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## Article

# Comparison of Different Radiological Risk Assessment Scenarios at a Coal Ash and Slag Disposal Site

Ana Getaldić <sup>1,\*</sup>, Marija Surić Mihić <sup>2</sup>, Želimir Veinović <sup>1</sup>, Božena Skoko <sup>3</sup>, Branko Petrinc <sup>4</sup>  
and Ivica Prlić <sup>4</sup>

<sup>1</sup> Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb, Pierottijeva 6, p.p. 390, HR-10000 Zagreb, Croatia; zelimir.veinovic@rgn.hr

<sup>2</sup> Civil Protection Directorate, Ministry of the Interior, Nehajska 5, HR-10000 Zagreb, Croatia; msuricmihic@mup.hr

<sup>3</sup> Ruđer Bošković Institute, Bijenička cesta 54, HR-10000 Zagreb, Croatia; bskoko@irb.hr

<sup>4</sup> Institute for Medical Research and Occupational Health, Ksaverska cesta 2, p.p. 291, HR-10001 Zagreb, Croatia; petrinc@imi.hr (B.P.); iprlc@imi.hr (I.P.)

\* Correspondence: agetaldic@rgn.hr

**Abstract:** Coal fly ash and slag waste residuals from coal combustion are an issue of importance as one of the possible sources of environmental contamination and exposure to NORM. This study compares the results of different radiological risk assessment scenarios targeting terrestrial biota at a legacy site in Croatia that contains large quantities of coal ash with an enhanced content of radionuclides originating from previous industrial activities. The ERICA assessment tool was used for a risk assessment, which included data from borehole samples with a maximum depth of 6 m and trees as the primary reference organisms. The results of the risk assessments from various depth ranges found the radiological risk to the reference organisms to be negligible, regardless of the depth range, since the screening dose rate of  $10 \mu\text{Gy h}^{-1}$  was not exceeded in any of the assessments. The risk assessment results from all depth ranges show higher total dose rate predictions when the tool's default CR values are used, compared to the site-specific ones, which is in agreement with previous studies on the application of the ERICA tool. A comparison of results from different spatial radiological risk assessments showed that sample depth does not affect the estimated total dose rate to biota.

**Keywords:** NORM; coal ash and slag; radiological risk assessment; ERICA tool; environmental protection



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## 1. Introduction

Coal combustion residue disposal is considered a major environmental issue due to its significant influence on the environment, financial constraints of residue management, and potential health risks related to disposal. One of the possible sources of exposure to naturally occurring radioactive materials (NORM) is coal fly ash and slag waste materials resulting from coal burning [1,2]. Consequently, the increase in world energy demands has resulted in an ongoing increase in generated residue quantities and the necessary development of responsible and efficient waste management strategies, including disposal options that offer possible revenue generation [3]. The usual methods of fly ash disposal, including wet and dry disposal, most often result in landfilling of the material, a disposal method which, given the environmental and health implications and costs, has been under scrutiny. Recent developments of sustainable design approaches in the mineral industry focus on post-utilization phases, including recovery of useful material and reconcentration, encouraging innovative solutions and integrative circular economy objectives [3–5].

The effects of coal burning affect the land use and aesthetics of the environment and present a potential source of health hazards and environmental danger to air, soil, and water [1,6,7]. The revegetation of disposal sites containing coal fly ash and slag is

important, not only for aesthetic purposes but also to prevent wind and water erosion of fly ash and to reduce water leaching through deposit layers [8]. In this context, radiological risk assessment findings are relevant in order to estimate not only the potential detrimental effect on the environment but also the potential for land reclamation and the effects of possible revegetation of coal fly ash and slag disposal sites.

Due to the potentially hazardous effect of radiation, the inclusion of non-human biota, i.e., the environment, in the radiation protection framework, presents one of the key changes in the field of radiation protection [9,10]. The crucial aspect of radiation protection is the assessment of the potential radiological impacts arising from the exposure of non-human biota to ionizing radiation. Currently, different approaches and models are being used in conducting assessments. These approaches differ in focusing on individuals or populations [11], radionuclide transfer in biota [12], use of activity concentrations [13], and available radiation effects data [14]. The existing assessment models include concentration ratios, kinetic models, compartment models, and allometry approaches [13,15]. One of the methods of performing a radiological risk assessment is using the ERICA approach and the ERICA tool, which were used in this study based on their availability and applicability to this specific research context. The ERICA approach addresses needs related to environmental exposure to ionizing radiation, including scientific, managerial, and social aspects of the assessment, while the ERICA tool covers the practical aspect of the assessment [16–19].

The ERICA tool is an impact assessment software with a three-tiered structure, available online free of charge, that combines data on environmental transfer and dosimetry to provide a measure of exposure, which is then compared to exposure levels associated with known detrimental effects of radiation [17,20–22]. Since the ERICA tool was developed as a part of the European Union co-funded 6th Framework Program EURATOM project “Environmental Risk from Ionising Contaminants Assessment and Management” (ERICA), it is especially applicable to European biota and has been used in Europe for risk assessments in various radiological risk assessment scenarios, including NORM-related industries and activities. The dose rate to biota received due to exposure to Cesium-137 was calculated using the ERICA tool in a study by Sotiropoulou et al. [23]. Babić et al. [24] performed dose rate assessments related to exposure of wildlife in a forest ecosystem using the ERICA tool. Vetikko and Saxén [25] used the ERICA tool for dose rate assessment in freshwater ecosystems in Finland. Aryanti et al. [26] used the ERICA tool to calculate dose rates in marine biota near a coal-fired power plant. A study from Čujić and Dragović [27] compared the results of dose rate assessments to terrestrial biota in the area around a coal-fired power plant using both the ERICA tool and RESRAD BIOTA assessment tool. Mrdakovic Popic et al. [28] evaluated the environmental impact at a NORM legacy mining site and used ERICA for dose rate assessments. Oughton et al. [29] used the ERICA tool for ecological risk assessment at several mining sites in Central Asia. Research from Vandenhove et al. [30] focused on the assessment of the potential radiological impact of the phosphate industry on wildlife.

Previous radiological studies conducted at the location include research of the chemical and radiological profile of the coal ash landfill [31], studies focused on research aspects of plant uptake of radionuclides from coal ash and slag [32], investigations of the radioactivity of the Mediterranean flora [33], a risk assessment of the legacy disposal site [34], research on natural and anthropogenic radionuclides in the karstic coastal part of the location [35,36], and an assessment of the environmental risk related to polycyclic aromatic hydrocarbons [37].

The main objective of this research was to study the potential effect of the depth of samples used in the radiological risk assessment on the actual risk assessment results. The assessment scenarios included samples collected at the same coal and ash disposal site from three boreholes with a maximum depth of 6 m. By determining the radiological risk in each of these scenarios, the potential effects of sampling depth can be closely studied in the context of specific radionuclides and reference organisms.

Findings from this study are expected to contribute to the future use of the ERICA tool in environmental risk assessments and facilitate the research design, selection of sampling methodologies, and comparison of different study results. An additional advantage of this study can be observed in this research addressing the practical context of the new ERICA tool version that enables complex spatial and temporal assessments, in this case, the vertical aspect of sample depth.

## 2. Materials and Methods

This study compares the results of radiological risk assessment scenarios targeting terrestrial biota at a legacy disposal NORM site in Croatia. This location contains large quantities of NORM originating from previous industrial activities, mainly from the coal-fired power plant used at the industrial complex.

### 2.1. Assessment Site

The researched NORM legacy disposal site is located in Kaštela Bay. The bay area is populated, and there are several cities in relatively close proximity to the disposal site, as well as the site being in contact with seawater. The disposal site is a part of a larger industrial complex, a remnant of a chemical factory that used a coal-powered thermo-electric unit to generate electricity for industrial activities. The remaining coal fly ash and slag were disposed of using the “wet method”, ending up in a settling basin [34]. As the material was disposed of during the 1980s and 1990s, the accumulated material in the basin comprises a much larger disposal site in the eastern part of the industrial complex. A detailed layout of the location is shown in Figure 1. During the industrial operation, various types of coal were used, namely, lignite, anthracite, and brown coal, originating from mining sites with increased natural radioactivity [31]. Since the site was not subjected to any treatment for more than a decade, spontaneous revegetation occurred, currently consisting of different species of Mediterranean terrestrial flora, providing a research opportunity to conduct studies and assessments in specific environmental conditions [32,34–37]. A previous study by Skoko et al. [33] focused on the radioactivity of the soil in Kaštela Bay and provided data on background activity concentrations based on samples from a control site which was not affected by previous industrial activities. The reported data includes average values and standard deviations of activity concentration for  $^{238}\text{U} = 53.5 \pm 23.8 \text{ Bqkg}^{-1}$ , activity concentration for  $^{226}\text{Ra} = 57.9 \pm 32.8 \text{ Bqkg}^{-1}$ , and activity concentration for  $^{232}\text{Th} = 47.1 \pm 19.7 \text{ Bqkg}^{-1}$  [33].

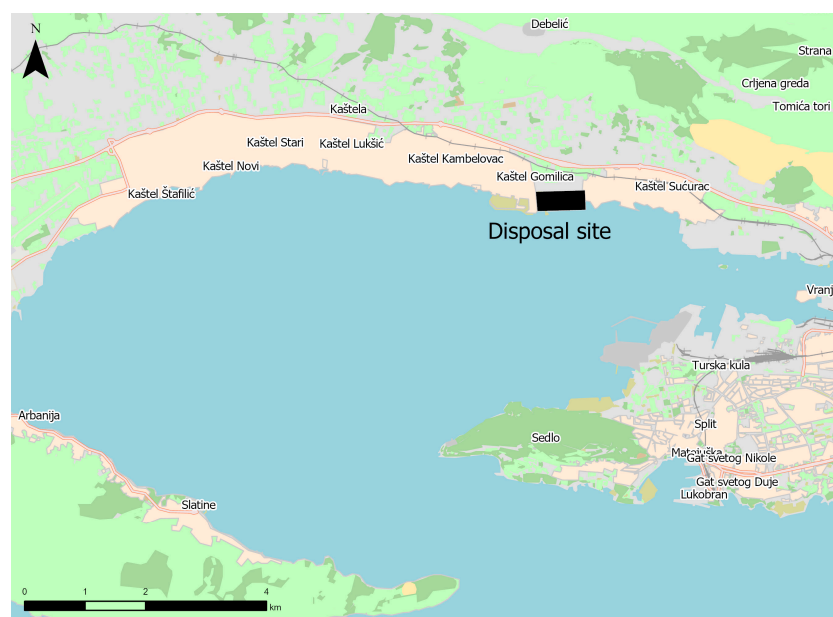


Figure 1. Site location.

## 2.2. ERICA Assessment Tool

The ERICA assessment tool (version 2.0) was used to calculate dose rates to terrestrial biota from exposure to radionuclides identified at the research site. The tool is available online at no charge and uses activity concentrations in the environmental media (sediment, soil, water, and air) as input data.

To estimate radionuclide transfer to biota, the ERICA tool uses concentration ratio (CR) values [16,17,21,22,38]. CRs are specific for each element and are defined by the ratio between activity concentrations of radionuclides in the biota (whole body) and activity concentrations in the environmental media (soil, water, and air) [16]. The ERICA tool assesses potential effects arising from both internal and external exposure to ionizing radiation by interpreting data on activity concentration in both media and biota through the use of dose conversion coefficients ( $DCC_{int}$  and  $DCC_{ext}$ ) in  $\mu\text{Gyh}^{-1}$  per  $\text{Bqkg}^{-1}$  fresh weight [16,21,22,39]. The ERICA tool relies on three (one for each ecosystem) radioecology-related databases to derive CRs and  $K_d$  values (distribution coefficients used for aquatic environments) [17,22]. In order to estimate the total absorbed dose rate, the ERICA tool uses weighting factors to address different components of radiation (low  $\beta$ ,  $\beta + \gamma$ , and  $\alpha$ ) [16,17]. The values used in this study are default values available in the ERICA tool: 10 for alpha, 3 for beta, and 1 for gamma radiation. The default screening dose rate in the ERICA tool is  $10 \mu\text{Gyh}^{-1}$  [17]. This value was chosen based on the analysis results available from the FRED effects database and is also in accordance with EC recommendations and more stringent than the value proposed by the US Department of Energy [16]. The tool allows users to select different screening dose rate values in tiers 1 and 2 of the assessment. Uncertainty factors are used to assure conservatism between tiers 1 and 2, whose values should correspond. Beresford et al. [16] define the uncertainty factor (UF) as “the ratio between the 95<sup>th</sup> and 99<sup>th</sup> percentile of risk quotient and the expected value of the probability distribution of the dose rate” [16,17]. Proposed values for UFs are 3 and 5, enabling the assessment for a 5% and 1% probability of exceeding the dose rate screening value, respectively [39]. These values were used in all assessment scenarios.

The ERICA tool tier 2 risk assessment also provides the risk quotient as an assessment output. The risk quotient (RQ) is a unitless value derived by comparing the selected assessment screening dose rate and the total estimated whole-body absorbed dose rate for each organism [17,39]. The tool also calculates a conservative risk quotient by multiplying the expected value of the RQ and uncertainty factor [16].

Both the ERICA tool's default list of radionuclides and the use of reference organisms as generalized ecosystem representations are in line with ICRP's propositions [9,40]. The use of ERICA in the context of planned or existing exposure situations applies to various scenarios, including decommissioning of a nuclear facility, radioactive waste disposal, remediation, NORM/TENORM, and clearance [16]. The newest version of the ERICA tool, used in this study, enables one to conduct the assessments by taking into account both daughter radionuclides, whose physical half-life is on the order of tens of days or less, and parent radionuclides. The assessment tool assumes that parent and daughter radionuclides in a particular decay chain are in secular equilibrium. Since the assessment results showed that the contribution of certain daughter radionuclides to the total dose rate was insignificant, a threshold of 1% of contribution to the total dose rate was established and only radionuclides contributing more than 1% to the total dose rate are included in results.

## 2.3. Assessment Input Data

The study used data on coal ash and slag samples from different depths from three boreholes (B2, B3, and B4) collected in a separate study in 2010 at the legacy site as input. The distance between boreholes was approximately 600 m. High-pressure drilling equipment with a pipe diameter of 11 cm was used for drilling. Samples were taken from several depths (0–2 m, 2–4 m, and 4–6 m) and packed in plastic bags. Although samples from deeper levels were available, samples from up to 6 m were selected based

on the rationale that for the reference organisms selected, i.e., trees, the majority of the root system is contained in the upper soil layers. For borehole B4, sampling at a depth range of 0–2 m was not conducted as the material mainly consisted of stones and deposited material transported from other locations. The radionuclide  $^{210}\text{Pb}$  was not detected in several samples from boreholes B3 and B4.

Laboratory preparation of the samples included drying at 105 °C. The dry sample masses were weighed and stored in 200 mL containers. The samples were measured in a gamma-spectrometric laboratory at the Radiation Protection Unit of the Institute for Medical Research and Occupational Health after 30 days, to ensure secular equilibrium. Radioactivity in samples was determined using a high-resolution gamma-spectrometry HP GMX ORTEC photon detector system with the following characteristics: a resolution of 2.2 keV at 1.33 MeV  $^{60}\text{Co}$  and a relative efficiency of 74.3% at 1.33 MeV  $^{60}\text{Co}$ . Activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{232}\text{Th}$  were determined from their decay products. Photopeaks at 609 keV, 1120 keV, and 1764 keV for  $^{214}\text{Bi}$  and 295 keV and 352 keV for  $^{214}\text{Pb}$  were used to determine the activity concentration of  $^{226}\text{Ra}$ , those of  $^{228}\text{Ac}$  at 338 keV, 911 keV, and 968 keV were used to determine the activity of  $^{232}\text{Th}$ , and photopeaks at 63 keV and doubled 93 keV for  $^{234}\text{Th}$  were used to determine the activity of  $^{238}\text{U}$ , where the activity of  $^{210}\text{Pb}$  was determined from its  $\gamma$ -ray photopeak at 46 keV [34]. The relative measurement uncertainty in the gamma-spectrometric measurements used to determine the soil activity concentrations in this study was below 10%. The measurement method was accredited in compliance with the HRN EN ISO/IEC 17025:2007 standard, and the efficiency calibration was carried out by the standards from the Czech Metrological Institute, covering the energy range from 40 to 2000 keV. The radionuclide determination quality assurance was conducted through participation in comparative measurements organized by the International Atomic Energy Agency (IAEA), the World Health Organization (WHO), and the European Commission Joint Research Centre (JRC) [41].

Site-specific CR values for this location were determined by Skoko et al. [32], and in the current study were used to compare site-specific assessment results with the results from an assessment using the tool's default CR values. A list of radionuclides used in the assessments is given in Table 1. The ERICA tool allows users to select between the tool's default reference organisms or generate specific ones. Given the fact that the location is in the Mediterranean climate and that revegetation of the location occurred, considering the depth aspect, trees were selected as the main reference organisms. Although the total sampling depth of the boreholes was up to 13 m, based on the data available in the literature [42] it was decided to use borehole sampling data up to 6 m, as the maximum rooting depth for Mediterranean flora was estimated to be not more than 5 m. The study by Canadell et al. [43] lists a maximum rooting depth of 5 m for *Pinus Pinea*. The data on average activity concentrations ( $\text{Bqkg}^{-1}$  dry mass) in three borehole samples (B2, B3, and B4) is presented in Table 2. For practical purposes, the available sampling data from different depths were grouped into three depth ranges (0–2 m, 2–4 m, and 4–6 m) and used in three separate risk assessment scenarios. Table 3 lists the tool's default and site-specific CR values from [32,34] used in the assessment scenarios.

**Table 1.** Assessment input data.

Ecosystem Type	Radionuclides	Reference Organism
Terrestrial	$^{238}\text{U}$	Tree
	$^{232}\text{Th}$	
	$^{235}\text{U}$	
	$^{226}\text{Ra}$	
	$^{210}\text{Pb}$	

**Table 2.** Activity concentrations (Bqkg<sup>-1</sup> dry mass) measured in borehole samples (B2, B3, and B4) from different depth ranges.

<b>B2</b>		<b>Activity Concentration (Bqkg<sup>-1</sup>)</b>			
	<sup>238</sup> U	<sup>232</sup> Th	<sup>235</sup> U *	<sup>226</sup> Ra	<sup>210</sup> Pb
<b>0–2 m</b>	1307 ± 203 **	36 ± 6 **	60	1065 ± 14 **	641 ± 13 **
<b>2–4 m</b>	1128	32	51	947	622
<b>4–6 m</b>	1265	47	58	1106	1951
<b>B3</b>		<b>Activity concentration (Bqkg<sup>-1</sup>)</b>			
	<sup>238</sup> U	<sup>232</sup> Th	<sup>235</sup> U *	<sup>226</sup> Ra	<sup>210</sup> Pb
<b>0–2 m</b>	1134 ± 28 **	67 ± 6 **	52	790 ± 38 **	
<b>2–4 m</b>	1175	54	54	845	
<b>4–6 m</b>	1224	61	56	1121	909
<b>B4</b>		<b>Activity concentration (Bqkg<sup>-1</sup>)</b>			
	<sup>238</sup> U	<sup>232</sup> Th	<sup>235</sup> U *	<sup>226</sup> Ra	<sup>210</sup> Pb
<b>2–4 m</b>	1290	62	59	932	
<b>4–6 m</b>	1374	71	63	1257	1136

\* <sup>235</sup>U activity concentration was estimated based on <sup>235</sup>U/<sup>238</sup>U natural activity ratio of 0.04. \*\* For locations where more than 1 sample was taken, the average mean value and standard deviation of activity concentration is shown.

**Table 3.** U, Th, Ra, and Pb concentration ratio (CR) values of trees used in risk assessments by ERICA tool (AM ± SD).

<b>Isotope</b>	<b>ERICA Tool Default CR Value</b>	<b>Site-Specific CR (From [32,34])</b>
U	0.006473 ± 0.014064	0.001 ± 0.0002
Th	0.001151 ± 0.001489	0.007 ± 0.005
Ra	0.01653 ± 0.02893	0.002 ± 0.001
Pb	0.0495 ± 0.1397	0.013 ± 0.003

In all of the assessment scenarios, the screening dose rate selected was the default tool value of 10 µGyh<sup>-1</sup>. The uncertainty factor (UF) selected was 3. The percentage of the dry weight of media used was its default value (100%), as well as weighting factors for alpha, high energy beta/gamma, and low energy beta radiation (10, 1, and 3, respectively).

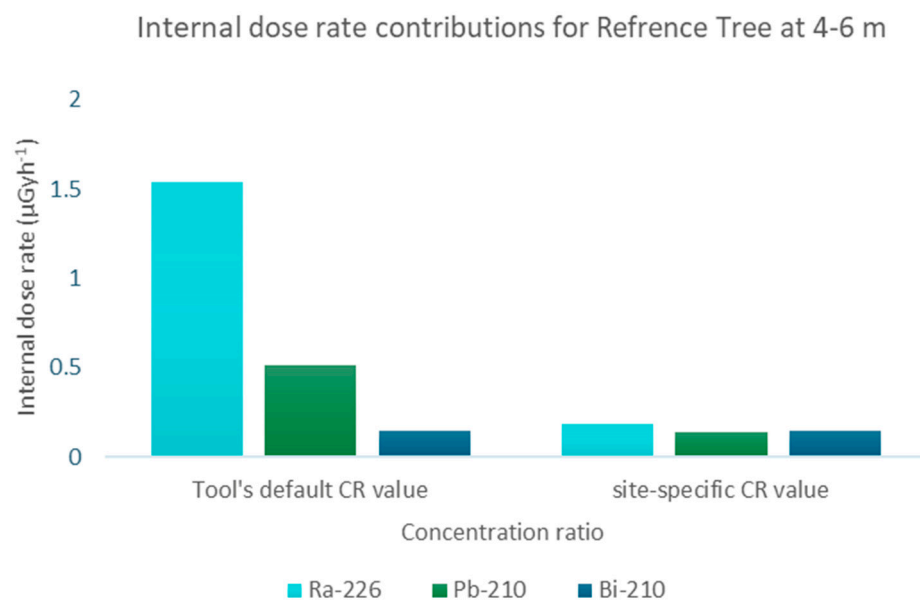
### 3. Results and Discussion

The radiological risk assessments were conducted based on the data from samples collected from boreholes B2, B3, and B4 at three depths: 0–2 m, 2–4 m, and 4–6 m. For each depth range, one risk assessment was performed.

In the assessments that used the tool's default CR values, the results for all three assessment scenarios (depth ranges) showed the resulting risk quotient (RQ) to be below 1. The tool's conservative RQ value was slightly above the value of 1 in three scenarios, mainly related to samples from greater depths (>4 m). For the assessments that used site-specific CR values, the resulting risk quotient was below 1, with the conservative risk quotient also below 1 in all assessment scenarios.

Data on the estimated dose rates for the reference tree showed that in the assessment using default CR values, the main contributor to the external dose rate in scenarios concerning all depth ranges was <sup>226</sup>Ra. This was also the case in the assessments using site-specific CR values at all depth ranges. However, the total dose rate mainly resulted from the internal dose rate in all assessments, contributing, on average, 90% to the total dose rate. In the context of the internal dose rate, in risk assessments that relied on the tool's default CR values and included depth ranges 0–2 m and 2–4 m, the main contributors were <sup>226</sup>Ra and <sup>238</sup>U. The results concerning the depth range of 4–6 m showed that in addition to

<sup>226</sup>Ra, <sup>210</sup>Pb and <sup>210</sup>Bi were also key contributors to the internal dose rate. Since the ERICA tool includes short-lived radionuclides with half-lives under 10 days in the assessment, <sup>210</sup>Bi, with a half-life of 5 days, is listed here as a direct progeny of <sup>210</sup>Pb. At this depth range, the radionuclide distribution of the internal dose rate in all samples showed that <sup>226</sup>Ra was the dominant radionuclide, accounting for approximately 70% of the internal dose rate, followed by <sup>210</sup>Pb and <sup>210</sup>Bi. This was also noticed in the analysis of the internal dose rate results from assessments using site-specific CR values at the depth range 4–6 m, where, in addition to <sup>226</sup>Ra, both <sup>210</sup>Pb and <sup>210</sup>Bi were detected by the tool as contributors to the internal dose rate, but distributed more evenly, with <sup>226</sup>Ra accounting for around 40% of the internal dose rate, and <sup>210</sup>Pb and <sup>210</sup>Bi each contributing with approximately 30%. Figure 2 shows a comparison of the distributions for radionuclides that contribute to the internal dose rate for the tree in the assessments related to a depth range of 4–6 m that relied on the tool’s CR and site-specific CR values. <sup>226</sup>Ra and <sup>210</sup>Pb primarily contribute to the total dose rate, and specific data is presented in Table 4.



**Figure 2.** Comparison of distributions for radionuclides that contribute to the internal dose rate for reference tree.

**Table 4.** Radionuclide dose rate contribution (µGyh<sup>-1</sup>) to ionizing radiation exposure of the reference tree and comparison of the tool’s output data obtained by the use of default and site-specific CR values.

Isotope	Total Dose Rate per Radionuclide [µGy h <sup>-1</sup> ] for Reference Tree in Assessments using Tool’s Default CR Values			Total Dose Rate per Radionuclide [µGy h <sup>-1</sup> ] for Reference Tree in Assessments using Site-Specific CR Values (Adopted from [34])		
	B2	B3	B4	B2	B3	B4
<sup>238</sup> U	0.574	0.585	0.594	0.099	0.101	0.076
<sup>232</sup> Th	0.004	0.008	0.014	0.020	0.037	0.024
<sup>235</sup> U	0.037	0.034	1.619	0.010	0.009	0.006
<sup>226</sup> Ra	7.497	7.117	11.700	1.581	1.501	1.193
<sup>210</sup> Pb	0.514	0.750	0.514	4.204	6.138	7.672

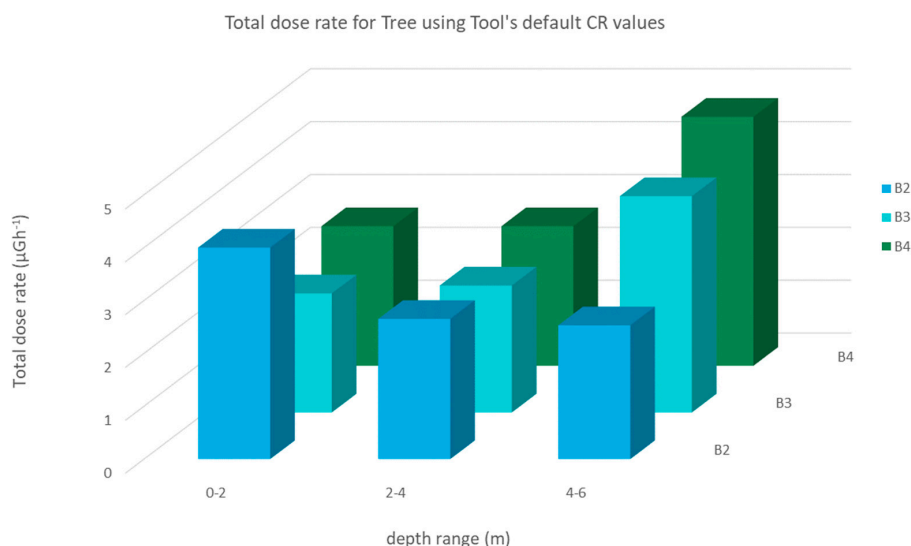
The presence of <sup>210</sup>Pb in plants is related to two main pathways that explain the uptake and content of lead in plants, one related to direct deposition from the atmosphere and the other via an indirect route through the root system [1]. Additionally, the plant radionuclide uptake and accumulation mechanisms are affected by a number of different



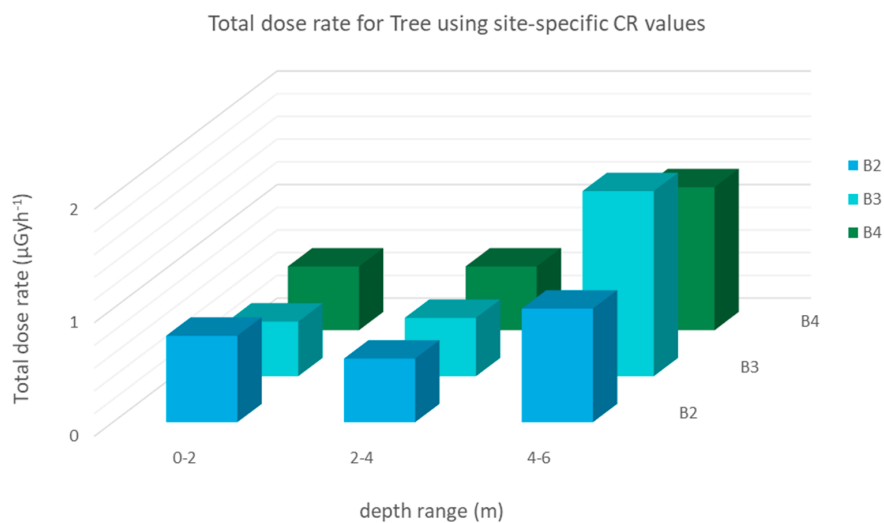
factors related to both soil type and its traits, plant species and characteristics, and climate features [28,44–46].

According to research from Pietrzak-Flis and Skowrońska-Smolak [47], the <sup>210</sup>Pb uptake by plants is primarily attributable to atmospheric deposition (mainly wet deposition), while the transfer through the root system can be considered insignificant. Consequently, since both the risk assessment scenarios using default CR and site-specific CR values that detected <sup>210</sup>Pb as a contributor to the total dose rate relate to assessments performed at a depth deeper than 4 m, and considering the estimated depths of root systems, the overall radiological risk from <sup>210</sup>Pb root uptake can be regarded as negligible. This assumption of the importance of atmospheric deposition in relation to the root uptake of <sup>210</sup>Pb is in line with the conclusions from a previous study at the exact location using surface soil samples, where similar activity concentrations of <sup>210</sup>Pb were detected in both plants from the disposal site and the control site, indicating atmospheric deposition as a major pathway for <sup>210</sup>Pb accumulation [32].

The total dose rates calculated by the tool using the default CR values and site-specific CR values in relation to the sample depth ranges are given in Figures 3 and 4.

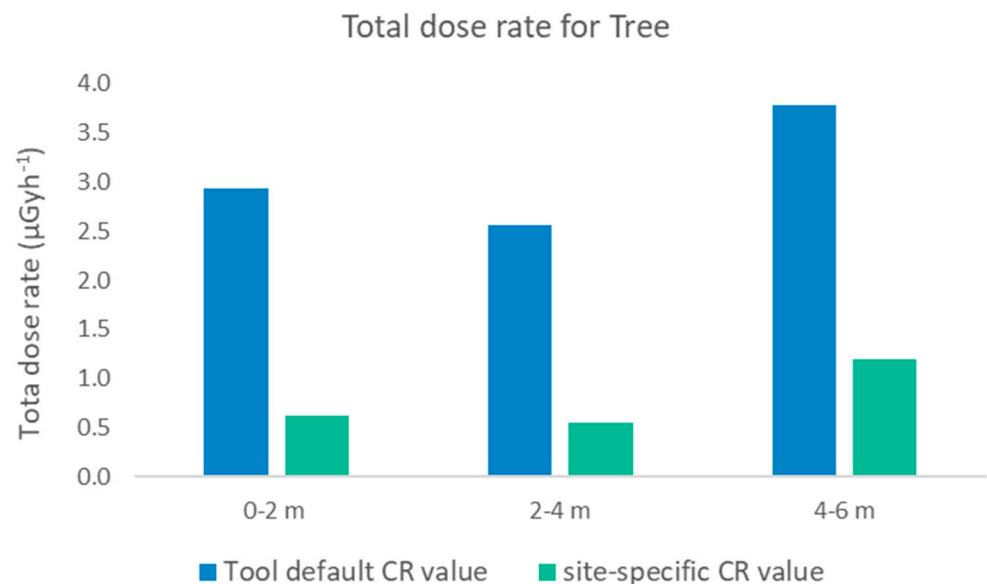


**Figure 3.** The total dose rate ( $\mu\text{Gy h}^{-1}$ ) for the reference tree at different depth ranges calculated using the tool’s default CR values.



**Figure 4.** The total dose rate ( $\mu\text{Gy h}^{-1}$ ) for the reference tree at different depth ranges calculated using site-specific CR values.

Figure 5 shows a comparison between the total dose rate for the reference organism—a tree—in assessments that used the default CR values and the ones using site-specific CR values. For practical purposes, these results show the average total dose rate for all samples taken from each sampling borehole, i.e., summarized assessment data from assessments conducted at three different depths. Since CR values are known to correlate the most with the estimated value of the total dose rate, as expected the tool estimated a higher total dose rate when a default CR value was used.



**Figure 5.** Comparison of estimated total dose rate for reference organism by different input data and used CR values.

The increase in the total dose rate when the tool’s default CR values are used in the assessments, as opposed to the site-specific CR values, ranged from 218 to 372%. Since the calculation of total dose rates is very sensitive to the CR values used [16], the use of the tool’s default CR values can often lead to overestimation of the dose rates and associated risks.

A previous study from Skoko et al. [34] used a control area in proximity to the disposal site to estimate dose rates to reference organisms. The results presented in Figure 5 show that the assessment results based on the use of site-specific CR values at the depth range 0–2 m correlate with the estimation results of the total dose rate for the tree at the control site of 0.5 μGyh<sup>-1</sup> [34], while for larger depths (4–6 m) the estimated total dose rates are twice as high (1.19 μGyh<sup>-1</sup>).

Assessment scenarios at various depth ranges found the radiological risk to the reference organism to be negligible, regardless of the depth range, as the screening dose rate of 10 μGyh<sup>-1</sup> was not exceeded in any of the assessments. The risk assessment results from all depth ranges show higher total dose rate predictions when the tool’s default CR values are used, which is an observation that was also made by other authors and is supported by previous research and assessments [28,34,38]. Our study, although focused on one reference organism (reference tree), confirms the risk assessment results of previous studies [34] that used a surface layer of coal ash (approximately the first 15 cm of the surface layer), finding both the total dose rate and the radiological risk predictions to be below predefined assessment values that assume no detrimental effects arising from potential exposure.

The study results need to be observed keeping in mind the assessment uncertainties related to a relatively small number of radionuclides included in the assessments and a limited number of samples available. Study limitations relate to the use of only gamma-ray spectrometry as an analytical method and, consequently, lack of data for radionuclides that are alpha emitters, such as <sup>230</sup>Th and <sup>210</sup>Po, that can considerably contribute to the exposure

of an organism and can be highly radiotoxic. Therefore, estimated dose rates might be an underestimation of the actual exposure of the tree roots to ionizing radiation due to the limited experimental data on radionuclide activity concentrations in the studied coal ash. Furthermore, data for  $^{232}\text{Th}$  in the coal ash were estimated from activity concentrations of its progeny ( $^{228}\text{Ra}$  and  $^{228}\text{Ac}$ ) under the assumption of secular equilibrium. However, considering that radionuclides from the thorium decay chain in the studied coal ash do not exceed background levels, it was considered that such an assumption can be acceptable. Another source of uncertainty that might affect the study results arises from the ERICA tool's inherent features related to assumptions on the homogeneous distribution of radionuclides in reference organisms and assumptions related to the occupancy factors. In several assessment scenarios, site-specific CR values from previous studies were used. Regarding the use of site-specific CR values for  $^{210}\text{Pb}$ , in previous research, Skoko et al. [34] noted that plant roots were not included in their study, so the resulting radiological effects to vegetation from our study could be underestimated. This observation is in agreement with the findings of the study from Mrdakovic Popic et al. [28], who noted that soil-to root transfer parameters for  $^{210}\text{Pb}$  are higher than transfer in above-ground plant parts. Since the ERICA tool is known for its conservative approach in predicting the possible effect on the biota, mainly when default CR values are used in the assessment, assuming the tool's calculation overestimated the total dose rate, the overall radiological risk can still be considered insignificant.

#### 4. Conclusions

The assessment of the radiological risk arising from exposure to NORM and the potentially hazardous effect of radiation on biota presents an initial step in the decision process on the need for implementation of radiation protection measures. Coal combustion residues are known for their environmental burden, and legacy disposal sites containing coal ash and slag provide specific research contexts for radiological and environmental studies.

In this study, the ERICA tool was used for dose rate assessments in a terrestrial ecosystem. The study used samples from a coal ash and slag disposal site that were collected as a part of previous extensive radiological research work at the location but still needed to be studied in detail. The main difference in relation to previous research conducted at the researched site is the depth range from which the samples were taken. To determine the possible effect of depth on the exposure of the selected reference organism, spatial risk assessment scenarios were performed using data from samples collected from boreholes from various depth ranges, as well as the tool's default CR values and site-specific CR values. The assessment results were compared and analyzed considering the sample depth, calculated risk quotient, resulting total dose rate, and distribution and contribution of internal and external dose rates.

The assessment results for all three selected depth ranges showed that radiological risk is negligible for the tree, as a main reference organism associated with these depths. This finding remains true also for both the assessment that relied on the tool's default CR values and the one that used the site-specific ones, although the total dose rate estimations were higher when the assessment included the tool's default CR values.

The results of our research imply that the use of soil surface samples, as opposed to the use of samples from deeper layers, is reasonable since the assessment results from our study did not exceed the set screening dose rate of  $10 \mu\text{Gyh}^{-1}$ , much less the limit of  $400 \mu\text{Gyh}^{-1}$ , a limit set by the UNSCEAR. The results can be useful for the optimization of future environmental monitoring and assessment design for different sites affected by NORM and general environmental radioprotection.

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## References

1. Vandenhove, H.; Olyslaegers, G.; Sanzharova, N.; Shubina, O.; Reed, E.; Shang, Z.; Velasco, H. Proposal for new best estimates of the soil-to-plant transfer factor of U, Th, Ra, Pb and Po. *J. Environ. Radioact.* **2009**, *100*, 721–732. [[CrossRef](#)] [[PubMed](#)]
2. International Atomic Energy Agency (IAEA). *Extent of Environmental Contamination by Naturally Occurring Radioactive Material (NORM) and Technological Options for Remediation*; Technical Report Series 419; International Atomic Energy Agency (IAEA): Vienna, Austria, 2003.
3. Osborne, D.; Jahandari, S.; Tao, Z.; Chen, Z.; Khazaie, A.; Rahme, M. Creating Additional Revenue Streams Prior to the Disposal of Tailings. *Int. J. Energy Clean Environ.* **2023**, *24*, 1–14. [[CrossRef](#)]
4. Tayebi-Khorami, M.; Edraki, M.; Corder, G.; Golev, A. Re-thinking mining waste through an integrative approach led by circular economy aspirations. *Minerals* **2019**, *9*, 286. [[CrossRef](#)]
5. Corder, G.D. Insights from case studies into sustainable design approaches in the minerals industry. *Miner. Eng.* **2015**, *76*, 47–57. [[CrossRef](#)]
6. Asokan, P.; Saxena, M.; Asolekar, S.R. Coal combustion residues—Environmental implications and recycling potentials. *Resour. Conserv. Recycl.* **2005**, *43*, 239–262. [[CrossRef](#)]
7. Popov, O.; Iatsyshyn, A.; Kovach, V.; Artemchuk, V.; Kameneva, I.; Radchenko, O.; Nikolaiev, K.; Stanytsina, V.; Iatsyshyn, A.; Romanenko, Y. Effect of Power Plant Ash and Slag Disposal on the Environment and Population Health in Ukraine. *J. Health Pollut.* **2021**, *11*, 210910. [[CrossRef](#)]
8. Haynes, R.J. Reclamation and revegetation of fly ash disposal sites—Challenges and research needs. *J. Environ. Manag.* **2009**, *90*, 43–53. [[CrossRef](#)]
9. International Commission on Radiological Protection (ICRP). *ICRP Publication 103: The 2007 Recommendations of the International Commission on Radiological Protection*; Annals of the ICRP; Elsevier: Oxford, UK, 2007; Volume 37.
10. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). *Effects of ionizing radiation on non-human biota*. In *Sources and Effects of Ionizing Radiation*; UNSCEAR 2008 Report to the General Assembly with Scientific Annexes; United Nations: New York, NY, USA, 2011; Volume II.
11. Copplestone, D.; Howard, B.J.; Bréchnignac, F. The ecological relevance of current approaches for environmental protection from exposure to ionising radiation. *J. Environ. Radioact.* **2004**, *74*, 31–41. [[CrossRef](#)]
12. Higley, K.A.; Bytwerk, D.P. Generic approaches to transfer. *J. Environ. Radioact.* **2007**, *98*, 4–23. [[CrossRef](#)]
13. Beresford, N.A.; Barnett, C.L.; Brown, J.E.; Cheng, J.J.; Copplestone, D.; Gaschak, S.; Hosseini, A.; Howard, B.J.; Kamboj, S.; Nedveckaite, T.; et al. Predicting the radiation exposure of terrestrial wildlife in the Chernobyl exclusion zone: An international comparison of approaches. *J. Radiol. Prot.* **2010**, *30*, 341–373. [[CrossRef](#)]
14. Beresford, N.A.; Barnett, C.L.; Beaugelin-Seiller, K.; Brown, J.E.; Cheng, J.-J.; Copplestone, D.; Gaschak, S.; Hingston, J.L.; Horyna, J.; Hosseini, A.; et al. Findings and recommendations from an international comparison of models and approaches for the estimation of radiological exposure to non-human biota. *Radioprotection* **2009**, *44*, 565–570.
15. Pentreath, R.J.; Woodhead, D.S. A system for protecting the environment from ionising radiation: Selecting reference fauna and flora, and the possible dose models and environmental geometries that could be applied to them. *Sci. Total Environ.* **2001**, *277*, 33–43. [[CrossRef](#)] [[PubMed](#)]
16. Beresford, E.N.; Brown, J.; Copplestone, D.; Garnier-Laplace, J.; Howard, B.; Larsson, C.; Oughton, D.; Pröhl, G.; Zinger, I. *D-ERICA: An Integrated Approach to the Assessment and Management of Environmental Risks from Ionising Radiation, 2007*; Deliverable of the ERICA Project (FI6R-CT-2004-508847); Swedish Radiation Protection Authority: Stockholm, Sweden, 2007; Available online: <https://wiki.ceh.ac.uk/download/attachments/115017395/D-Erica.pdf> (accessed on 1 March 2023).
17. Brown, J.E.; Alfonso, B.; Avila, R.; Beresford, N.A.; Copplestone, D.; Pröhl, G.; Ulanovsky, A. The ERICA Tool. *J. Environ. Radioact.* **2008**, *99*, 1371–1383. [[CrossRef](#)] [[PubMed](#)]
18. Larsson, C.M. An overview of the ERICA Integrated Approach to the assessment and management of environmental risks from ionising contaminants. *J. Environ. Radioact.* **2008**, *99*, 1364–1370. [[CrossRef](#)]
19. Howard, B.J.; Larsson, C.-M. The ERICA Integrated Approach and its contribution to protection of the environment from ionising radiation. *J. Environ. Radioact.* **2008**, *99*, 1361–1363. [[CrossRef](#)]
20. International Atomic Energy Agency (IAEA). *Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment*; Safety Reports Series No. 19; International Atomic Energy Agency (IAEA): Vienna, Austria, 2001.

21. Beresford, N.A.; Barnett, C.L.; Howard, B.J.; Scott, W.A.; Brown, J.E.; Copplestone, D. Derivation of transfer parameters for use within the ERICA Tool and the default concentration ratios for terrestrial biota. *J. Environ. Radioact.* **2008**, *99*, 1393–1407. [[CrossRef](#)]
22. Beresford, N.A.; Balonov, M.; Beaugelin-Seiller, K.; Brown, J.; Copplestone, D.; Hingston, J.L.; Horyna, J.; Hosseini, A.; Howard, B.J.; Kamboj, S.; et al. An international comparison of models and approaches for the estimation of the radiological exposure of non-human biota. *Appl. Radiat. Isot.* **2008**, *66*, 1745–1749. [[CrossRef](#)]
23. Sotiropoulou, M.; Mavrokefalou, G.; Florou, H.; Kritidis, P. Determination and mapping of the spatial distribution of cesium-137 in the terrestrial environment of Greece, over a period of 28 years (1998 to 2015). *Environ. Monit. Assess.* **2021**, *193*, 591. [[CrossRef](#)]
24. Babić, D.; Skoko, B.; Franić, Z.; Senčar, J.; Šoštarić, M.; Petroci, L.; Avdić, M.; Kovačić, M.; Branica, G.; Petrinec, B.; et al. Baseline radioecological data for the soil and selected bioindicator organisms in the temperate forest of Plitvice Lakes National Park, Croatia. *Environ. Sci. Pollut. Res.* **2020**, *27*, 21040–21056. [[CrossRef](#)]
25. Vetikko, V.; Saxén, R. Application of the ERICA Assessment Tool to freshwater biota in Finland. *J. Environ. Radioact.* **2010**, *101*, 82–87. [[CrossRef](#)]
26. Aryanti, C.A.; Suseno, H.; Muslim, M.; Prihatiningsih, W.R.; Aini, S.N. Potential Radiological Dose of  $^{210}\text{Po}$  to Several Marine Organisms in Coastal Area of Coal-Fired Power Plant Tanjung Awar—Awar, Tuban. *Ilmu Kelaut.* **2022**, *27*, 73–87. [[CrossRef](#)]
27. Čujić, M.; Dragović, S. Assessment of dose rate to terrestrial biota in the area around coal fired power plant applying ERICA tool and RESRAD BIOTA code. *J. Environ. Radioact.* **2018**, *188*, 108–114. [[CrossRef](#)]
28. Mrdakovic Popic, J.; Oughton, D.H.; Salbu, B.; Skipperud, L. Transfer of naturally occurring radionuclides from soil to wild forest flora in an area with enhanced legacy and natural radioactivity in Norway. *Environ. Sci.* **2020**, *22*, 350–363. [[CrossRef](#)]
29. Oughton, D.H.; Strømman, G.; Salbu, B. Ecological risk assessment of Central Asian mining sites: Application of the ERICA assessment tool. *J. Environ. Radioact.* **2013**, *123*, 90–98. [[CrossRef](#)]
30. Vandenhove, H.; Vives i Batlle, J.; Sweeck, L. Potential radiological impact of the phosphate industry on wildlife. *J. Environ. Radioact.* **2015**, *141*, 14–23. [[CrossRef](#)]
31. Oreščanin, V.; Barišić, D.; Mikelić, L.; Lovrenčić, I.; Rožmarić-Mačefat, M.; Pavlović, G.; Lulić, S. Chemical and radiological profile of the coal ash landfill in Kaštel Gomilica. *Arh. Hig. Rada Toksikol.* **2006**, *57*, 9–16.
32. Skoko, B.; Marović, G.; Babić, D.; Šoštarić, M.; Jukić, M. Plant uptake of  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  from a coal ash and slag disposal site and control soil under field conditions: A preliminary study. *J. Environ. Radioact.* **2017**, *172*, 113–121. [[CrossRef](#)]
33. Skoko, B.; Marović, G.; Babić, D. Radioactivity in the Mediterranean flora of the Kaštela bay, Croatia. *J. Environ. Radioact.* **2014**, *135*, 36–43. [[CrossRef](#)]
34. Skoko, B.; Babić, D.; Marović, G.; Papić, S. Environmental radiological risk assessment of a coal ash and slag disposal site with the use of the ERICA Tool. *J. Environ. Radioact.* **2019**, *208–209*, 106018. [[CrossRef](#)]
35. Lovrenčić Mikelić, I.; Barišić, D. Radiological risks from  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in urbanised and industrialised karstic coastal area (Kaštela Bay, Croatia). *Environ. Sci. Pollut. Res.* **2022**, *29*, 54632–54640. [[CrossRef](#)]
36. Lovrenčić Mikelić, I.; Barišić, D. Natural and anthropogenic radionuclides in karstic coastal area (Kaštela Bay, Adriatic Sea, Croatia) exposed to anthropogenic activities: Distribution, sources, and influencing factors. *Radiochim. Acta* **2023**, *111*, 147–157. [[CrossRef](#)]
37. Mandić, J.; Veža, J.; Kušpilić, G. Assessment of environmental risk related to the polycyclic aromatic hydrocarbons (PAH) in the sediments along the eastern Adriatic coast [Određivanje toksičnosti sedimenta povezane s policikličkim aromatskim ugljikovodicima—PAH duž istočne obale Jadranskog mora]. *Acta Adriat.* **2022**, *63*, 135–150.
38. Brown, J.E.; Beresford, N.A.; Hosseini, A. Approaches to providing missing transfer parameter values in the ERICA Tool—How well do they work? *J. Environ. Radioact.* **2013**, *126*, 399–411. [[CrossRef](#)] [[PubMed](#)]
39. Brown, J.E.; Alfonso, B.; Avila, R.; Beresford, N.A.; Copplestone, D.; Hosseini, A. A new version of the ERICA tool to facilitate impact assessments of radioactivity on wild plants and animals. *J. Environ. Radioact.* **2016**, *153*, 141–148. [[CrossRef](#)] [[PubMed](#)]
40. International Commission on Radiological Protection (ICRP). *ICRP Publication 108: Environmental Protection—The Concept and Use of Reference Animals and Plants*; Annals of the ICRP; Elsevier: Oxford, UK, 2008; Volume 38.
41. Petrinec, B.; Franić, Z.; Bituh, T.; Babić, D. Quality Assurance in Gamma-Ray Spectrometry of Seabed Sediments. *Arh. Hig. Rada Toksikol.* **2011**, *62*, 17–22. [[CrossRef](#)]
42. Silva, J.S.; Rego, F.C. Root distribution of a Mediterranean shrubland in Portugal. *Plant Soil* **2003**, *255*, 529–540. [[CrossRef](#)]
43. Canadell, J.; Jackson, R.B.; Ehleringer, J.B.; Mooney, H.A.; Sala, O.E.; Schulze, E.D. Maximum rooting depth of vegetation types at the global scale. *Oecologia* **1996**, *108*, 583–595. [[CrossRef](#)]
44. Černe, M.; Smodiš, B.; Strok, M.; Jećimović, R. Plant Accumulation of Natural Radionuclides as Affected by Substrate Contaminated with Uranium-Mill Tailings. *Water Air Soil Pollut.* **2018**, *229*, 371. [[CrossRef](#)]
45. Madruga, M.J.; Brogueira, A.; Alberto, G.; Cardoso, F.  $^{226}\text{Ra}$  bioavailability to plants at the Uregiriça uranium mill tailings site. *J. Environ. Radioact.* **2001**, *54*, 175–188. [[CrossRef](#)]

46. Vandenhove, H.; Van Hees, M. Predicting radium availability and uptake from soil properties. *Chemosphere* **2007**, *69*, 664–674. [[CrossRef](#)]
47. Pietrzak-Flis, Z.; Skowrońska-Smolak, M. Transfer of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  to plants via root system and above-ground interception. *Science* **1995**, *162*, 139–147.

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