSITE code was used to develop a radionuclide transport model with radionuclide sorption properties in degraded concrete. The most recent version of the code, Version 4.0, has additional capabilities that allow for a more comprehensive simulation of radionuclide emissions from radioactive waste confined within engineered barriers. In this case, the contaminated zone is considered to have two domains: the nuclide bearing waste domain, which consists of the radioactive waste and fill materials contained within the designed barriers, and the surrounding domain, which is represented by different levels of biospheres surrounding the disposal facility, as shown [19].

In addition, the improved source-term model allows for the entry of data describing the change in the cumulative releasable fraction over time. Consequently, the source-term model permits the choice of up to nine time periods to monitor the steady degradation of an engineered barrier. The model provides two methods for specifying the change in the releasable fraction across time periods: stepwise at time or linearly over time. This allows for flexibility in specifying the degradation rate. In this analysis, it was assumed that concrete deterioration would continue linearly after 300 years of regulatory control. The diffusive transport model is selected because it is significantly slower than the advective transport; as a result, it restricts the rate at which radionuclides are released into the surrounding soil and, subsequently, into a hypothesized 300-m receptor well [19].

## 2.3. Radionuclide Release Modeling

The diffusion-controlled release conceptual model implies that a radionuclide is discharged at the surface and that the entire 6.2 m thickness of the concrete waste vault is



evenly contaminated. In the RESRAD code, the contaminated medium represents the inner composition of the concrete waste vault, and the contaminated zone represents the area of the 84 by 10 m concrete vault. A total mass of 44,100 kg of concrete and waste is assumed, together with a volume of 17,640 cubic meters. Concrete is conservatively estimated to have a porosity of 25 percent and a density of 2380 kg/m<sup>3</sup>. The unsaturated and saturated zones of the near-surface disposal site are positioned beneath the contaminated concrete waste vault, where radionuclides discharged from the contaminated medium come into contact with groundwater. Additional site-specific RESRAD-OFFSITE input parameters are listed in Table 2.

This study considers four radionuclides (with half-life greater than 20 years) from the Gyeongju LILW site source term inventory (<sup>14</sup>C, <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>99</sup>Tc), which represent a fair percentage of the radionuclide inventory of the Gyeongju LILW site. The diffusion coefficient of concrete is determined by the water-to-cement ratio used in its creation, as well as its aggregates. This analysis employs the sorption values of typical Portland cement concrete with a water-to-cement ratio of 0.45 [7]. To model the effect of concrete degradation, the sorption values for states II and IV are selected. State I is not considered because its duration is only three years. States II and III are considered to be the same state of degradation due to the fact that their sorption values are not significantly different. State IV is the most limiting because most radionuclides have low K<sub>d</sub> values. The sorption values for the selected radionuclides are listed in Table 1. A sensitivity analysis was conducted on the distribution coefficient, the diffusion coefficient, and the dry bulk density to further understand the parameters that influence the release of radiation from degraded concrete.

**Table 2.** Site-specific RESRAD-OFFSITE input parameters.

RESRAD Parameters	Value
Cover thickness	5.8 m
Area of contaminated zone	16,400 m <sup>2</sup>
Thickness of contaminated zone	6.2 m
Density of contaminated zone	1890 kg/m <sup>3</sup>
Total volume of waste	33,600 m <sup>3</sup>
Total Porosity of contaminated zone	0.00001
Hydraulic conductivity of contaminated zone	30 m/yr
Unsaturated zone thickness	79.3 m
Unsaturated zone density	1770 kg/m <sup>3</sup>
Hydraulic conductivity of unsaturated zone	12.54 m/yr
Saturated zone thickness	100 m **
Saturated zone density	3060 kg/m <sup>3</sup>
Drinking water intake	730 L/yr **
Well pumping rate	50,000 m <sup>3</sup> /yr
Inhalation rate	7400 m <sup>3</sup> /yr
Wind speed	2.45 m/s
Precipitation	1.3 m/yr
Distance parallel to aquifer (Well)	300 m
Distance parallel to aquifer (Surface water)	450 m

The site-specific parameters listed above were gathered from [3]. \*\* For the parameters not included in the table, the RESRAD code's default values were used.

### 3. Results

Figures 3 and 4 illustrate the radiation dose profiles of all radionuclides associated with the drinking water pathway, which were modeled using stage II and state IV concrete degradation sorption data. The distinct morphologies of the dose peaks in both figures indicate a balance between the code's release rate, radionuclide decay rate, and sorption properties. By comparing their dose curves, one may evaluate the impact of each radionuclide's sorption properties. For instance, the narrowness of the peaks representing <sup>137</sup>Cs and <sup>90</sup>Sr was primarily impacted by the short half-life of the two radionuclides compared to the lengthy half-life exhibited by the broad peaks representing <sup>14</sup>C and <sup>99</sup>Tc. Consequently,

it can be deduced from the two figures that radionuclides with shorter half-lives, such as  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , have narrower widths, while those with longer half-lives have wider widths. Despite having nearly identical short half-lives of 30 yrs and 28.8 yrs, respectively,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  have significantly different dose peaks due to their distinct distribution and diffusion coefficient values. The projected  $^{137}\text{Cs}$  dose contribution is higher than that of  $^{90}\text{Sr}$ . This is due to high concentration of  $^{137}\text{Cs}$  in well water, suggesting that  $^{137}\text{Cs}$  with a low  $K_d$  value of zero desorb from concrete faster than  $^{90}\text{Sr}$  with a  $K_d$  value of 1. Overall  $^{14}\text{C}$  has the lowest dose contribution in both states II and IV because of its high  $K_d$  values of 5000 and 100  $\text{cm}^3/\text{g}$  respectively. Due to its low  $K_d$  value of 1,  $^{99}\text{Tc}$  had the maximum dose contribution in state IV. Although both figures depict a high dose profile for  $^{137}\text{Cs}$ , it is reasonable to assume that its activity level will decrease significantly by the time state II and state IV degradation of concrete barriers occurs due to its short half-life of 30 years. Table 3 depicts the dose for all radionuclides at various RESRAD reporting periods.

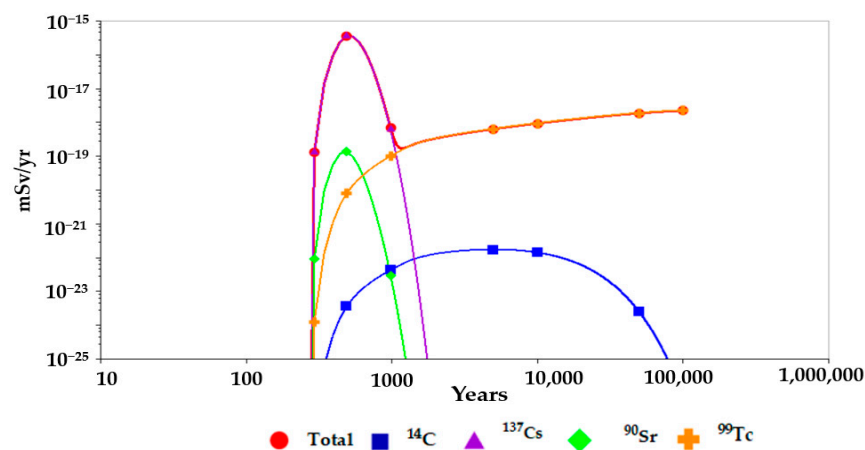


Figure 3. Radiation dose profiles associated with state II of concrete degradation.

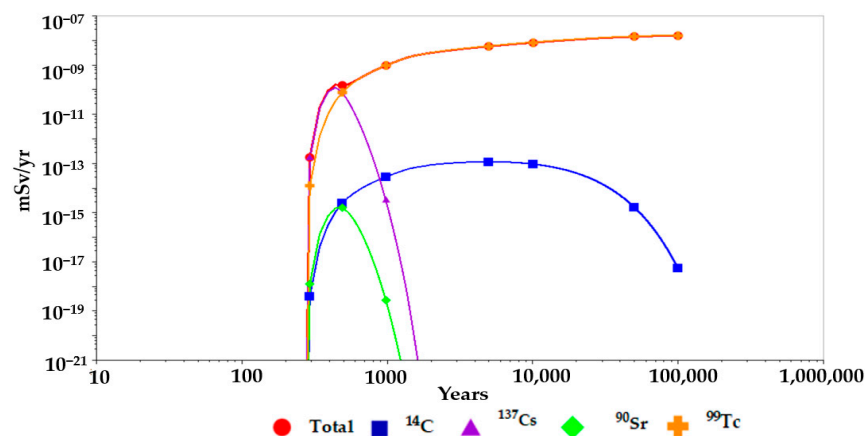
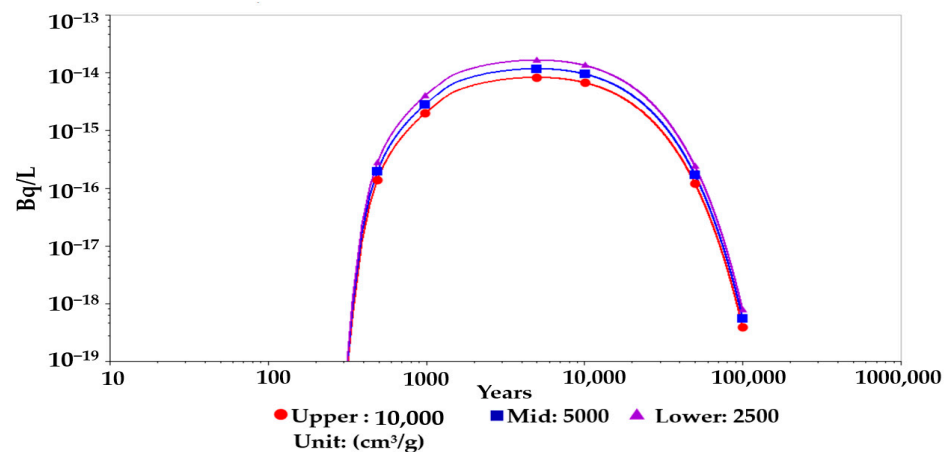
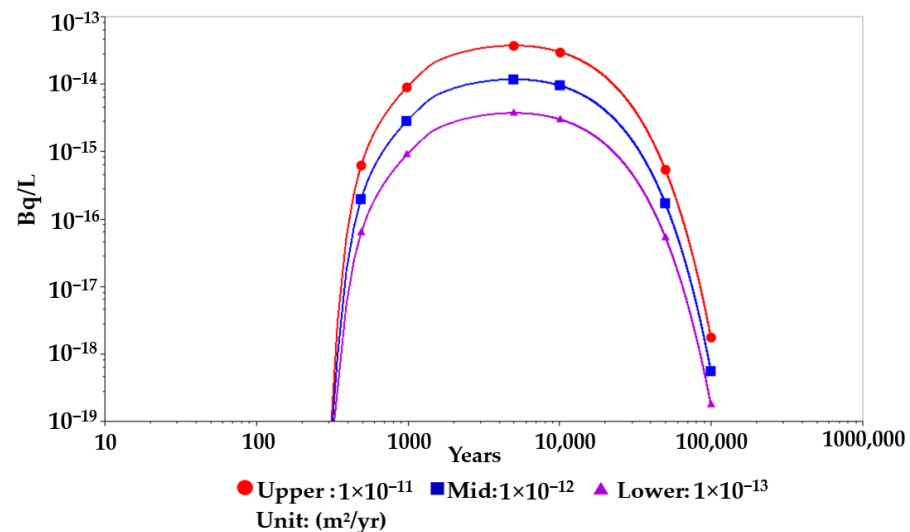


Figure 4. Radiation dose profiles associated with state IV of concrete degradation.

**Table 3.** Radionuclide dose (mSv/yr) contribution in drinking water pathway.

Year	<sup>14</sup> C		<sup>137</sup> Cs		<sup>90</sup> Sr		<sup>99</sup> Tc	
	K <sub>d</sub> = 5000	K <sub>d</sub> = 100	K <sub>d</sub> = 20	K <sub>d</sub> = 0	K <sub>d</sub> = 30	K <sub>d</sub> = 1	K <sub>d</sub> = 3000	K <sub>d</sub> = 1
300	$1.2 \times 10^{-21}$	$3.9 \times 10^{-19}$	$8.4 \times 10^{-20}$	$1.7 \times 10^{-13}$	$3.0 \times 10^{-20}$	$1.3 \times 10^{-18}$	$4.0 \times 10^{-19}$	$1.2 \times 10^{-14}$
500	$7.6 \times 10^{-18}$	$2.4 \times 10^{-15}$	$1.1 \times 10^{-16}$	$7.9 \times 10^{-11}$	$3.7 \times 10^{-17}$	$1.6 \times 10^{-15}$	$2.5 \times 10^{-15}$	$7.7 \times 10^{-11}$
1000	$1.0 \times 10^{-16}$	$3.2 \times 10^{-14}$	$1.1 \times 10^{-20}$	$1.1 \times 10^{-15}$	$2.2 \times 10^{-21}$	$9.4 \times 10^{-20}$	$3.5 \times 10^{-14}$	$1.1 \times 10^{-9}$
5000	$3.6 \times 10^{-16}$	$1.1 \times 10^{-13}$	0	0	0	0	$2.0 \times 10^{-13}$	$5.8 \times 10^{-9}$
10,000	$2.9 \times 10^{-16}$	$9.2 \times 10^{-14}$	0	0	0	0	$2.9 \times 10^{-13}$	$8.2 \times 10^{-9}$
50,000	$5.2 \times 10^{-18}$	$1.6 \times 10^{-15}$	0	0	0	0	$5.8 \times 10^{-13}$	$1.4 \times 10^{-8}$
100,000	$1.7 \times 10^{-20}$	$5.3 \times 10^{-18}$	0	0	0	0	$6.9 \times 10^{-13}$	$1.5 \times 10^{-8}$

The results of a sensitivity assessment on the key parameters of distribution coefficient, diffusion coefficient, and density of the contaminated zone are shown in Figures 5–7, respectively. Figures 5 and 6 illustrate how the distribution coefficient and diffusion coefficient of <sup>14</sup>C impact its concentration in well water in distinct ways, with high K<sub>d</sub> values resulting in low concentrations and large D<sub>s</sub> values resulting in high concentrations. The concentration of the four radionuclides in well water (Bq/L) based on a sensitivity analysis on distribution coefficient is shown in Tables 4 and 5.

**Figure 5.** Sensitivity analysis of the distribution coefficient of <sup>14</sup>C.**Figure 6.** Sensitivity analysis of the diffusion coefficient of <sup>14</sup>C.



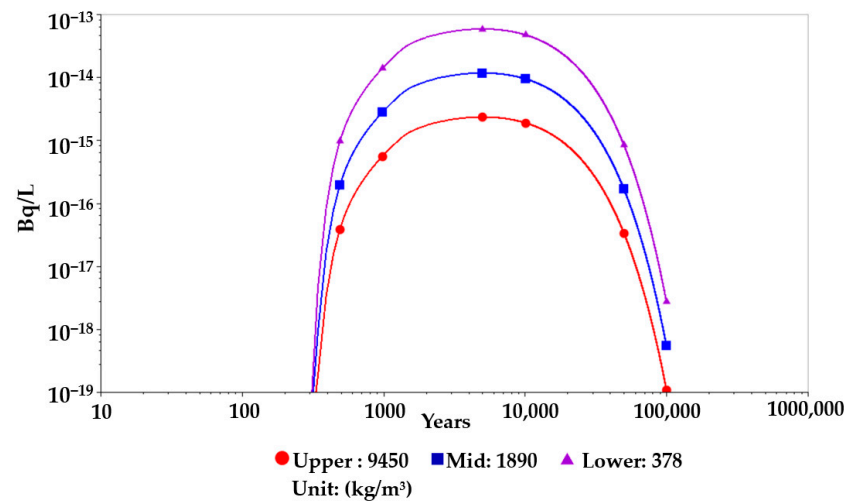


Figure 7. Sensitivity analysis of the density of concrete contaminated zones.

Table 4. Radionuclide Concentration in Well water (Bq/L) with sensitivity on distribution coefficient.

Year	<sup>14</sup> C		<sup>137</sup> Cs		<sup>90</sup> Sr		<sup>99</sup> Tc	
	Upper: 10,000	Lower: 2500	Upper: 200	Lower: 2	Upper: 300	Lower: 3	Upper: 30,000	Lower: 300
300	$2.9 \times 10^{-17}$	$5.8 \times 10^{-17}$	$6.1 \times 10^{-17}$	$1.7 \times 10^{-16}$	$6.8 \times 10^{-18}$	$2.2 \times 10^{-17}$	$3.0 \times 10^{-15}$	$2.8 \times 10^{-14}$
500	$1.8 \times 10^{-15}$	$3.7 \times 10^{-15}$	$8.2 \times 10^{-16}$	$2.3 \times 10^{-15}$	$8.5 \times 10^{-17}$	$2.8 \times 10^{-16}$	$1.9 \times 10^{-13}$	$1.8 \times 10^{-12}$
1000	$2.5 \times 10^{-14}$	$4.9 \times 10^{-14}$	$8.6 \times 10^{-20}$	$2.4 \times 10^{-19}$	$5.1 \times 10^{-21}$	$1.7 \times 10^{-20}$	$2.7 \times 10^{-12}$	$2.6 \times 10^{-11}$
5000	$8.8 \times 10^{-14}$	$1.8 \times 10^{-13}$	0	0	0	0	$1.5 \times 10^{-11}$	$1.4 \times 10^{-10}$
10,000	$7.1 \times 10^{-14}$	$1.4 \times 10^{-13}$	0	0	0	0	$2.2 \times 10^{-11}$	$2.1 \times 10^{-10}$
50,000	$1.3 \times 10^{-15}$	$2.5 \times 10^{-15}$	0	0	0	0	$4.5 \times 10^{-11}$	$4.1 \times 10^{-10}$
100,000	$4.1 \times 10^{-18}$	$8.2 \times 10^{-18}$	0	0	0	0	$5.4 \times 10^{-11}$	$4.8 \times 10^{-10}$

Table 5. Radionuclide Concentration in Well water (Bq/L) with sensitivity on diffusion coefficient.

Year	<sup>14</sup> C		<sup>137</sup> Cs		<sup>90</sup> Sr		<sup>99</sup> Tc	
	Upper: $1 \times 10^{-11}$	Lower: $1 \times 10^{-13}$	Upper: $5 \times 10^{-9}$	Lower: $5 \times 10^{-11}$	Upper: $5 \times 10^{-10}$	Lower: $5 \times 10^{-12}$	Upper: $1 \times 10^{-7}$	Lower: $1 \times 10^{-9}$
300	$1.3 \times 10^{-16}$	$1.3 \times 10^{-17}$	$4.3 \times 10^{-16}$	$4.3 \times 10^{-17}$	$5.1 \times 10^{-17}$	$5.1 \times 10^{-18}$	$3.0 \times 10^{-14}$	$3.0 \times 10^{-15}$
500	$8.2 \times 10^{-15}$	$8.2 \times 10^{-16}$	$5.7 \times 10^{-15}$	$5.7 \times 10^{-16}$	$6.5 \times 10^{-16}$	$6.5 \times 10^{-17}$	$1.9 \times 10^{-12}$	$1.9 \times 10^{-13}$
1000	$1.1 \times 10^{-13}$	$1.1 \times 10^{-14}$	$5.9 \times 10^{-19}$	$6.0 \times 10^{-20}$	$3.8 \times 10^{-20}$	$3.8 \times 10^{-21}$	$2.7 \times 10^{-11}$	$2.7 \times 10^{-12}$
5000	$3.9 \times 10^{-13}$	$3.9 \times 10^{-14}$	0	0	0	0	$1.5 \times 10^{-10}$	$1.5 \times 10^{-11}$
10,000	$3.2 \times 10^{-13}$	$3.2 \times 10^{-14}$	0	0	0	0	$2.2 \times 10^{-10}$	$2.2 \times 10^{-11}$
50,000	$5.7 \times 10^{-15}$	$5.7 \times 10^{-16}$	0	0	0	0	$4.2 \times 10^{-10}$	$4.5 \times 10^{-11}$
100,000	$1.8 \times 10^{-17}$	$1.8 \times 10^{-18}$	0	0	0	0	$4.9 \times 10^{-10}$	$5.4 \times 10^{-11}$

When the sensitivity of the density of the contaminated concrete zone was compared to that of the distribution coefficient, a similar pattern was observed. The radionuclide concentration of all four radionuclides increased with increasing density. Figure 7 depicts a concentration profile of <sup>14</sup>C with sensitivity analysis based on varying densities of the concrete-contaminated zone.

#### 4. Discussion

The ability to demonstrate that properly disposed radioactive waste radionuclides will not expose an average member of the critical group to radiation doses greater than 0.1 mSv per year is a key safety element of engineered designed barriers at near-surface radioactive waste disposal sites [2,3]. This safety measure acknowledges that radionuclides will migrate from the disposal site into the surrounding ecosystem due to a variety of factors, but the

one of interest in this study is concrete degradation. Thus, the aim of this study was to determine whether radionuclide migration caused by concrete degradation can result in significant radiation exposure to the public. This effect of radionuclide migration due to concrete deterioration is assessed in terms of the exposure dose from four radionuclides ( $^{14}\text{C}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , and  $^{99}\text{Tc}$ ) via RESRAD-offsite ground water pathways, assuming drinking water will be fetched from a well located 300 m from the disposal site.

The four radionuclides were chosen based on their long half-lives (more than 20 years) and the availability of sorption data. By restricting the half-life to at least 20 years, the specified radionuclides can be expected to be present in the disposal site after 300 years of regulatory control. Previous studies have shown that concrete will deteriorate in four phases over its lifetime, with each stage defined by a decreasing pH and C/S ratio in concrete pore water. Sorption data in the form of distribution coefficients and diffusion coefficients have been published on the behavior of numerous radionuclides during the four stages of degradation.

The results of this study show that when concrete deteriorates, the amount of radiation released reflect changes in radionuclide sorption properties. As indicated in Table 1, the distribution coefficient of most radionuclides is generally higher during state II of concrete deterioration than in state IV. This difference in  $K_d$  between the two stages is depicted by the radiation dose profile of state II, which is lower than that of state IV, as shown in Figures 3 and 4. In other words, radiation exposure is greater during state IV than during state II. This suggests that radionuclides are substantially more absorbed during state II than state IV of concrete degradation. Thus, the migration of radionuclides from the disposal facility will be significant during state IV of concrete degradation. Nonetheless, the risk of significant radiation exposure during state IV is mitigated by the fact that the radionuclide activity levels will have decreased to insignificant levels by the time the concrete reaches State IV. This knowledge is critical for the encapsulation of radioactive waste in the concrete matrix because the leaching rates of radionuclides with longer half-lives, such as  $^{99}\text{Tc}$  and  $^{14}\text{C}$ , can be prioritized, as these radionuclides will survive in the most deteriorated state of concrete.

This study also revealed the limitations of the RESRAD-OFFSITE radionuclide transport model for simulating the geochemical deterioration of concrete. The RESRAD-OFFSITE release model considers both radionuclide decay rate and leaching (due to concrete degradation) factors when estimating released radioactivity. The resulting concentration/dose peak for each radionuclide indicates a balance between the release rate of the code, the radionuclide decay rate, and the sorption properties. A sharp peak is observed early on for radionuclides with short half-lives ( $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ), whereas a broad peak develops over time for radionuclides with extended half-lives ( $^{14}\text{C}$  and  $^{99}\text{Tc}$ ). Thus, it was difficult to determine how much the sorption properties affect the overall dose peak in comparison to other factors, such as the code's inbuilt release rate and radionuclide decay rate. In addition, it was difficult to predict, based on sorption parameters, when each radionuclide reached its maximum concentration in ground water. It is worth noting that the application of  $K_d$  and  $D_s$  in evaluating the long-term safety of a radioactive waste site is limited to cement-based concrete and its aggregates [17]. Thus, the selection of a single distribution coefficient value for each radionuclide was a crucial task that required a thorough evaluation of sorption data information from various sources [1–3]. This constraint was mitigated by conducting a sensitivity study on  $K_d$  and  $D_s$ , which provided a wide range of sorption values for each radionuclide. This analysis revealed a significant pattern in which a decrease in  $K_d$  results in an increase in released radiation (Figure 5), supporting the hypothesis that concrete deterioration leads to radionuclide discharge from disposal facilities due to a decrease in the sorption capabilities (such as low  $K_d$ ) of the radionuclides.

## 5. Conclusions

For the long-term safety of a near-surface disposal facility, concrete-designed barriers are still sufficient safety features. The simulation of concrete degradation in this study

attempted to prove this assertion. The findings showed that even in their most deteriorated form, concrete structures do not cause a significant release of radionuclides into the environment. The most degraded condition IV of the concrete emitted a maximum dose of  $1.4 \times 10^{-8}$  mSv/yr in drinking water drawn from a hypothetical well 300 m from a disposal site. Aside from being significantly lower than the regulatory limit of 0.1 mSv/yr, the most degraded state of concrete poses no significant radiation exposure risk, as the bulk of radionuclide activity will have decreased by the time the concrete reaches its worst-case condition of degradation. Certain key variables were subjected to a sensitivity analysis ( $K_d$ ,  $D_s$  and density) and radionuclide concentration in well water was found to be inversely related to distribution coefficient ( $K_d$ ) and density, but directly proportional to diffusion coefficient ( $D_s$ ). The findings of this study also highlight the importance of radionuclide sorption in designing barriers to preventing their migration to the environment. The general view is that engineered features with higher sorption properties will increase the safety of disposal facilities.

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