INVESTIGATION OF RESIDUAL RADIOACTIVITY IN THE SOIL OF DECOMMISSIONED NUCLEAR FACILITIES

by

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According to the Soil Pollution Prevention and Control Law, the decommissioning sites of nuclear facilities can be divided into three open scenarios – agricultural land, class I construction land and class II construction land. Relevant parameters (including soil density, contamination depth, effective porosity of contaminated soil, personal water intake, personal breathing volume, etc.) were selected for calculation. Based on the dose screening value of 0.01 mSv per year, the residual level in the soil was calculated and the main nuclide pathways were analyzed. The study provides the principle of determining soil residual radioactivity level according to different land use types and irradiation routes, so that the treatment target of contaminated sites is more scientific and detailed, and at the same time, the environmental management of the management department can be adjusted. Compared with the recommended values in foreign guidelines, the rationality and variety of residual radioactivity levels can provide technical support for the revision of "Regulations on Acceptable Levels of Residual Radioactivity in Soil of Proposed Open Sites (Provisional)" (HJ53-2000).

Key words: *nuclear facilities, retirement site, residual radioactivity level*

Introduction

In order to determine the level of radioactivity in the soil of contaminated sites [1-5] and then standardize the use of decommissioned sites of nuclear facilities, China formulated *The Provisions on the Acceptable Level of Remaining Radioactivity in the Soil of the Proposed Open Site (Interim) (HJ 53-2000)* in 2000. With the decommissioning of various types of nuclear facilities in China, the decommissioning situation has become increasingly complex. The scope and limits of residual radionuclides in the previous standard can no longer meet the needs of existing decommissioning practices, so it is urgent to revise the standard to meet the needs of existing and future decommissioning projects in China.

According to the principle of optimizing radiation protection and the existing decommissioning practices at home and abroad, the dose constraint value of the final state of unrestricted open sites in China should be 0.25 mSv per year. Meanwhile, the lowest limit of dose optimization should be 0.01 mSv per year. This value will also be used as the screening level in the revision process.

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The screening level is the primary basis for determining whether the soil needs to be treated if the site contamination level is equal to or less than the screening level. If the site contamination level is higher than the screening level, based on the determined use of the site after it is opened, derivation calculations and dose criteria can be used to further determine whether treatment is needed. The use of the screening level can simplify the process of decommissioning treatment. For sites with light contamination, the decision not to clean the soil can be made without repeated iterative calculations.

The Regulation on the Acceptable Level of Residual Radioactivity in the Proposed Site (Interim) (HJ 53-2000) considers only the conservative and comprehensive irradiation path and gives the residual level of radionuclides in the soil based on 0.1 mSv per year. Therefore, according to the requirements of the Soil Pollution Prevention and Control Law, the principle of determining the level of residual radioactivity in soil should be given according to different types of land use and irradiation routes, so that the control target of contaminated sites can be more scientific and refined, and at the same time, the environmental management of the inspection department can be adjusted.

According to *The recommended methods and scenarios in the Scenarios, Calculation Mode and Parameters of Acceptable Activity Concentrations of Radiation Residues in the Soil after Decommissioning (EJ1191-2005)*, the Residual radioactivity (RESRAD) program developed by Argonne Laboratory was used to calculate the screening level (dose optimization lower limit of 0.01 mSv per year), the corresponding soil radioactivity levels were compared with the values recommended in foreign regulatory guidelines to provide technical support for the revision of *The Provisions on the Acceptability Level of Residual Radioactivity in the Soil of the Proposed Open Site (Interim) (HJ 53-2000).*

Methodology

Selection of the calculation method

This study will use the US RESRAD-onsite program to carry out relevant calculations.

Analysis of the irradiation pathway

Based on the contamination situation of the decommissioning site of nuclear facilities and the proposed opening scenario after decommissioning, the key nuclides were screened out and the reasonably conservative irradiation routes were determined. The screening of the key residues was based on the investigation of the decommissioning sources, the operational history of the nuclear facility and the events/accidents. The selection of the irradiation route was determined by the proposed opening scenario after the decommissioning of the nuclear facilities.

For different major nuclear contamination and proposed opening scenarios, although the exposure pathways are different, most do not exceed the following exposure pathways, usually one or a combination of them: external surface exposure, inhalation exposure, internal food exposure related to contaminated soil (including vegetables, milk, meat, fish, *etc.*), water-related internal exposure, direct internal exposure of contaminated soil, and radon inhalation exposure. Due to the decommissioning approval practice in China in recent years, the radon inhalation route was not considered in this study. A schematic diagram of the radiation pathway is shown in fig. 1.

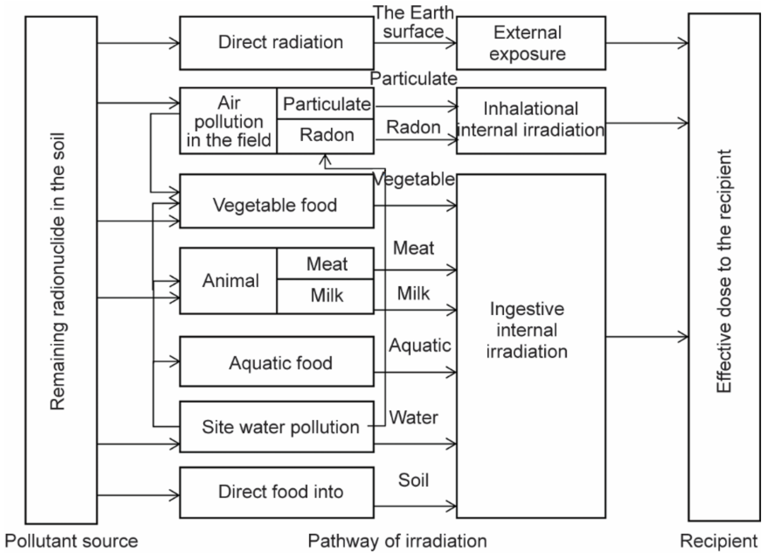

Figure 1. Possible route of residual radionuclide exposure in contaminated soil

The RESRAD program considers the main radiation path in the calculation. Possible primary pathways under various open scenarios are shown in tab. 1.

Table 1. Main exposure pass-ways under different open scenarios in RESRAD

Exposure pathway	Agricultural land	Construction land type I	Construction land type II
Gamma external exposure	√	√	√
Micro dust inhalation	√	√	√
Ingested by growing food	√	×	×
Ingestion through meat	√	×	×
Ingestion through milk	√	×	×
Ingestion through fish	√	×	×
Ingestion through soil	√	×	×
Drinking water	√	√	×

The first type of land use is characterized by long-term exposure risk for children and adults. The land use in question was predominantly residential land (R), as specified by GB50137. In consideration of the social sensitivity of the land use, the following types of land are included in the first category: schools, medical care, and pension, including primary and secondary schools (A33), medical and health land (A5), and social welfare facilities (A6) in public management and public service land; community park or children's park land in park green space (G1). The typical land type is farmers' homestead, focusing on children.

The second category of land use is associated with a higher risk of long-term exposure for adults. The second type of land use includes GB50137 urban construction land (M), logistics warehousing land (W), commercial services facilities land (B), road and transportation land (S), utilities land (U), public management and public service land (A) (A except A33, A5, A6), and green space and square land (G) (except community park or children park in G1), *etc.* (The typical land type is the factory, focusing on adults).

Agricultural land is defined as land that is used for the production of food and other agricultural products. The Land Management Law stipulates that agricultural land is not to be considered for residential construction. The typical land type is cultivated land, which is considered suitable for adults.

The RESRAD program offers nine distinct irradiation routes that can be selected or excluded to align with the specific circumstances of the site and the population under consideration. The nine irradiation routes can be broadly classified into three categories: external irradiation, internal irradiation, and the feeding route.

- External irradiation (direct exposure)
- Inhalation internal radiation (inhalation of particulates)
- Inhalation of radon gas released from contaminated soil (inhalation of radon).
- Plant food (ingestion of plant foods) grown in contaminated soil and irrigated with contaminated water.
- Meat produced by livestock fed with contaminated feed and water.
- The ingestion of milk (drinking milk) is another potential source of contamination.
- The ingestion of water from wells or ponds in the adjacent contaminated area.
- Consumption of pond aquatic foods (ingestion of aquatic foods).
- Inhalation of soil from contaminated areas is another potential route of exposure.

Parameter determination

Nuclide type

Five calculated nuclide species included Co-60, Sr-90, Cs-137, Pu-239, and Am-241.

Soil parameters of the contaminated site

The parameters pertaining to soil characteristics in a contaminated site are as follows:

- The pollution area is 10000 m^2, 100×100 m^2, and there are no other overlays.
- The soil density is 1.5 g/cm^3, as recommended by HJ 25.3-2014.
- The pollution depth is 30 cm.
- The depth of the tillage layer is 15 cm.
- The moisture content of the contaminated soil is 20% kg water per 1 kg soil.
- The effective porosity of the contaminated soil is 0.
- The thickness of the aquifer at the contaminated site is 5 m.
- The longitudinal and transverse dispersion coefficient of the groundwater at the contaminated site is 2.20 m squared per day, with a transverse dispersion coefficient of 0.09 m^2 per day.
- The groundwater flow rate at the contaminated site is 1.
- The annual precipitation infiltration capacity of the plant is 0.19 m per year.
- The deposition rate is 0.001 m/s.
- The dust concentration in the resuspended air is 0.0003 g/m^3.

Public-related parameters

The public can be classified into two groups: children (1-7) and adults ($\geq$ 18).

– Personal water intake [L per year], children (400), adult (730);
– Personal respiratory volume [m^3 per hours], children (0.61), adult (0.96);
– Recipe parameters. The unit of measurement is [kg per person per year]. The recipes for children and adults are presented in tab. 2.

Table 2. The amount of consumption for children and adult [kg per year]

Age group consumption amount	Children	Adult (by mean within range)
Commissariat	95.8	100-150
Vegetables	147.40	110-120
Meat	34.00	50-60
Aquatic product	130	40-60
Milk class	91.25	20-30
Fruit	68.44	50-60
Eggs	49.9	20-30

Results and discussion

Calculation results of the radioactivity level in the contaminated site

In accordance with the established irradiation route, the parameter value is the recommended value. During the calculation process, other values may be selected according to experience and divided into three open scenarios.

Agricultural land. The typical scenario considered in the evaluation was arable land, in which farmers worked and ate agricultural products. The route of exposure can be divided into three categories: external exposure, internal exposure, and feeding and drinking water. For the sake of argument, let us suppose that a person remains in the aforementioned scene for a period of 24 hours.

The first type of construction land. The typical scene considered in the evaluation was a rural homestead, with a particular focus on the long-term exposure risk of children and adults.

The exposure pathway is comprised of external exposure to surface deposition, internal exposure to inhalation, and drinking water.

For the purposes of this analysis, it is assumed that a person will remain in the scene for a period of 24 hours, with 12 hours spent outdoors.

The second type of construction land. The typical scene considered in the evaluation is the logistics and storage land, with a focus on the long-term exposure risk of adults.

The route of exposure is external exposure to surface deposition, inhalation, and internal exposure.

For the purposes of this analysis, it is assumed that the human presence in the scene is 12 hours, with 6 hours spent outdoors.

The dose contribution of 1 Bq of the nuclide in the three land-use types was calculated to be 0.01 mSv per year. The three scenario land uses are presented in tabs. 3-6.

Table 3. Dose contributions [mSv per year] of farmland radionuclide (1 Bq/g)

Pathway of irradiation	Co-60	Sr-90	Cs-137	Pu-239	Am-241
Gamma external exposure	$2.78\cdot10^{00}$	$3.30\cdot10^{-03}$	$6.30\cdot10^{-01}$	$4.00\cdot10^{-05}$	$5.90\cdot10^{-03}$
Micro dust inhalation	$3.91\cdot10^{-05}$	$4.03\cdot10^{-04}$	$4.91\cdot10^{-05}$	$1.51\cdot10^{-01}$	$1.21\cdot10^{-01}$
Ingested by growing food	$2.87\cdot10^{-03}$	$8.66\cdot10^{-01}$	$4.75\cdot10^{-02}$	$1.34\cdot10^{-03}$	$6.19\cdot10^{-03}$
Ingestion through meat	$6.19\cdot10^{-04}$	$2.35\cdot10^{-04}$	$2.91\cdot10^{-03}$	$1.02\cdot10^{-03}$	$1.00\cdot10^{-02}$
Ingestion through milk	$2.86\cdot10^{-05}$	$2.35\cdot10^{-02}$	$5.24\cdot10^{-03}$	$4.73\cdot10^{-05}$	$4.62\cdot10^{-04}$
Ingestion through soil	$3.52\cdot10^{-03}$	$5.11\cdot10^{-04}$	$2.37\cdot10^{-04}$	$4.56\cdot10^{-03}$	$3.65\cdot10^{-03}$
Drinking water	$6.21\cdot10^{-05}$	$3.36\cdot10^{-01}$	$6.92\cdot10^{-14}$	$2.73\cdot10^{-02}$	$1.65\cdot10^{-02}$
Integral dose	$2.78\cdot10^{00}$	$1.23\text{E}\cdot10^{-00}$	$6.86\cdot10^{-01}$	$1.88\cdot10^{-01}$	$1.64\cdot10^{-01}$

Table 4. Dose contributions [mSv per year] of each route of the first-class construction site radionuclide (1 Bq/g)

Pathway of irradiation	Co-60	Sr-90	Cs-137	Pu-239	Am-241
Gamma photo	$4.71\cdot10^{-00}$	$5.62\cdot10^{-03}$	$1.07\cdot10^{-00}$	$6.80\cdot10^{-05}$	$1.00\cdot10^{-02}$
Micro dust inhalation	$2.39\cdot10^{-05}$	$1.09\cdot10^{-04}$	$2.83\cdot10^{-05}$	$6.07\cdot10^{-02}$	$4.86\cdot10^{-02}$
Drinking water	$9.19\cdot10^{-03}$	$3.09\cdot10^{-01}$	$2.80\cdot10^{-14}$	$1.50\cdot10^{-02}$	$1.22\cdot10^{-02}$
Ingestion through soil	$8.57\cdot10^{-04}$	$2.37\cdot10^{-03}$	$4.84\cdot10^{-04}$	$1.26\cdot10^{-02}$	$1.36\cdot10^{-02}$
Integral dose	$4.72\cdot10^{-00}$	$3.17\cdot10^{-01}$	$1.07\cdot10^{-00}$	$8.84\cdot10^{-02}$	$8.44\cdot10^{-02}$

Table 5. Dose contribution [mSv per year] from each route (1 Bq/g) of Class II construction land

Pathway of irradiation	Co-60	Sr-90	Cs-137	Pu-239	Am-241
Gamma photo	$2.36\cdot10^{-00}$	$2.81\cdot10^{-03}$	$5.36\cdot10^{-01}$	$3.40\cdot10^{-05}$	$5.02\cdot10^{-03}$
Micro dust inhalation	$2.73\cdot10^{-05}$	$1.41\cdot10^{-04}$	$3.44\cdot10^{-05}$	$1.06\cdot10^{-01}$	$8.47\cdot10^{-02}$
Ingestion through soil	$4.34\cdot10^{-05}$	$3.58\cdot10^{-04}$	$1.66\cdot10^{-04}$	$4.22\cdot10^{-03}$	$2.56\cdot10^{-03}$
integral dose	$2.36\cdot10^{-00}$	$3.31\cdot10^{-03}$	$5.36\cdot10^{-01}$	$1.10\cdot10^{-01}$	$9.22\cdot10^{-02}$

Table 6. Calculated values based on 0.01 mSv of three land use types per year [Bg/g]

Nuclein	Agricultural land	First class construction land	Second class construction land
^{60}Co	$3.59\cdot10^{-02}$	$2.12\cdot10^{-03}$	$4.24\cdot10^{-03}$
^{90}Sr	$8.13\cdot10^{-03}$	$3.15\cdot10^{-02}$	$3.02\cdot10^{-00}$
^{137}Cs	$1.46\cdot10^{-02}$	$9.33\cdot10^{-03}$	$1.87\cdot10^{-02}$
^{239}Pu	$5.32\cdot10^{-02}$	$1.13\cdot10^{-01}$	$9.08\cdot10^{-02}$
^{241}Am	$6.10\cdot10^{-02}$	$1.18\cdot10^{-01}$	$7.81\cdot10^{-02}$

Aanalysis of foreign recommended values

The calculated value of soil residue level based on 0.01 mSv per year, in comparison with the foreign technical guidelines (including the comprehensive decommissioning guide (NUREG-1757) and the NCRP report No.129), is presented in tabs. 7-9.

Table 7. Analysis of soil residual level in agricultural land scenario [Bg/g]

Nuclein	Agricultural land	NUREG-1757
^{60}Co	$3.59\cdot10^{-03}$	$1.41\cdot10^{-01}$
^{90}Sr	$8.13\cdot10^{-02}$	$6.29\cdot10^{-02}$
^{137}Cs	$1.46\cdot10^{-02}$	$4.07\cdot10^{-01}$
^{239}Pu	$5.32\cdot10^{-02}$	$8.51\cdot10^{-02}$
^{241}Am	$6.10\cdot10^{-02}$	$7.77\cdot10^{-02}$

Table 8. Analysis of soil residual level [Bg/g]

Nuclein	First class construction land	NCRP No.129 reports Suburban (SU) views
^{60}Co	$2.12\cdot10^{-03}$	$1.76\cdot10^{-03}$
^{90}Sr	$3.15\cdot10^{-02}$	$3.36\cdot10^{-03}$
^{137}Cs	$9.33\cdot10^{-03}$	$8.00\cdot10^{-03}$
^{239}Pu	$1.13\cdot10^{-01}$	$4.80\cdot10^{-02}$
^{241}Am	$1.18\cdot10^{-01}$	$4.80\cdot10^{-02}$

Table 9. Analysis of soil residual level under category ii construction land situation [Bg/g]

Nuclein	Second class construction land	Construction, commercial, industrial (CC) scenes reported by NCRP 129
^{60}Co	$4.24\cdot10^{-03}$	$3.96\text{E}\cdot10^{-03}$
^{90}Sr	$3.02\cdot10^{-00}$	$1.24\cdot10^{-00}$
^{137}Cs	$1.87\cdot10^{-02}$	$1.80\cdot10^{-02}$
^{239}Pu	$9.08\cdot10^{-02}$	$1.88\cdot10^{-02}$
^{241}Am	$7.81\cdot10^{-02}$	$1.88\cdot10^{-02}$

The screening values in NUREG1757 were calculated by NRC using the DANDD program, and the parameters of the feature site were selected in the calculation. The difference between the calculated levels of soil residue is mainly due to the following reasons.

– The screening value of NRC is derived based on the dose of 0.25 mSv per year, so to facilitate the comparison of the calculation results of agricultural land, the data values of NRC in the table are based on the screening values in NUREG1757 divided by 25, respectively. However, the screening value in NUREG1757 considers a series of migration processes, so the data obtained divided by 25 and the real 0.01 mSv per year will have all differences.

– The open scene of agricultural land is a typical farming scene in China. The irradiation methods include external irradiation of cultivated land, inhalation and internal irradiation, edible agricultural products, edible soil and drinking water, without considering residence. The scenario identified in the screening values in NUREG1757 is agriculture, which assumes that the irradiation pathway includes external irradiation and inhalation and internal irradiation, and that children and infants may consume contaminated food. Eating soil and drinking water were not considered.
– The main dose route of gamma nuclide is gamma external irradiation. The actual situation of domestic and foreign agricultural land is different. The USA considers large-scale automated planting. China's agricultural land is still 12 hours per day, and the residence factors are 0.1 and 0.5.

One type of construction land considered in this study is the risk of long-term exposure to children and adults. Select the suburban (SU) scene reported by NCRP No.129. The scene assumes that there is some smaller food production, such as food production from vegetable gardens. The people may be adults who live and eat vegetables on the site, or children who play outdoors and inhale contaminated soil.

For ^{60}Co and ^{137}Cs, both calculations are in the same magnitude, and the difference in results is small and agrees well.

For ^{90}Sr, ^{239}Pu, and ^{241}Am, the two calculations are only one order of magnitude worse.

The main irradiation route of ^{90}Sr is the drinking water route. In the calculation, the full field leach is assumed and then the point source is released instantly. By finding the minimum dilution multiple, the maximum concentration of the nuclide in the groundwater at the *x*-distance in the downstream direction is obtained from the release concentration, and the well position is assumed to be 100 m from the release point. The NCRP in the calculation of drinking water way to consider the migration process is complex, respectively from the gas belt and water to carry out the calculation, and in the two media input a large number of parameters.

The main route of irradiation for ^{239}Pu and ^{241}Am is micro-dust inhalation. When calculating the two calculation methods, the calculation logic is basically the same, but the parameters are different: the conversion coefficient of the internal irradiation dose refers to GB18871-2002, while NCRP uses ICRP38 report, and assuming that the erosion of the contaminated soil is not considered, the default of the resuspended soil particles is the fixed value. The NCRP introduces the coverage and depth correction coefficient FCD, *t*, which corrects the share of soil particles that can be resuspended.

The second type of construction land in this study mainly considered the risk of long-term exposure in adults. For ^{60}Co, ^{137}Cs, ^{90}Sr, ^{239}Pu, and ^{241}Am, the two results are in the same order, and the difference of results is small and in good agreement.

At the same time, the results of the NCRP 129 are slightly lower than the results of this study, which is consistent with the report that, because the dose from building and earthwork may be short-term, the screening limits will be more conservative than long-term exposure.

Conclusions

In this study, three types of agricultural land were selected for investigation, with particular attention paid to the site and public parameters. The dosage criterion was set at 0.01 mSv per year, after which the soil residual level value was calculated.

In accordance with the stipulations of the Soil Pollution Prevention and Control Law, this study presents the fundamental principles governing the determination of soil residual radioactivity levels according to distinct land use categories and irradiation pathways. This approach enables the delineation of more precise and scientific treatment targets for contaminated sites, while simultaneously facilitating the alignment of environmental management strategies with the objectives of the relevant regulatory authorities.

In this study, the soil residual level was calculated according to the screening level (optimization lower limit of 0.01 mSv per year), which analyzed the rationality of the calculation results and the reason for the discrepancy, and provided strong technical support for the revision of the acceptable level of residual radioactivity in the proposed open site (provisional) (HJ 53-2000).

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