PROPOSAL FOR UNCONDITIONAL CLEARANCE LEVELS FOR CONTAMINATED SOIL FROM ITALIAN NPP

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1. ABSTRACT

The paper purpose is to propose criteria for the definition of unconditional clearance levels of excavated soil derived from Italian Nuclear Power Plants (NPP).
The clearance levels guideline refers to radionuclides derived from:
- radiological inventory of Italian sites (SOGIN sites);
- IAEA Technical Reports Series n° 389-1998. [8]

The list of radionuclides for which are defined the levels are presented table 2. The paper also reports an analysis on the levels of final release of land account that the definition of these levels must be analyzed case by case basis whereas the final destination of the site.

2. NATIONAL REFERENCES

The Italian law governing the Radiation Protection is the Legislative Decree no. 230/1995 and subsequent amendments. In particular the art. 154 established that the clearance levels of materials to be disposed, recycled or reused in installations or for activities which do not apply the rules of the mentioned Law, must meet the criteria established by Art. 1 and for this purpose take into account the guidelines, recommendations and the technical guidance provided by the European Union.
The proposed criteria refer to European legislation and, in particular, to the documents issued by European Commission Radiation Protection and publications of the IAEA, NRPB, German legislation and US legislation.

3. INTERNATIONAL REFERENCES

The European Directive 96/29 Euratom fixes that any clearance levels of radioactive materials set by national authorities must always take account of certain basic criteria with attention to individual radiological risk and collective radiological incidence.
Must not be exceeded:
- Effective dose of 10 microSv/year for public (critical group);
- Collective dose of 1 man-Sv/year during the period in which the practice is performed.

The European Commission produced a series of publications on materials from nuclear facilities, but none of them is related directly to the soil. To define the proposed levels, it is considered European Commission, IAEA, NRPB publications and German and US legislation on radiation protection.

European Commission

The clearance levels proposed by European Commission for metals are fixed in RP89 [1], for buildings and building rubble in RP113 [4] and the general clearances levels in RP122/I [6].
Methodologies and calculation methods to define the clearance levels for metals and buildings and building rubble materials are reported in RP 101 [2], RP 117 [3] and RP 114 [5].

IAEA

IAEA published a set of clearance levels in Safety Guide RS-G-1.7[11], while the technical approach, the scenarios and parameters are in the Safety Report No.44 [12]. Natural radionuclides are separated from artificial radionuclides. Natural radionuclides include ^{40}K and nuclides of ^{238}U, ^{235}U and ^{232}Th decay series. Others radionuclides are considered artificial.
The scenarios developed in the Safety Report No.44 deal with artificial radionuclides only and cover a wide variety of exposure conditions for both workers and public.
These conditions are associated with different radiological scenarios developed into two forms, even considering the same routes of exposure:
- "Realistic scenarios": parameters close to real exposure situation are used. The scenarios results are compared with an effective dose constrain equal to 10 microSv/year;

- "Low probability" scenarios: more conservative values of parameters are used. The results are compared with the effective limit dose equal to 1 milliSv/y.

The scenarios are in table 2 of the Safety Report No.44. Many of these scenarios include a number of simultaneous exposure situations which are listed in the table, although they are identify with a specific description. The dose for each scenarios is obtained summing the dose contribution of all exposure routes that define the scenario.
In Safety Report RG-G-1.7 were not limits on the types of materials considered. They include: metals, building materials, scrap, non-metallic solid materials and others.
The proposed clearance levels for natural radionuclides are based on global activity concentration in the soil, rocks and natural materials. The levels are not derived from exposure scenarios.

Germany

The German Radiation Protection Ordinance (July 2001) [13] contains specific instructions on removal and clearance levels. These guidelines are applied on materials release and re-use of originally nuclear installations. The clearance levels are based on radiological compatible models with international criterion (individual effective dose constrain equal to 10 microSv/y; collective dose constrain equal to 1 man-Sv /y).
The levels are set according to several clearance options:

1. Unconditional clearance of:
    a. Solids for reusing, recycling or disposal (including building materials with amount lower than 1000 ton/y);
    b. Liquids;
    c. Building rubble materials and soil with amount higher than 1000 ton/y;
    d. Buildings reuse;
    e. Site.
2. Clearance:
    a. Solid materials for disposal;
    b. Liquids …. ;
    c. Buildings for demolition;
    d. Scrap metal for recycling.

NRPB-W36

Publication NRPB-W36 of the National Radiological Protection Board [14] shows methodology and calculation method to determinate the doses per concentration unit in soil. The potential doses, arising from the use of the land, refer to members of the public. The dose contributions depend on both uniform contamination and non-uniform contamination of the soil.
The exposure scenarios include the use of the ground for agriculture, recreational activities and other purposes (houses, offices, schools). Are also taken into account workers involved in structures building.

The exposure routes are:

- External radiation;
- Internal exposure for soil radionuclides (resuspension inhalation, inadvertent soil ingestion);
- Internal exposure from eating foods grown in the field.

US References

In U.S. there are different rules governing the clearance of contaminated materials from nuclear facilities. U.S. Nuclear Regulatory Commission (USNRC), Department of Energy (DOE) and Environmental Protection Agency (EPA) have issued rules and/or guidelines in accordance with their statutory duties.

- USNRC

USNRC defines two different authorization approaches: "10 CFR Part 20" deals with sites release and sets two constrains: dose lower than 250 microSv/year and the application of ALARA in reducing radioactive wastes; in general, USNRC assesses the situation referring to license specifications and actual guidelines. In particular, the "Regulatory Guide 1.86" deals with materials release and defines clearance levels in terms of surface contamination. There isn't a guideline for mass contaminations: in this case, the assessment and decisions are made in such a way to ensure a maximum dose of a few percent to the limit of 1milliSv/y.

The USNRC has also published NUREG-1640 that deal with the estimated individual effective dose from re-use, recycling, or materials storage for various radionuclides that may be present in solid materials from nuclear facilities decommissioning.

- DOE

DOE issued Ordinance 5400.5 defines the standards for radiation protection of pubblic and environment. Very interesting is Chapter IV of the Ordinance, “Residual Radioactive Material”, which defines the requirements and guidelines for the management and release of contaminated material. The document incorporates the values of the table in regulatory Guide 1.86 release levels for surface contamination.

Subsequently, the DOE published an implementation guide, DOE G 441.1-XX [19], control and release of contaminated material in compliance with DOE 5400.5.

- EPA

EPA has the responsibility to establish radiation protection standards, which must meet USNRC and DOE Standards. EPA also published a guide on the levels of residual contamination allowed: “Comprehensive Environmental Response, Compensation and Liability Act (CERCLA)” (EPA, 1997b), applying a dose constrain of 150 microSv.

Among the technical documentation, it must also remember the American Standard ANSI/HPS N.13.12-1999. The document contains the values for contaminated materials release for the dose constrain of 10 microSv/y. Radionuclides are grouped by similar factor and assigned to dose levels in a range of 0.1 – 100 Bq/cm^2 (or Bq/g). The dose factors used to calculate the concentrations are similar to the IAEA factors.

4. DEFINITION OF THE CLERANCE LEVELS

Referring to described documents, it is possible to define the clearance levels for:

1) soil release;
2) land release.

In the first case the reference documents are: RP122-part. I, RP113, German norm, American norm and RS-G-1.7. In the last case the reference documents are: the German norm and the NRPB-W36.

The Radiation Protection criterion used for both situations is expressed by the no radiological importance: Individual Effective Dose is minor to 10 microSv/year for the person of the critical group of the population.

The clearance levels are defined for every radionuclide considering the contribution of the short time daughters and, therefore, for these last ones, they have not been defined specific limits. Table 2 resumes radionuclides for which the daughters contributions are added up the fathers radionuclides contribution.

Symbol “+” indicates that the derived clearance levels also includes daughter nuclides.

The daughters not in equilibrium have to be considered for some alpha emitters in the event currencies that the decay times are such to produce not negligible amounts. In particular this is valid for the parents of the three natural radioactive chains: Th-232, U-238 and U-235.

In Figure 1 the relative activities of the daughters activities with reference to the parents are presented to the 10th, 20th, 50th, and 100th year. The analysis of Figure 1 and Table 1 suggests that:

- for U-235 they could be considered all the daughters of the chain as after 100 years the concentrations of the Pa-231 and daughter turn out to be advanced to 1/1000 regarding that of the U-235; the levels would defined for U-235, Pa-231 and Ac-227;
- for the chain of the U-238 after a time of 100 years the concentrations of the Ra-226 and relative daughters turn out to be approximately 9 inferior orders of magnitude regarding the concentration in activity of the U-238, for which the daughters till the U-234 can themselves be taken in consideration; the levels would defined for U-238 and U-234;
- for the chain of the Th-232 the equilibrium with the daughters is caught up after approximately 50 years and in this case they would considered all the daughters of the same chain; the levels would defined for Th-232, Ra-228 and Th-228.

Parent	Progeny included in secular equilibrium with the parents
	Chains Th-232
^{232}Th	
^{228}Ra	^{228}Ac
^{228}Th	^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, ^{208}Tl, ^{212}Po
	Chains U-238
^{238}U	^{234}Th, ^{234m}Pa, ^{234}Pa
^{234}U	
^{230}Th	
^{226}Ra	^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po
^{210}Pb	^{210}Bi
^{210}Po	
	Chains U-235
^{235}U	^{231}Th
^{231}Pa	
^{227}Ac	^{227}Th, ^{223}Fr, ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Pb, ^{211}Bi, ^{207}Tl, ^{211}Po

Table 1 Natural Radioactive Chains

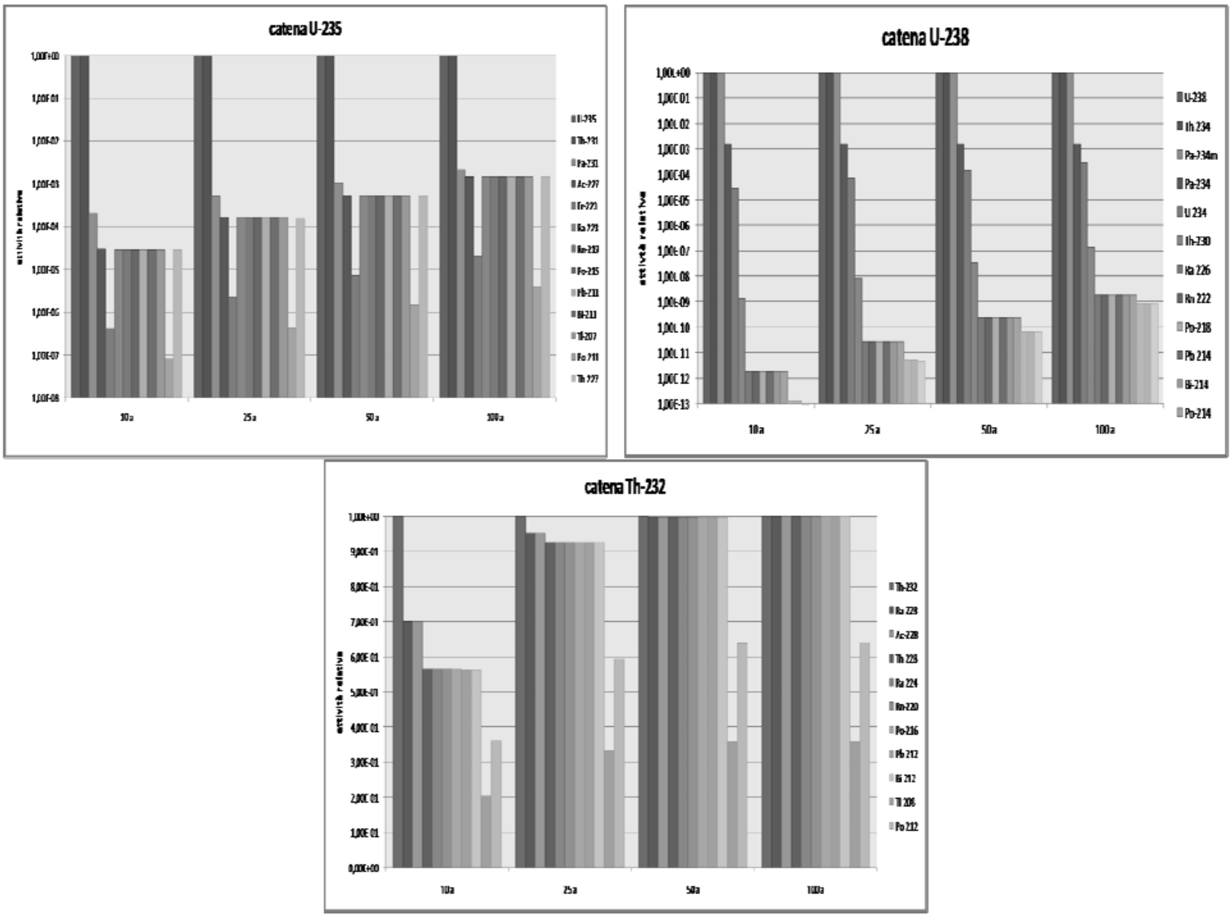

Figure 1 Relative activities of the daughters nuclides - Natural radioactive Chains (235U, 238U and 232Th)

Soil release

Proposals of soil release levels (Table 4) have been assumed equal to the lowest values between those defined in documents RS-G-1.7, RP122 part.I, RP 113, German norm , ANSI/HPS N.13.12-1999, NUREG-1640.

The values derived from the German norm and from the NUREG-1640, have been approximated with the criteria used in the RPs and the RS-G-1.7: if the calculated value is between $3 \cdot 10^{x}$ and $3 \cdot 10^{x+1}$, then it is rounded off to the value of 10^{x+1}.

The values of mass concentration turned out advanced to 1 Bq/g, in conformity with the levels of exemption of the Italian legislation, have been fixed to 1 Bq/g and have been indicated with the symbol "o" in Table 4.

Land release

Proposals of land release levels (Table 7), have been assumed like those more little ones between those derived from the publication NRPB-W36 and from the German norm, evaluating the values with the criteria used in the RPs and the RS-G-1.7, if the calculated value is between $3 \cdot 10^{x}$ and $3 \cdot 10^{x+1}$ then it is rounded off to the value of 10^{x+1}.

In Table 5 the clearance levels of the German norm and those calculated using the coefficients (Sv/year)/(Bq/g) from publication NRPB-W36, basing on a constrain of 10 dose of μSv/year. Table 6 presents the dose coefficients proposed in NRPB-W36 and the scenarios of exposure from which they have been assumed. The values of mass concentration higher to 1 Bq/g, for being conformed to exemption levels of the Italian legislation, are fixed to 1 Bq/g and are indicated with the symbol "o" in Table 7.

The proposed limits represent reference values and they are calculated in a conservative way and independently from the characteristics of every site.

Such limits can be reviewed in function of:

- the final destination of the site after the dismantling operations;
- the physical, geological and hydro geological of the site;
- the reference scenarios and the ways of exposure of the critical groups of the site.

5. CRITERIA FOR APPLICATION OF THE CLERANCE LEVELS

Soil

Applying the proposed limits (Table 4), they must be respected the following items:

- The levels are defined in terms of mass activity concentration (Bq/g) and they are applied to the total activity for mass unit. Notice that it's considered the total mass to remove. The levels are defined as average value on modest amounts (till some hundred of kilograms). For amounts of soil greater to 1000 ton/year, 1 ton is mass of material on which it is concurred to calculate the average value;
- Generally it is been involved more than a radionuclide. In order to determine if the concentration of a mixture of radionuclides is lower to the levels of removal it can be used the formula:

$$\sum_{i=1}^{n} \frac{C_i}{C_{Li}} < 1.0$$

  where:

  C_i is the activity concentration of the radionuclide *i* in the material (Bq/g);

  C_{Li} is the removal level of the radionuclide *i* in the material (Bq/g).
- They can be ignored the radionuclides that do not derive from the practice.
- The application of the levels of Table 4 isn't a constrain on the yearly amount of removed material.

Land

For applying the proposed limits (Table 7), these items are to be considered:

- The areas for the radiological control can be thus subdivided:
  - Areas of class 1: zones of the site that, on the base of the typical site's activities, are potentially contaminated: values are higher to the release limits;
  - Areas of class 2: premises and zones of the site in which, as a result of appropriate measures, were found values lower to the LODR;

- Areas of class 3: areas in which one previews not you are some residual radioactivity, or that they can introduce levels of residual radioactivity with equal values to some fraction of the LODR, on the base of the operating history and/or specific controls.

- The largeness of the "areas of measure" depends from its belongings to a determined class. The maximum extensions suggested for every area of measure are:
  Class 1: 2.000 m^2;
  Class 2: in a range of 2.000 – 10.000 m^2;
  Class 3: whitout limits.

Radionuclides	Radionuclides daughters
^{3}H	
^{14}C	
^{54}Mn	
^{55}Fe	
^{59}Ni	
^{63}Ni	
^{60}Co	
^{90}Sr	^{90}Y
^{93}Zr	
^{125}Sb	^{125m}Te
^{134}Cs	
^{137}Cs	^{137m}Ba
^{151}Sm	
^{152}Eu	
^{154}Eu	
^{155}Eu	
^{227}Ac	^{227}Th, ^{223}Fr, ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Pb, ^{211}Bi, ^{207}Tl, ^{211}Po
^{228}Th	^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, ^{208}Tl, ^{212}Po
^{231}Pa	
^{232}Th	
^{228}Ra	^{228}Ac
^{232}U	
^{233}U	
^{234}U	
^{235}U	^{231}Th
^{236}U	
^{238}U	^{234}Th, ^{234m}Pa, ^{234}Pa
^{237}Np	^{233}Pa
^{238}Pu	
^{239}Pu	
^{240}Pu	
^{241}Pu	
^{242}Pu	
^{241}Am	
^{243}Am	^{239}Np
^{242}Cm	
^{244}Cm	

Table 2 Considered radionuclides for clearance levels

Radionuclides	Activity concentration (Bq/g)						
	RS-G-1.7	*RP113*	*RP122*	*German norm*		*ANSI/HPS N.13.12-1999*	*NUREG 1640 (concrete materials)*
				Solids and Liquids	Concrete, excavation soil, etc (> 1000 ton/anno)		
^{3}H	100	100	100	1000	60	100	100
^{14}C	1	100	10	80	10	100	100
^{54}Mn	0.1	0.1	0.1	0.4	0.3	1	0.1
^{55}Fe	1000	1000	100	200	200	100	1000
^{59}Ni	100	1000	100	800	800	10	10000
^{63}Ni	100	1000	100	300	300	100	10000
^{60}Co	0.1	0.1	0.1	0.1	0.09	1	0.1
$^{90}Sr+$	1	1	1	2	2	1	10
^{93}Zr	10	100	10	10	10	--	1000
$^{125}Sb+$	0.1	1	1	0.8	0.5	--	0.1
^{134}Cs	0.1	0.1	0.1	0.2	0.1	1	0.1
$^{137}Cs+$	0.1	1	1	0.5	0.4	1	0.1
^{151}Sm	1000	1000	100	500	500	--	10000
^{152}Eu	0.1	0.1	0.1	0.2	0.2	1	0.1
^{154}Eu	0.1	0.1	0.1	0.2	0.2	--	0.1
^{155}Eu	1	10	10	30	8	--	10
$^{227}Ac+$	--	--	0.01	--	--	--	0.1
$^{228}Th+$	--	0.1	0.1	0.1	0.07	0.1	0.1
^{231}Pa	--	0.1	0.01	0.007	0.004		1
^{232}Th	--	0.1	0.01	0.03	0.03	0.1	0.1
$^{228}Ra+$	--	0.1	0.01	0.07	0.1	0.1	0.1
^{232}U	0.1	0.1	0.1	0.06	0.05	--	1
^{233}U	1	1	1	0.4	0.3	--	1
^{234}U	--	1	1	0.5	0.4	1	1
$^{235}U+$	--	1	1	0.5	0.3	1	1
^{236}U	10	1	1	0.5	0.4	--	1
$^{238}U+$	--	1	1	0.6	0.4	1	1
$^{237}Np+$	1	0.1	0.1	0.09	0.2	0.1	0.1
^{238}Pu	0.1	0.1	0.1	0.04	0.08	--	1
^{239}Pu	0.1	0.1	0.1	0.04	0.08	0.1	1
^{240}Pu	0.1	0.1	0.1	0.04	0.08	0.1	1
^{241}Pu	10	1	1	2	2	10	100
^{242}Pu	0.1	0.1	0.1	0.04	0.04	--	1
^{241}Am	0.1	0.1	0.1	0.05	0.05	0.1	1
$^{243}Am+$	0.1	0.1	0.1	0.05	0.09	--	1
^{242}Cm	10	1	1	0.8	0.7	--	10
^{244}Cm	1	0.1	0.1	0.08	0.08	0.1	1

Table 3 Soil clearance levels comparison

Radionuclides	Activity concentration [1] (Bq/g)
^{3}H	1º
^{14}C	1
^{54}Mn	0.1
^{55}Fe	1º
^{59}Ni	1º
^{63}Ni	1º
^{60}Co	0.1
^{90}Sr+	1
^{93}Zr	1º
^{125}Sb+	0.1
^{134}Cs	0.1
^{137}Cs+	0.1
^{151}Sm	1º
^{152}Eu	0.1
^{154}Eu	0.1
^{155}Eu	1
^{227}Ac+	0.01
^{228}Th+	0.1
^{231}Pa	0.01
^{232}Th	0.01
^{228}Ra+	0.01
^{232}U	0.1
^{233}U	0.1
^{234}U	1
^{235}U+	0.1
^{236}U	1
^{238}U+	1
^{237}Np+	0.1
^{238}Pu	0.1
^{239}Pu	0.1
^{240}Pu	0.1
^{241}Pu	1
^{242}Pu	0.1
^{241}Am	0.1
^{243}Am+	0.1
^{242}Cm	1
^{244}Cm	0.1

Table 4 Soil clearance level

Radionuclides	Activity concentration (Bq/gr)	
	NRPB-W36[2]	German Norm
^{3}H	667	3
^{14}C	--	0.04
^{54}Mn	--	0.09
^{55}Fe	185	6
^{59}Ni	--	8
^{63}Ni	26	3
^{60}Co	0.009	0.03
^{90}Sr+	0.01	0.002
^{93}Zr	--	20
^{125}Sb+	--	0.08
^{134}Cs	0.016	0.05
^{137}Cs+	0.04	0.06
^{151}Sm	108	40
^{152}Eu	--	0.07
^{154}Eu	0.019	0.06
^{155}Eu		2
^{227}Ac+	0.007	--
^{228}Th+	0.013	--
^{231}Pa	0.003	--
^{232}Th	--	--
^{228}Ra+	0.007	--
^{232}U	--	--
^{233}U	1.1	--
^{234}U	1.1	--
^{235}U+	0.19	--
^{236}U	1.17	--
^{238}U+	0.56	--
^{237}Np+	0.08	--
^{238}Pu	0.09	0.06
^{239}Pu	0.08	0.04
^{240}Pu	0.08	0.04
^{241}Pu	4.8	4.00
^{242}Pu	--	0.04
^{241}Am	0.1	0.06
^{243}Am+	--	0.05
^{242}Cm	--	0.40
^{244}Cm	0.16	0.08

Table 5 Land clearance level comparison - German norm vs. NRPB-W36

[1] The symbol "o" identifies radionuclides that have values higher to 1Bq/g.

[2] The levels are calculated through the (Sv/y)/(Bq/g) of the NRPB-W36 for an effective dose of 10 μ Sv/y.

Radionuclides	**(Sv/y)/(Bq/g)** NRPB-W36	**Scenarios**	**Group reference**
^{3}H	1.50E-08	agriculture	infants
^{14}C	--	--	--
^{54}Mn	--	--	--
^{55}Fe	5.40E-08	agriculture	infants
^{59}Ni			
^{63}Ni	3.86E-07	agriculture	infants
^{60}Co	1.11E-03	building	adults
$^{90}Sr+$	9.04E-04	agriculture	children
^{93}Zr	--	--	--
$^{125}Sb+$	--	--	--
^{134}Cs	6.34E-04	building	adults
$^{137}Cs+$	2.41E-04	building	adults
^{151}Sm	9.22E-08	agriculture	infants
^{152}Eu	--	--	--
^{154}Eu	5.19E-04	building	
^{155}Eu	--	--	--
$^{227}Ac+$	1.44E-03	building	
$^{228}Th+$	7.78E-04	building	adults
^{231}Pa	3.76E-03	agriculture	adults
^{232}Th	--	--	--
$^{228}Ra+$	1.37E-03	agriculture	children
^{232}U	--	--	--
^{233}U	9.35E-06	agriculture	adults
^{234}U	8.89E-06	agriculture	adults
$^{235}U+$	5.28E-05		
^{236}U	8.57E-06	agriculture	adults
$^{238}U+$	1.79E-05	buildinging	adults
$^{237}Np+$	1.23E-04	buildinging	adults
^{238}Pu	1.07E-04	buildinging	adults
^{239}Pu	1.17E-04	buildinging	adults
^{240}Pu	1.17E-04	buildinging	adults
^{241}Pu	2.10E-06	buildinging	adults
^{242}Pu	--	--	--
^{241}Am	1.01E-04	building	adults
$^{243}Am+$	--	--	--
^{242}Cm	--	--	--
^{244}Cm	6.30E-05	building	adults

Table 6 Dose for unit of concentration in the land for the scene predominated and critical group of publication NRPB-W36

Radionuclides	**Activity concentration**[3] *(Bq/g)*
^{3}H	1º
^{14}C	0.1
^{54}Mn	0.1
^{55}Fe	1 º
^{59}Ni	1 º
^{63}Ni	1º
^{60}Co	0.01
$^{90}Sr+$	0.001
^{93}Zr	1º
$^{125}Sb+$	0.1
^{134}Cs	0.01
$^{137}Cs+$	0.1
^{151}Sm	1 º
^{152}Eu	0.1
^{154}Eu	0.01
^{155}Eu	1
$^{227}Ac+$	0.01
$^{228}Th+$	0.01
^{231}Pa	0.001
^{232}Th	0.001
$^{228}Ra+$	0.01
^{232}U	0.1
^{233}U	1
^{234}U	1
$^{235}U+$	0.1
^{236}U	1
$^{238}U+$	1
$^{237}Np+$	0.1
^{238}Pu	0.1
^{239}Pu	0.1
^{240}Pu	0.1
^{241}Pu	1 º
^{242}Pu	0.1
^{241}Am	0.1
$^{243}Am+$	0.1
^{242}Cm	1
^{244}Cm	0.1

Table 7 Land clearance level

[3] The symbol "or" the radionuclides is indicated with for which they have been proceeds advanced values to 1 Bq/g.

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