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DOCTORAL DISSERTATION

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PROCJENA UTJECAJA DUBINE I VREMENA UZORKOVANJA TLA NA REZULTATE KVANTIFIKACIJE RADIOLOŠKOG RIZIKA

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“One never notices what has been done; one can only see what remains to be done.”

- Marie Skłodowska-Curie

ABSTRACT

Given that most mineral resources and raw materials contain radionuclides of natural origin, the exploitation of these resources, mining and mineral processing can result in an increased concentration of radionuclide activity in NORM (naturally occurring radioactive material) waste and residues, which can lead to potential exposure to ionizing radiation. This research focused on activities associated with potential radiation exposure, including coal combustion and natural gas processing. Since the generated waste and residues can contain significant amounts of NORM with often long-lived radionuclides and can adversely affect human health, safety and the environment, environmental monitoring and assessment of the impact of these industrial activities are immensely important. Radiological risk assessment is imperative to assess the possible radiological effects of these industrial processes on the environment.

The ERICA Assessment Tool was used to quantify the radiological risk at three research locations related to the mining and oil and gas industry in Croatia. In all samples collected, radioactivity was determined using high-resolution gamma-spectrometry with a method accredited in compliance with the HRN EN ISO/IEC 17025 standard. The performed assessments focused on different spatial and temporal data sets, considering the specifics of each research location.

The results of radiological risk assessments confirmed that the overall radiological risk is negligible at all three selected research locations. In all risk assessment scenarios conducted by this research, lichens and bryophytes were the most sensitive organisms with generally the highest predicted dose rates. Indoor exposure was the most significant contributor to the total dose rate in all scenario assessments, with ^{226}Ra as the key factor. The overall results of this spatial assessment suggest that the use of surface soil samples, as opposed to the use of samples from deeper layers, is reasonable since the radiological risk assessment results did not exceed the ERICA Assessment Tool dose rate limit of 10 μGyh^{-1}. A time assessment based on data from the location of a natural gas processing facility showed that neither singular time assessments nor assessments based on time-averaged radiological data resulted in a significant risk to the environment. The results suggest that reliable monitoring and assessment should be used to continuously confirm radiological protection and environmental protection on the site. The results indicate the importance of environmental monitoring in ensuring long-term radiological protection and safety and environmental protection, and also demonstrate the applicability of the ERICA Assessment Tool to confirm the effects of remediation.

PROŠIRENI SAŽETAK

S obzirom da većina mineralnih resursa i sirovina sadrži radionuklide prirodnog podrijetla, uglavnom iz lanaca raspada urana i torija, eksploatacija ovih resursa, rudarstvo i oplemenjivanje mineralnih sirovina mogu rezultirati povećanom koncentracijom aktivnosti radionuklida u NORM (engl. *naturally occuring radioactive material*) otpadu i reziduima, što može dovesti do potencijalne izloženosti ionizirajućem zračenju. Ovo se istraživanje usredotočilo na aktivnosti povezane s potencijalnom izloženošću biote zračenju, uključujući spaljivanje ugljena te proizvodnju i preradu plina. S obzirom da nastali otpad i rezidui mogu sadržavati značajne količine NORM-a, a sadržani radionuklidi često su dugovječni i mogu negativno utjecati na ljudsko zdravlje, sigurnost i okoliš, monitoring okoliša i procjena radiološkog utjecaja ovih industrijskih aktivnosti od iznimne su važnosti. Procjena radiološkog rizika imperativ je za procjenu mogućih radioloških učinaka ovih industrijskih procesa na okoliš.

Ciljevi i hipoteze

Glavni ciljevi ovog istraživanja bili su utvrditi moguću vezu između kvantifikacije radiološkog rizika i dubine uzorkovanja tla, utvrditi moguću vezu između kvantifikacije radiološkog rizika i učestalosti uzorkovanja tla te istražiti potencijalne učinke vrsta proizvodnih aktivnosti na odabranim lokacijama istraživanja na rezultate procjene ukupnog radiološkog rizika.

Ovi su se ciljevi temeljili na dvije glavne hipoteze: (i) dubina s koje se uzima uzorak tla za analizu utječe na rezultat procjene radiološkog rizika zbog mehanizama transporta radionuklida u okolišu, (ii) učestalost uzorkovanja (jednokratno ili povremeno praćenje) na određenoj lokaciji, unatoč dugovječnosti radionuklida, utječe na rezultat procjene radiološkog rizika zbog mehanizama transporta radionuklida u okolišu.

Znanstveni doprinos

Rezultati ovog istraživanja korištenjem *ERICA Assessment Tool*-a omogućuju bolje razumijevanje ovisnosti rezultata procjene radiološkog rizika o dubini i učestalosti uzorkovanja tla na različitim lokacijama istraživanja. U okviru rada, na lokacijama rudarske i naftne i plinske industrije u Hrvatskoj po prvi put je istražen odnos između dubine i učestalosti uzorkovanja tla i rezultirajućeg radiološkog rizika za okoliš (biotu). Dobiveni rezultati doprinose području zaštite od zračenja i daju uvid u pristup odabiru uzoraka, planiranju budućih istraživanja procjene radiološkog rizika i interpretaciji dobivenih rezultata.

Metode i postupci

U svrhu kvantifikacije radiološkog rizika i moguće povezanosti rezultata radiološkog rizika i dubine te učestalosti uzorkovanja tla korišten je *ERICA Assessment Tool* koji provodi probabilističku kvantifikaciju radiološkog rizika za okoliš kombinirajući radiološke podatke i podatke o prijenosu radionuklida, pritom se oslanjajući se na dostupne baze podataka o referentnim organizmima i radionuklidima (**Beresford i dr., 2007; Brown i dr., 2008; Brown i dr., 2016**). *ERICA Assessment Tool* oslanja se na upotrebi referentni organizama (**Brown i dr., 2008; Larsson, 2008**) što je u skladu s preporukama International Commission on Radiological Protection (**ICRP, 2007**). Primjena *ERICA Assessment Tool*-a uključuje širok raspon mogućih izloženosti biote, uključujući izloženosti koje proizlaze iz odobrenih sustava za ispuštanje, potencijalnih ispuštanja radionuklida iz skladišta radioaktivnog otpada, proizvodnje i procesa sanacije koji uključuju rezidue i prirodne radioaktivne materijale te scenarije koji uključuju moguće radiološke ili nuklearne nesreće (**Brown i dr., 2008.; Brown i dr., 2016.).** Na svim uzorcima s lokacija istraživanja, s obzirom na način prisutnosti ili odlaganje prirodno nastalih radioaktivnih materijala i/ili rezidua, provedena je radiološka karakterizacija metodama visokorezolucijske gamaspektrometrije.

Rezultati i zaključci

Procjene radiološkog rizika, korištenjem *ERICA Assessment Tool*-a, koje su provedene u sklopu ove disertacije, uključile su tri lokacije povezane s NORM-om u Hrvatskoj, uključujući lokacije na kojima se odlažu rezidui, i to na tzv. *legacy* odlagalištu ili na saniranom odlagalištu te postrojenje za preradu prirodnog plina. Rezultati provedenih procjena radiološkog rizika potvrdili su da je ukupni radiološki rizik zanemariv na sve tri odabrane lokacije istraživanja. U svim scenarijima procjene rizika provedenim ovim istraživanjem, lišajevi i briofiti su bili najosjetljiviji organizmi s općenito najvećim predviđenim brzinama doze. Unutarnja izloženost najviše je pridonijela ukupnoj brzini doze u svim scenarijima procjene, s ^{226}Ra kao ključnim čimbenikom.

Budući da ERICA Assessment Tool-a omogućuje provođenje procjene radiološkog rizika s obzirom na specifičan prostorni i vremenski kontekst pojedinih istraživačkih lokacija, izvršene procjene usmjerene su na različite prostorne i vremenske skupove podataka, s obzirom na specifičnosti svake lokacije. Procjena koja se odnosi na tzv. *legacy* odlagalište ugljenog pepela i šljake koristila je različite dubine uzorkovanja tla kao ulazne podatke za procjenu u prostornom kontekstu. Ovisno o referentnim organizmima uključenim u procjenu i njihovom staništu, kao što je bio slučaj s dubinom korijena mediteranske flore, dubina uzorka tla može neznatno utjecati na rezultate procjene rizika i akumulaciju radionuklida, ali potrebna su daljnja terenska istraživanja kako bi se razjasnio utjecaj dubine s koje je uzet uzorak u unosu radionuklida korijenjem. Vrijednosti koncentracijskih omjera (CR) korištene u procjeni rizika značajno su utjecale na rezultate procjene, pri čemu su procjene ukupne brzine doze bile veće kada je procjena uključivala zadane konzervativne vrijednosti CR alata *ERICA Assessment Tool*, za razliku od CR vrijednosti specifičnih za lokaciju. Ukupni rezultati ove prostorne procjene upućuju na to da je korištenje površinskih uzoraka tla, za razliku od korištenja uzoraka iz dubljih slojeva, razumno budući da rezultati procjene radiološkog rizika nisu premašili graničnu brzinu doze za procjenu *ERICA Assessment Tool*-a od 10 μGyh^{-1}.

U kontekstu procjena radiološkog rizika na temelju različitih vremenskih skupova podataka, rezultati procjene koji se odnose na sanirano odlagalište ugljenog pepela i šljake pokazali su da su procijenjeni radiološki rizik i odgovarajuće brzine doze za referentne organizme nakon sanacije lokacije bili značajno niži u usporedbi s razdobljem prije sanacije lokacije. Rezultati ukazuju na važnost monitoringa okoliša u osiguravanju dugoročne radiološke zaštite i sigurnosti i zaštite okoliša te također ukazuju na primjenjivost *ERICA Assessment Tool*-a za potvrdu učinaka sanacije.

Vremenska procjena temeljena na podacima s lokacije postrojenja za preradu prirodnog plina pokazala je da niti pojedinačne vremenske procjene niti procjene temeljene na vremenski usrednjenim radiološkim podacima nisu rezultirale značajnim rizikom za okoliš. U svim scenarijima procjene nije uočen određeni trend u procijenjenim brzinama doza. Međutim, rezultati sugeriraju da radiološka zaštita i zaštita okoliša trebaju biti kontinuirano potvrđivani pouzdanim monitoringom i s njim povezanom procjenom. Učinak učestalosti uzorkovanja na odabranim istraživačkim lokacijama na rezultate procjene radiološkog rizika može se smatrati neznatnim, s obzirom na specifičnu lokaciju istraživanja, tj. bez fluktuacija u ispuštanju ili razinama kontaminacije i odsutnosti organizama s kratkim životnim vijekom koji bi mogli biti pogođeni izlaganjem zračenju.

U kontekstu vrste industrijskih aktivnosti koje se provode na određenoj lokaciji istraživanja, rezultati procjene radiološkog rizika pokazali su da su predviđanja ukupne brzine doze veća na lokacijama povezanim sa sagorijevanjem ugljena u usporedbi s preradom prirodnog plina, posebno u kontekstu *legacy* odlagališta ugljenog pepela i šljake koje se ne nadzire. Industrijske aktivnosti prerade prirodnog plina, iako imaju značajan potencijal za izloženost zračenju, zbog primijenjenih strogih standarda zaštite okoliša utvrđene procijenjene ukupne brzine doze nisu bile značajne.

KEYWORDS

NORM (Naturally Occurring Radioactive Material)

Radiological risk assessment

Coal ash and slag

Oil and gas industry

Environmental monitoring

Environmental protection

KLJUČNE RIJEČI

Materijali s povišenom prirodnom radioaktivnošću

Procjena radiološkog rizika

Ugljeni pepeo i šljaka

Naftna i plinska industrija

Monitoring okoliša

Zaštita okoliša

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1. INTRODUCTION

1.1. Naturally occurring radioactive materials

Many natural resources contain radionuclides of natural origin, while increased concentrations of naturally occurring radionuclides are often found in different geological materials, igneous rocks and ores (**IAEA, 2022; IAEA, 2003**). From a radiation protection standpoint, the most important radionuclides are those from the ^{238}U and ^{232}Th decay series and ^{40}K. While many human activities outside the nuclear fuel cycle involving minerals and raw materials do not result in increased levels of exposure to ionising radiation, some activities might result in significantly enhanced exposures due to naturally occurring radioactive materials (**Michalik et al., 2023; ICRP, 2019; IAEA, 2013**). NORM – naturally occurring radioactive materials – can be differently defined depending on the national context and regulatory approaches in particular countries, but generally, NORM can be defined as "all naturally occurring radioactive materials where human activities have increased the potential for exposure in comparison with the unaltered situation" (**IAEA, 2019; IAEA, 2003**).

The potential effects of NORM to humans and the environment are related to the following:

1) certain exploitation processes and operations that result in wastes and residues with enhanced activity concentrations, sometimes by order of magnitude of the original material (**IAEA, 2013**) and
2) increased availability of wastes and residues released into the biosphere as a result of their physicochemical changes or due to the residues' management method (**Garcia-Tenorio et al. 2015; IAEA, 2003**).

The abundance and presence of radionuclides in some natural resources are shown in **Table 1-1.**, which presents activity concentrations of key radionuclides in major rock types and soil, where **Table 1-2.** summarises activity concentrations of radionuclides in natural resources that, when exploited, can lead to enhanced concentrations in resulting waste and residues. NORM waste and residues generation concerning coal production is determined by the geological formation of coal seams, and excavation usually results in large quantities of waste rock and wastewater (**Wysocka et al., 2019; Skubacz et al., 2011; Michalik et al., 2002; IAEA, 2003**). In the context of resulting activity concentrations, coal combustion in coal-fired power plants is related to the generation of bottom ash, fly ash, and sludge with higher activity concentrations than coal excavation and production alone (**Papastefanou,** 2010; IAEA, 2003**). The activity concentrations of coal combustion residues are directly related to the activity concentration of the origin coal used as fuel (**Habib et al., 2019; Lauer et al., 2015; Walencik-Łata & Smołka-Danielowska, 2020; Hasani et al., 2014; IAEA; 2003**). **Table 1-3.** shows measured activity concentrations in coal combustion residues.

Table 1-1. Summary of activity concentrations (Bq/kg) of key radionuclides in major rock types and soil (modified according to **IAEA, 2003**)

Rock Type	**^{238}U** Bq/kg	**^{232}Th** Bq/kg	**^{40}K** Bq/kg	**^{87}Rb** Bq/kg
Igneous rocks				
Basalt				
crustal average	7 - 10	10 - 15	300	30
mafic	7,1	7,1	70 - 400	1 - 40
salic	50,6	60,8	1100 - 1500	150 - 180
Granite				
crustal average	40	70	>1000	150 - 180
Sedimentary rocks				
Shale				
sandstones	40	50	800	110
Clean quartz	<10	<8	<300	<40
Dirty quartz	40*	10-25*	400*	90*
Arkose	10 - 25*	<8*	600 - 900	80 - 120
Beach sands				
(unconsolidated)	40	25	<300*	<40*
Carbonate rocks	25	8	70	8
Continental upper crust average	36	44	850	110
Soils	66	37	50	400

*Estimations in the absence of measured values

Table 1-2. Naturally occurring radionuclides in mineral resources (modified according to **IAEA, 2003)**

Element/mineral resource	**Source**	**Radioactivity**
Natural gas	Gas, average for groups of US and Canadian wells	2 – 17 000 Bq/(m^3 Rn)
	Gas, individual US and Canadian wells	0,4 – 54 000 Bq/(m^3 Rn)
	Scale, residue in pumps, vessels and residual gas pipelines	100 – 50 000 Bq/(kg ^{210}Pb/^{210}Po)
Oil	Brines or produced water	Ranging from mBq to 100 Bq/(L Ra)
	Sludge	Raging up to 70 000 Bq/(kg Ra)
	Scale	Ranging up to 4 × 10^6 Bq/(kg Ra)
Uranium	Ore	15000 Bq/(kg Ra)
	Slime	10^5 Bq/(kg Ra)
	Tailings	10 000 – 20 000 Bq/(kg Ra)

Table 1-3. Activity concentrations (in Bq/kg) in different coal combustion residues (modified according to **IAEA, 2003)**

Material	^{238}U	^{226}Ra	^{228}Ra	^{235}U	^{40}K
Polish coal-fired power stations					
Ash (average)		131	102		631
Slag (average)		108	79		654
Croatian coal-fired power stations					
Fly ash	8700	2400	20	400	150
Bottom ash and slag	3400	2000	60	200	290
Brazilian coal and combustion products					
Fly ash	144	192	144		
Bottom ash and slag	156	120	84		
US coal combustion wastes					
Fly ash	96	111	96	5	
Bottom ash and slag	26	26	22	1	

The origin of NORM in the oil and gas industry relates to radionuclides from uranium and thorium decay series but also includes smaller amounts of radionuclide parents and more significant amounts of radium isotopes (^{224}Ra, ^{226}Ra, and ^{228}Ra) which are contained in the formation water (**Michalik et al., 2023; Xhixha et al., 2015; IAEA, 2010**). Gas production is associated with the occurrence of ^{210}Pb, also known as radiolead, which is found in the form of sludge, and stable lead deposited in the form of thin films, coatings, and plating on the inside of the production equipment (**Barros et al., 2018; Jodłowski et al., 2017; IAEA, 2010**). Radon gas, ^{222}Rn, emanation is also related to gas production where radon is found in the gas phase and in the form of films on the gas handling equipment **(Michalik et al., 2023; IAEA, 2003**). Hard radioactive scale and sludge usually relate to extraction and production stages. Scales consist of carbonate and sulphate mixtures, and compared to the natural environment and other NORM residues, they can contain ^{226}Ra in elevated activity concentrations (**Gäfvert et al., 2006; Hamlat et al., 2001**). Table 1-4. lists forms and activity concentrations of NORM residues that are found in the oil and gas industries.

Table 1-4. Examples of NORM activity concentrations in oil and gas residues (modified according to **IAEA, 2003)**

Material	**Activity concentration (Bq/kg)**
Scale in downhole tubing, pipes and other equipment for handling oil/gas and formation waters	^{226}Ra: background to 15 000 000 (average 1000 to hundreds of thousands)
Sludges in separations and production equipment	^{226}Ra: 10 000 to 1 000 000
Sludges, films in natural gas supply equipment	^{210}Pb: background to about 40 000
Sludges from soils beneath ponds of produced water	^{226}Ra: 10 000 to 40 000

An essential step for industries is understanding when and where NORM occurs in the industrial processes, in which amounts and activity concentrations, and how it should be managed. Effective management of wastes and residues, including NORM, is also stipulated by the United Nations Conference on Environment and Development Agenda 21, stating that all wastes should be managed to protect human health and the environment (**IAEA, 2003**). In the context of regional law, the European Basic Safety Standards Directive (Council Directive 2013/59/Euratom) includes planned exposure from new sources or new pathways of exposure resulting from industrial activities processing naturally occurring radioactive materials (NORM) (**Michalik, 2009**).

In the Republic of Croatia, several existing industrial sites are associated with NORM, remediated disposal sites, and legacy disposal sites. The national regulatory framework also provides a list of activities and industrial sectors that use naturally radioactive materials, including research and relevant secondary processes in the Ordinance on environmental radioactivity monitoring ("Official Gazette", no. 6/22) that includes oil and gas production and coal-fired power plants.

1.2. Overview of radiological risk assessment, its development and implementation

Over the past three decades, protecting the environment from ionising radiation and radiological assessment of radiological risk have received increasing attention (**ICRP, 1990**). There is a continuous scientific interest and need for the development of a comprehensive framework for radiation protection that ensures a credible radiological risk assessment system. Various international organisations have focused their efforts on developing methods and approaches to protect the environment from ionising radiation that is recognised and approved internationally, thus promoting major advances in environmental radiation protection (**Howard and Larsson, 2008; Stark et al., 2017**).

A fundamental paradigm shift in ionising radiation protection was the one including the assumption that if humans are protected, the non-human biota is equally protected, made in 1991 by the International Commission on Radiological Protection (ICRP) (**ICRP, 1991**). In their recommendations from 2007, the same Commission referred to the environmental impact of ionising radiation, particularly on biota and the environment as a whole (**ICRP, 2007**). The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) also contributed to the paradigm shift, with their report emphasising that ecosystems consist of different organisms with different radiological sensitivities (**UNSCEAR, 2008**). In 2015 the International Union of Radioecology (IUR) made seven consensus statements regarding the impact of radiation on the environment, populations and ecosystems, thus moving toward an eco-centric approach to radiation protection (**Bréchignac et al., 2016)**. According to **Oughton (2003**), ionising radiation should be treated the same way as other environmental stressors in support of a holistic approach to radiation protection. The approach taking into consideration only humans cannot ensure the protection of all biota in all situations, as stated by **Bréchignac (2003**). Hence, radiation protection must include non-human biota in its approaches and implementation of its framework.

A reliable system of assessing radiation impacts and potential dose rates is essential for protecting both humans and the environment, including animals and plants, from risks related to radionuclides in the environment (**Stark et al., 2017**). **Bréchignac et al. (2016**) provide a list of sites of particular importance for understanding the effects of radiation at the population and ecosystem levels, such as accidentally contaminated sites, locations with high concentrations of natural radionuclides such as uranium-rich mining sites, hydrocarbon deposits, sites where residues are discharged, former military sites, landfills, storage facilities, etc. The estimation of risks to humans and human radiation dosimetry is highly advanced, demonstrated, and supported by an internationally accepted framework where the probability of detrimental effects can be attributed to a unit of a dose rate received (**Stark et al., 2017**). The biological effects of radiation are determined by different parameters, including the type of radiation, amount and rate of exposure, area of the body irradiated, etc. On the other hand, environmental radiation protection and radiation dosimetry for animals and plants are still developing, and the current levels are not as advanced as the ones applicable to human exposure (**ICRP, 2008**). Studies on risk quantification to biota are necessary to expand the knowledge of potential effects and improve the dose-effect relationship (Hinton et al., 2013). The detrimental effects of radiation to humans refer to deterministic (reproduction attributes) and stochastic effects (induction of cancer) (**Bréchignac, 2003**). However, biological effects to biota are determined by observing endpoints, including mortality, morbidity, sterility, fecundity, reproduction, physiology, and genetic damage (**Bréchignac, 2003**). At population levels, these effects are being researched, considering reproduction decrease as one of the key biological effects of radiation exposure. In their review of dose assessment approaches, **Stark et al. (2017**) list parameters that affect the radiation dose received by biota, which includes the external distribution of the source (type of radiation, spatial distribution and medium composition), the internal distribution of the source (variations in radionuclide uptake – inhalation, ingestion, and absorption, life stage differences, physiological differences, seasonal variations, weighting factors for tissues and organs); organism location (movement, life stage, seasonal variations); and organism characteristics (shape and size, life stage, life span, and physiological differences).

Currently, different models and approaches are being used to estimate potential environmental impacts related to the exposure of non-human biota to ionizing radiation. Those include the use of concentration ratios, kinetic models, compartment models, and allometric approaches (**Higley and Bytwer, 2007; Beresford et al., 2010; Pentreath and Woodhead, 2001**). Some of these models and approaches have a tiered structure, where the initial screening level is usually straightforward, requiring minimal inputs and having a conservative output. Still, the assessment complexity increases as the tiers advance. The overall aim of these models is to identify sites of negligible concern with a high degree of confidence (**Copplestone et al., 2009**), and typically the assessment methodology is designed to provide conservative dose rate estimations for the worst-case scenario in a particular research context (**Stark et al., 2017**).

1.3. Use and application of the ERICA Assessment Tool

One of the crucial incentives for the development of the ERICA Integrated Approach and ERICA Assessment Tool was the need for the radiological protection framework to include non-human biota (**Brown et al., 2008**). Both the ERICA Integrated Approach and the ERICA Assessment Tool were developed through the ERICA project (Environmental Risk from Ionising Contaminants: Assessment and Management) funded under the 6th Framework Euratom Programme in the period 2004-2007 as a collective work of fifteen institutions in seven EU countries. Significant scientific contributions to the development of radiation protection at the EU level were also derived from two previous international projects, the FASSET (Framework for Assessment of Environmental Impact) project and the EPIC (Environmental Protection from Ionising Contaminants) project. The ERICA project goal was to develop an integrated approach to the scientific, regulatory and social context of the problem of the effects of ionising radiation on humans and biota, with an emphasis on the impact of technology on ecosystems, primarily through the interaction of ionising radiation and chemical agents (**Brown et al., 2008; Brown et al., 2016**). According to **Larsson (2008**), the paradigm shift that preceded the ERICA project development contributed to the fact that the project also included an aspect of decision-making and the development of regulatory frameworks, enabling an extensive practical context for implementing the ERICA Integrated Approach and the ERICA Assessment Tool. **Brown et al. (2016)** consider that the ERICA project not only incorporated the radiological protection paradigm change but also provided a robust radiological protection framework that includes far-reaching environmental impacts.

The importance of a verified and demonstrated, rather than presumed, protection of non-human biota from ionising radiation has been previously emphasised (**Bréchignac, 2003; Copplestone et al., 2004; Hinton et al., 2013; Stark et al., 2017**) and both the ERICA Integrated Approach and Assessment Tool comprehensively address these issues. The ERICA Assessment Tool performs a probabilistic quantification of environmental risks by combining radiological and radionuclide transport data while relying on the available organism and radionuclide databases (**Beresford et al., 2007; Brown et al., 2008; Brown et al., 2016**). The basis of the ERICA Integrated Approach itself is the use of generalised ecosystem representations in the form of reference organisms, which are defined as "a series of entities that provide a basis for the estimation of radiation dose rate to a range of organisms which are typical, or representative, of a contaminated environment and form a basis for assessing the likelihood and degree of radiation effects" (**Brown et al., 2008; Larsson, 2008**). The use of reference plants and animals concept aligns with the proposed ICRP methodology (**ICRP, 2008**) and the ERICA Assessment Tool's radionuclide data following the ICRP's environmental protection framework (**ICRP, 2007**). In addition, the radionuclides available in the ERICA Assessment Tool database have been selected to cover a wide range of potential biota exposures, including exposures arising from approved residue release systems, potential releases of radionuclides from radioactive waste storage, production, and remediation processes involving residues and naturally occurring radioactive materials and scenarios involving possible radiological or nuclear accidents (**Brown et al., 2008; Brown et al., 2016**). The ERICA Assessment Tool quantifies radiological risk through three different levels of assessment, with the complexity of the assessment process itself and the complexity of the required input data increasing with each transition to a higher level enabling the various users to conduct risk assessments according to their specific needs (**Beresford et al., 2007; Brown et al., 2008**). **Figure 1-1.** outlines the ERICA Integrated Approach and the interaction between assessment, risk characterisation, and management. According to **Brown et al. (2008)**, the ERICA Tool has been used to consider the potential environmental impacts of geological disposal facilities in different European countries and to assess the impacts of near-surface radioactive waste repositories in Europe and Australia, to analyse the impacts related to new environmental regulations, to quantify the environmental effects from operational and planned nuclear facilities, for the assessment of discharges from medical facilities and the assessment of biota exposure following accidents.

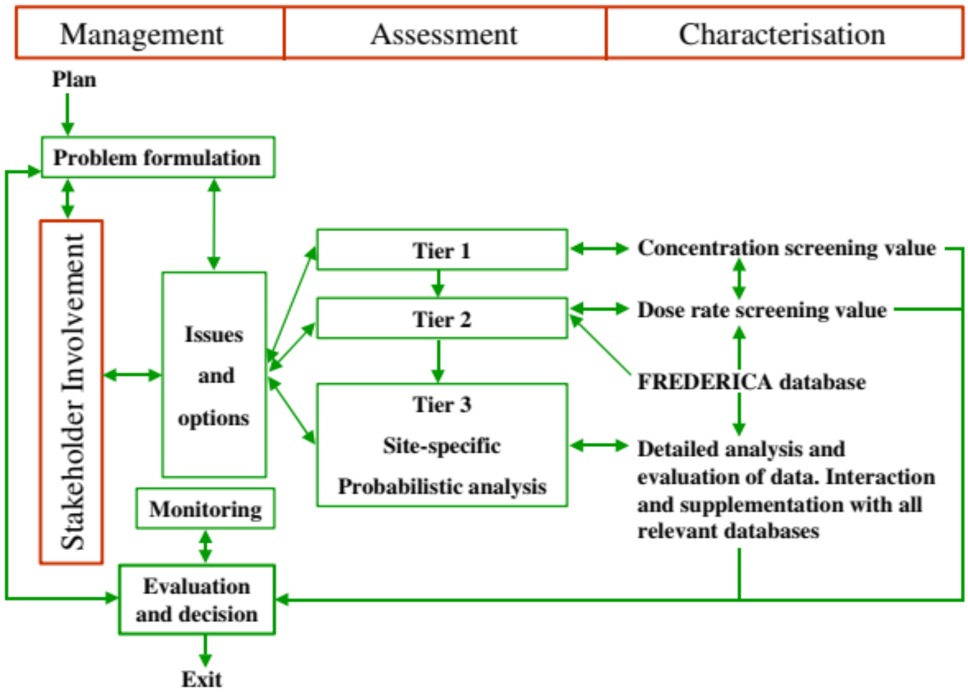

Figure 1-1. The ERICA Approach and Assessment Tool overview (adopted from **Beresford et al., 2007**)

Exposure situations are defined by the ICRP as planned, existing, and emergency exposure situations (**ICRP, 2007**). Various scientific studies have demonstrated the applicability of the ERICA Integrated Approach and the ERICA Assessment Tool at different research sites and exposure contexts. **Oughton et al. (2013)** used the ERICA Assessment Tool to estimate radiological risk at multiple mining sites in Central Asia. The effects on freshwater biota in Finland as a result of post-accident radionuclide discharges in Chornobyl were investigated by **Vetikko and Saxén (2010)**. ERICA Assessment Tool was used to compare dynamic models in radionuclide transfer in scenarios developed after the Fukushima accident (**Vives i Batlle et al., 2016a**). The effects on marine and terrestrial biota in hypothetical accidents involving the recovery of the dumped Russian submarine K-27 in Norway were estimated in the study by **Hosseini et al. (2017)**. The potential effects of radionuclide discharges from radioactive waste landfills in Belgium were estimated by **Vives i Batlle et al. (2016b)**, while **Vandehove et al. (2013)** assessed environmental risks and effects to biota in the potential release of radionuclides from Belgian nuclear installations. The impact of radionuclides in abandoned mines in Greece using the ERICA Assessment Tool was researched by **Pappa et al. (2019)**. **Ćujić and Dragović (2018)** assessed dose rates to terrestrial biota around a coal-fired power plant using ERICA Assessment Tool. **Mrdakovic Popic et al. (2020)** used ERICA to estimate the dose rate at the NORM legacy mining site. A study by **MacIntosh et al. (2023)** on radiological risk assessment to marine biota from exposure to NORM related to decommissioning offshore oil and gas pipelines.

Previous relevant radioactivity research at sites in Croatia includes studies focused on the measurement and modelling of the radiological impact of phosphogypsum landfills (**Bituh et al., 2015**), investigations of the absorption of radionuclides from coal and slag landfills into plant biota (**Skoko et al., 2017**) as well as the radiological risk to biota (**Skoko et al., 2019**), and the impact of radionuclides in the Kopački rit Nature Park (**Petrinec et al., 2018**). An example of the presentation of risk assessment results from the ERICA Assessment Tool is shown in **Figure 1-2.**

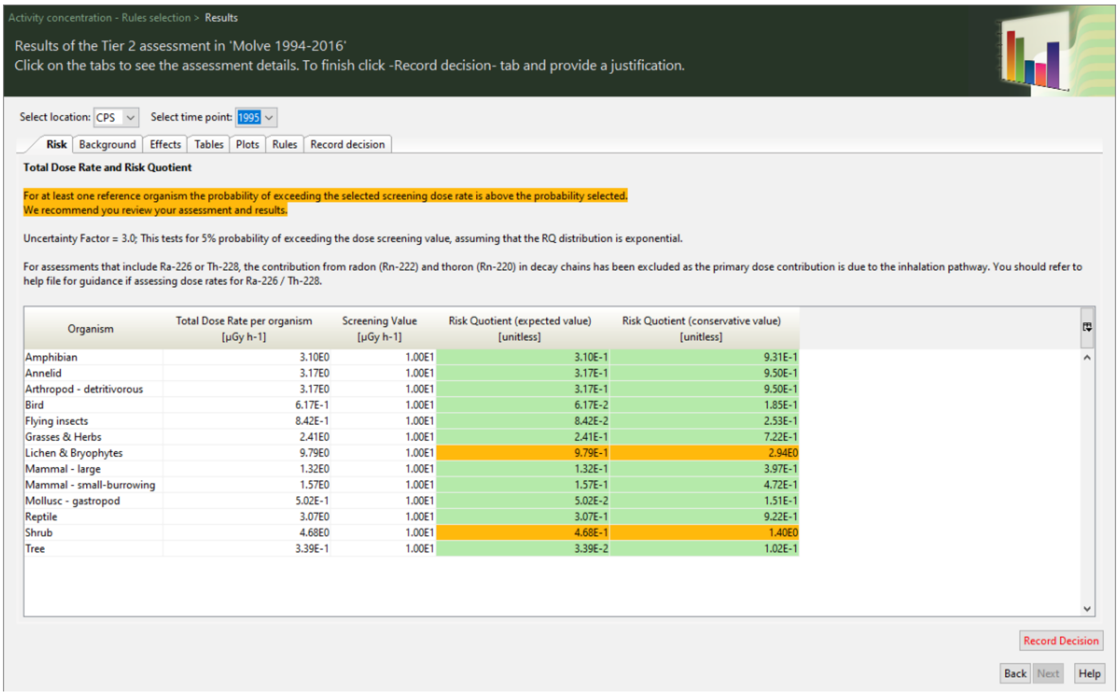

Figure 1-2. The ERICA Assessment interface

1.4. Objectives and hypotheses of research

The main objectives of this research were: (1) to determine possible connection between radiological risk quantification and soil sampling depth, (2) to determine possible relation between radiological risk quantification and soil sampling frequency, and (3) to detect potential effects of types of production activities at selected research locations on the overall radiological risk results.

These objectives were based on two main hypotheses:

1. the depth from which the soil sample is taken for analysis affects the result of radiological risk assessment due to the transport mechanisms of radionuclides in the environment;

2. the sampling frequency (once or through a periodic monitoring) at a specific location, despite the longevity of the radionuclides, affects the result of the radiological risk assessment due to the transport mechanisms of the radionuclides in the environment.

1.5. Scientific contribution

The results of this research enable better understanding of the dependence of the ERICA Tool radiological risk assessment results on the soil sampling depth and frequency at different research locations. For the first time the relationship between soil sampling depth and frequency and the resulting radiological risk to environment (biota) was investigated at the locations of the mining and oil and gas industries in Croatia. The obtained results and conclusions contribute to the field of radiological protection and provide insight to the sample selection approaches, design of future radiological risk assessment research, and interpretation of results.

2. ORIGINAL SCIENTIFIC PAPERS

Paper 1: Prlić, I., Mostečak, A., Surić Mihić, M., Veinović, Ž. & Pavelić, L. (2017) Radiological risk assessment: an overview of the ERICA Integrated Approach and the ERICA Tool use. Archives of Industrial Hygiene and Toxicology, 68 (4), 298–307.

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Radiological risk assessment: an overview of the ERICA Integrated Approach and the ERICA Tool use

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The ERICA project (Environmental Risk from Ionising Contaminants: Assessment and Management) was co-funded by the European Union as part of the 6th Framework Programme (FP EURATOM). The project was carried out between 2004 and 2007 as the collective work of 15 organisations in seven European countries. Two significant outputs of the project are the ERICA Integrated Approach and the ERICA Tool. The ERICA Integrated Approach consists of three elements: assessment, risk characterisation and management. The ERICA Tool is a practical implementation of the assessment component of the ERICA Integrated Approach and has a three-tier structure. The aim of this review paper is to give a concise overview of ERICA project outputs and their structure, updates done since their first release in 2007, as well as to provide a context for their practical application in environmental radiation protection and radiological risk assessments for various engineering scenarios.

KEY WORDS: *biota; environment; ionising radiation; radionuclides*

Environmental radiation protection and radiological risk assessment have received a lot of attention in the last two decades, partially due to the contentious nature of facilities emitting radionuclides and encouraged by accidental contaminations of the environment. There is an increasing interest and need to develop an environmental protection framework and set up a credible radiological risk assessment system. Several international organisations have invested efforts into developing methods and approaches for environmental protection from ionising radiation that would be recognised and approved at the international level (1). The initial assumption, stated by the International Commission on Radiological Protection (ICRP) in 1991, and often quoted, saying that if humans were adequately protected, non-human biota would generally be protected as well, however, lacks explicit scientific evidence to support it [Stone, 2002 as quoted in Delistraty (2)]. In addition, ICRP Recommendations from 2007 consist of considerations of the environment and furthermore, include impacts and effect on the non-human biota and environment as a whole (3, 4). Annex E (Article 280) of the United Nations Scientific Committee on the Effects of Atomic Radiation Report (5) states that ecosystems consist of various organisms with different radiosensitivities and that effects at the community level should be evaluated by mathematical modelling, model ecosystem experiments and field irradiation experiments. The output from a consensus symposium organized by the International Union of Radioecology (IUR) in November 2015 offered seven consensus statements regarding the ecological effects of radiation on populations and ecosystems while moving towards an *ecocentric* approach to environmental protection [for more details see Bréchignac (6)]. In a different paper, Bréchignac et al. (7) stated that the approach taking into consideration only humans cannot ensure the protection of all biota in all situations. Furthermore, he suggested the implementation of an ecosystem approach as a basis to support the argument for a more holistic system approach. In a paper by Oughton (8), ethical issues regarding the protection of the environment from radiations were discussed and the conclusion was that, all other things being equal, there is no reason to treat ionising radiation differently from other environmental stressors.

There have been two multinational projects relevant for the area of environmental radiation protection preceding the ERICA project. Both FASSET (Framework for Assessment of Environmental Impact) and EPIC (Environmental Protection from Ionising Contaminants) projects were supported by the European Commission, under the 5th Framework Programme (FASSET) or by the Inco-Copernicus Programme (EPIC).

The ERICA project (Environmental Risk from Ionising Contaminants: Assessment and Management) was co-funded by the European Union as part of the 6th Framework Programme (FP EUROATOM). The project was carried out between 2004 and 2007 as the collective work of 15 institutions in seven European countries. Larsson (3)

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mentioned that a shift in focus following the launch of the 6th FP made possible to include support for decisions and policy-making beside the usual pure assessment. This is highlighted in the main objective of the project: "provide and apply an integrated approach of addressing scientific, managerial, and societal issues surrounding environmental effects of ionising contamination, at a community level, with emphasis on biota and ecosystems" (1). Additionally, emphasis was put on the environmental dimension of ionising radiation i.e. ensuring that decisions related to environmental issues give appropriate weight to the exposure, effects, and risks from ionising radiation (3). Another shift in focus, adding value to the ERICA project and its outputs, concerns the radiological protection framework based not solely on humans but including overall impacts on the environment (4). This agrees with several international guidelines and recommendations mentioned earlier. Corresponding to the project's objectives, there are two significant outputs of the project: the ERICA Integrated Approach and the ERICA Tool. The ERICA Integrated Approach incorporates elements related to environmental management, risk characterisation, and impact assessment (9) where the ERICA Tool is a supportive software programme that facilitates the use of the ERICA Integrated Approach.

The aim of this review paper is to give a concise overview of ERICA project outputs, the ERICA Integrated Approach, and the ERICA Tool and the updates made since their first release in 2007, as well as to provide a context for their practical application in environmental radiation protection and radiological risk assessments for various engineering scenarios.

ERICA project

The ERICA project is successor to two other multinational EU projects: FASSET and EPIC. On the European level, the aspect of wildlife exposure to ionising radiation was first addressed in the FASSET project, which developed FRED (the FASSET Radiation Effects Database). One of the first steps in the ERICA project was to evaluate the outputs from the FASSET project using case studies. Under the ERICA project, the FRED database was extended to FREDERICA – a valuable compilation of scientific literature on radiation effect experiments and field studies, organised around different wildlife groups and, for most data, categorised according to four umbrella endpoints: morbidity, mortality, reproduction, and mutation (3). In short, FREDERICA is a radiation effects database. Project EPIC provided information on environmental transfer and radionuclide behaviour in aquatic and terrestrial ecosystems in the Arctic.

As listed in Howard and Larsson (1) the key aims of the ERICA projects were: 1) to provide clear and consistent guidance in the form of deliverables and the Tool whose Help section is extensive and provides support at each stage; 2) to ensure transparency in the derivation of information achieved by the development of the Tool; 3) to provide flexibility for the user to consider different situations than those available through default values; 4) to provide detailed information on effects via the FREDERICA base; 5) to provide the ability to address issues regarding uncertainty by using probabilistic calculations; 6) to ensure that the Tool is user-friendly and appropriate for use by people outside its development circle, and 7) to ensure free access to different outputs from the ERICA project. Essential to the ERICA Integrated Approach is the quantification of environmental risk. Data on environmental transfer and dosimetry are combined to provide a measure of exposure, which is compared to exposure levels at which detrimental effects are known to occur, and those data sets are used in calculations supported by a computer-based ERICA Tool (9). A table with a full list of project deliverables is available in Larsson (3). The D-ERICA deliverable (10), which describes the ERICA Integrated Approach and the ERICA Tool, is freely available online[1], as are all project deliverables. D-ERICA helps the user (the assessor) to formulate the problem, perform an impact assessment, and interpret and evaluate data. For most user purposes, consulting the D-ERICA and using the Tool is sufficient. The basis of using the ERICA Integrated Approach is usually an environmental situation that calls for a plan of action. Defined by ICRP Recommendations from 2007 (11), as stated in Larsson (3), situations can be categorised as planned, emergency, or existing exposure situations (Table 1).

ERICA Integrated Approach

The ERICA Integrated Approach consists of three elements: *assessment, risk characterisation, and management* (Figure 1).

Assessment is the process of estimating exposure of biota and involves the estimation or measurement of activity

Table 1 *Examples of exposure situations (10)*

Planned	Existing	Emergency
• siting a new facility • re-assessing the authorisation of an existing facility • decommissioning a nuclear facility and disposing of radioactive waste • remediation • NORM/TENORM • clearance	• exposure after an accident • residues from past or existing practices	• accidents in nuclear facilities • accidents in the transport of radioactive materials • deliberate/malevolent uses, including terrorism

[1]D-ERICA deliverables download: *https://wiki.ceh.ac.uk/display/rpemain/ERICA+reports*

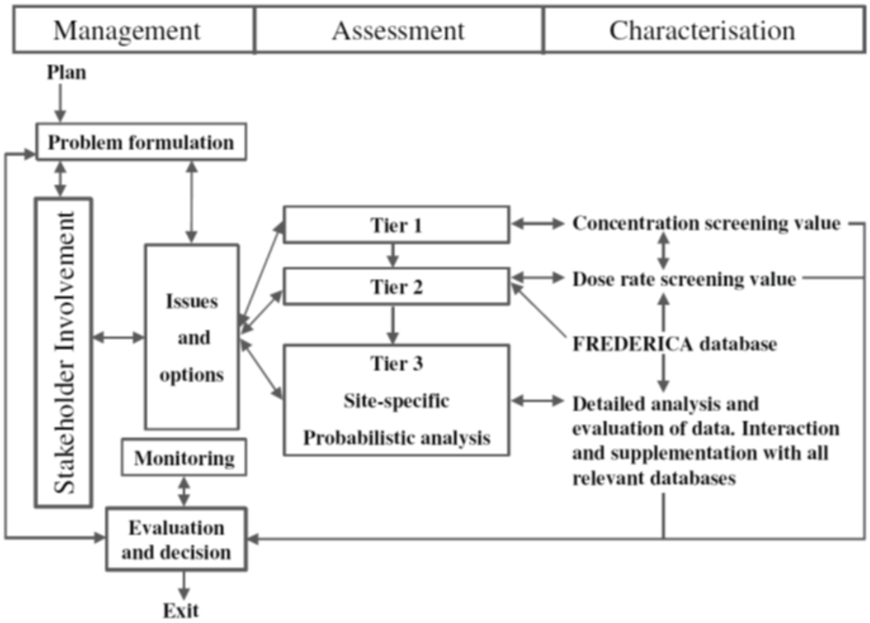

Figure 1 *Structure of the ERICA Integrated Approach (10)*

concentrations in environmental media and organisms, definition of exposure conditions, and estimation of radiation dose rates to selected biota. The proposed assessment process (which uses the ERICA Tool) has a three-tiered structure, depending on the level of concern or regulatory demand, with the highest tier (Tier 3) being the most complex, specific, and data-consuming (3). More details on the tiered assessment structure will be mentioned in the next section. Risk characterisation includes estimation of the probability and magnitude of adverse effects in biota, together with identification of uncertainties to prioritise risks as a basis for further action. Risk characterisation is based primarily on the FREDERICA database as a source of scientific information. Firstly, it relates to the assessment process in a way that it offers a scientific basis for advocating the exit of the assessment process when there are strong arguments that the situation is of negligible concern. Secondly, in cases of potential or existing concern, it provides a necessary basis for probability assessments of the effects and their possible severity. Management used in the context of the ERICA Integrated Approach refers to the process of taking decisions before, during, and after an assessment. The term covers an aspect diverse from defining the purpose of the assessment, decisions on technical issues associated with the assessment execution, general decisions related to the stakeholder interaction, and post-assessment decisions (3). In general, the ERICA Integrated Approach advises the user on the issues and options available not just during the assessment but also before and after assessment.

The basis for the ERICA Integrated Approach are generalised ecosystem representations, termed Reference Organisms. The definition of a reference organism originates from the FASSET project and refers to “a series of entities that provide a basis for the estimation of the radiation dose rate to a range of organisms which are typical or representative of a contaminated environment. These estimates, in turn, would provide a basis for assessing the likelihood and degree of radiation effects” (10). The ERICA Integrated Approach uses Reference Organisms complementary to the proposition by the ICRP (12) and the Reference Animals and Plants – RAPs methodology adopted by the ICRP (1, 3). Each reference organism has its own specified geometry and is representative of terrestrial, freshwater, or marine ecosystems. An original reference organism list is available in Larsson (3) with a list of updates from the newest version of the Tool available in Brown et al. (4).

The default radionuclides list available in the Tool has been updated in the newest version of the Tool and is consistent with ICRP's developing environmental protection framework (4). The Tool provides default information for a whole range of radionuclides chosen to cover a wide variety of conceivable exposure situations including those arising from authorised discharge regimes, potential releases from repositories for radioactive waste (including High Level Waste), operations involving NORM, and accident scenarios (9).

ERICA Tool development and structure

As mentioned earlier, the ERICA tool is a practical implementation of the assessment component of the ERICA Integrated Approach and has a three-tier structure. In the newest publication on the ERICA Tool by Brown et al. (4), tiered approaches are mentioned as a standard means of structuring risk assessments for chemicals and radioactivity. The approach used in the ERICA Tool consists of two generic screening tiers and a third site-specific tier. Three separate tiers allow the user to exit the assessment process (after satisfying certain criteria in Tiers 1 and 2) while being confident that the effects on biota are low or negligible and that no further action is necessary. In the case where the effects are not negligible, the assessment should continue. Besides guiding the user through the assessment procedure, ERICA Tool also provides a logical format for documenting the assessment procedure and recording information and decisions.

There are two basic calculation steps included in the assessment process:

1) estimation of the activity concentrations in biota and environmental media and

2) estimation of the dose rates to biota.

The Tool requires user to:

- provide a detailed description of the assessment;
- list the transfer pathways and assessment endpoints;
- upload a conceptual model; select the ecosystem to be considered;
- select the reference organisms; select radionuclides to include in the assessment;
- provide information on media activity concentrations;
- select the screening dose rate against which the results from Tiers 1 and 2 will be compared (10).

Estimation of the activity concentrations in biota and environmental media

The radionuclide activity concentrations in media (water, sediment, soil or air) are the basic inputs required in all three tiers of the ERICA Tool. In cases where sufficient data is not available from environmental monitoring, media activity concentrations need to be estimated using dispersion models (10). Users can use their own models, but screening transport models adopted from International Atomic Energy Agency (IAEA), known as the SRS 19 models (13), are part of the ERICA Tool in Tiers 1 and 2 (10). These models are generic and refer to the dilution and dispersion in the environment, requiring a minimum of site-specific input data. Transport models available within the Tool: Small lake (<400 km^2); Large lake (≥400 km^2); Estuarine; River; Coastal and Air.

Tier 2 and 3 require radionuclide activity concentrations in biota. In the ERICA Tool, whole body activity concentrations of radionuclides in biota are predicted from media activity concentrations by using equilibrium concentration ratios (CRs). Equations [1] and [2] for terrestrial and eq. [3] and [4] for aquatic ecosystems are given below (9, 10). The distribution coefficient (K_d), in equation [4] is used to relate equilibrium activity concentrations in sediments with those in water.

$$CR = \frac{\text{Activity concentration in biota whole body (Bq kg}^{-1}\text{ f.w.)}}{\text{Activity concentration in soil (Bq kg}^{-1}\text{ d.w.)}} \quad [1]$$

$$CR = \frac{\text{Activity concentration in biota whole body (Bq kg}^{-1}\text{ f.w.)}}{\text{Activity concentration in air (Bq kg}^{-3}\text{)}} \quad [2]$$

$$CR = \frac{\text{Activity concentration in biota whole body (Bq kg}^{-1}\text{ f.w.)}}{\text{Activity concentration of filtered water (Bq L}^{-3}\text{)}} \quad [3]$$

$$CR = \frac{\text{Activity concentration in sediment (Bq kg}^{-1}\text{ d.w.)}}{\text{Activity concentration in water (Bq L}^{-1}\text{)}} \quad [4]$$

The ERICA Tool relies on three default radioecology databases (one for each ecosystem) containing a complete set of CR and K_d values for all reference organisms and default radionuclides within ERICA. When it was first released in 2007, the Erica Tool contained the most comprehensive $CR_{wo\text{-}media}$ database available for wildlife (4). If adequate measured data are unavailable, the ERICA Tool calculates the activity concentrations of radionuclides in biota by multiplying the corresponding media activity concentrations with equilibrium concentration ratios (CRs). For details on the derivation of transfer parameters see Beresford et al. (14) . For aquatic environments, K_d values are used to derive activity concentrations in sediment from water concentrations and *vice versa*. Where there are no CR values available from empirical data, derivation methods are used. Since most data were available for European environments, the default reference organisms (and their characteristics) address mostly the species protected in Europe. However, in Tiers 2 and 3 of the assessment, the user (assessor) can define their own organism and its associated parameter. Therefore, the ERICA Tool can be used for assessing situations on a broader geographical scale if that representative region or site-specific data for the organism is available (15).

Estimation of the dose rates to biota

Estimation of the dose rates to biota is explained in detail in Brown et al. (9) and Beresford et al. (10). In order to calculate the dose-rate, activity concentration data are used in equations [5] and [6] given below. Through equations we derive the internal (D_{int}) and external (D_{ext}) absorbed dose rates in μGyh^{-}1. The total absorbed dose rate is the sum of internal and external absorbed dose rates derived through application of dose conversion coefficients (DCC). Equations [7] and [8] show the method of calculating weighted total dose rates for alpha, low beta, and high-beta- gamma radiation.

$$\dot{D}^b_{int} = \Sigma_i C^b_i D\, CC^b_{int,i} \quad [5]$$

where:

C^b_i is the average concentration of radionuclide *i* in the reference organism *b* (in Bq kg^{-1} fresh weight) and $DCC^b_{int,i}$ is radionuclide-specific dose conversion coefficient defined as the ratio between the average activity concentration of radionuclide *i* in the organism *b* and the dose rate to the organism *b* (in μGy h^{-1} per Bq kg^{-1} fresh weight).

$$\dot{D}^b_{ext} = \Sigma_z v_z \Sigma_i C^{ref}_{zi} DCC^b_{ext,zi} \quad [6]$$

where:

v_z is the occupancy factor (i.e. fraction of time that the organism *b* spends at a specified position *z* in its habitat). $\dot{D}^b_{ext}$ is the average activity concentration of radionuclide *i* in the reference media of a given location z in (in Bqkg^{-1} fresh weight (soil or sediment) or Bq L^{-1} water). $DCC^{b}_{ext,zi}$ is the dose conversion coefficient for external exposure defined as the ratio between the average activity concentration of radionuclide i in the reference media corresponding to the location z and the dose rate to organism b (in µGy h^{-1} per Bq kg^{-1} fresh weight or µGyh^{-1} per Bq L^{-1}).

$$DCC_{int}=wf_{low\beta}DCC_{int,low\beta}+wf_{\beta+\gamma}DCC_{int,\beta+\gamma}wf_{\alpha}DCC_{int,\alpha} \quad [7]$$

$$DCC_{ext}=wf_{low\beta}DCC_{ext,low\beta}+wf_{\beta+\gamma}DCC_{ext,\beta+\gamma}wf_{\alpha}DCC_{ext,\alpha} \quad [8]$$

where:

wf are the weighting factors for various components of radiation (low β, $\beta+\gamma$, and α) and are dimensionless.

For more details on Dose Conversion Coefficient (DCC) calculations see chapter 4.4. in Beresford, et al. (10).

Assessment process in Tiers 1, 2, and 3

An outline of specifics, uses and results in each of the three tiers is given below.

Tier 1 assessment

The Tier 1 assessment is simple, highly conservative, and requires a minimum of input data. If assessment meets a predefined screening criterion, the user can exit the process. It is assumed that many situations will be exempt from further evaluation in this tier. The default screening criterion in the ERICA Integrated Approach, for all ecosystems and organisms, is an incremental dose rate of 10 µGy h^{-1}. This value was derived through a pioneering use of the species sensitivity distribution analysis performed on chronic exposure data in the FREDERICA database (1, 3). User-defined values and other screening dose rate values can be used if necessary.

An essential step in Tier 1 is the calculation of the Environmental Media Concentration Limits (EMCLs). The EMCL is the activity concentration in the selected media that would result in a dose rate to the most exposed reference organism equal to that of the screening dose rate, see equation [9]. In other words, the screening dose rate is back-calculated to yield an EMCL value for all reference organism/radionuclide combinations.

$$\text{EMCL}=\frac{\text{SDR}}{\text{F}} \quad [9]$$

where:

F is the maximum dose rate that an organism will receive for a unit activity concentration of a given radionuclide in an environmental medium (in µGy h^{-1} per Bq kg^{-1} dry weight, µGy h^{-1} per Bq l^{-1} or µGy h^{-1} per Bq m^{-3} air) and *SDR* is the screening dose rate (in µGy h^{-1}) which is by default set to a value of 10 µGy h^{-1}. For *F*, the default location within the habitat is selected based on the configuration that will result in the maximum exposure of the reference organism (e.g. for the terrestrial soil invertebrate this is soil, hence the index *si*), see equation [10]. F values are calculated using information on CR and DCC values probabilistically by performing a Monte Carlo approach (4, 9).

$$F=[DCC_{int,si}CR_{si}+DCC_{ext,si}] \quad [10]$$

Across all reference organisms, the minimum EMCL value is selected to define the value for a particular radionuclide *n*, i.e. radionuclides have a single value but can have different limiting organisms. Therefore, in Tier 1, the user cannot select reference organisms.

After the most restrictive EMCL for each radionuclide *n* is determined, the Tool compares the input media activity concentrations, whether they are site-specific values or derived through the use of models, with a risk quotient (RQ_n) for each specific radionuclide *n*. The risk quotient can be expressed as an assumed value divided by the screened value. The total risk quotient *RQ* is a sum of risk quotients RQ_n for each radionuclide *n*, see equation [11].

$$RQ=\Sigma_n RQ_n=\Sigma_n \frac{M_n}{EMCL_n} \quad [11]$$

where:

M_n is the measured or predicted maximal activity concentration for radionuclide *n* in the medium (in Bql^{-1} for water, Bqkg^{-1} dry weight for soil or sediment or Bqm^{-3} for air), $EMCL_n$ is the Environmental Media Concentration Limit for radionuclide *n* (in same units as the media).

If the sum of risk quotients is less than one, the user can be assured that there is very little probability that the assessment dose rate to any organism exceeds the screening dose rate i.e. the risk to non-human biota is negligible. If the RQ is greater than one, the user is advised to continue with the assessment since a deep study of the situation is required.

Tier 2 assessment

Where Tier 1 is conservative, Tier 2 allows the user to be more interactive: to change the default parameters and/or to select specific reference organisms. Estimated total weighted absorbed doses (sums of internal and external doses) for each reference organism in the assessment are compared with dose rate screening values selected by the assessor. The risk quotient that is derived is shown in equation [12].

$$RQ_{org}=\frac{DR_{org}}{SDR} \quad [12]$$

where:

RQ is the risk quotient for reference organism *org*; *DR* is the estimated dose rate for reference organism *org* (in µGy h^{-1}); SDR is the screening dose rate selected by the assessor (in µGy h^{-1}). User interaction in this tier refers to the user's flexibility in the selection of parameters used in equations [1]-[3] and [5]-[6]: CR values, K_d values, percentage dry weight soil or sediment, occupancy factors, and radiation weighting factors. In Tier 2 and 3, users can add organisms and isotopes if they are not represented in ERICA. The main difference between Tiers 1 and 2 is the value for activity concentration, which is very conservative in Tier 1, i.e. maximum activity concentrations are used, whereas in the Tier 2 the recommended values used are expected (or best estimated) values, i.e. the most representative of an area. Depending on the amount of user interaction, the Tool follows a certain set of rules (available in the Help section) in data calculation and extrapolation. Tier 2 differs from Tier 1 regarding risk quotient values. In Tier 2, RQs are based on estimated values, although conservative RQs are also available. Conservative values are obtained by introducing the Uncertainty Factor (UF), which is an approximation applied to account for the uncertainty of the dose rate estimation. The exact definition is: the ratio between the 95th, 99th, or any other percentile (above the expected value), and the expected value of the probability distribution of the dose rate (and RQ) (9). Assessors can define their own UF values. The uncertainty factor also has a role in maintaining conservativism between Tiers 1 and 2. In the case where the same values are used in both tiers, conservative estimates from tiers should correspond to one another but would not be identical. Brown et al. (9) explain this by different distributions that characterise values used in the tiers. In Tier 1, the EMCL values are derived from uncertainty propagation based on real probability density function (PDF); e.g. CR values are often characterised by lognormal distributions. However, in Tier 2, UF is applied to the expected value in order to derive the RQ. In other words, Tier 2 assumes the PDFs of the RQ and can be approximated using an exponential distribution where in Tier 1 the derived PDFs display a combination of different functions that may or may not be of exponential form. For a detailed explanation on the use of exponential distribution in deriving UFs see Brown et al. (9). The criteria suggested for Tier 2 results evaluations are shown in Table 2.

The calculated values and other available information allow the assessor to decide whether to proceed with the assessment. In certain cases, automatic progression to Tier 3 is not necessary e.g. if refined or new data is available. Nonetheless, Brown et al. (9) mention that the use of site-specific data instead of generic data might not always prove to be justified. To help the assessor, the Tool provides a context for decision-making in the form of tabs labelled as "Background" and "Effects". The background tab offers information on background exposure rates and Effects tab contains a summary of information on known biological effects of ionising radiation for every reference organism included in the assessment (based on the FREDERICA database).

Tier 3 assessment

Tier 3 consists of a probabilistic risk assessment in which uncertainties within the results may be determined using sensitivity analysis. Situations that call for full Tier 3 assessment are often complex and unique. Therefore, it is difficult to provide straightforward guidance on how Tier 3 assessment should be implemented. The specific context necessary for decision-making requires an experienced, knowledgeable assessor or consultation with an appropriate expert. User flexibility is present in Tier 3 as well as in Tier 2. Apart from editing various parameters, users can assign a probabilistic density function (PDF) to them. The tool supports exponential, normal, triangular, uniform, lognormal, logtriangular, and loguniform distribution. Additional details regarding Monte Carlo probabilistic simulations used in the ERICA Tool are given in Brown et al. (4, 9). Data and numerical, model and scenario uncertainties in the ERICA Integrated Approach and Tool are further discussed in Oughton et al. (16), as well as conceptual, societal, and ethical uncertainties. Results available from Tier 3 offer no information on risk quotients since at this stage of the assessment; screening dose rates are no longer suitable. The results tab includes deterministic data (in the tabulated form) and probabilistic data (related to PDFs and in the form of figures). Supporting information for interpretation can be found in the FREDERICA database. Together, these allow the user to estimate the probability and magnitude of the environmental effects likely to occur. Finally, the acceptability of the risk to non-human species can be determined through discussion and agreement with stakeholders. More information on decision-making and stakeholder interaction within the ERICA project is given in the following section.

Stakeholder engagement aspect in ERICA

As mentioned by Zinger et al. (17), there is an emphasis on the importance of stakeholder involvement and public

Table 2 *The criteria and recommendations for Tier 2 results (9)*

$RQ_{cons} < 1$	$RQ_{cons} \geq 1$ / $RQ_{exp} < 1$	$RQ_{exp} \geq 1$
• low probability that the screening dose rate is exceeded • environmental risk is arguably negligible	• substantial probability that screening dose rate is exceeded • assessment should be reviewed (Tier 2)	• screening dose rate is exceeded • assessment should continue (Tier 3)

participation in policy-making, especially concerning environmental issues and technology assessment. Additionally, the requirement for stakeholder participation in decision-making has been stated in several official publications, legislation, and implementation documents, on both EU and worldwide level. The term »stakeholder« is used in the ERICA project in its broadest sense; i.e. an individual or a group affected by or having an interest in a specific issue. The method used in stakeholder interaction was to include stakeholders as early as possible and for the engagement to be continuous and ongoing (17). One of the most innovative aspects of the ERICA project was the central role of stakeholders by their participation in the End-Users Group (EUG) events. There were seven EUG categories: regulatory, national advisory body, academia, non-governmental organisation, industry, consultants, and inter-governmental organisation with 60 organisations registered as EUG members [for more details see Zinger et al., (17)]. Besides the consultation regarding the development of the ERICA Integrated Approach, stakeholders contributed to the development of the ERICA Tool, its quality, and application. Many experts, policy makers, and decision-makers in different areas provided views from the user's perspective (3). We should point out the conclusion from Zinger et al. (17) that, in the UK and Sweden, the ERICA Integrated Approach and Tool will be used as part of their regulatory practice. The ERICA Tool contains a generic list of stakeholders that can be used to help group stakeholders into different classes.

Decision-making in ERICA

Decision-making in the context of the potential, perceived, or actual environmental concern is usually governed by the Environmental Impact Assessment (EIA) where, if relevant and depending on the circumstances, consideration of effects (or potential effects) of ionising radiation can be a minor or major concern within the overall EIA (3). Due to the nature of facilities related with radionuclide emission, substantial stakeholder attention is likely to be present. Aspects of decision-making in environmental radiation protection and use of the ERICA Integrated Approach in a hypothetical case study are discussed in detail in Zinger et al. (18). It is important to emphasise that ERICA's tiered approach to risk assessment does not provide a straightforward yes/no decision, especially if the situation requires Tier 3 to be implemented. The necessary flexibility in the assessment procedure results, *inter alia*, from a difference in legislation between countries and national standards and/or criteria. As mentioned in the Tiered assessment overview, problem formulation in ERICA is essential and directly affects how the assessment will be carried out. The factors mentioned by Zinger et al. (18) are susceptible to modification and revision as assessment progresses or in post-assessment. In some cases, a decision that has been taken after a full Tier 3 assessment might need to be reconsidered in the light of new information, a new problem formulation or a change in uncertainty. Deliverable D8 Considerations for applying the ERICA Integrated Approach (19) states "decisions regarding the acceptability of a plan or project will necessarily involve consideration of a range of consequences, including potential impacts on human health, and environmental, economic, ethical, and societal factors" (17). The selection of the approach for socio-economic analysis depends on the specific situation. Furthermore, Zinger et al. (18) mention a stepped approach to socio-economic analysis recommended by the Nordic Council of Ministers that is consistent with the ERICA Integrated Approach recommendations. The conclusion regarding the decision-making aspect of the use of the ERICA Integrated Approach is that, although three tiers guide decision-makers in determining whether there is likely to be an impact on non-human species, once the assessment is complete and one of three outcomes is identified, other factors may still influence which actions are to take place. Data and results are not a standalone factor, but do however represent an important piece in the overarching context of responsible and transparent decision-making.

The newest version of the ERICA Tool

The newest version of ERICA Tool, to date, is the release from February of 2016 (ERICA Assessment Tool 1.2 updated). Changes mostly refer to the updates and amendments of the $CR_{wo\text{-}media}$ database to provide consistency with the IAEA and ICRP, changes in the reference organism list, dosimetric parameters, distribution coefficients, and EMCL values. Methods of missing data derivation in the Tiers have been improved as well. The limitations that are still present concern the assessment of impacts from certain radionuclides in gaseous forms, single location and time data-entry option of the ERICA Tool and dealing with radionuclide decay series (i.e. system being too rigid in this aspect) (4).

Examples of practical use

Bréchignac et al. (6) list a number of sites with particular relevance to the topic of understanding radiation effects on both population and ecosystem levels: accidentally contaminated sites; sites with a high level of natural radioactivity, well-characterised sites which may include: uranium mining sites, gas and oil sites, marine sites receiving exhaust pipes; former nuclear test sites; waste management/waste disposal sites, etc.

The importance of demonstrated, rather than assumed, protection of non-human biota from the effects of ionising radiation was mentioned by Doering (15). In his report for the Australian Radiation Protection and Nuclear Safety Agency, a review of ICRP's framework and ERICA is given with specific regard to its applicability to the Australian context (especially the uranium mining industry). The use

Table 3 *Concise summary of the Tiers (9)*

Tier 1
• highly conservative
• requires minimal data input (maximum measured media concentrations suggested as input)
• simple and can be used by non-specialist users
• compares input media concentrations to Environmental Media
• activity concentration limits calculated for the most limiting reference organism for each radionuclide
• if the Tool recommends that the assessment can be exited, the situation can be considered to be of negligible radiological concern
Tier 2
• less conservative screening tier
• the user can edit transfer parameters
• media and biota activity concentrations can be an input (best estimate values are recommended)
• estimated whole body absorbed dose rates compared directly to the screening dose rate
• "Traffic light" system indicates if the situation is: - of negligible concern (with a high degree of confidence) the user is recommended to exit the assessment process - of potential concern – the user is recommended to review and amend the assessment - of concern – the user is recommended to continue the assessment
• results can be assessed against summarised tables of effects and exposure due to naturally occurring radionuclides
Tier 3
• not a screening tier so no screening dose rate
• not prescriptive and does not have "yes/no" answers
• provides the user with guidance, template, and tool to help conduct a more detailed assessment
• probabilistic and sensitivity analyses
• access to up to date on-line database of radiological effects

of ERICA is suggested as part of a development of national guidance on the protection of non-human species, with necessary adaptations for Australian situations.

According to Brown et al. (4), following its release, the ERICA Tool has been widely used in numerous applications worldwide. Some of the examples include: for consideration of potential environmental impacts from deep geological disposal facilities in various European countries and assessments of the impact of near-surface radioactive waste repositories in Europe and Australia; scoping analyses in line with newly-introduced environmental regulations; quantifying environmental impacts from operating and planned nuclear power stations; assessing releases from medical facilities; for exposure estimates of biota following accidents.

Application of ERICA in the ecological risk assessment of Central Asian mining sites was studied by Oughton et al. (20), where assessment results proved useful for identifying priority areas for future field studies. Vetikko and Saxén (21) studied the application of the ERICA Assessment Tool in freshwater biota in Finland, focusing on incremental dose rate resulting from Chernobyl-derived radionuclides. ERICA Tool was used to assess the impacts on both marine (22) and terrestrial (23) environments in case of a hypothetical accident involving the recovery of a dumped Russian submarine K-27. In their recent analysis of the impacts of radiation on the environment, the United Nation's Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) approved components of the ERICA approach following the 2011 accident at the Fukushima Dai-Ichi Nuclear Power Plant (4). Details on inter-comparison of dynamic models for radionuclide transfer in Fukushima accident scenario are available in Vives and Batlle et al. (24). The impact of releases from a Belgian LLW repository to local biota using the ERICA approach is discussed in Batlle et al. (25). Prediction of environmental risks of radioactive discharge from Belgian nuclear power plants and impacts on wildlife was evaluated by using the ERICA Tool in Vandenhove et al. (26).

CONCLUSION

The advantages of using the ERICA Integrated Approach and ERICA Tool can be summarised by stating that it offers an affordable, accessible, and user-friendly method of conducting radiological risk assessments, while still providing a highly significant scientific basis for a complex decision-making process in the interdisciplinary context of environmental issues.

A short overview of the projects and work preceding the ERICA project shows the amount of effort invested in the development of both Integrated Approach and Tool and their role and contribution to the protection of the environment from ionising radiation. Updated versions of the Tool give credibility to continuous improvement and its importance in the area of environmental risk assessments, as well as encourage users to rely on the ERICA Integrated Approach in their work.

The various papers listed herein present the variety of ERICA Integrated Approach and Tool's uses and applicability to a whole range of different environmental challenges that can be answered in a clear and comprehensive manner. The approach used by ERICA provides an improvement in radiological risk assessment methodologies since the protection threshold for radiological substances was, for the first time, set using a transparent and objective process (27). In general, the outputs of the ERICA project substantially improved the ability of a wide range of users to carry out assessments and are making significant contributions to key international initiatives in this field (1).

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Radiološka procjena rizika: pregled uporabe ERICA integriranog pristupa i ERICA alata

Projekt ERICA (ekološki rizik od ionizirajućih onečišćivača: procjena i upravljanje) sufinanciran je od Europske unije u sklopu Šestog okvirnog programa (FP Euroatom). Projekt je proveden između 2004. i 2007. godine kao kolektivni rad 15 organizacija u sedam europskih zemalja. Dva su značajna rezultata projekta: ERICA integrirani pristup i ERICA alat. ERICA integrirani pristup sastoji se od triju elemenata: procjene, karakterizacije rizika i upravljanja. ERICA alat je praktična primjena komponente procjene unutar ERICA integriranoga pristupa te ima trorazinsku strukturu. Cilj je ovoga rada dati ne samo kratak pregled rezultata projekta ERICA i njihove strukture nego i ažuriranja rezultata od njihova prvog objavljivanja 2007. godine, te pružiti kontekst za njihovu praktičnu primjenu u zaštiti okoliša od zračenja i procjeni radiološkoga rizika za razne inženjerske primjene.

KLJUČNE RIJEČI: *biota; ionizirajuće zračenje; okoliš; radionuklidi*

Paper 2: Getaldić, A., Surić Mihić, M., Veinović, Ž., Skoko, B., Petrinec, B. & Prlić, I. (2023) Comparison of Different Radiological Risk Assessment Scenarios at a Coal Ash and Slag Disposal Site. Minerals, 13, 832.

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Article

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

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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 μGyh^{-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 Ćujić 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}U = 53.5 ± 23.8 Bqkg^{-1}, activity concentration for ^{226}Ra = 57.9 ± 32.8 Bqkg^{-1}, and activity concentration for ^{232}Th = 47.1 ± 19.7 Bqkg^{-1} [33].

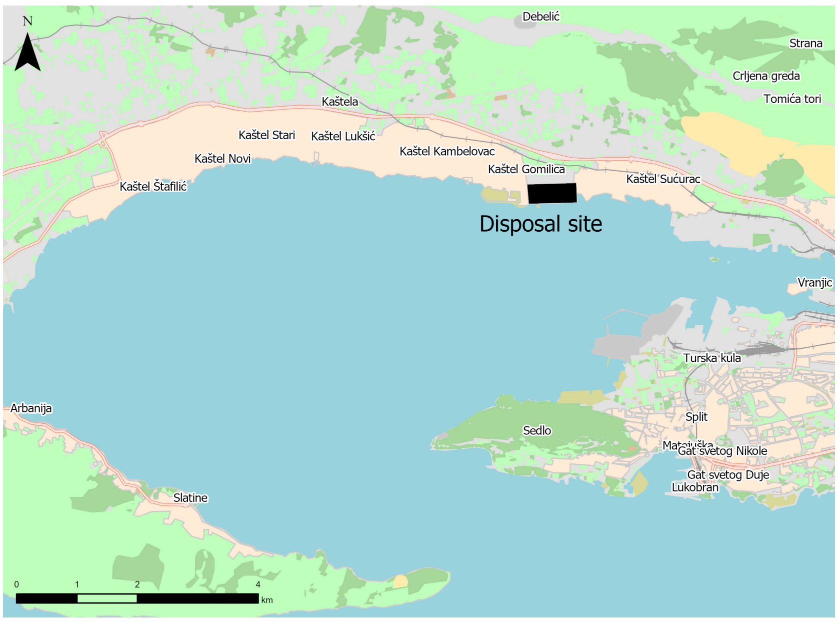

Figure 1. Site location.

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 μGyh^{-1} per $Bqkg^{-1}$ 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 β, $\beta + \gamma$, and α) [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 μGyh^{-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 conservativism between tiers 1 and 2, whose values should correspond. Beresford et al. [16] define the uncertainty factor (UF) as "the ratio between the 95^{th} and 99^{th} 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}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}Co and a relative efficiency of 74.3% at 1.33 MeV ^{60}Co. Activity concentrations of ^{238}U, ^{226}Ra, and ^{232}Th were determined from their decay products. Photopeaks at 609 keV, 1120 keV, and 1764 keV for ^{214}Bi and 295 keV and 352 keV for ^{214}Pb were used to determine the activity concentration of ^{226}Ra, those of ^{228}Ac at 338 keV, 911 keV, and 968 keV were used to determine the activity of ^{232}Th, and photopeaks at 63 keV and doubled 93 keV for ^{234}Th were used to determine the activity of ^{238}U, where the activity of ^{210}Pb was determined from its γ-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 (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}U ^{232}Th ^{235}U ^{226}Ra ^{210}Pb	Tree

Table 2. Activity concentrations (Bqkg^{-1} dry mass) measured in borehole samples (B2, B3, and B4) from different depth ranges.

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

* ^{235}U activity concentration was estimated based on ^{235}U/^{238}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).

Isotope	ERICA Tool Default CR Value	Site-Specific CR (From [32,34])
U	0.006473 ± 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^{-1}. 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 ^{226}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 ^{226}Ra and ^{238}U. The results concerning the depth range of 4–6 m showed that in addition to

^{226}Ra, ^{210}Pb and ^{210}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, ^{210}Bi, with a half-life of 5 days, is listed here as a direct progeny of ^{210}Pb. At this depth range, the radionuclide distribution of the internal dose rate in all samples showed that ^{226}Ra was the dominant radionuclide, accounting for approximately 70% of the internal dose rate, followed by ^{210}Pb and ^{210}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 ^{226}Ra, both ^{210}Pb and ^{210}Bi were detected by the tool as contributors to the internal dose rate, but distributed more evenly, with ^{226}Ra accounting for around 40% of the internal dose rate, and ^{210}Pb and ^{210}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. ^{226}Ra and ^{210}Pb primarily contribute to the total dose rate, and specific data is presented in Table 4.

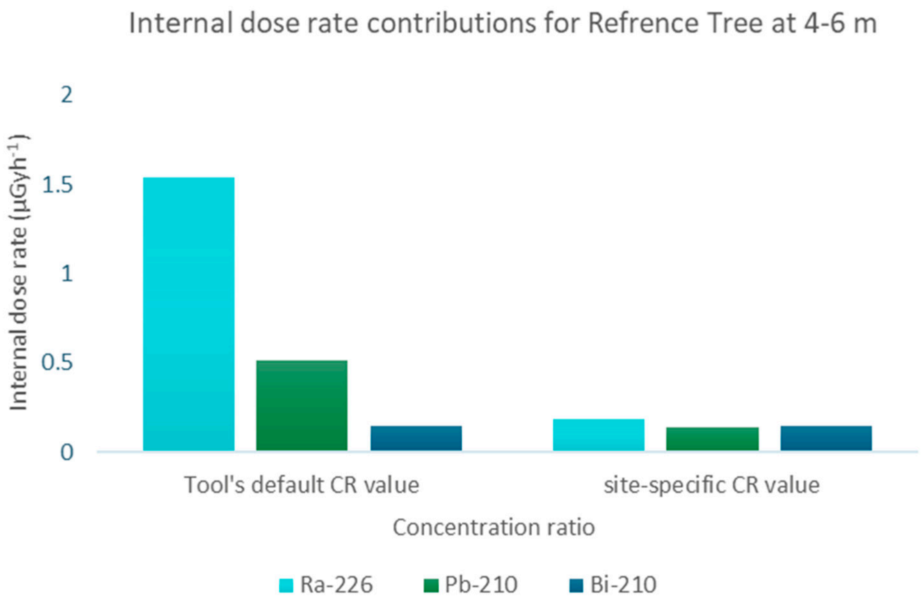

Figure 2. Comparison of distributions for radionuclides that contribute to the internal dose rate for reference tree.

Table 4. Radionuclide dose rate contribution (μGyh^{-1}) 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^{-1}] for Reference Tree in Assessments using Tool's Default CR Values			Total Dose Rate per Radionuclide [μGy h^{-1}] for Reference Tree in Assessments using Site-Specific CR Values (Adopted from [34])		
	B2	**B3**	**B4**	**B2**	**B3**	**B4**
^{238}U	0.574	0.585	0.594	0.099	0.101	0.076
^{232}Th	0.004	0.008	0.014	0.020	0.037	0.024
^{235}U	0.037	0.034	1.619	0.010	0.009	0.006
^{226}Ra	7.497	7.117	11.700	1.581	1.501	1.193
^{210}Pb	0.514	0.750	0.514	4.204	6.138	7.672

The presence of ^{210}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 Skowrohska-Smolak [47], the ^{210}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 ^{210}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 ^{210}Pb root uptake can be regarded as negligible. This assumption of the importance of atmospheric deposition in relation to the root uptake of ^{210}Pb is in line with the conclusions from a previous study at the exact location using surface soil samples, where similar activity concentrations of ^{210}Pb were detected in both plants from the disposal site and the control site, indicating atmospheric deposition as a major pathway for ^{210}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.

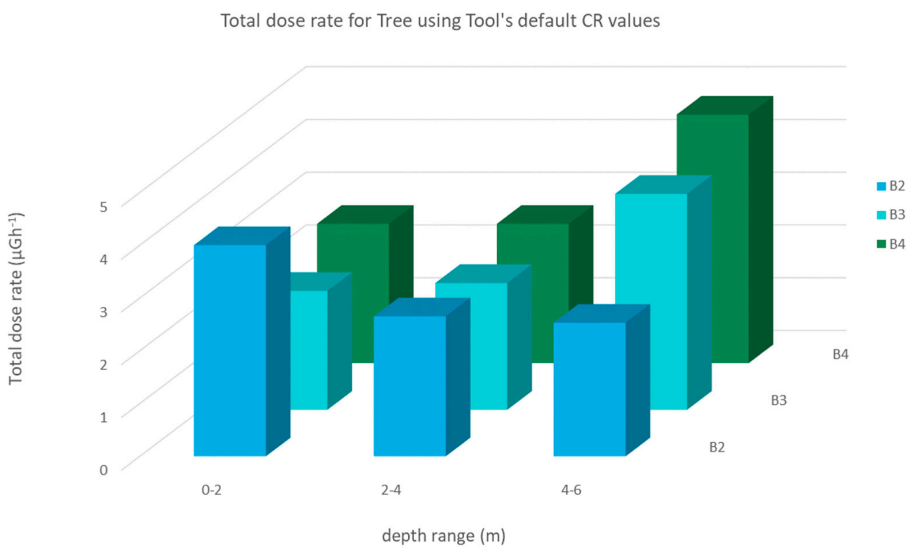

Figure 3. The total dose rate (μGyh^{-1}) for the reference tree at different depth ranges calculated using the tool's default CR values.

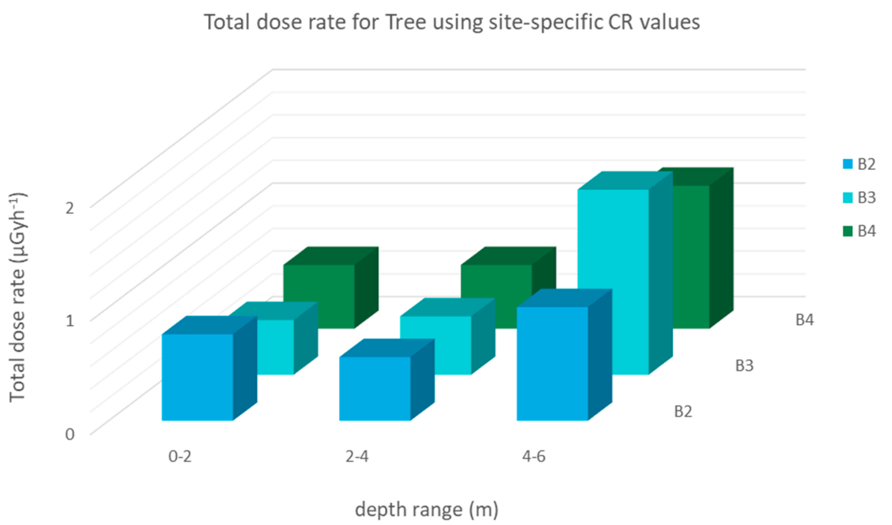

Figure 4. The total dose rate (μGyh^{-1}) for the reference tree at different depth ranges calculated using site-specific CR values.

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.

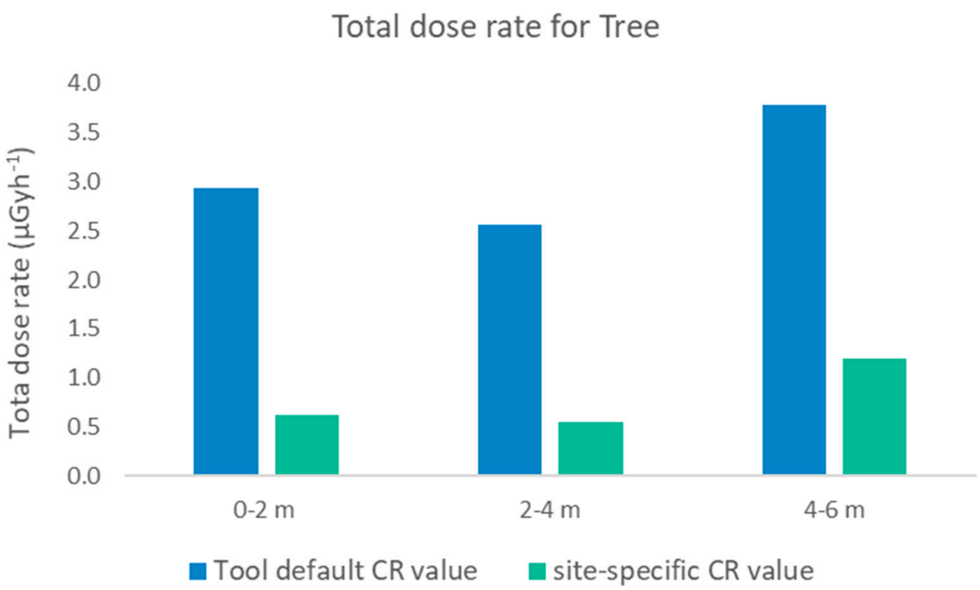

Figure 5. Comparison of estimated total dose rate for reference organism by different input data and used CR values.

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^{-1} [34], while for larger depths (4–6 m) the estimated total dose rates are twice as high (1.19 μGyh^{-1}).

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^{-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, 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 ^{230}Th and ^{210}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}Th in the coal ash were estimated from activity concentrations of its progeny (^{228}Ra and ^{228}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}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}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 μGyh^{-1}, much less the limit of 400 μGyh^{-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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Original scientific paper

Remediation of coal ash and slag disposal site: Comparison of radiological risk assessments

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Abstract

Residuals from coal combustion are known as a potential source of radiation exposure, especially in cases where the coal used in the combustion is characterized by increased radioactivity, resulting in coal ash and slag with potentially high activity concentration of radionuclides. This paper presents the results of the radiological risk assessments based on the ERICA Tool approach, used to estimate dose rates to terrestrial biota in the proximity of a coal fired thermal power plant in Croatia. The study consists of three radiological risk assessments using environmental data on activity concentration (Bqkg^{-1}) from samples collected prior to the remediation of the disposal site and samples after the remediation implementation was completed. The resulting total dose rate to biota derived using data prior to the remediation ranged from 3.28 μGyh^{-1} to 147.68 μGyh^{-1}. Assessment results of total dose rate based on the data from the studied area after remediation ranged from 0.23 μGyh^{-1} to 18.06 μGyh^{-1}. The results showed that after the remediation only the total dose rate for lichens and bryophytes slightly exceeded ERICA Tool conservative screening value of 10 μGyh^{-1}, which implies that environmental risks in relation to exposure to the disposal site can be considered negligible. The study results confirm the applicability of the ERICA Tool for the assessment of potential radiological impact and the effective remediation implementation at the coal and ash slag disposal site.

Keywords:
radiological risk assessment; NORM; coal fired power plant; remediation; environmental monitoring

1. Introduction

Although the use of renewable energy sources is on the rise, worldwide statists show that coal use as a primary energy source still accounts for approximately a quarter of the global energy mix (**Ritchie et al., 2022**). In addition, European countries still significantly rely on fossil fuels (**Martins et al., 2018**). The disposal of residues related to coal use is associated with different environmental challenges, engineering solutions, and resource management strategies. The disposal of coal combustion residues (coal fly ash and slag) is often related to large waste quantities and requires that specific environmental and safety standards are met (**Hirschi and Chugh, 2019**).

Naturally occurring radioactive materials (NORM) are found in different natural resources. Various industrial processes generate NORM residues and present a potential for radiation exposure. The international community has recognized the risk associated with natural radioactivity and radiation exposure through different legal acts (e.g. The European Council 2013/59/Euratom) and international guidance documents (**IAEA, 2003; IAEA, 2013; IAEA, 2022; ICRP, 2019**). Coal combustion presents a potential source of radiation exposure, where the resulting coal ash and slag can contain considerable activity concentrations of radionuclides, which are usually related to the activity concentrations present in the parent coal used in the combustion in the first place (**IAEA, 2003**). These radionuclides, contained in coal ash, can later be transported to the environment by different pathways, like dispersion and leaching, and can be associated with detrimental environmental and health effects. In order to mitigate potential adverse effects, NORM-related industries are required to establish and implement radiological protection principles, including the principle of justification and, in different industrial stages, optimization through the use of a graded approach (**Lecomte, 2020**).

The international community provided recommendations on radiation protection, including non-human biota (**ICRP, 2007; NEA, 2007**), and the need for it to be sci-

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entifically and independently assured through the use of ecological risk assessment paradigm, defined dose limits and reference organisms, and considering the geographic region (**Delistraty, 2008**). Potential environmental impacts related to the exposure of non-human biota to ionizing radiation can be estimated using different models and approaches, including concentration ratios, kinetic models, compartment models, and an allometry approach (**Higley and Bytwer, 2007; Beresford et al., 2010; Pentreath and Woodhead, 2001**). The ERICA Tool was developed as a part of the European Union co-funded 6th Framework Program EURATOM project named Environmental Risk from Ionising Contaminants Assessment and Management (ERICA). The ERICA Tool uses concentration ratios to calculate activity concentrations in the whole organism, and together with the activity data from environmental media, estimates dose rates (internal, external, and total) to organisms (**Johansen et al., 2012**). In the ERICA Tool, the radiological risk assessment is defined by comparing the calculated total dose rate and the exposure levels associated with known detrimental radiation effects (**Brown et al., 2008; Brown et al., 2013**). The ERICA Tool and Approach is applicable to various exposure situations, including planned and existing exposure situations and activities, including NORM/TENORM (**Beresford et al., 2007**). Several studies have used the ERICA Tool to assess the radiological impacts of NORM-related industries. A study by **Ćujić and Dragović (2018)** assessed dose rates to terrestrial biota around a coal-fired power plant in Serbia. Research from **Mrdakovic Popic et al. (2020)** used ERICA for the estimation of dose rate at the NORM legacy mining site in Norway. ERICA Tool was used for radiological risk assessment at different mining sites in Central Asia in a study by **Oughton et al. (2013)**. **Skoko et al. (2019)** used the ERICA Tool for risk assessment of the coal ash and slag legacy disposal site in Croatia.

Previous research conducted at the location includes studies of radionuclides in the soil and their distribution (**Kovac and Bajlo, 1996; Ernečić et al., 2014; Radolić et al., 2019; Dvoršćak et al., 2019**), investigations related to coal used at the power plant and resulting waste (**Marović et al., 2008**), and environmental impacts studies of the disposal site (**Skanata et al., 1996a; Marović et al., 1997; Marović et al., 2004; Bituh et al., 2017**).

Although the management of NORM residues is increasingly focusing on approaches other than disposal, such as recycling and use as by-products, the disposal of NORM residues is still very much present. In the context of NORM residue disposal, the implementation of remediation must include principles of optimization and justification and ensure that the radiological and environmental impacts of these activities are within the acceptable limits, and provide long-term protection and safety (**IAEA, 2013**).

The aim of this paper is to estimate potential impacts to the terrestrial biota from coal ash and slag disposal using the Erica Tool and compare the estimations in relation to the implemented remediation of the disposal site. The study used several spatial and temporal data sets before and after the site remediation was performed. The results from different assessments provide insight into the degree to which the deposited material is contained and can be used as a reference in the design of future estimations and assessments of radiological impacts at similar locations. In addition, the study confirms the importance of environmental monitoring in the implementation of radiological protection in NORM-related industries.

2. Materials and Methods

This study compares the results of three radiological risk assessments performed with the ERICA Tool for terrestrial biota at a disposal site near a coal-fired power plant in Croatia. The disposal site contains large quantities of residues resulting from coal combustion.

2.1. Assessment site

The study focused on the location of the coal-burning power plant “TE Plomin” in Croatia, situated on the eastern coast of the Istrian Peninsula, in the northern part of the Adriatic Sea (see **Figure 1**). Areas with slightly elevated natural background radioactivity are present in the Istrian region (**Marović et al., 2004**), studies on radioactivity in the soil also showed that activity concentrations of soil were above the national average (**Šoštarić et al., 2021**).

The site consists of two facilities: Plant I and Plant II. Plant I has been operational since the 1970s and is known for using local coals (anthracite, lignite, and brown coal) until 1999 when Plant II was constructed. Anthracite coal, also known as Raša coal, was characterized by elevated levels of radioactivity and, owing to its high content of organic sulphur (up to 14%), was classified as a superhigh-organic-sulphur coal (**Medunić et al., 2016**). The radioactivity of the resulting coal ash and slag from coal combustion was increased. Local mines were eventually closed during the 1990s due to environmental unacceptability, insufficient reserves, and lack of profit (**Medunić et al., 2016**). Hence, Plant II used imported coal with low sulphuric content and low radioactivity (**Marović et al., 2004**).

Waste from the plant’s routine operation was continuously disposed of at the site. The disposal site was remediated during the 1990s. The remediation included the use of geo-synthetic material as a ground sealing layer, a protective cover consisting of an earth layer and grass, and the implementation of rainwater channels and a settling tank (**Marović et al., 2008**).

Figure 1: Study site location

2.2. Use of the ERICA Assessment Tool

Estimation of potential dose rates to terrestrial biota from exposure to radionuclides detected in samples collected from the research site, before and after the remediation, was done using the ERICA Tool (version 2.0), freely available to users online. The ERICA Tool relies on activity concentrations in the environmental media (sediment, soil, water, and air) as input data, activity concentrations in organisms, and uses reference organisms as generalised ecosystem representations of animal and plant species (**Beresford et al., 2007; Brown et al., 2008**). Radionuclides available in the Tool and the concept of reference organisms follow the guidelines proposed by the International Commission on Radiological Protection (**ICRP, 2007; ICRP, 2008**). The ERICA Tool is suited for environmental assessments related to the potential impacts of radiation due to planned or existing exposure situations, including scenarios related to NORM/TENORM, remediation, radioactive waste disposal, decommissioning of various nuclear facilities, and nuclear accidents (**Beresford et al., 2007**).

In the ERICA Tool, the estimation of environmental transfer of radionuclides to the biota is performed by using the concentration ratio (CR) values which represent the ratio between activity concentrations of radionuclides in the biota (whole body) and activity concentrations in the selected environmental media (soil, water, and air) (**Beresford et al., 2007; Brown et al., 2008; Brown et al., 2016**). In assessing the potential effects of internal and external exposure to ionizing radiation, the ERICA Tool uses Dose Conversion Coefficients (DCC_{int} and DCC_{ext}) in μGyh^{-1} per $Bqkg^{-1}$ fresh weight and compares the data on activity concentration in the biota and the environmental media (**Beresford et al., 2007; Brown et al., 2008; Brown et al., 2013**). Other parameters and values used to perform the assessment include weighting factors, used to address different components of radiation (low β, β + γ, and α) (**Brown et al., 2008; Brown et al., 2016**). The assessor conducting the assessment can select one of the three ecosystems (freshwater, terrestrial, and marine) and either the Tool's default screening dose rate value of 10 μGyh^{-1} (**Brown et al., 2008**), 400 μGyh^{-1} (**UNSCEAR, 1996**), or a custom as-

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sessment screening dose rate value. The Tool uses uncertainty factors (UF), 3 and 5, to ensure conservativism between Tier 1 (a simple assessment requiring minimal data input) and Tier 2 assessments, which are defined as the ratio between the 95th and 99th percentile of the risk quotient and the expected value of the probability distribution of the dose rate (**Beresford et al., 2007**). The default UF values of 3 and 5 have the role of ensuring that the assessment is run for a 5% and 1% probability of exceeding the dose rate screening value (**Brown et al., 2016**). One of the risk assessment outputs is the risk quotient (RQ), which is a unitless value that the Tool calculates by comparing the selected assessment screening dose rate and the total estimated whole-body absorbed dose rate for each organism (**Beresford et al., 2007; Brown et al., 2008**). A conservative risk quotient is calculated by multiplying the expected RQ value and the uncertainty factor (**Beresford et al., 2007**). All radiological risk assessments performed in this study used the default values of weighting factors, occupancy factors, screening dose rate value, and uncertainty factors.

2.3. Assessment input data

The overview of assessment input data for scenarios before and after the remediation is given in **Table 1**. Radiological risk assessment related to the environmental scenario before the remediation of the disposal site was based on laboratory gamma-spectrometric measurements of the samples collected at the disposal site published in previous studies by **Marović and Bauman (1986)** and **Skanata et al. (1996b)**. The average values of the activity concentrations in coal ash and slag samples and activity concentration ranges are presented in **Table 2**. For the risk assessment of the potential environmental impact of the disposal site after the remediation, available data from a previous study by **Marović et al. (2008)** was used. This study included data from in situ gamma-spectrometry measurements and gamma-spectrometry measurements in the laboratory that were carried out using an HPGe detector. Details on the measurement methods and sampling are available in **Marović et al. (2008)**. **Table 3** presents the activity concentrations in the media that were used as assessment input data.

An additional radiological risk assessment scenario for the plant site was performed using the extensive data on 50 surface soil samples taken at the plant perimeter in 2015 as a part of environmental monitoring conducted by the Institute for Medical Research and Occupational Health, Zagreb, Croatia. Sampling was carried out in accordance with the procedures proposed by International Atomic Energy Agency (**IAEA, 1989**). The sampling method included the removal of vegetation and sampling of the surface layer of soil (0 - 10 cm). The samples were oven-dried at 105°C and then sieved. The dried and sieved samples were sealed with PVC in 1000 mL volume Marinelli containers. To ensure radioactive equilibrium the samples were stored for at least 30 days before conducting measurements. Radionuclide activity concentrations were determined by high-resolution gamma-ray spectrometry using HPGe detectors. The activity concentration of ^{238}U was determined based on the activ-

Table 1: Assessment parameters for scenarios before and after* the remediation

Ecosystem type	Radionuclides	Reference organisms
Terrestrial	^{238}U ^{226}Ra ^{238}U* ^{226}Ra* ^{232}Th*	Amphibian Annelid Arthropod - detritivorous Bird Flying insects Grasses & Herbs Lichen & Bryophytes Mammal - large Mammal - small-burrowing Mollusc - gastropod Reptile Shrub Tree

Table 2: Average activity concentrations (Bqkg^{-1} dry mass) in samples collected prior to the remediation (AM ± SD, range) (adopted from **Marović and Bauman, 1986** and **Skanata et al., 1996b**)

Radionuclide	Activity concentration (Bqkg^{-1})
^{238}U	1344 ± 653 (882 – 1806)
^{226}Ra	1180 ± 543 (796 – 1565)

Table 3: Average activity concentrations (Bqkg^{-1} dry mass) in samples collected after the remediation (AM ± SD, range) (adopted from **Marović et al., 2008**)

Radionuclide	Activity concentration (Bqkg^{-1})
^{238}U	105 ± 35 (69 – 139)
^{226}Ra	79 ± 33 (49 – 115)
^{232}Th	57 ± 1 (56 – 59)

Table 4: Average activity concentrations (Bqkg^{-1} dry mass) in surface soil samples collected in 2015 (AM ± SD, range)

Radionuclide	Activity concentration (Bqkg^{-1})
^{238}U	96 ± 65 (17 – 304)
^{226}Ra	106 ± 64 (18 – 299)
^{232}Th	37 ± 24
^{210}Pb	(3 – 96) 115 ± 147 (15 – 710)

ity concentration of ^{234}Th (photopeaks at 63.29 keV and 92.38 keV) under the assumption that secular equilibrium had been established. Activity concentration of ^{226}Ra was determined from that of ^{214}Pb (photopeaks at 295.22 keV and 351.93 keV) and activity concentration of ^{232}Th from the activity of ^{228}Ac based on photopeaks at 338.32 keV, 911.20 keV and 968.97 keV. ^{210}Pb activity concentration was obtained from photopeak at 46.54 keV. The average activity concentrations data for this assessment scenario are given in **Table 4**.

3. Results and Discussion

The results from the radiological risk assessment based on the data from 1990s before the remediation (adopted from **Marović and Bauman, 1986; Skanata et al., 1996b**), showed that the overall expected risk quotient (unitless) and the conservative risk quotient values were the highest in lichen and bryophytes, with a risk quotient of 14.77 and a conservative risk quotient of 44.3. Regarding the data set from 2008, the overall calculated risk quotient values were much lower, with the highest risk quotient value estimated for lichen and bryophytes equal to 1.92. A comparison of the risk quotient for all reference organisms in both assessment scenarios is given in **Figure 2**.

The estimated total dose rate in the assessment scenario before the remediation exceed the screening value of 10 μGyh^{-1} for 9 out of 13 reference organisms included in the risk assessment, with the highest estimated to-

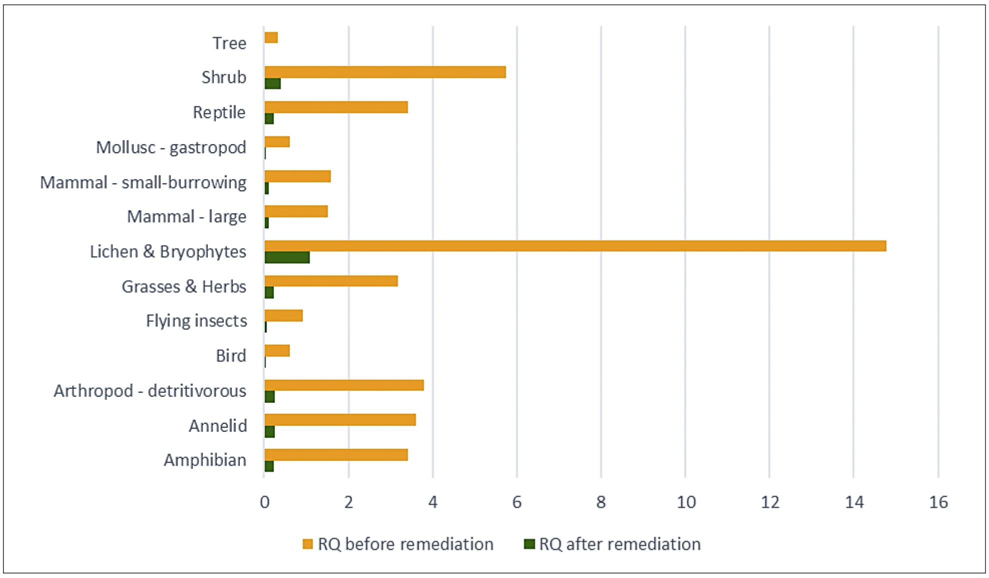

Figure 2: Comparison of the RQ results in assessment scenarios before and after the remediation

Table 5: Comparison of estimated total dose rates (μGyh^{-1}) to reference organisms in assessment scenarios before and after the remediation

Reference organism	Total Dose Rate (μGyh^{-1}) before remediation	Total Dose Rate (μGyh^{-1}) after remediation
Amphibian	34.10	2.30
Annelid	35.94	2.45
Arthropod - detritivorous	37.97	2.62
Bird	6.17	0.42
Flying insects	9.27	0.63
Grasses & Herbs	31.66	2.40
Lichen & Bryophytes	147.68	10.92
Mammal - large	15.15	1.02
Mammal - small-burrowing	15.82	1.07
Mollusc - gastropod	6.02	0.44
Reptile	34.11	2.30
Shrub	57.43	3.98
Tree	3.28	0.23

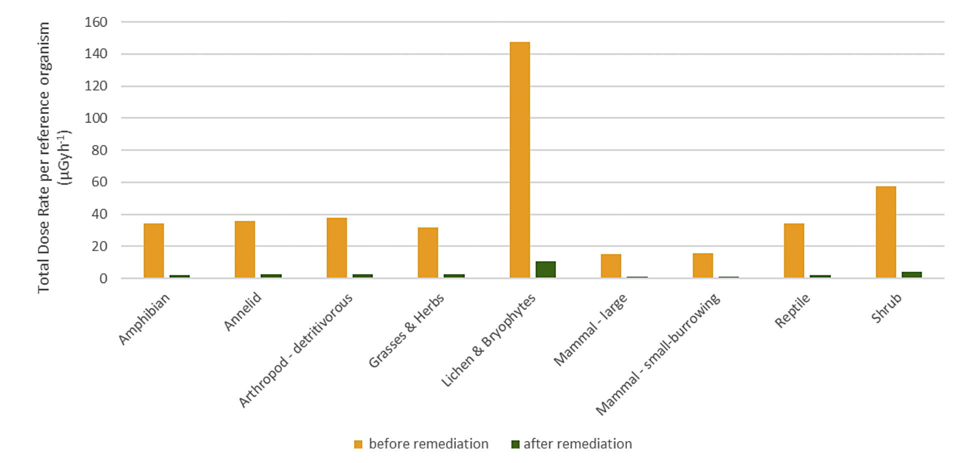

Figure 3: Comparison of total dose rate to most affected reference organisms from assessment scenarios before and after the remediation

tal dose rate for lichen and bryophytes being 147.68 µGyh^{-1}. In the assessment scenarios referring to post-remediation, estimated total dose rates were much lower, with the total dose rate for lichen and bryophytes of 10.92 µGyh^{-1}, almost equal to the selected screening value. **Table 5** presents the comparison of data on the estimated total dose rate to all reference organisms in assessment before and after remediation of the disposal site, where **Figure 3** shows the comparison of total dose rates to 9 reference organisms for which the screening dose rate in the assessment scenario before the remediation of the site was exceeded.

The results of the assessment scenario before the remediation showed that the total dose rate estimation can primarily be attributed to the internal exposure, with ^{226}Ra as the main contributor, especially for lichen and bryophytes and a shrub as reference organisms. The distribution of radionuclides that contribute to the external dose rate also includes ^{226}Ra as a key contributor and amphibian, annelid, arthropod, mammal (small-burrowing) and reptile as the most affected reference organisms.

The total dose rate results for the post-remediation assessment scenario also show that internal exposure contributes the most to the total dose rate to all reference organisms. Again, ^{226}Ra is the main contributor to the internal dose rate, with the highest dose internal rate in lichen and bryophytes and a shrub. **Sotiropoulou et al. (2016)** from Greece also found ^{226}Ra to be the main contributor to the internal dose rate to lichen and bryophytes. External dose rate can primarily be attributed to ^{226}Ra, with the highest dose rates in amphibian, annelid, arthropod, mammal (small-burrowing) and reptile. The distribution of internal and external dose rates from exposure to ^{226}Ra before and after the remediation for the most affected reference organisms is given in **Figure 4** and **Figure 5**.

The assessment results from additional assessment based on the data on soil samples from 2015 are in line with the results of the assessment scenario based on the data from 2008, although the activity concentrations from 2015 resulted in slightly higher dose rate estimations. The overall highest estimated value was found in lichen and bryophytes 18.06 µGyh^{-1}, where data from 2008 resulted in predicted total dose rate to lichen and bryophytes of 10.92 µGyh^{-1}. ^{226}Ra was found to contribute the most to both internal and external dose rates to reference organisms.

As lichen and bryophytes were found to be the most affected organisms in the scenario before the remediation, **Table 6** presents the distribution of total, internal and external dose rate to lichen and bryophytes from an assessment run with data before the disposal site remediation.

The results from the assessment that relied on data before the disposal site remediation are in line with re-

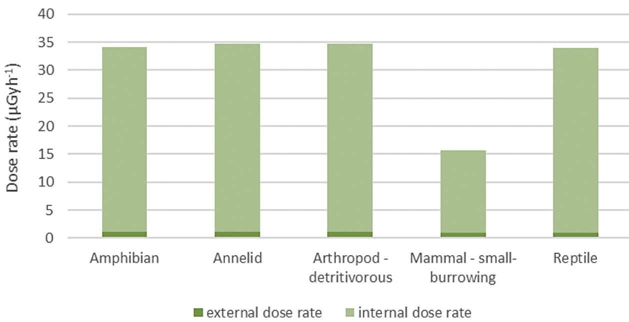

Figure 4: ^{226}Ra contribution to external and internal dose rates to the most affected reference organisms before disposal site remediation

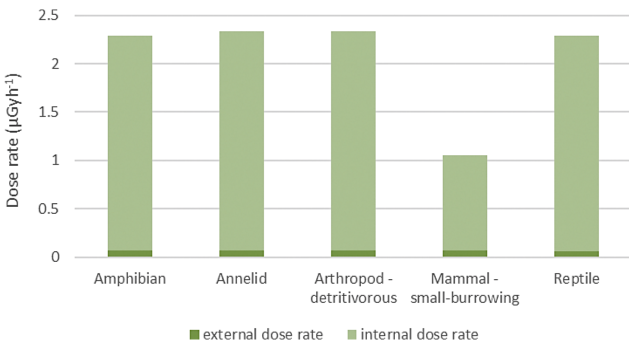

Figure 5: ^{226}Ra contribution to external and internal dose rates to the most affected reference organisms after remediation disposal site

Table 6: Distribution of internal and external dose rate to lichen and bryophytes assessed based on the sample data before the disposal site remediation

Isotope	External Dose Rate ($\mu Gy\ h^{-1}$)	Internal Dose Rate ($\mu Gy\ h^{-1}$)	Total Dose Rate* ($\mu Gy\ h^{-1}$)
^{238}U	0.01	30.35	30.36
^{226}Ra	0.31	116.9	147.68

* The total dose rate presented includes contributions from other radionuclides besides the ^{238}U and ^{226}Ra listed in this table

sults from previous studies on total dose rates to terrestrial biota that focused on NORM legacy site and mining. The study by **Mrdakovic Popic et al. (2020)**, at the legacy NORM site in Norway, reported the highest predicted total dose rate of 206 μGyh^{-1} to lichen and bryophytes when default CR values were used and 23 μGyh^{-1} when site-specific soil and plant activity concentrations were used. **Oughton et al. (2013)** conducted risk assessments at several mining sites in Central Asia. Risk assessments used site-specific data and included calculated dose rates to aquatic and terrestrial biota. Findings from the study include assessment results related to disposal site containing tailings with the highest total dose rate value of 660 μGyh^{-1} predicted to lichen and bryophytes. Additionally, this study also reported ^{226}Ra as the main contributor to the internal and external dose rates (**Oughton et al., 2013**), which was also the case in our assessments that used data before the site remediation. The assessment of dose rate to terrestrial biota in the area around coal fired power plant in Serbia also resulted in screening dose rate being exceeded only for lichen and bryophytes (**Ćujić and Dragović, 2018**).

Regarding the results of the total dose rate after the site remediation, which only slightly exceeded the ERICA Tool conservative screening value of 10 μGyh-1 for lichen and bryophytes but were still below the value of 40 and 400 μGyh^{-1} for terrestrial biota for which no effects on population levels should be expected (**UNSCEAR, 2008**), the overall risk can be regarded as negligible. This conclusion is also supported by the fact that lichen and bryophytes are considered highly radiosensitive organisms and, as a result, are often used as biomonitors of potential contamination and concerning both artificial and natural radionuclides (**Marović et al., 2008; Garty et al., 2003; Kirchner and Daillant, 2002; Loppi et al., 2003**).

Both assessment scenarios and respective results should be observed considering several uncertainties. The study used two sets of data on soil activity concentrations that included a limited number of soil samples and relatively small number of radionuclides. Although soil samples are from the same disposal site, given the remediation of the disposal site, they were taken from different sampling locations. Hence, the data sets should be regarded as different spatial and temporal sets of data. An additional source of uncertainty is the lack of other site-specific data, such as CR values and activity concentrations in plants or animals. Considering that the ERICA tool is known to use a conservative approach when the Tool's default CR values are used, an overestimation of the total dose rate results in both assessment scenarios is possible.

4. Conclusions

In cases where coal with elevated levels of natural radioactivity is used as a primary energy source, coal combustion can be a source of exposure to radiation due to the resulting coal ash and slag accumulating significant activity concentrations of radionuclides. Remediation of such coal and ash slag disposal site provided research context for our study of radiological risk assessment for terrestrial biota. In order to assess the potential impacts of the disposal site and the effects of the remediation, the environmental data on activity concentration in the soil before and after the disposal site remediation was used to conduct radiological risk assessments using the ERICA tool.

The results from the assessment related to the period before the remediation showed that for several reference organisms, the estimated total dose rate exceeded the default screening value, with the highest predicted value of 147.68 μGyh^{-1} to lichens and bryophytes, which is not surprising as they are considered as most radiosensitive organisms. The assessed radiological risk and respective dose rates to reference organisms after the site remediation were significantly lower, with the total dose rate to lichens and bryophytes being 10.92 μGyh^{-1}, which is almost equal to the assessment's conservative screening dose rate of 10 μGyh^{-1}. In both assessment scenarios, internal exposure attributed the most to the estimated total dose rate, with ^{226}Ra contributing the most to both the internal and external dose rates, around 80%. This finding is in line with results from similar studies conducted at different locations from other authors.

Assessment results indicate that remediation of the site was adequate and that the overall radiological risk to terrestrial biota from the disposal site can be considered negligible, and that the estimated total dose rates to biota are below the levels that can be associated with detrimental effects. It should be stressed that environmental monitoring of the site is required to ensure reliable long-term radiological and environmental protection and safety. The results from both radiological risk assessment scenarios can serve as an example for the future estimation of potential radiological impacts of similar disposal sites and radiological risk assessment design.

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SAŽETAK

Sanacija odlagališta ugljenoga pepela i šljake: usporedba procjena radiološkoga rizika

Ostatci od spaljivanja ugljena poznati su kao potencijalni izvor izloženosti zračenju, posebno u slučajevima kada je ugljen koji se koristi u spaljivanju karakteriziran povećanom radioaktivnošću, što rezultira pepelom i šljakom s potencijalno visokom aktivnošću koncentracije radionuklida. U radu su prikazani rezultati procjene radiološkoga rizika temeljeni na pristupu ERICA alata korištenim za procjenu brzine doza za kopnenu biotu u blizini termoelektrane na ugljen u Hrvatskoj. Studija se sastoji od triju procjena radiološkoga rizika korištenjem podataka o koncentraciji aktivnosti ($Bqkg^{-1}$) u okolišu iz uzoraka prikupljenih prije sanacije odlagališta i uzoraka nakon završetka sanacije. Rezultirajuća ukupna brzina doze za biotu dobivena korištenjem podataka prije sanacije kretala se od 3,28 μGyh^{-1} do 147,68 μGyh^{-1}. Rezultati procjene ukupne brzine doze na temelju podataka s istraživanoga područja nakon sanacije kreću se od 0,23 μGyh^{-1} do 18,06 μGyh^{-1}. Rezultati su pokazali da je nakon sanacije samo ukupna brzina doze za lišajeve i briofite neznatno premašila konzervativnu vrijednost provjere ERICA alata od 10 μGyh^{-1}, što implicira da se rizici za okoliš u odnosu na izloženost odlagalištu mogu smatrati zanemarivima. Ovi rezultati studije potvrđuju prikladnost korištenja ERICA alata za procjenu potencijalnoga radiološkog utjecaja i učinkovite provedbe sanacije odlagališta ugljena i šljake.

Ključne riječi:
procjena radiološkoga rizika, NORM, elektrana na ugljen, sanacija, monitoring okoliša

Author's contribution

Ana Getaldić (MEng, PhD Candidate) proposed and defined the idea for the manuscript, conducted all assessments, and prepared the original manuscript draft. **Marija Surić Mihić** (PhD, Senior Research Associate, Radiation Protection Expert), **Želimir Veinović** (PhD, Associate Professor, Radiation Protection Expert) provided supervision, performed the formal analysis of the results and the factual review. **Božena Skoko** (PhD) performed the formal analysis of the results and the factual review. **Branko Petrinec** (PhD, Senior Research Associate, Radiation Protection Expert) provided the data and performed the formal analysis of the results.

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ENVIRONMENTAL PROTECTION IN NATURAL GAS INDUSTRY
Comparison of Different Spatio-Temporal Radiological Risk Assessment Scenarios

by

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The paper analyses results of spatio-temporal radiological risk assessment scenarios based on existing in-situ long-term monitoring data from a natural gas processing plant to analyse the effect of different input data on the assessment outcome. The ERICA Assessment Tool was used to estimate the dose rates to biota and potential impacts due to the exposure to ionising radiation. The input data for radiological risk assessment scenarios included annual data on activity concentration of radionuclides in soil from *in-situ* measurements performed from 1994 to 2016 and laboratory gamma-spectrometric data related to the period from 2014 to 2019. Predicted total dose rate to biota was generally below the ERICA Tool's screening dose rate of 10 μGyh^{-1} or slightly above, with the highest total dose rate estimated for lichen and bryophytes. Total dose rates to lichen and bryophytes in the studied period show certain temporal variation, but a specific trend was not detected. Estimated total dose rates to biota from different assessment scenarios were below internationally proposed reference levels for which no detrimental effects are expected. The overall potential radiological risk to terrestrial biota from the operation of the natural gas processing plant was found to be negligible.

Key words: NORM, natural gas, radiological risk assessment, environmental protection, Erica tool

INTRODUCTION

Natural gas is being used worldwide as a primary energy source, with global data showing it constitutes as more than a quarter in the global energy mix [1] and similarly in the energy mix of the EU [2]. Natural gas also has an important role in the global energy security [3-5]. Consequently, the natural gas industry has a significant impact on the quality of the overall environment.

Naturally occurring radioactive material (NORM) results from different industrial processes as an industrial by-product where radionuclides accumulate in different types of waste. Industrial activities that may lead to the enhanced levels of radioactivity have been gaining attention in the last decades. The European Council 2013/59/Euratom recognizes possible risks arising from natural radioactivity, *i. e.*, NORM, while possible environmental contamination risks associated with NORM-related industries were documented in detail by international community as well [6-10]. Different aspects of NORM generation in industries, its emissions, and possible effects on health and the environment have been studied in the last two decades [11, 12]. Since industrial NORM releases can be associated with detrimental effects on populations and environment, radiation protection in the context of industries related to NORM aims to mitigate adverse effect by using radiation protection principles of justification, and optimization in occupational exposure [13-16].

The importance of oil and gas industries as NORM-related industries in establishing standards and ensuring adequate protection of both populations and the environment has been researched in several specific studies. Koppel *et al*. [17] stress the potential role of oil and gas facilities that are to be decommissioned, risks associated with decommissioning options, and possible ecological impacts. In their paper Cowie *et al*. [18] present a practical industrial experience in developing a NORM management strategy in oil and gas industry. Jodlowski *et al*. [19] studied waste from gas exploration

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and production including drill cuttings, drilling muds, fracking fluids, return, fracking fluids, and waste proppants, while Gafvert *et al*. [20] sampled produced water from offshore platforms in Norway. Al-Masri and Haddad [21] used fly and bottom ash samples from a natural gas power plant to study NORM emissions. Several studies were conducted on soil and sludge samples, Xhixha *et al*. [22] conducted an extensive study using soil and sludge samples in order to identify areas for strategical plan of future radiological assessments in Albania, where Barros *et al*. [23] sampled scale in industrial pipelines in Venezuela. Garner *et al*. [24] explored oil and gas producing region in the United Kingdom, Attallah *et al*. [25] studied scale samples from petroleum industry in Egypt, and study from Taheri *et al*. [26] used samples of soil and sludge from a gas field in Iran. There are also studies that include characterization of waste arising from oil and natural gas production [27] and geochemical signature of NORM waste from oil industry [28]. The study from Husain and Sakhnini [29] focused on radiological impacts of NORM from oil and gas industry in Bahrein. All these studies demonstrate the importance of robust environmental monitoring and proper attention paid to NORM waste and assessments of its potential radiological risks to the environment in all production phases. Lazarus *et al*. [30] investigated presence of mercury, and other stable metalloids and radionuclides in biota as a part of the extensive monitoring of soil, earthworms, moss, livestock and wildlife animals at the natural gas treatment plant.

The main goal of environmental monitoring is the quantification of radioactive substances or ionising radiation that arise from human activities and natural sources in different environmental media [31]. Regarding the practical context of environmental monitoring programmes, Article 35 of the Euratom Treaty implies implementation of comprehensive national programmes of monitoring the environmental radioactivity. These programmes aim at monitoring main pathways of potential exposure of population and include sampling and analyses of the environmental media [32]. These programmes might not include particular industrial sites related to NORM, but environmental monitoring of NORM-related industrial locations aligns with the overarching goals of national environmental radioactivity programmes. Environmental radioactivity monitoring also has a role in effective risk preparedness and prevention [33]. Study from Riberio *et al*. [34] presents an example of extensive environmental monitoring programme implementation. Sun *et al*. [35] focused on optimization of long-term monitoring of radiation air-dose rates, while including the goals of long term environmental monitoring *i.e*., detecting possible changes of contaminant mobility and validating the reduction of hazard levels. Michalik [12] emphasizes the importance of environmental radioactivity monitoring including non-human species representatives, and possible radiation dose and effects on biota. Soil radioactivity was also studied to establish baseline data for future radiation impact assessments [36], to estimate possible pollution with industry as a source of radionuclides and heavy metals [37], and to estimate possible use of organisms as biomonitors [38].

The assessment of potential impacts arising from exposure of non-human biota to ionising radiation can be performed using different approaches and models [39, 40]. The ERICA Integrated Approach and ERICA Tool were developed through EU co-funded 6th Framework Program EURATOM project Environmental Risk from Ionising Contaminants Assessment and Management (ERICA). The key characteristics of the ERICA Tool is the assessment-based risk quantification through use of data on environmental transfer and dosimetry, resulting in the measure of exposure that is further compared to exposure levels associated with known detrimental effects of radiation [41-44]. The use of ERICA Tool can be used for planned, emergency or existing exposure situation, where NORM-related activities are regarded as planned exposure situations [45, 46].

This paper compares different spatio-temporal radiological risk assessment scenarios based on existing in-situ long-term monitoring data from a natural gas processing plant to analyse the effect of different input data on the assessment outcome. Additionally, a risk assessment using laboratory gamma-spectrometric data from the same site was conducted, and results from both studies were compared. The results of these comparisons could provide valuable feedback for design of future radiological risk assessments in NORM-related industries and general insight in justifiability of conducting long-term radioactivity monitoring and using the resulting data to perform radiological risk assessments, as opposed to using more concise environmental radioactivity data sets.

MATERIALS AND METHODS

Assessment site

The research area included the natural gas processing plant site Molve, located in Croatia, Europe. The site is part of Podravina reservoir and presents one of the largest natural gas and gas condensate reserves in the Republic of Croatia that accounts for the majority of the national natural gas production [47]. After initial research in 1974, as a part of the project *Podravina* the production at the natural gas field Molve first started in 1981 with two gas wells and was later further developed in several phases [47, 48]. The ongoing production of natural gas and gas condensate for the last 40 years makes this the most complex energy project related to hydrocarbon exploration and production in Croatia, as well as an example of a project implementation that effectively combined energy-related goals and environmental protection principles [49]. The ongoing activities at the site include the production and purification of gas and gas condensate for transport.

Assessment data

In-situ gamma-ray spectrometry measurements were performed by Radiation Protection Unit of the Institute for Medical Research and Occupational Health in the period from 1994 to 2016 on three locations at the plant site which included the location of the central gas station (CPS) and locations of two gas wells, station M-9 and station M-10, fig. 1.

In-situ gamma-spectrometric measurements were carried out to determine the sources of basic radiation, both cosmic and terrestrial, by direct measurements in the field using a semiconductor detector ORTEC HPGe, a multi-channel analyser (with 16000 channels) and the associated electronic circuit with a computer. The characteristics of the HPGe detector included resolution of 1.75 keV at 1.33 MeV ^{60}Co and relative efficiency of 21 % at 1.33 MeV ^{60}Co. All *in-situ* measurements were conducted during 1000 seconds and ORTEC Gamma Vision software was used to analyse the resulting spectra. The activity concentrations of natural radionuclides in the soil were calculated assuming their uniform distribution in the soil.

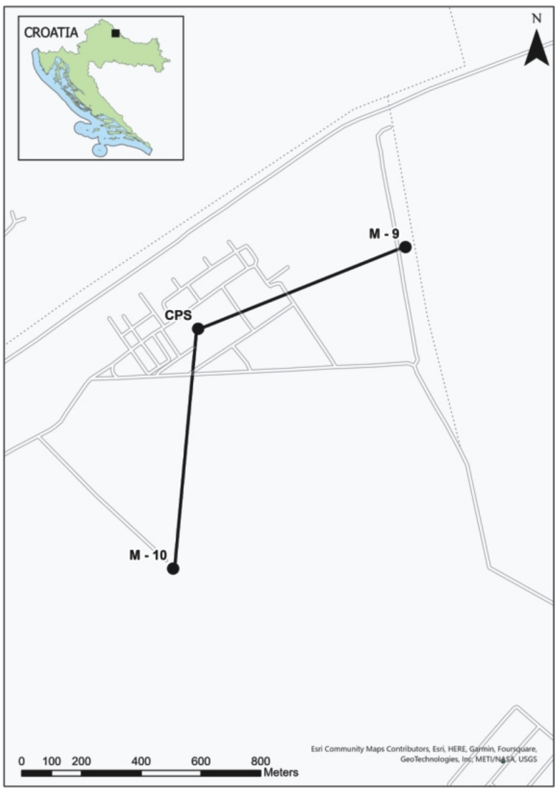

Figure 1. Assessment site and sampling locations (CPS, M-9, and M-10) layout

In the period from 2014 to 2019, samples of soil (0-10 cm) were taken from the location of central gas station CPS, station M-9, and station M-10. All the samples were prepared in the laboratory and analysed using gamma-ray spectrometry. The sample preparation included sample sieving, drying of samples at 105 °C, and then ashing at 450 °C in a muffle furnace. The samples prepared in this manner were then packed in sealed containers of 200 ml volume. The samples were measured in a gamma-spectrometric laboratory after 66 days to ensure the secular equilibrium within the uranium and thorium decay chains. Determining radioactivity in soil samples was performed using high-resolution gamma-spectrometry with a method accredited according to HRN EN ISO/IEC 17025. HP GMX ORTEC detector system was used with the following characteristics: resolution of 2.2 keV at 1.33 MeV ^{60}Co and a relative efficiency of 74.3 % at 1.33 MeV ^{60}Co. Efficiency calibration was carried out by the standards from the Czech Metrological Institute covering the energy range from 40 to 2000 keV. Data on ^{238}U, ^{226}Ra, and ^{232}Th activities were determined from those of their decay products. Activity of ^{226}Ra was determined from that of ^{214}Bi (photopeaks at 609 keV, 1120 keV, and 1764 keV), activity of ^{232}Th from that of 228Ac (photopeaks at 338 keV, 911 keV, and 968 keV), and activity of ^{238}U from those of ^{234}Th (photopeak at 63 keV). The measured activity in all the samples was above the detection limit. The quality assurance of radionuclide determination was performed through systematic participation in comparative measurements organized by the International Atomic Energy Agency (IAEA), the World Health Organization (WHO), as well as the EU's Joint Research Centre (JRC) [50].

Use of the ERICA tool

The ERICA Assessment Tool (version 2.0) was used to calculate dose rates to terrestrial biota from exposure to radionuclides. The assessment can be performed by selecting different default ecosystems: terrestrial, marine and freshwater. The ERICA Tool uses activity concentrations in environmental media *i.e.*, sediment, soil, water and air as input data for the assessment. The estimation of radionuclide transfer to the environment is performed using the concentration ratio (CR) values [41, 43, 45]. The ERICA Tool assesses potential effects arising from both internal and external exposure by interpreting activity concentration data in environmental media and biota which is done through the use of internal and external dose conversion coefficients (DCC_{int} and DCC_{ext}) [44, 45]. The Tool also uses weighting factors to address different components of radiation, 10 for alpha, 3 for beta and 1 for gamma radiation [43]. The default list of radionuclides in the ERICA Tool in line with the environmental protection framework of the International Commission on Radiological Protection, as well as the use of reference organisms as generalised ecosystem representations [43]. For reference organisms, the occupancy factors define the fraction of time that the organism spends in a certain environmental media, and these values can be modified by the user if necessary [44, 45].

The ERICA Assessment Tool allows users to run assessment in different assessment contexts, *i.e.*, different tiers. Tier 1 presents the basic highly conservative assessment that requires minimal user data input. The Tier 2 assessment context allows users to input site-specific media concentrations and to use single point or more complex temporal and spatial data series. Tier 2 also offers users to perform a less conservative assessment and comparison of results against tables of radiological effects and exposure due to naturally occurring radionuclides [45]. The default screening dose rate proposed by the ERICA Tool is 10 μGyh^{-1}, and suggested uncertainty factors (UF) are 3 and 5 that enable the assessment for 5 %, and 1 % probability of exceeding the dose rate screening value, respectively [43-45].

All risk assessment scenarios using the *in-situ* gamma-spectrometric measurements from the long- term monitoring data were run at Tier 2 of the ERICA Tool for a terrestrial ecosystem. The reason for this is that only Tier 2 allows users to input multiple series data and specific combination of spatial and temporal series of data.

The input data included annual activity concentration of radionuclides in soil (in Bqkg^{-1}) from samples collected at three sampling locations, at a natural gas processing plant, in the period from 1994 to 2016. Table 1 summarizes activity concentrations in the soil samples for the studied period.

The assessments included all ERICA Tool's default terrestrial reference organisms, and the default occupancy factors, assuming that the selected organisms spend 100 % of their time at the site, which could be regarded as a conservative approach. The selected screening dose rate for all the assessment scenarios was the ERICA Tool's default value of 10 μGyh^{-1}. Other default parameters included UF of 3, percentage of dry weight of media of 100 %, and the default weighting factors for alpha, high energy betta/gamma and low energy beta radiation of 10, 1, and 3, respectively. The CR values used in the assessments were default values provided by the assessment Tool, as site-specific CR values were not available. The use of site-specific CR values by the ERICA Tool in a NORM-related assessment context was researched in detail by other authors and generally, the results show lower dose rate estimations as opposed to assessments that use ERICA Tool's default CR values [51-53]. Table 2 summarizes parameters used in the risk assessments: list of radionuclides and reference organisms.

The study performed multiple radiological risk assessment scenarios using the *in-situ* gamma-spectrometric measurements. The first risk assessment scenario used the complete long-term data set on annual activity concentrations per sampling location (CPS, M-9, and M-10) from 1994 to 2016. For the same data set separate risk assessment were performed using an annual average radionuclide concentration from all three sampling locations. Additional assessment used the maximum measured activity concentrations from all the sampling locations in the studied period. In order to assess the potential cumulative effects, a separate assessment scenario used tripled maximum measured activity concentrations from the sampling locations.

A second radiological risk assessment scenario using the data from the laboratory gamma-spectroscopic measurements, for the period from 2014 to 2019, was also performed using the Tier 2 assessment context with data on activity concentration of radionuclides in soil (in Bqkg^{-1}) from three sampling locations at the Molve site. This assessment also included the ERICA Tool default reference organisms and default parameters of the screening dose rate, occupancy factors, UF of 3, the percentage of dry weight of media of 100 %, and the default weighting factors for an alpha, high energy betta/gamma, and low energy beta radiation. Again, Tool's default CR values were used. Table 3 summarizes all assessment input data, and tab. 4 lists activity concentrations of soil samples used in the assessment scenario.

Table 1. Activity concentrations (Bqkg^{-1} dry mass) in soil the samples from in-situ gamma spectrometric measurements in the period 1994-2016, (N-number of measurements, the range is given in parenthesis)

Activity concentrations SD [Bqkg^{-1}]			
Sampling location	N	^{232}Th	^{226}Ra
CPS	18	40 15 (11-61)	44 19 (26-97)
M-10	16	30 20 (11-90)	36 12 (23-77)
M-9	18	34 32 (5-128)	38 14 (20-69)

Table 2. Assessment parameters in terrestrial assessments using *in-situ* gamma-spectrometric data

Radionuclides	Reference organisms
^{232}Th ^{226}Ra	Grasses and Herbs
	Shrub
	Tree
	Amphibian
	Annelid
	Arthropod – detritivorous
	Bird
	Flying insects
	Lichen & Bryophytes
	Mammal – large
	Mammal – small burrowing
	Mollusc – Gastropod
	Reptile

RESULTS AND DISCUSSION

Risk quotient (RQ), a unitless value calculated by the Tool, using the data on selected screening dose rate and the total estimated whole body absorbed dose rate for each individual organism [45], did not exceed 1 in risk assessment scenario related to the *in-situ* gamma spectrometric temporal data set. The risk assessment scenario that used laboratory gamma-spectrometric data detected a RQ slightly above 1 and resulting in lichen and bryophytes as the most affected reference organisms. These results could be explained by the laboratory gamma-spectrometric data including more radionuclide data which then increases the estimated dose rates and consequently the estimated RQ.

In assessment scenarios using the *in-situ* gamma spectrometric data and laboratory gamma spectrometric data, Tool's output data on external and internal dose rate was analysed to determine the dominant exposure route and key contributors to the dose rate. The assessments based on the *in-situ* gamma spectrometric data resulted in external dose with ^{226}Ra as the main contributor, with amphibian, annelid, arthropod, small burrowing mammals and reptile as the most affected organisms. The internal dose rate was also primarily associated with exposure to ^{226}Ra, with the highest internal dose rate to lichen and bryophytes and shrub.

Table 3. Assessment parameters in terrestrial assessments using laboratory gamma-spectrometric data

Radionuclides	Reference organisms
^{238}U ^{232}Th ^{226}Ra ^{210}Pb	Grasses and Herbs
	Shrub
	Tree
	Amphibian
	Annelid
	Arthropod – detritivorous
	Bird
	Flying insects
	Lichen and Bryophytes
	Mammal – large
	Mammal – small burrowing
	Mollusc – Gastropod
	Reptile

Table 4. Activity concentrations (Bqkg^{-1} dry mass) in soil samples from laboratory gamma-spectrometric measurements in the period 2014-2019, (N-number of measurements, the range is given in parenthesis)

Sampling location	N	Activity concentrations SD [Bqkg^{-1}]			
		^{238}U	^{232}Th	^{226}Ra	^{210}Pb
CPS	5	38 5 (33-45)	44 17 (31-74)	45 10 (35-60)	53 21 (28-76)
M-10	5	44 12 (28-57)	52 19 (31-83)	49 15 (31-73)	49 21 (28-77)
M-9	5	48 16 (30-67)	54 23 (14-70)	33 22 (5-57)	38 28 (11-84)

Again, the internal dose rate was the parameter that affected the estimated total dose rate the most irrespective of the temporal aspect of the input data or if the maximum activity concentrations was used in the assessment. The calculation of dose rate in assessment scenarios using laboratory gamma-spectrometric data showed that the main contributor to the external dose rate for all reference organisms was ^{226}Ra, with the highest contribution to following reference organisms: amphibian, annelid, arthropod, mammals (small -burrowing), mollusc, and reptile. The data on the internal dose rate showed that ^{226}Ra contributes the most to the internal dose rate, primarily observed in reference to lichen and bryophytes and shrub. The total dose rate estimation can almost entirely be attributed to internal dose rate. The contribution of different radionuclides, specifically ^{226}Ra, to the total dose rate from our study is in accordance with results from previous studies related to exposure to naturally occurring radionuclides from other authors [51, 54-57]. Additionally, the presence of ^{226}Ra and importance of its activity concentration for the assessment results is related to the fact that ^{226}Ra is a prevalent radionuclide in scales and deposits found in equipment of the oil and gas industry and discharges, and as such, is a major source of radiation exposure [13, 20, 24].

Individual temporal assessments that relied on the annual *in-situ* data from 1994 to 2016 resulted in estimated dose rates between the lowest of 0.1 μGyh^{-1} to the tree as a reference organism and the highest total dose rate of 10.13 μGyh^{-1} to lichen and bryophytes. The same data set, that was temporally averaged before calculation, resulted in an estimated total dose between 0.1 μGyh^{-1} for tree and 4.39 μGyh^{-1} for lichen and bryophytes.

Results from assessment scenario that used laboratory gamma-spectrometric data from 2013 to 2019 showed that the total dose rate to biota ranges from 0.05 μGyh^{-1} for tree to 15.20 μGyh^{-1} to lichen and bryophytes. Figure 2 shows the comparison of the average values of the estimated total dose rate in the studied period for two most affected reference organisms (lichen and bryophytes and shrub) from individual assessment scenario using in-situ and laboratory gamma spectrometric data from 1994 to 2016.

Additional assessment scenario, performed using the maximum soil activity concentrations from the period 1994 to 2016, estimated the total dose rate from 0.26 μGyh^{-1} for tree and 10.87 μGyh^{-1} to lichen and bryophytes. In order to conduct an assessment considering the highest input values, maximum measured activity concentrations from all the sampling locations were used. A comparison of estimated total dose rates to all reference organism from assessment scenarios that used temporal average and maximum activity concentrations from *in-situ* gamma-spectrometric measurements is presented in fig. 3. To estimate the potential cumulative radionuclide concentration effect, the maximum mea-

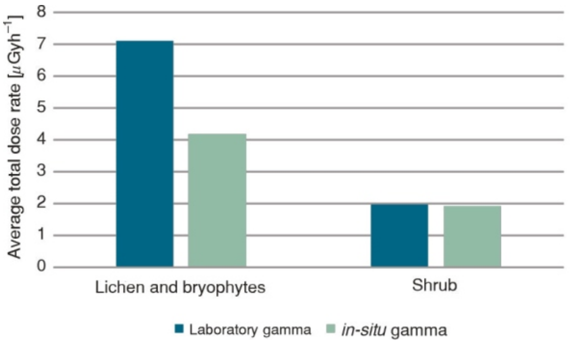

Figure 2. Comparison of average total dose rates to most affected reference organisms from assessments based on *in-situ* and laboratory gamma-spectrometric data sets

sured activity concentrations per radionuclide from all three sampling locations were tripled and another assessment scenario was run with these parameters. In this case, the predicted total dose rate to lichen and bryophytes was 32.5 μGyh^{-1}, which exceeded the ERICA Tool's default screening value, but was below the reference values of 400 μGyh^{-1} for terrestrial plants [57]. These results would imply that even in the case of cumulative contamination the predicted effects to the biota in the proximity of the facility would be below internationally recognized reference levels.

Since lichen and bryophytes were found to be the most affected reference organisms in all the assessment scenarios, and given they are often used as biomonitors of potential environmental contamination [58-60]. Figure 4 presents the estimated total dose rate to lichen and bryophytes based on the in-situ gamma spectrometric data from 1994 to 2016. Total dose rates to lichen and bryophytes in the studied period show certain temporal variation, but our analysis did not detect a specific trend.

Estimated total dose rate value in the studied period was below the selected screening dose rate of 10 μGyh^{-1}, which together with the assessments results, based on the maximum input activity concentrations, implies that the potential radiological risk to terrestrial biota arising from the operation of the natural gas processing plant is not significant. The overall results from various temporal assessments, including in-situ and laboratory data, are in accordance with the results from previous studies. Study by Ćujić and Dragović [55] assessed NORM-related total dose rate to lichen and of 14.4 μGyh^{-1}. Lazarus *et al*. [30] reported estimated dose rates to terrestrial biota up to 3.7 μGyh^{-1} to mosses and lichen. Study by MacIntosh *et al*. [61] on radiological risk assessment to marine biota from exposure to NORM related to decommissioning offshore oil and gas pipeline, estimated a potential dose rate from external exposure up to 33 μGyh^{-1}.

Presented results from risk assessment scenarios need to be observed keeping in mind certain uncertainties associated with performed assessments. One of the possible contributors to the uncertainty is a minimal data gap in available in-situ gamma spectrometric data, *i.e.*, missing data points for a specific radionuclide in a

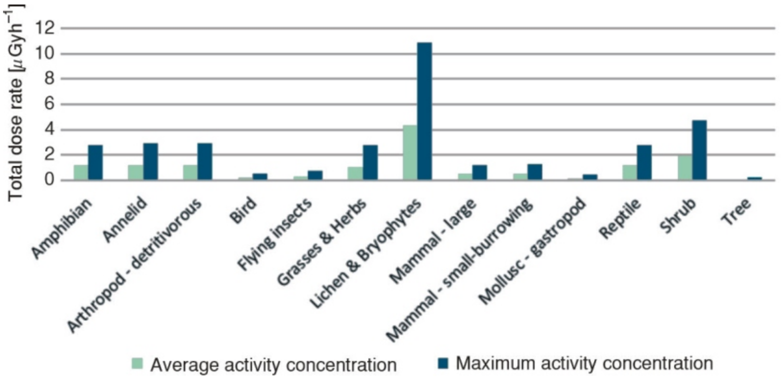

Figure 3. Comparison of total dose rates [μGyh^{-1}] to reference organisms estimated by different input data on activity concentrations (average or maximum values)

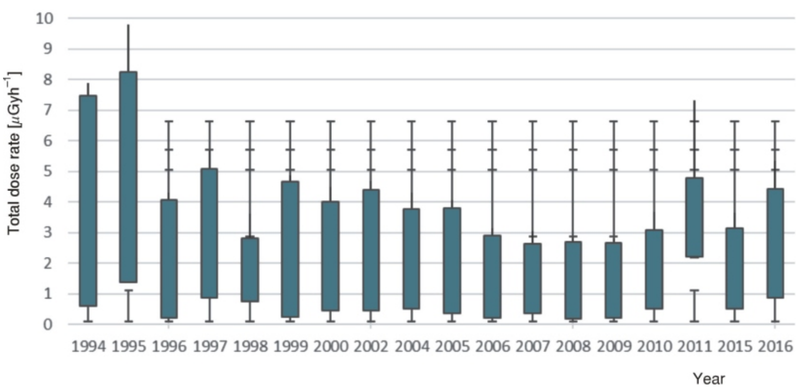

Figure 4. Estimated total dose rate to lichen and bryophytes based on the *in-situ* gamma-spectrometric data from the period 1994 to 2016

certain year, but given the observed temporal variations of the available data, extreme activity concentrations of the missing data are unlikely. Other uncertainties are associated with the lack of experimental data on site-specific transfer values. The ERICA Tool uses a conservative approach to assessments, especially when Tool's default CR values are used, as was the case in all risk assessment scenarios conducted in this study. Hence, the chance of assessment results underestimating the radiological effects and risks should be minimal, but an overestimation of the total dose rates due to the use of Tool's default CR values is possible.

CONCLUSIONS

The assessment results from temporal assessments using *in-situ* gamma spectrometric data showed that the same reference organisms, lichen and bryophytes, were the most affected for in all performed assessment scenarios, irrespective of the time period selected, with the highest estimated total dose rate of 10.13 μGyh^{-1}. The effect of using average activity concentrations in temporal assessments resulted in total dose rates generally below the assessment screening dose rate of 10 μGyh^{-1}. Assessments that relied on maximum activity concentrations as input resulted in total dose rate only slightly exceeding the default screening dose rate for lichen and bryophytes. The assessment scenario that used gamma-spectrometric laboratory data from soil samples from the same location, resulted in the highest total dose rate to lichen and bryophytes of 15.20 μGyh^{-1}. In this context, the results correlate with the previous studies related to NORM-related exposure scenarios, recognizing the lichen and bryophytes as organisms most sensitive to potential radiological hazards. Given the Tool's inherent conservativism and the effect of using the Tool's default CR values, which are known to lead to overestimation of the potential dose rates, the overall radiological risk in all assessment scenarios can be considered negligible. Nonetheless, the continuation of environmental monitoring is encouraged. The conclusions of this study should be observed in a particular research context, where the assessment results identifying the exposure situation as posing no significant risk to the environment could also be attributed to the gas industry in question setting and implementing robust standards of both radiological and environmental protection that are continuously being confirmed through monitoring and assessment.

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AUTHORS' CONTRIBUTIONS

Conceptualization of the paper was done by A. Getaldić and methodology by A. Getaldić and B. Skoko. B. Skoko conducted the formal analysis. B. Petrinec and T. Bituh provided the resources and data curation. Original draft was prepared by A. Getaldić, and review and editing was carried out by A. Getaldić, M. Surić Mihić, B. Skoko, B. Petrinec and T. Bituh. Supervision was performed by M. Surić Mihić and Ž. Veinović.

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ЗАШТИТА ОКОЛИНЕ У ИНДУСТРИЈИ ПРИРОДНОГ ГАСА – УПОРЕЂЕЊЕ РАЗЛИЧИТИХ ПРОЦЕНА ПРОСТОРНО-ВРЕМЕНСКИХ РАДИОЛОШКИХ РИЗИКА

Рад анализира резултате просторно-временских сценарија процене радиолошког ризика на темељу постојећих *in-situ* података дуготрајног мониторинга на локацији постројења за прераду природног гаса, да би се анализирао учинак различитих улазних података на исход процене. ERICA Assessment Tool коришћен је за процену доза на биоту и потенцијалних утицаја због изложености јонизујућем зрачењу. Улазни подаци за сценарије процене радиолошког ризика укључивали су годишње податке о концентрацији активности радионуклида у тлу из *in-situ* мерења обављених од 1994. до 2016. године те лабораторијске гамаспектрометријске податке који се односе на период од 2014. до 2019. године. Предвиђена укупна брзина дозе на биоту генерално је била испод јачине дозе алата ERICA од 10 μGyh^{-1} или незнатно повишена, с највећом укупном јачином дозе процењеном за лишајеве и бриофите. Укупне јачине доза на лишајеве и бриофите у посматраном раздобљу показују одређене временске варијације, али у раду није уочен одређени тренд. Процењене укупне јачине дозе за биоту из различитих сценарија процене биле су испод међународно предложених референтних нивоа за које се не очекују никакви штетни учинци. Утврђено је да је укупни потенцијални радиолошки ризик за копнену биоту због рада постројења за прераду природног гаса занемарив.

Кључне речи: NORM, природни гас, процена радиолошког ризика, заштита околине, алат Erica

3. DISCUSSION

In order to investigate the potential effect of soil depth on the radiological risk assessments, spatial sets of radiological data from legacy coal ash and slag disposal site and a remediated coal ash and slag disposal site were used in radiological risk assessments. Radiological risk assessment at the legacy coal ash and slag disposal site was conducted on the data from samples collected from three boreholes (B2, B3, and B4) at three depth ranges: 0–2 m, 2–4 m, and 4–6 m. For each depth range, one risk assessment was performed. In this research context, depth was used as a spatial risk assessment component. An additional comparison of the risk assessment results was based on using ERICA Assessment Tool's default CR values and site-specific CR values (adopted from **Skoko et al., 2019; Skoko et al., 2017**).

Another spatial assessment was conducted at a remediated coal and ash disposal site based on the radiological data before and after the remediation. Part of this data was adopted from the literature (**Marović and Bauman, 1986; Skanata et al., 1996; Marović et al., 2008**), and another data set included extensive soil samples from environmental monitoring conducted at the disposal site perimeter. The spatial component of this assessment refers to three different spatial datasets used, one prior to the remediation and two after it was completed.

The risk quotient results for three assessment scenarios (i.e. depth ranges) from the assessments that used the tool's default CR values at the legacy site showed the resulting risk quotient (RQ) to be below 1. The ERICA Assessment Tool's conservative RQ value was slightly above the value of 1 in three assessment scenarios, mainly those related to samples from greater depths (>4 m). Using site-specific CR values in the assessments resulted in the risk quotient and the conservative risk quotient below 1 in all assessment scenarios. Assessment at the remediated disposal site showed that both the overall expected risk quotient and the conservative risk quotient values were the highest in lichen and bryophytes, with a risk quotient of 14,77 and a conservative risk quotient of 44,3.

At the legacy coal and ash disposal site, the risk assessment results were analysed in relation to depth ranges based on the estimations for internal, external, and total dose rates for the selected reference organism – tree. In the assessment using default CR values for scenarios concerning all depth ranges, the main contributor to the external dose rate was ^{226}Ra. This was also the case in the assessments using site-specific CR values at all depth ranges. However, in all assessments, the total dose rate mainly resulted from the internal dose rate, contributing, on average, 90% to the total 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 ^{226}Ra and ^{238}U. The results concerning the depth range of 4–6 m showed that in addition to ^{226}Ra, ^{210}Pb and ^{210}Bi were also key contributors to the internal dose rate. The reason for this is the inherent feature of the ERICA Assessment tool, which includes short-lived radionuclides with half-lives under ten days in the assessment. Hence, ^{210}Bi, with a half-life of 5 days, was included as a direct progeny of ^{210}Pb. At this depth range, the radionuclide distribution of the internal dose rate in all samples showed that ^{226}Ra was the dominant radionuclide, accounting for approximately 70% of the internal dose rate, followed by ^{210}Pb and ^{210}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 ^{226}Ra, the Tool detected both ^{210}Pb and ^{210}Bi, as contributors to the internal dose rate. In this assessment scenario, the radionuclides contributing to the internal dose rate were distributed more evenly, with ^{226}Ra accounting for around 40% of the internal dose rate and ^{210}Pb and ^{210}Bi, each contributing approximately 30%. Since the assessment detected ^{210}Pb, two main pathways further elaborated its presence in plants: direct deposition from the atmosphere and an indirect route through the root system (**Vandehove et al., 2009**). The plant radionuclide uptake and accumulation mechanisms are also affected by several different factors related to both soil type and its traits, plant species and characteristics, and climate features (**Mrdakovic Popic et al., 2020; Černe et al., 2018; Madruga et al., 2001; Vandehove et al., 2007**). The atmospheric deposition was found to be a significant pathway for ^{210}Pb accumulation based on the fact that both the radiological risk assessment scenarios using default CR and site-specific CR values that detected ^{210}Pb as a contributor to the total dose rate relate to assessments performed at a depth deeper than 4 m. Considering that the estimated depths of root systems is up to 6 m, the research found the overall radiological risk from ^{210}Pb root uptake can be considered as negligible. This finding is in accordance with previous research where **Pietrzak-Flis and Skowrohska-Smolak (1995)** found that ^{210}Pb uptake by plants is primarily attributable to atmospheric deposition, mainly wet deposition, while the transfer through the root system can be regarded as insignificant. A study by **Skoko et al. (2017)** from the same coal ash and slag disposal site, using surface samples, detected similar activity concentrations of ^{210}Pb in both plants from the disposal site and the control site.

Analysis of the ERICA Assessment Tool estimations for internal, external, and total dose rates for the selected reference organism at the remediated site, but before the remediation took place, found the internal exposure to ^{226}Ra contributed the most to the total dose rate, especially in lichen and bryophytes and shrub as reference organisms. ^{226}Ra also contributed to the external dose rate, with amphibian, annelid, arthropod, mammal (small-burrowing) and reptile as the most affected reference organisms. The post-remediation assessment scenario results on the total dose rate showed that internal exposure contributes the most to the total dose rate to all reference organisms, with ^{226}Ra as the primary contributor. The highest internal dose rate was estimated in lichen and bryophytes, and shrub. This finding aligns with previous research; a study by **Sotiropoulou et al. (2016)** also found ^{226}Ra to be the main contributor to the internal dose rate to lichen and bryophytes in a similar research context. The external dose rate was primarily associated with ^{226}Ra, with the highest dose rates in amphibian, annelid, arthropod, mammal (small-burrowing) and reptile.

At the legacy site, regardless of the depth range, the radiological risk to the reference organism was found to be negligible, as the screening dose rate of 10 μGyh^{-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, which is an observation that was also made by other authors and is supported by previous research and assessments (**Mrdakovic Popic et al., 2020; Skoko et al., 2019; Brown et al., 2013**). The results of this study confirmed the risk assessment results of previous studies that used a surface layer of coal ash (**Skoko et al., 2019**), 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.

Although the assessment results from data before the disposal site remediation only slightly exceeded the ERICA Tool conservative screening value of 10 μGyh^{-1} for lichen and bryophytes, they were still below the value of 40 and 400 μGyh^{-1} for terrestrial biota for which no effects on population levels should be expected (**UNSCEAR, 1996**). Hence, the overall risk associated with the disposal site was found to be negligible. The overall assessment results that relied on data before the disposal site remediation are in line with results from previous studies on total dose rates to terrestrial biota that focused on NORM legacy site and mining. The study by **Mrdakovic Popic et al. (2020)**, at the legacy NORM site in Norway reported the highest predicted total dose rate of 206 μGyh^{-1} to lichen and bryophytes when default CR values were used and 23 μGyh^{-1} when site-specific soil and plant activity concentrations were used. **Oughton et al. (2013)** conducted risk assessments at several mining sites in Central Asia. Risk assessments used site-specific data and calculated aquatic and terrestrial biota dose rates. Findings from the study include assessment results related to disposal site containing uranium tailings with the highest total dose rate value of 660 μGyh^{-1} predicted to lichen and bryophytes. Additionally, this study also reported ^{226}Ra as the main contributor to the internal and external dose rates (**Oughton et al., 2013**), which was also the case in our assessments that used data before the site remediation. Assessment of dose rates to terrestrial biota around the coal-fired power plant in Serbia also resulted in screening dose rates being exceeded only for lichen and bryophytes (**Ćujić and Dragović, 2018**).

Temporal data sets were used for radiological risk assessments at two research locations: remediated coal ash and slag disposal site and a natural gas processing plant site.

The radiological risk assessment based on the data set from 2015, after the remediation of the coal and ash disposal site, resulted in much lower calculated risk quotient values, with the highest risk quotient value of 1,92 estimated to lichen and bryophytes. Based on the available data before and after the remediation, a comparison of internal and external dose rate estimations from exposure to ^{226}Ra for most affected reference organisms indicated a fifteen times decrease in the total dose rate estimations.

In the temporal context, assessment results from a risk assessment based on the data on soil samples from 2015 were in line with the results of the assessment scenario based on the data from 2008, although the activity concentrations from 2015 resulted in a slightly higher dose rate estimations. This could be explained by the 2015 dataset, including more extensive radiological data, thus increasing the overall risk quotient and estimated dose rates. The highest estimated value was found in lichen and bryophytes 18,06 μGyh^{-1}, where data from 2008 resulted in a predicted total dose rate to lichen and bryophytes of 10,92 μGyh^{-1}. ^{226}Ra contributed the most to reference organisms' internal and external dose rates.

Extensive temporal datasets, including in-situ gamma spectrometric data from 1994 to 2016 and laboratory gamma spectrometric data from 2013 to 2019, were used to conduct assessments at a natural gas processing plant site. The ERICA Assessment Tool's external and internal dose rate estimations were analysed to determine the dominant exposure route and key contributors to the dose rate. The assessments based on the in-situ gamma spectrometric data resulted in an external dose with ^{226}Ra as the main contributor, with amphibian, annelid, arthropod, small burrowing mammals and reptile as most affected organisms. The internal dose rate was also primarily associated with exposure to ^{226}Ra, with the highest internal dose rate to lichen and bryophytes and shrub. Again, the internal dose rate was the parameter that affected the estimated total dose rate the most, irrespective of the temporal aspect of the input data or whether the maximum activity concentrations were used in the assessment. The calculation of dose rate in assessment scenarios using laboratory gamma-spectrometric data showed that the main contributor to the external dose rate for all reference organisms was ^{226}Ra, with the highest contribution to amphibian, annelid, arthropod, mammals (small-burrowing), mollusc, and reptile. The data on the internal dose rate showed that ^{226}Ra contributes the most to the internal dose rate, primarily observed in reference to lichen and bryophytes and shrub. The total dose rate estimation was almost entirely attributed to the internal dose rate. The contribution of different radionuclides, specifically ^{226}Ra, to the total dose rate from our study is in accordance with results from previous studies related to exposure to naturally occurring radionuclides from other authors (**Oughton et al., 2013; Hosseini et al., 2011; Čujić and Dragović, 2018**).

Additionally, the presence of ^{226}Ra and the importance of its activity concentration for the assessment results is related to the fact that ^{226}Ra is the most prevalent radionuclide in scales and deposits found in the equipment of the oil and gas industry and discharges, and as such, can be a source of radiation exposure (**Hamlat et al., 2001; Gäfvert et al., 2006; Garner et al., 2015**). Individual temporal assessments that relied on the annual in-situ data from 1994 to 2016 resulted in estimated dose rates between the lowest of 0,1 μGyh^{-1} to the tree as a reference organism and the highest total dose rate of 10,13 μGyh^{-1} to lichen and bryophytes. The same data set, which was temporally averaged before calculation, resulted in an estimated total dose between 0,1 μGyh^{-1} for tree and 4,39 μGyh^{-1} for lichen and bryophytes.

Results from the assessment scenario that used laboratory gamma-spectrometric data from 2013 to 2019 showed that the total dose rate to biota ranges from 0,05 μGyh^{-1} for the tree to 15,20 μGyh^{-1} for lichen and bryophytes. An additional assessment scenario, performed using the maximum soil activity concentrations from 1994 to 2016, estimated the total dose rate from 0,26 μGyh^{-1} for tree and 10,87 μGyh^{-1} to lichen and bryophytes. In order to conduct an assessment considering the highest input values, maximum measured activity concentrations from all sampling locations were used. A comparison of estimated total dose rates to all reference organisms from assessment scenarios that used temporal average activity concentrations and maximum activity concentrations from in-situ gamma-spectrometric measurements showed an average increase of 160%. The maximum measured activity concentrations per radionuclide from all three sampling locations were tripled, and another assessment scenario was run with these parameters to estimate the potential cumulative radionuclide concentration effect. In this case, the predicted total dose rate to lichen and bryophytes was 32,5 μGyh^{-1}, which exceeded the ERICA Tool's default screening value, but was below the reference values of 400 μGyh^{-1} for terrestrial plants (**UNSCEAR, 1996**). These results would imply that even in the case of cumulative contamination, the predicted effects to the biota in the proximity of the facility would be below internationally recognized reference values.

Lichen and bryophytes were found to be the most affected reference organisms in all assessment scenarios at the natural gas processing plant site, which was also noticed in other assessments at other research sites in this study. This finding should be observed considering that lichen and bryophytes are highly radiosensitive organisms and are, as a result, often used as biomonitors of potential contamination and concerning both artificial and natural radionuclides (**Marovic et al., 2008; Garty et al., 2003; Kirchner and Daillant, 2002; Loppi et al., 2003**).

The estimated total dose rate value in the studied period was below the selected screening dose rate of 10 μGyh^{-1}, which, together with the assessment results based on the maximum input activity concentrations, implies that the potential radiological risk to terrestrial biota arising from the operation of the natural gas processing plant is not significant. The overall results from various temporal assessments, including in-situ and laboratory data, are in accordance with the results from previous studies. A study by **Ćujić and Dragović (2018)** assessed NORM-related total dose rate to lichen and of 14,4 μGyh^{-1}. **Lazarus et al. (2022)** reported estimated dose rates to terrestrial biota up to 3,7 μGyh^{-1} to mosses and lichen. A study by **MacIntosh et al. (2023)** on radiological risk assessment to marine biota from exposure to NORM related to decommissioning offshore oil and gas pipelines estimated a potential dose rate from external exposure up to 33 μGyh^{-1}.

Based on the types of production activities, the research locations where radiological risk assessments were conducted can be referred to as legacy coal and ash disposal site without monitoring, remediated coal and ash disposal site, and industrial site with ongoing activities related to natural gas processing. The overall radiological risk at all three sites was found to be negligible. The study results suggest that in cases where there are no discharges to the environment nor significant discharge fluctuations, ongoing industrial activities require environmental monitoring. This assumption might change in cases where there are substantial changes to the contaminant levels, in which case the radiological risk assessment would need to include the specific lifespan of certain organisms associated with the site that could be exposed to the contamination. Although the results of risk assessments related to coal ash and slag disposal in this study suggest minimal radiological effects and risks, assessment and monitoring of the sites where NORM waste and residues are disposed of is recommended, even in the case of a remediated disposal site.

This study includes several limitations and uncertainties associated with performed assessments. Studies at certain research locations were based on a limited number of samples available or had a relatively small number of radionuclides in the assessments. Other uncertainties are associated with the lack of experimental data on site-specific transfer values for certain research locations. Since the ERICA Assessment Tool is known to use a conservative approach when the Tool's default CR values are used, the underestimation of the assessed radiological effects and risks should be minimal.

Study limitations also relate to the use of only gamma-ray spectrometry as an analytical method and, consequently, the lack of data for radionuclides that are alpha emitters, such as ^{230}Th and ^{210}Po, which could point out a direction of future research. The activity of certain radionuclides concentrations was estimated based on its progeny under the assumption of secular equilibrium. Another source of uncertainty that might affect the study results arises from the ERICA Assessment Tool's inherent features related to assumptions on the homogeneous distribution of radionuclides in reference organisms and assumptions related to the occupancy factors.

4. CONCLUSION

Exploitation of mineral resources and raw materials that contain radionuclides of natural origin, mining, and mineral processing operations can result in enhanced activity concentrations of radionuclides in NORM waste and residues, which can lead to potential exposure to ionising radiation. This research focused on activities related to potential exposure to radiation, including coal combustion and oil and gas production. Environmental monitoring and assessment of these industrial activities is essential, considering that resulting waste and residues can contain significant amounts of NORM; the radionuclides in question are often long-lived, and can adversely impact human health, safety, and the environment. The assessments using the ERICA Assessment Tool, that were conducted as a part of this thesis, included three NORM-related locations in Croatia where NORM waste and residues are disposed, either at a legacy site or at a remediated disposal, site and natural gas processing plant site. The results of performed radiological risk assessments confirmed the overall radiological risk to be negligible at all three selected research locations. Across all risk assessment scenarios conducted through this research, lichen and bryophytes were found to be the most radiosensitive organisms with generally the highest predicted dose rates. The internal exposure contributed the most to the estimated total dose rate in all assessment scenarios, with ^{226}Ra as the key contributor.

Since the ERICA Assessment Tool enables to conduct radiological risk assessment considering the specific spatial and temporal context of particular research locations, performed assessments focused on different spatial and temporal data sets, given the specifics of each research site. An assessment related to the coal ash and slag legacy disposal site used different soil sampling depths as assessment input data in a spatial context. Depending on the reference organisms included in the assessment and their habitat, as was the case with the root depth of the Mediterranean flora, the research found that sample soil depth slightly affect the risk assessments results and the radionuclide accumulation. Further field research would be needed to clarify the exact role of sample depth in the radionuclide root uptake for specific organisms and radionuclides. The CR values used in the risk assessment significantly affected the assessment results, with the total dose rate estimations higher when the assessment included the ERICA Assessment Tool’s default CR values, as opposed to site-specific CR values. The overall results from spatial assessment at this research location imply that using soil surface samples, as opposed to using samples from deeper layers, is reasonable since the radiological risk assessment results did not exceed the selected screening dose rate.

In the context of radiological risk assessments based on the different temporal data sets, the assessment results related to a remediated coal ash and slag disposal site showed that the assessed radiological risk, and respective dose rates to reference organisms, after the site remediation were significantly lower when compared to the period before the remediation of the site. The results indicate the importance of environmental monitoring in ensuring long-term radiological and environmental protection and safety, and demonstrate the applicability of the ERICA Assessment Tool for confirmation of remediation effects.

Temporal assessment based on the data from the natural gas processing plant site showed that neither individual temporal assessments nor assessments based on the temporally averaged radiological data resulted in a significant risk to the environment. Across all assessment scenarios, a specific temporal trend was not noticed in the estimated dose rates. However, the results suggest that radiological and environmental protection should continuously be confirmed through reliable monitoring and assessment. The effect of sampling frequency at selected research locations on the radiological risk assessment results can be regarded as insignificant, given the research site specifics, namely, no fluctuations in discharge or contamination levels, and the absence of organisms with a short life span that could be affected by the exposure to radiation.

In the context of the type of industrial activities being performed at the particular research site, the radiological risk assessment results showed that total dose rate predictions were higher related to coal combustion in comparison to natural gas processing, especially in the context of a coal ash and slag legacy disposal site that is not being monitored. The industrial activities of natural gas processing, although having the potential to result in substantial exposure to radiation, due to the robust environmental protection standards practised at the site, the potential total dose rates estimations to biota were not found to be significant.

Considering the retrospective nature of the conducted risk assessments at given research locations and their stable contamination levels, although sampling depth and sampling frequency did not significantly contribute to the resulting risk assessment results, spatial and temporal assessments should be considered in the design of prospective radiological risk assessments that could comprehensively include spatial and temporal datasets, such as monitoring, as they can provide valuable insight.

5. LITERATURE

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