Derivation of Activity Concentration Upper Limits for Low Level Solid Radioactive Waste

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Abstract. The derivation method of activity concentration upper limits for low level radioactive waste is put forward. The activity concentration upper limits for low level solid radioactive waste will be ascertained, on the basis of safety assessment of near surface disposal. On the premise of basic safety requirements about radioactive waste near surface disposal, taking Yaotian disposal site and Feifengshan disposal site as the reference sites, the drilling water scenario, drilling scenario, post-drilling scenario, and housing scenario after the institutional control period when the disposal sites have been closed are considered. The radionuclide transfer process and exposure pathway of various scenarios are analyzed, the conceptual model and mathematical model of radionuclide transfer are established, and the effective dose to human from various scenarios is calculated. Assuming a linear relationship between the activity concentration and the dose, the activity concentration upper limits of various scenarios are then derived for each radionuclide that meet the appropriate dose criteria. The smallest upper limit is chosen, by the approximate integer principle, the magnitude of upper limit of each radionuclide for low level solid waste is then ascertained.

Keywords: Activity Concentration Upper Limit · Safety Assessment · Near Surface Disposal

1 Introduction

In 2009, International Atomic Energy Agency (IAEA) released the latest Safety Guidelines for the Classification of Radioactive Waste (IAEA-GSG-1, 2009) [1], which provides a dispose-based classification scheme of radioactive waste, classifying radioactive waste into exempt waste, very short-lived waste, very low level waste, low level waste, intermediate level waste and high-level waste. Low level waste is defined as the waste that is above clearance levels, but with a limited amounts of long-lived radionuclides. This class of waste requires robust isolation and containment for periods of up to a few hundred years. It is suitable for disposal in engineered near-surface disposal facilities and covers a very broad range of waste. The activity concentration of short-lived radionuclides in LLW may be high, while the activity concentration of long-lived radionuclides is relatively low, but IAEA has not given a specific upper limits. Quantitative waste classification values need to be determined on the basis of safety assessment of disposal

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facilities. The aim of this study is to derive the upper limits of the activity concentration of radionuclides in the low level solid waste based on the safety assessment of the near surface disposal of radioactive waste.

2 Approach to Deriving Activity Limits

Safety assessment is required in the derivation of the near-surface disposal activity concentration limits for specific radionuclides to ensure that the results of safety assessment for both operational and post-closure periods of disposal facilities meet the appropriate safety criteria [2]. The derivation process of the activity concentration limit is shown in Fig. 1. First, take a unit activity concentration of each radionuclide, consider all the potential scenarios, calculate the dose caused by each scenario, compare the peak dose of each nuclide in each scenario, that is, the maximum dose. In order to obtain the activity concentration limit, the maximum dose of each nuclide was compared with the dose limit. Assuming that there is a linear relationship between the activity concentration value and the dose, the activity concentration limit of each radionuclide can be derived in each scenario meeting the appropriate radiological protection criteria. For any given scenario, the activity concentration limits of each radionuclide in the waste can be calculated using formula (1):

$$Cp_i = \frac{Dose_{\lim} \cdot C_i}{MaxDose_{iu}} \tag{1}$$

where, Cp_i is the activity concentration limit of radionuclide i in the waste (Bq/kg), $Dose_{\lim}$ is the relevant dose limit (Sv/a), $maxDose_{iu}$ is the peak dose resulting from the initial activity of radionuclide i in the waste (Sv/a), C_i is the initial activity concentration of the radionuclide i in the waste (Bq/kg).

The minimum value of the activity concentration limit calculated by each scenario is selected to obtain the activity concentration upper limit of the nuclide in the waste, and the corresponding scenario is the key scenario.

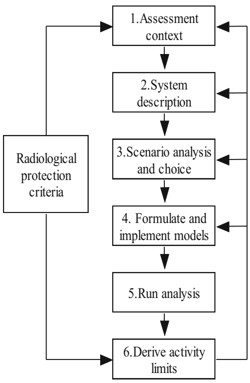

Fig. 1. Activity limits derivation process

2.1 Assessment Context

The purpose of the assessment is to derive the activity concentration limit of the radionuclide for the disposal of radioactive waste in the near surface disposal site, so as to determine the activity concentration upper limit of the low level solid waste, and to ensure that the radiation impact of low level solid waste in near surface disposal on human beings and the environment remains at an acceptable level. In the derivation of the activity concentration limit, 300 years of institutional control period was assumed. Therefore, a possible inadvertent human intrusion should occur 300 years after closure of the facility.

2.2 Dose Limit

The derivation of the activity concentration upper limit of radionuclide in radioactive waste is to consider the long-term safety of the disposal of waste, that is, to consider its potential harm to current and future human beings. The derivation of the activity concentration upper limit of radionuclide for low level solid waste follows the following dose criteria:

The annual effective dose of radionuclides released into the environment by various pathways from the near surface disposal facility to key groups of individuals in the public before the expiration of 300 years of institutional control does not exceed 0.25 mSv [3].

The annual effective dose to the individual public of radionuclides released into the environment by various pathways from the near surface disposal facility at any time after 300 years of institutional control has been released from the disposal facility does not exceed 0.01 mSv [4]. For intruders who breach engineering barriers, the annual effective dose of the radionuclides released from disposal facility to intruders through various pathways does not exceed 1 mSv.

2.3 Disposal Site and Nuclide Selection

In the derivation of the activity concentration limit of the radionuclide in waste, the Yaotan disposal site [5] and Feifengshan disposal site [6] were taken as the objects to conduct evaluation and derivation. The selected nuclides are as follows:

The nuclides with half-life of 5–30 years: ^{3}H, ^{90}Sr and ^{137}Cs. ^{3}H represents the transferable nuclide; ^{90}Sr and ^{137}Cs represent nuclides with half-lives of 30 years, which are about one-tenth of the institutional control period.

β nuclides with long half-life: ^{63}Ni, ^{14}C and ^{99}Tc.

α nuclides with long half-life: ^{239}Pu, ^{240}Pu, ^{237}Np and ^{241}Am.

2.4 Scenario Analysis and Choice

Scenario analysis is an analytical method to determine and quantify the phenomena that can facilitate the release of radionuclides from near surface repository, or influence the release rate. According to the scenarios selected for the environmental impact assessment of the disposal site, the scenarios considered in the derivation of the upper limit value of the activity concentration of low level solid waste can be divided into two categories:

Operational scenarios: in the operational scenarios (such as waste package dropping, fire and other accidents), the risk of radioactivity may be generated for a short duration. The presence of operators and government supervision agencies minimizes the release of radioactivity during operations. Therefore, when determining the limit of the activity concentration of radionuclides, this kind of scenario is of little significance, so the operational scenario is not calculated in this derivation.

Post-closure scenarios:

After the expiration of the institutional control period, the exposure scenario caused by drilling wells and drinking water at the boundary of the disposal site is mainly related to the migration of groundwater, and depends on the environmental characteristics and facility design of the specific site. As this scenario is dependent on the total activity of the whole repository, and has nothing to do with the nuclide concentration in the waste, is called the total activity scenario, the scenario can be used to calculate the total activity limit in the whole site, divided by the waste volume of the entire disposal site can be concluded that nuclide activity concentration limit related to groundwater migration mechanism [7].

After the expiration of the institutional control period, the occurrence of human intrusion scenario, that is, the exposure scenario caused by drilling on the disposal site, living after drilling, and housing. These scenario analysis results are generally less dependent on the environmental characteristics of the site and restrict the concentration level of nuclides, so they are called concentration scenarios. Because the intrusion scenario has little dependence on the characteristics of a particular site, the derived limits are of general significance.

3 Derivation and Calculation of the Upper Limit of Activity Concentration

3.1 Assessment Context

Conceptual Model

After the repository is closed, the performance of the engineering barrier will gradually deteriorate over time. Due to the effect of seepage water, the nuclides will be leached from the waste form, then released from the disposal unit floor into the unsaturated zone under the disposal unit, and then enter the aquifer and transfer along the direction of underground water flow. It is assumed that after the closure of the repository, the public dug a well at 100m downstream of the disposal site to drink the well water and use it for agricultural irrigation. The internal exposure caused by drinking the well water, ingesting the agricultural products irrigated by the well water, and ingesting the poultry and livestock products fed by the crops irrigated by well water is considered. For the drilling water scenario, the radionuclides transfer process and exposure pathways are shown in Fig. 2.

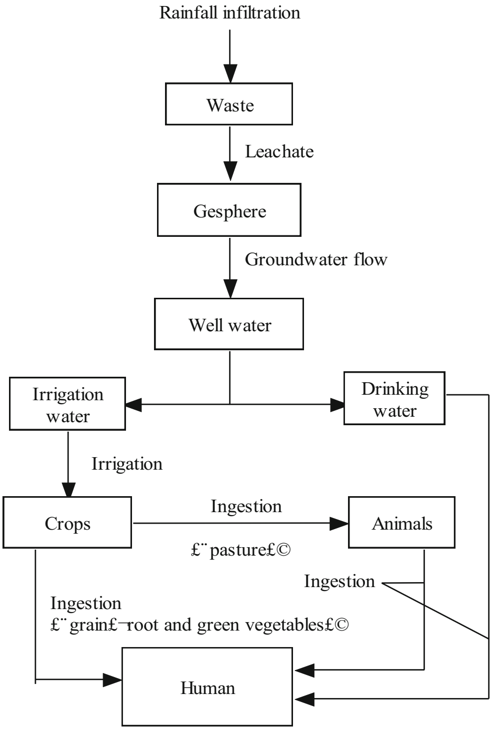

Fig. 2. Radionuclide transfer process and exposure pathway of drilling water scenario

Mathematical Model

(1) Migration of nuclides in the disposal unit

Decay, adsorption and vertical downward migration of nuclides are considered in the calculation of nuclide migration in the disposal unit. The vertical downward transfer rate ($\lambda_{\text{inf, a}-1}$) of nuclides caused by infiltration water is given by the following formula [8]:

$$\lambda_{\text{inf}} = \frac{q_{in}}{L_w \theta_{w,1} R} \tag{2}$$

where, q_{in} is the Darcy flow rate (m/a) through the disposal unit, namely infiltration rate; L_w is the total length of radionuclide migration (m), namely the height of the disposal unit; is the effective porosity of the medium in the disposal unit; R is the retention coefficient of the media in the disposal unit for nuclides, and its value is given by the following formula:

$$R = 1 + \frac{\rho Kd}{\theta_{w,1}} \tag{3}$$

where, ρ is the density of the medium in the disposal unit (kg/m^3), Kd is the distribution coefficient of nuclides in the medium in the disposal unit (m^3/kg).

(2) Migration of nuclides in unsaturated zone

The nuclides released from the bottom of the disposal unit enter the unsaturated zone, and the same model is used to calculate the migration of nuclides in the unsaturated zone. The migration process of nuclides vertically downward is shown in Eqs. (2) and (3).

(3) Migration of nuclides in aquifer

The nuclides migrate into the aquifer through the unsaturated zone. Decay, adsorption, convection and dispersion are considered in the calculation of the nuclides migration in the aquifer. It is assumed that: there is no other leakage source term; the nuclides are uniformly mixed vertically in the aquifer; ignoring molecular diffusion.

When calculating the migration of nuclides in aquifer, for convective transport, the transfer rate is $\lambda_{A,ij}$, and for dispersion, the transfer rate $\lambda_{D,ij}$ is:

$$\lambda_{A,ij} = \frac{q}{\theta_w L_i R} \tag{4}$$

$$\lambda_{D,ij} = \frac{a_x}{\Delta_x} \lambda_{A,ij} \tag{5}$$

where, q is the Darcy flow rate of groundwater, m/a; θ_w is the effective porosity of the aquifer; $q\big/\theta_w$ is the actual velocity of groundwater, m/a; L_i is the length of the compartment in the aquifer, m; α_x is longitudinal dispersion, m; $a_x = \frac{D_x \theta_w}{q}$; D_X is longitudinal dispersion coefficient, m^2/a; Δ_X is the migration distance of the nuclide in the medium, m.

(4) Dose estimation

The total individual dose for drinking water from drilling Wells is calculated as follows:

$$D_{well} = D_{W,P} + D_g + D_{g,1} \tag{6}$$

where, $D_{W,P}$ is annual individual effective dose from drinking well water, Sv/a; D_g is annual individual effective dose from ingestion of contaminated agricultural products, Sv/a; $D_{g,1}$ is annual individual effective dose from ingestion of poultry and livestock products, Sv/a.

The annual individual effective dose from drinking well water can be calculated by the following formula:

$$D_{W,P} = Q \times C_{W,i} \times DC_{ing} \tag{7}$$

where: Q is the amount of drinking water, m^3/a; $C_{W,i}$ is the concentration of nuclide i in well water, Bq/m^3; DC_{ing} is the dose coefficient of nuclides for ingestion, Sv/Bq.

The annual individual effective dose from ingestion of contaminated agricultural products can be calculated by the following formula:

$$D_g = DC_{ing} \times v_p \times f_p \times C_{p,i} \tag{8}$$

where, v_p is an individual's annual intake of P products, kg/a; f_p is the share of P products consumed in the relevant area, conservatively taking FP = 1; $C_{p,i}$ is the activity concentration of nuclide i in P agricultural products, Bq/kg, where $C_{p,i}$ is calculated by the following formula:

$$C_{p,i} = \frac{C_{G,i} \times B_V}{P} \times e^{-\lambda t_h} \tag{9}$$

where, B_V is the concentration factor of nuclides absorbed by the edible part of crops from soil, Bq·kg^{-1}(fresh crops)/Bq·kg^{-1}(dry soil); P is the effective surface density of soil, kg(dry soil)/m^2, P = 240; t_h is the time from harvest to consumption of crops, a; $C_{G,i}$ is the concentration of nuclides deposited on the soil surface caused by irrigation water, Bq/m^2, and its value can be calculated by the following formula:

$$C_{G,i} = \frac{C_{W,i} \times I}{\lambda_e^s} \times (1 - exp(-\lambda_e^s t_b)) \times P_p \tag{10}$$

where, I is the average irrigation rate in the growing season, m^3/(m^{2}a); t_b is the accumulation time of radionuclides on the soil surface, a; P_p is the share of well water irrigation, 0.1; λ_e^s is the effective rate constant for the removal of radionuclides from the soil surface, a^{-1}, which can be calculated by the following formula:

$$\lambda_e^s = \lambda + \lambda_s \tag{11}$$

where, λ_s is the rate constant for other removal processes of nuclide other than decay, a^{-1}.

The annual individual effective dose from ingestion of poultry and livestock products can be calculated by the following formula:

$$D_{g,1} = DC_{ing} \times v_p \times f_p \times C_{p,i,1} \tag{12}$$

where, $C_{p,i,1}$ is the activity concentration of nuclide i in animal products, Bq/kg, which can be calculated by the following formula:

$$C_{p,i,1} = F_A \times C_F \times Q_F \times \exp(-\lambda t_f) \tag{13}$$

where, F_A is the average share of daily intake of radionuclides in each kilogram of animal products, d/kg; C_F is activity concentration of nuclide in animal feed, Bq/kg(dry weight); Q_F is the daily feed consumption of animals, kg(dry weight)/d; t_f is the time from the slaughter of the animal to the consumption of the animal product, a.

3.2 Drilling Scenario Calculation

Conceptual Model

It is assumed that 300 years after the closure of the repository, intruders drill above the disposal unit with a core diameter of D = 0.1 m, a waste length of L = 8 m, a uniform distribution of nuclides, a drilling operator 0.5 m away from the core and the contact time of 8 h. Consider the dose to drilling operators caused by external irradiation from the waste and internal exposure of radioactive dust inhalation. In the drilling scenario, the radionuclide transfer process and exposure pathway are shown in Fig. 3.

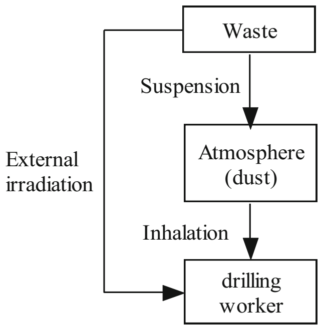

Fig. 3. Radionuclide transfer process and exposure pathway of drilling scenario

Mathematical Model

The total annual individual effective dose to a intruder caused by drilling scenario is calculated by the following formula:

$$D_{drill} = D_{ex} + D_{inh} \tag{14}$$

where, D_{ex} is the external exposure dose, Sv; D_{inh} is the dose of internal exposure due to inhalation, Sv.

(1) External exposure dose

Simplified as a line source, the operator's dose due to external exposure of a certain nuclide is calculated by the following formula:

$$D_{ex} = t \times \frac{47.4 \times A \times \Gamma}{L_{rock} \times R} \times tg^{-1} \frac{L_{rock}}{2R} \tag{15}$$

where, t is the time that the operator contacts the core containing waste, s; Γ is the exposure rate constant, $\mathrm{C \cdot m^2 \cdot kg^{-1} \cdot Bq^{-1} \cdot s^{-1}}$, and the radionuclide of gamma rays is $^{137}\mathrm{Cs}$. L is the length of the core containing waste, m; R is the distance between the operator and the core, m; A is the activity of radionuclide in the core of 8 m long, Bq.

$$A = C_{rock} \times V_{rock} \times \rho_{waste} \tag{16}$$

where, C_{rock} is the activity concentration of the nuclide in the core, Bq/kg; V_{rock} is the volume of the core, m^3; ρ_{waste} is the density of waste in the core, kg/m^3.

(2) Internal exposure dose

Dose due to internal exposure of inhalation is calculated by the following formula:

$$D_{inh} = t \times \eta \times C_i \times DC_{inh} \tag{17}$$

where, η is the air respiration rate of personnel, m^3/min; t is the contact time, min; DC_{inh} is the dose coefficient of nuclides for inhalation, Sv/Bq; C_i is the activity concentration of nuclides in the air, Bq/m^3.

$$C_i = C_{rock} \times S \tag{18}$$

where, S is the dust content in the air (kg/m^3).

3.3 Post-drilling Scenario Calculation

Conceptual Model

The post-drilling scenario refers to the drilling that occurred 300 years after the closure of the repository. The 8 m-long core brought out by the drilling scattered and contaminated the soil. Then the intruder will be engaged in farming on the contaminated soil, and will be exposure by various pathways. It is assumed that the core is evenly distributed in the soil of 2500 m^2 with a thickness of 0.15 m and a soil density of 2000 kg/m^3. Inhalation internal irradiation through resuspension of contaminated soil, direct external irradiation through surface deposition of contaminated soil, and internal exposure through ingestion of crops and vegetables growing in contaminated soil were considered. In the post-drilling scenario, radionuclide transfer process and exposure pathways are shown in Fig. 4.

Mathematical Model

The total annual individual effective dose to an intruder caused by post-drilling scenario is calculated by the following formula:

$$D_{afterdrill} = D_{soil} + D_{inh,1} + D_{ing} \tag{19}$$

where, D_{soil} is the dose caused by surface deposition external exposure, Sv; $D_{inh,1}$ is the dose of internal exposure due to inhalation, Sv; D_{ing} is ingestion dose, Sv.

(1) External exposure dose

The annual individual effective dose due to surface deposition external exposure is calculated by the following formula:

$$D_{soil} = C_{i,soil} \times s \times DC_{ext} \times t \tag{20}$$

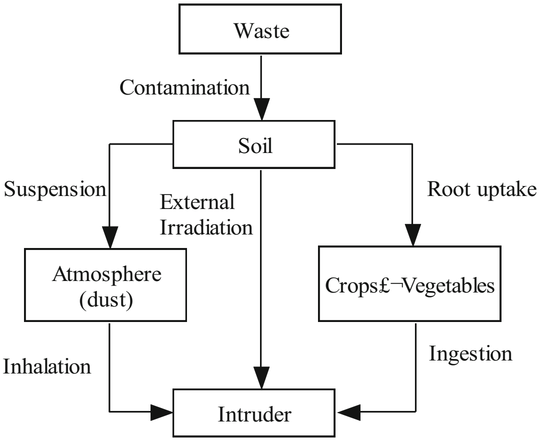

Fig. 4. Radionuclide transfer process and exposure pathway of post-drilling scenario

where, $C_{i,\ soil}$ is the surface concentration of nuclide i in the soil, Bq/m^2; $C_{i,soil} = A/S_{soil}$, A is the activity of radionuclide in the core of 8 m long, Bq; S_{soil} is the area of soil, m^2; s is the building shielding factor, s = 1; DC_{ext} is the dose coefficient for external irradiation from soil, $(Sv/a)/(Bq/m^2)$; t is the time of exposure in a year, a.

(2) Inhalation internal exposure dose

Inhalation internal exposure due to suspension of soil into the air. The resulting dose is calculated by the following formula:

$$D_{inh,1} = t \times \eta \times C_{i,air} \times DC_{inh} \tag{21}$$

where, η is the air respiration rate of personnel, m^3/a; t is the contact time, a; $C_{i,air}$ is the concentration of nuclide in air, Bq/m^3.

$$C_{i,air} = \frac{A}{V_{soil} \times \rho_{soil}} \times S \tag{22}$$

where, V_{soil} is the volume of soil, m^3; ρ_{soil} is soil density, kg/m^3; S is the dust content capacity in the air, kg/m^3.

(3) Ingestion internal exposure dose

Individual dose due to consumption of crops grown on soil contaminated by scattered cores is expressed in the following formula:

$$D_{ing} = I_j \times f \times C_{i,SOIL} \times B_i \times DC_{ing} \tag{23}$$

where, I_j is the annual intake of crop type j, kg; f is the proportion of crops produced in contaminated areas in the annual crop intake, where 1 is taken; B_i is the concentration factor of crops, Bq·kg^{-1}(fresh weight)/Bq·kg^{-1} (dry soil); $C_{i,SOIL}$ is the activity concentration of radionuclide i in the soil, Bq/kg, calculated by the following formula:

$$C_{i,SOIL} = \frac{A}{V_{soil} \times \rho_{soil}} \tag{24}$$

3.4 Housing Scenario Calculation

Conceptual Model

It is assumed that the building and living above the disposal unit takes place 300 years after the closure of the repository, and there is 0.5 m concrete between the house and the waste due to the stripping of all 5 m of the covering layer by digging the foundation. The exposure pathway considered was γ external irradiation. In the housing scenario, the exposure pathway of radionuclide is shown in Fig. 5.

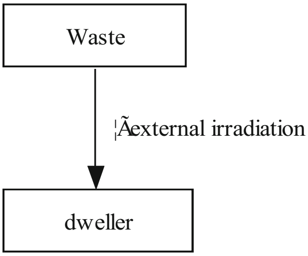

Fig. 5. Radionuclide exposure pathway of housing scenario

Mathematical Model

The waste form is regarded as an infinite flat body source with a certain thickness and uniform distribution of nuclides. After adding 0.5 m concrete shielding, the exposure rate can be calculated by the following formula:

$$X_B = B\exp(-\mu L)\frac{2\pi M\Gamma}{\mu}(1 - E_2(\mu d)) \tag{25}$$

where, X_B is the exposure rate with shielding, C·kg^{-1}·s^{-1}; M is the volume specific activity of a certain nuclide in the disposal unit 300 years after the closure of the repository, Bq/m^3; Γ is the exposure rate constant, C·m^2·kg^{-1}·Bq^{-1}·s^{-1}; μ is the attenuation coefficient of concrete to γ-ray of some energy, m^{-1}; d is the thickness of the body source, m; E_2 is a constant, looking up the table, it can be found that the value of E_2 is in the order of magnitude of 10^{-6}, which is far less than 1 and can be ignored. L is the shielding thickness, m; B is the exposure accumulation factor, $B = A\exp(-a_1\mu L) + (1 - A)\exp(-a_2\mu L)$,, A, a_1 and a_2 are constants.

The air absorbed dose rate calculated from the exposure rate is:

$$D_1 = 33.85X_B \tag{26}$$

where, D_1 is the absorbed dose rate of air, Gy/s. The radionuclide of gamma ray is ^{137}Cs. For ^{137}Cs, the ratio of effective dose rate to air absorbed dose rate is expressed by 1Sv/Gy. H is the effective dose rate, Sv/s, H = D1.

The individual dose to an intruder caused by housing scenario is calculated by the following formula:

$$D_{house} = H \times t_{juzhu} \tag{27}$$

where, t_{juzhu} is the residence time, s.

4 Determination of Nuclide Activity Concentration Upper Limit for Low Level Radioactive Waste

Based on the upper limit value of the activity concentration of each nuclide derived from the above scenarios, the minimum upper limit value was selected and the magnitude of the upper limit value of the activity concentration of each nuclide was determined by the rule for rounding of numbers. According to the rule for rounding of numbers in IAEA RS-G-1.7, that is, if $3 \times 10^{x} < \text{result} < 3 \times 10^{x+1}$, it is $1 \times 10^{x+1}$. The rounding result of the upper limit value is shown in Table 1.

Table 1. Activity concentration upper limits for low level solid radioactive waste

Nuclide	Half-life (a)	Activity concentration upper limit of drilling scenario (Bq/kg)	Activity concentration upper limit of post-drilling scenario (Bq/kg)	Activity concentration upper limit of housing scenario (Bq/kg)	Activity concentration upper limit of drilling water scenario (Yaotian disposal site) (Bq/kg)	Activity concentration upper limit of drilling water scenario (Feifengshan disposal site) (Bq/kg)	Activity concentration upper limit (Bq/kg)
^{3}H	12.3	7.66E+18	8.98E+17		9.92E+07	**3.78E+07**	**1E+08**
^{90}Sr	29.1	8.27E+11	2.30E+09		**4.62E+07**	4.93E+07	**1E+08**
^{137}Cs	30.0	2.47E+10	3.30E+09	**4.36E+08**	2.23E+15	2.04E+16	**1E+09**
63Ni	96	6.38E+11	**4.45E+10**		1.24E+12	3.03E+12	**1E+11**
^{241}Am	4.32E+02	**1.76E+06**	1.14E+07		9.90E+07	3.92E+06	**1E+06**
^{14}C	5.73E+03	1.86E+10	2.70E+07		**1.97E+06**	3.78E+06	**1E+06**
^{240}Pu	6.54E+03	8.96E+05	6.10E+06		4.63E+05	**3.35E+05**	**1E+05**
^{239}Pu	2.41E+04	8.75E+05	5.69E+06		9.20E+04	**5.34E+04**	**1E+05**
99Tc	2.13E+05	8.02E+09	2.59E+06		**5.93E+04**	7.25E+04	**1E+05**
^{237}Np	2.14E+06	2.08E+06	1.21E+07		9.01E+04	**1.74E+03**	**1E+03**

As can be seen from Table 1, the key scenario of ^{137}Cs is housing scenario, the key scenario of ^{63}Ni is post-drilling scenario, the key scenario of ^{241}Am is drilling scenario, and the key scenario of other nuclides is drilling water scenario.

For low level solid waste containing a variety of artificial radionuclides, the sum of the ratio of the activity concentration value of each artificial radionuclide to the upper limit value of their respective activity concentration should not be greater than 1, that is, it should satisfy Eq. (28):

$$\sum_{i=1}^{n} \frac{C_i}{C_{io}} \leq 1 \tag{28}$$

where, C_i is the activity concentration of nuclide i in low level solid waste, Bq/kg; C_{io} is the upper limit of activity concentration of nuclide i in low level solid waste, Bq/kg; n is the number of artificial radionuclides in low level solid waste.

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