Radiological Modeling for Determination of Derived Concentration Levels of an Area with Uranium Residual Material – 13533

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ABSTRACT

As a result of a pilot project developed at the old Spanish "Junta de Energía Nuclear" to extract uranium from ores, tailings materials were generated. Most of these residual materials were sent back to different uranium mines, but a small amount of it was mixed with conventional building materials and deposited near the old plant until the surrounding ground was flattened. The affected land is included in an area under institutional control and used as recreational area. At the time of processing, uranium isotopes were separated but other radionuclides of the uranium decay series as Th-230, Ra-226 and daughters remain in the residue. Recently, the analyses of samples taken at different ground's depths confirmed their presence. This paper presents the methodology used to calculate the derived concentration level to ensure that the reference dose level of 0.1 mSv y-1 used as radiological criteria. In this study, a radiological impact assessment was performed modeling the area as recreational scenario. The modelization study was carried out with the code RESRAD considering as exposure pathways, external irradiation, inadvertent ingestion of soil, inhalation of resuspended particles, and inhalation of radon (Rn-222). As result was concluded that, if the concentration of Ra-226 in the first 15 cm of soil is lower than, 0.34 Bq g-1, the dose would not exceed the reference dose. Applying this value as a derived concentration level and comparing with the results of measurements on the ground, some areas with a concentration of activity slightly higher than latter were found. In these zones the remediation proposal has been to cover with a layer of 15 cm of clean material. This action represents a reduction of 85% of the dose and ensures compliance with the reference dose.

INTRODUCTION

The CIEMAT is developing since 2000 a plan to improve its facilities, in order to recover the infrastructure for conventional uses, giving up its status as nuclear facility. The scope of this project is broad and one of its tasks is the remediation of land with uranium residual materials. This is the case of an affected land area where, as a result of a past project to extract uranium from ores, a small amount of residual tailings materials mixed with conventional building residues were deposited to level the surrounding ground. This area is under institutional control and is used as recreational zone for the public.

The radiation protection criteria to implement at the CIEMAT restoration program (PIMIC) have been analyzed by an expert group and subsequently summarized together with the implementable methodology in a project report [1]. This includes an analysis of national and international references on basic radiological criteria expressed in terms of reference levels of annual radiation doses.

Referring to that, there are several international recommendations from the International Atomic Energy Agency and from the International Commission on Radiological Protection in its publication ICRP-82 [2]. In both cases, dose of the order of a few mSv per year are considered as acceptable, and particularly in the ICRP-82 is established as difficult to justify intervening in situations where the total dose (including the natural background) are under a Reference Generic Level of 10 mSv per year. Obviously, the authorities may choose different values depending on their own situations. Actually, in practice, relevant authorities for European countries (include Spain), have decided as values for various "de facto" situations in the range of reference dose of 0.1 to 1 mSv per year. It is important to point out that the Spanish regulatory body (CSN) has decided for PIMIC, in the year 2002, a reference dose value of 0.1 mSv per year as "radiological approach" to implement, indicating that higher values would be acceptable in situations arising from the past which must be justified with an optimization study approved by the CSN [3].

The establishment of Derived Concentration levels (DCLs) has become an important part of remedial actions at contaminated facilities. Any media with radioactivity levels in excess of the DCLs would require remediation to or below the DCL prior to release of the site for unrestricted use. The main premise in the derivation of this concentration levels is to ensure that the site will not represent a threat to human health and the environment after remediation has been completed. These levels must not only protect the human and the environmental but must also be acceptable to regulators, local authorities, and nearby communities responsible for overseeing the restoration. Also, it is a starting point to guide the technology selection to remediation.

The determination of DCLs to assess the potential impact of radionuclides on general public requires the scenario definition take into account the major pathways trough which a contaminant may become available to a receptor. A scenario can consider a group of radionuclides, the different pathways that may lead to irradiation of individuals, site characteristics and the use made of it. The development of the scenarios is based in modeling of migration and dispersion of different radionuclides as basis for the remediation, which ultimate goal is to achieve acceptable levels of concentration of residual activity.

In this study the disposal of uranium residual materials in a specific area was modeled in order to evaluate the potential radiological dose to general public and to determine the site specific derived concentration level (DCLs) in soil according to the radiation protection criteria applied. The dose modeling approach used to develop site specific DCLs for the CIEMAT site are described below. The scenario, and applicable exposure pathways used to develop site specific DCLs, along the assign parameters values and computer code selected to represent the dose models are described. Dose modeling results and the calculate DCLs are discussed.

DOSE ASSESSMENT METHODOLOGY

Radiological Site Characterization

For a dose assessment, it is important to determine the present radionuclides, as well as relations of activity among them. To accomplish the radiological characterization of the site, several surface and drilling samples were taken and the Laboratory for Radiological Protection Measures (LMR) at CIEMAT made their radioactivity analysis. As a result of this characterization, it was observed that the naturally occurring radionuclides that were deposited in the site were essentially: Ra-226 and its descendants, Th-230 and a lower concentration of Uranium (U-238, U-235 and U-234) and its progeny. According to these results of the analyzed samples from the characterization [4], it is concluded that the origin of the contamination of the site came from mining and tailing material.

As a result of the extraction of Uranium from raw material, it can be assumed that only a 10% of the initial activity concentration of natural Uranium remains in the tailing material. However, for the Uranium sub-series with a precursor of long half-life period, the same activity concentration and the radioactive equilibrium with their descendants initially present in the ore material is considered.

The Th-230 presents a special situation because it is a radionuclide with alpha radiation emissions. To estimate the dose it is assumed that it is in equal proportion with its descendent Ra-226. This consideration corresponds with some analytical results of surface samples of the area as reported by the Laboratory for Radiological Protection Measures at CIEMAT [5]. The Ra-226 concentrations in deposited materials are highly variable, ranging from near background levels to several hundred thousand of Bq g^{-1}. Besides, several samples from the vicinity of the contaminated area were analyzed in order to determine the average background concentration of natural radionuclides in soil.

Scenario and Exposure Pathways

For this study, the disposal of uranium residual materials in a zone was modeled to evaluate potential dose for the public for a recreational scenario and dose calculations were conducted for the maximally exposed receptor. The principal exposure pathways considered for the public that visit the recreational was: external irradiation, inhalation of particles due to resuspension of soil, inadvertent ingestion of soil particles and inhalation of Rn-222 and descendents.

Knowing the restrictive use of the parcel as recreational scenario, have been made some queries regarding the spend time for recreational activities that take place there. On a conservative way, take in to account a person who could make more intensive use of the parcel, and estimated length of stay of 20 hours per week throughout the year have been considered. It time represent a visitor who spends a time 1040 hours per year (12% of the hours of the year). It is understandable that, in the summer season the spend time will be higher than winter, but on average means about 8 hours every weekend and about 4 hours extra each week. The exposure time of 1040 hours per year is reasonable, considering also that, recreational opportunities would not include fishing and swimming activities, because the area is limited in size and does not locate in the vicinity of surface water.

Input Parameters and Dose Model Computer Code

Dose models are necessary to derive levels of residual radioactivity in the soil at the time of site release that would result in a 0.1 mSv y^{-1} to the public. Radiological doses were modeled by using the RESRAD computer code, Version 6.3. The RESRAD code is a pathways analysis code that implements the methodology for determining concentrations of residual radioactivity in soil. The exposure pathways available for analysis included external radiation; inhalation of resuspended dust and Rn-222; ingestion of crops, milk, and meat grown on the contaminated property; incidental ingestion of contaminated soil; ingestion of fish from a nearby pond; and ingestion of surface water or groundwater. Doses were projected over a period of 30 years following initial residual materials placement. This methodology allowed the assessment to account for radioactive decay and erosion of the cover material over time.

Inputs to the code were mostly based on the site specific data. They were obtained from the measurements made by CIEMAT and from the reports made available. Any unavailable local data were adopted from default values recommended by RESRAD. These default values were assessed and chosen to be the most realistic for the conditions on site. However, as a normal practice in impact assessment, the values were chosen in such a way that use of these values in any situation would not result in underestimation of the dose. It has been decided then that the parameters used RESRAD default characterize enough and even with some degree of conservatism that is with certain safety margins the situation in the CIEMAT area.

Listed below Table I, some of the parameters with their values for their importance in exposure pathways considered in this scenario. Calculations were based on a 26157 m^2 area of land; contamination was assumed to be distributed homogeneously throughout the contaminated zone. The thickness of the contaminated zone was assumed to be 15 cm.

The definition and justification of all parameters as well as dose conversion factors for different pathways of radiation are detailed in references [6, 7].

Table I. Input parameters for calculating the DCLs for Recreational Scenario.

Parameter	Value
Non adverted Ingestion of soil	100 mg d^{-1}
Dust load in air	100 µg m-3
Inhalation rate	8400 m3 y^{-1}
Exposure time	30 years
Precipitation rate	0.436 m y^{-1}
Occupancy factor	12 % annual
Density of soil	1.4 g cm^{-3}

RESULTS

Variation of Dose with Contamination Depth

In order to evaluate the dose variation with the tailing materials depth, it has been assumed that there is a uniform vein of 15 cm of thickness throughout the area of the contaminated site. Besides, it has been considered a concentration of activity of 1 Bq g^{-1} of ^{226}Ra in equilibrium with their descendants and equal to its precursor, ^{230}Th. The concentration of activity for the ^{238}U and ^{234}U is, 0.1 Bq g^{-1}, for the ^{235}U and their descendants a concentration 0.005 Bq g^{-1} is considered. The results obtained with these values as the source term are shown in Figure 1.

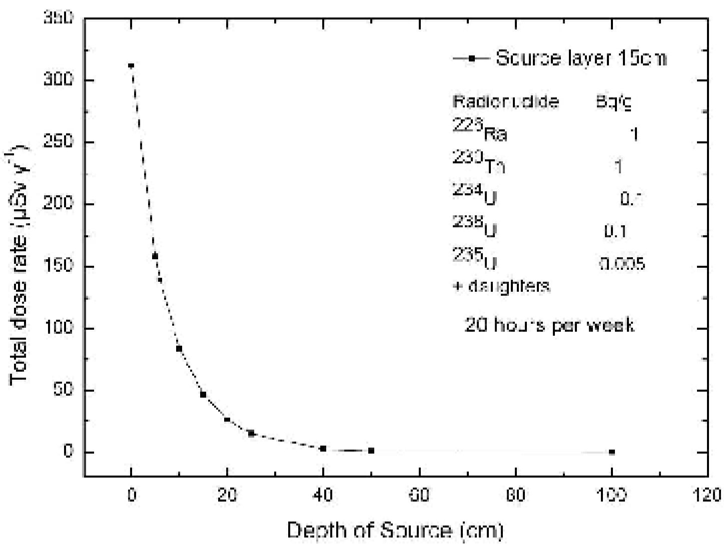

Fig. 1. Changes in the Total dose rate with contamination depth for a vein of 15cm thickness.

This graph shows that with soil coverage of 10 cm or more, the obtained dose is lower than the reference dose for all pathways of exposure. This mean that, if there is no higher concentration than natural background in surface soil, this coverage thickness would be enough to shield the additional radiation caused for the presence of radionuclides in deeper areas.

Figure 2 shows the results, expressed as annual doses as a function of time, considering the whole area with a uniform surface contamination in the first 15 cm of soil, with similar concentrations to those used in the exercise of variation of dose rate with depth. The obtained results for the whole area showed that the external gamma irradiation produces the greatest contribution to the dose and the responsible of that is the ^{226}Ra with its descendants.

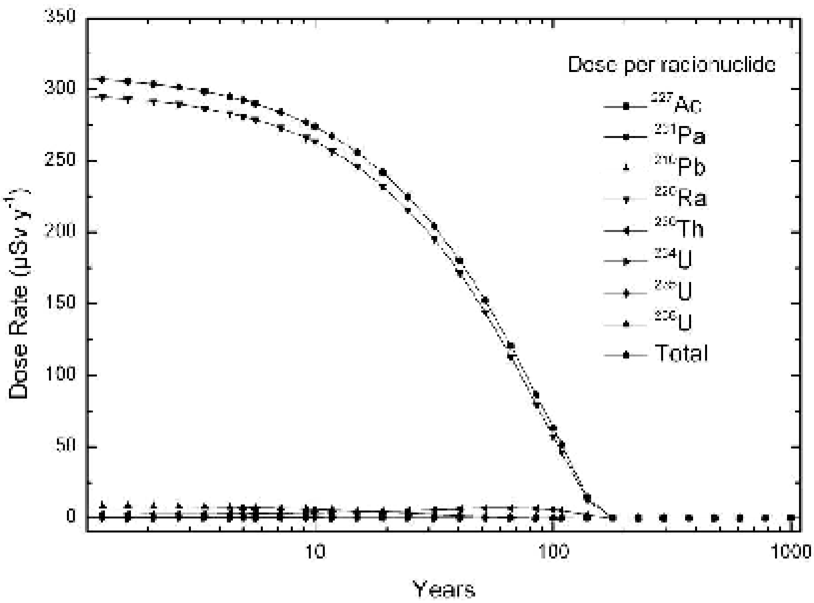

Fig. 2. Variation of annual dose rate as a function of time for radionuclides present in soil.

The external gamma dose rate for a continuous vein, located from the surface soil that increases its thickness in depth and contains 1 Bq g^{-1}of Ra-226 and its descendants is shown in Figure 3. The graph shows that, for depths greater than 40 cm approximately, the presence of tailing materials not affects the external gamma dose received at the surface. Also, assuming natural land coverage, the resulting dose would be, substantially lower, decreasing by a factor of about six.

Table II, presented bellow includes the values obtained of annual doses at a concentration of activity in the surface area of 1 Bq g^{-1} Ra-226, its descendants and the radionuclides listed above at the initial current time (year one in Fig. 2).

The effect of the coverage layer in the reduction of the gamma external dose can be observed, as well as the lack of influence of the ingestion and inhalation pathways. In other words, the influence of gamma external dose due to Ra-226 and its descendants is determining if the topsoil does not contain additional contamination to natural background.

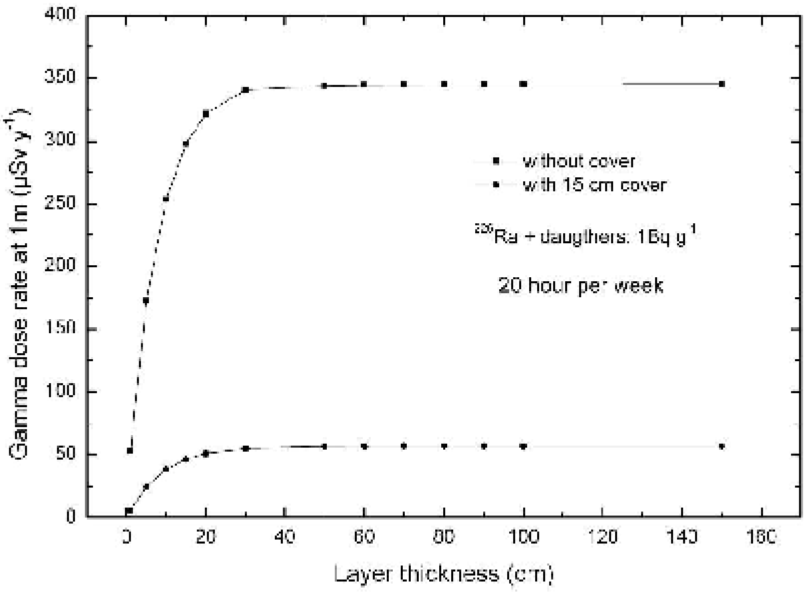

Fig. 3. Gamma dose rate variation, at 1 m of soil, for a continuous source layer of 15 cm with concentration of 1 Bq g^{-1} of Ra-226 at different depths, with and without clean soil cover.

Table II. Calculate doses for CIEMAT site with and without clean soil cover.

Exposure Pathways	Without cover		With cover	
	Dose Rate ($\mu Sv\ y^{-1}$)	Fraction (%)	Dose Rate ($\mu Sv\ y^{-1}$)	Fraction (%)
Gamma External Radiation	298.9	95.73	46.68	99.65
Soil Ingestion	11.0	3.52	0	0
Inhalation	2.15	0.69	0	0
Radon Inhalation	0.17	0.05	0.17	0.35
Total Dose	312.2	100	46.84	100

Calculation of Derived Concentrations Levels

Knowing the irradiation pathways, the involved radionuclides and the reference dose criteria considered acceptable from radiation protection point of view (0.1 mSv per year), it is possible to derive the concentration values that can be acceptable in the topsoil.

The soil concentrations for each radionuclide to ensure that individually, do not provide an exceeding dose of 0.1 mSv per year was obtained by a modeling assessment and the obtained DCLs values are shown in Table III.

These values can be used as departure values for calculation of derived guidelines concentration levels to apply the MARSSIM methodology [8]. This methodology has been recommended to implement the release of radiological contaminated sites. That methodology has been developed through a consensus among the regulatory agencies in U.S., mentioned as a methodology for its possible application to CIEMAT restoration program and also prompted by the CSN.

Table III. Calculate Derive Concentration Levels from radionuclides present in CIEMAT site.

Radionuclide	DCLs (Bq g^{-1})
Ra-226	0.34
Th-230	41.50
U-238	19.52
U-234	99.14
U-235	4.24

It should be noted that it is applied to surface contamination in the first 15cm and does not include scenarios derived from depth contamination. It is estimated that the derivate concentration values, in the absence of influence of the deeper areas, can be acceptable to consider the recreational use of the parcel, fulfilling the requirement of a reference dose of 0.1 mSv per year.

CONCLUSIONS

The involved radionuclides in some measurements of higher values than natural background are typical of tailing material once the uranium has been extracted, and they represent natural series, also present in all types of soils. The external gamma exposure due to Ra-226 and its descendants is considerably predominant. All depths greater than 50 cm, the presence of materials containing Ra-226 and its descendant has no influence on the external gamma dose at the surface, due to the shield provided by the soil.

This study recommends the use of the value of 0.1 mSv per year, determined by the regulatory body (CSN) as radiological criteria for the release of contaminated soil sites at CIEMAT. In this case, it is intended for use as recreational area.

The derived radionuclides concentrations value (DCLs) in the soil to ensure this radiological criteria fulfillment has been deduced. It is important to highlight that this is not and unconditional release of the area. Applying this value as a derived concentration level and comparing with the results of measurements on the ground, some areas with a concentration of activity slightly higher than latter were found. In these zones the remediation proposal has been to cover with a layer of 15 cm of clean material. This action represents a reduction of 85% of the dose and ensures compliance with the reference dose.

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