Heliyon 10 (2024) e27788

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Assessment of the radionuclide remediation potential of novel miscanthus hybrids

Vanja Jurišić[a], Davor Rašeta[b], Mislav Kontek[c], John Clifton-Brown[d,e], Luisa M. Trindade[f], Isabelle Lamy[g], Annie Guerin[h], Andreas Kiesel[i], Ana Matin[a], Tajana Krička[a], Branko Petrinec[b,*]

[a] University of Zagreb Faculty of Agriculture, Svetošimunska cesta 25, 10000, Zagreb, Croatia
[b] Institute for Medical Research and Occupational Health, Ksaverska cesta 2, 10000, Zagreb, Croatia
[c] Energovizija d.o.o., Ilica 42, 10000, Zagreb, Croatia
[d] Justus Liebig University Giessen, Ludwigstrasse 23, 35390, Giessen, Germany
[e] Institute of Biological, Environmental and Rural Sciences, Aberystwyth University, Aberystwyth, UK
[f] Wageningen University & Research, Plant Breeding, Droevendaalsesteeg 4, 6708, PB Wageningen, the Netherlands
[g] University Paris-Saclay, INRAE, AgroParisTech, UMR EcoSys, 91120, Palaiseau, France
[h] US 0010 Laboratoire d'Analyses des Sols (LAS), 62000, Arras, France
[i] University of Hohenheim, Institute of Crop Science, 70599, Stuttgart, Germany

ARTICLE INFO

Keywords:
Miscanthus
Radionuclides
Phytoremediation
Sustainability
Construction materials

ABSTRACT

There are few studies related to the radionuclide remediation options, which comply to the demands of the environmentally non-destructive physical remediation methods. So far, most of the research was conducted on the phytoremediation capacity of different energy crops, as well as the established miscanthus hybrids which involved metal and heavy metal contaminants. Hence, the objective of this research was the radioecological characterization of the examined agro-ecosystem, including the initial source of the radionuclides (soil) as well as different miscanthus hybrids grown on the same soil. The results have shown that the radioactive content of soil was similar to the global averages. All measurements of the activity concentration of ^{137}Cs in miscanthus samples were below the detection limits. There is also an indication that ^{210}Pb is leaching into the lower layers (or is being taken up by miscanthus plant from the upper layers). Moreover, transfer factors (TFs) for radionuclides, as a more precise parameter for evaluating the phytoremediation potential, were calculated; the TFs were found to be very low for ^{226}Ra ($\leq$0.07), TFs for ^{40}K ($\leq$0.39) and for ^{232}Th ($\leq$0.21) were in the lower limits, whereas the TFs for ^{238}U were found to be the highest ($\leq$0.92). For ^{210}Pb, the TFs were not calculated, since the expectation was that a significant part of the measured quantity came from the air, and not through the soil. Having in mind the sustainability and the circularity aspect of the radionuclide phytoremediation system, the appropriate management method should be applied for the disposal and utilization of the biomass contaminated with radionuclides. This research has shown that the radiological content in miscanthus is high enough and the ash content is low enough that miscanthus ash could be considered as a NORM (Naturally Occurring Radioactive Material), and it can be further

---

[*] Corresponding author.

E-mail addresses: vjurisic@agr.hr (V. Jurišić), draseta@imi.hr (D. Rašeta), mislav.kontek@energovizija.hr (M. Kontek), John.Clifton-Brown@agrar.uni-giessen.de (J. Clifton-Brown), luisa.trindade@wur.nl (L.M. Trindade), isabelle.lamy@inrae.fr (I. Lamy), annie.guerin@inrae.fr (A. Guerin), a.kiesel@uni-hohenheim.de (A. Kiesel), amatin@agr.hr (A. Matin), tkricka@agr.hr (T. Krička), petrinec@imi.hr (B. Petrinec).

https://doi.org/10.1016/j.heliyon.2024.e27788
Received 17 August 2023; Received in revised form 14 January 2024; Accepted 6 March 2024

used for the construction industry (i.e. concrete, tiles), in mixtures with other materials with certain limitations, similar to the utilization of ash from other sources such as coal or wood.

---

1. Introduction

Soil contamination, as a result of different human activities during the last several decades, is a major environmental issue worldwide. Assessment of the natural radionuclides' activity and heavy metals concentrations in the environment is of benefit to human health. It is imperative to establish and know the background levels of both, radionuclides, and heavy metals in different sources [1], as well as to find appropriate measures for their remediation. So far, most of the research was conducted on the phytoremediation capacity of different energy crops, as well as the established miscanthus genotypes which involved metal and heavy metal contaminants [2–5]. *Miscanthus x giganteus* [3,6–11] and *Miscanthus sinensis* [7,11] were mostly investigated, and have shown a great potential to stabilize and possibly remove the potentially toxic elements over the time, especially zinc, mercury, arsenic, cobalt, and manganese. Aside the metal contaminants, several studies have examined the possibility of phytoremediation of soils contaminated with polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs) and ^{232}Th radionuclides. *M. x giganteus* has shown the ability to maintain a degrading rhizosphere microflora thus enhancing the dissipation of recalcitrant PAH compounds in polluted soils [12]. *M. sinensis* could grow satisfactorily in the soil contaminated with OCPs and accumulate them in the plant's tissues [13]. *M. floridulus* has the potential for phytoremediation of ^{232}Th radionuclides, compared to other plants used in the research described by Yan [14].

However, there is only a scarce number of studies related to the radionuclide remediation options, which comply to the demands of the environmentally non-destructive physical remediation methods. Naturally occurring materials generally contain radioactive nuclides from the main decay chains for ^{238}U, ^{235}U, ^{232}Th, and their daughter products, and from the long-lived radioactive nuclides such as ^{40}K [15]. However, the occurrence of anthropogenic (artificial) radioactivity and technologically enhanced natural radioactivity causes the alteration of the natural ecological characteristics of the biosphere [16]. The commonly encountered radionuclides include ^{232}Th, ^{239}Pu, ^{226}Ra, ^{60}Co, ^{222}Rd, ^{99}Tc, and ^{238}U [17]. Primordial radionuclides, which emit ionizing radiation, are present all around us in the environment, especially in soil. So is ^{137}Cs, following nuclear fallout from the atmospheric testing and the Chernobyl accident. Radionuclides such as ^{192}Ir, ^{201}Tl, ^{90}Sr, and ^{137}Cs are also produced during the splitting of elemental atoms in the nuclear power plant and they impose a serious hazard, especially to ecosystems [17]. Hence, monitoring radionuclides in the soil, water and air is important to protect human health and the environment, since the ionizing radiation carries enough energy to ionize the atoms of the material it passes through, including human tissue, by knocking the electrons out of those atoms [18].

Sources of contamination are many. Parts of Europe and Japan are still contaminated by the fallout from the Chernobyl and Fukushima accidents. Control measures for last sheep farms in Great Britain were lifted only in 2012 [19]. Countries with military nuclear programs have areas contaminated by those programs. Uranium mines are also considered contaminated areas. Contaminated areas are also often the result of some industries, such as energy industry (coal ash dumps, even areas around some coal-fired power plants before the filters were mandatory), oil and gas industry (pipes and drills are often contaminated, and drilling often brings soil with higher radionuclide content to the surface), fertilizer industry (phosphogypsum), ash and refuse from ceramic industry, decorative stone industry etc. Mines and quarries used as a source material for those industries can also be contaminated areas. Rare earths are often found in rocks that contain elevated levels of ^{232}Th, so mining and refinement of rare earths can create highly contaminated areas.

Given this, it's important to implement remediation strategies to prevent radionuclides from spreading into various environments, including land, air, and water, with the aim of reducing contamination either partially or completely. Various remediation approaches have been made in the past which include mechanical techniques such as soil incineration, excavation and land fill, soil washing, solidification, and electric field application [17]. However, conventional remediation technologies are costly and less suitable for removing the large-scale contamination. Phytoremediation, which uses plants to remove radionuclides *in situ* from the contaminated sites, is a promising alternative for environmental remediation [20]. Plants are useful even for remediating the environment with low concentration of contaminant due to their extensive root system [17].

Currently, despite the major steps done in this regard (e.g. RED II directive), the majority of biofuels still originate from food crops grown on the agricultural land, whereas the most of the wooden biomass used for energy production are of forestry origin. In order to avoid the conflict between food and fuel production, energy crops should be grown on the low-quality land wherever possible. It makes marginal and degraded land a favourable place for cultivating energy crops while dedicating the high-quality arable lands for food production [21,22]. Moreover, the cultivation of energy crops on marginal and contaminated areas provides opportunities for the *in situ* remediation which could also have a significant economic and social impact [22,23].

Today, miscanthus is one of the leading perennial energy crops in Europe due to its high dry matter yields under a wide range of agroclimatic conditions [24,25]. The most common commercially available cultivar is *Miscanthus x giganteus*, a natural sterile hybrid of *M. sinensis* and *M. sacchariflorus*. However, commercially obtained yields are often much lower than those scientifically determined and are commonly referred to as the 'commercial yield gap' [26]. There are several breeding programs in Europe to develop new varieties with improved traits [26–28]. The general objective of these programs is to optimize the production systems of miscanthus genotypes for different applications, mainly for production of bioenergy and bioproducts [29]. In addition to technological advances, the breeding programs can indirectly improve the other capacities of certain genotypes, for example, promote the phytoremediation capacity and revitalization of polluted soils. Miscanthus has been shown to be a useful crop for decontamination of those soils [13].

Fig. 1. Experimental station Šašinovec, miscanthus trial and soil cores.

Table 1
Soil analyses (n = 8).

Physical properties					
Coarse sand (g kg^{-1})	Fine sand (g kg^{-1})	Coarse silt (g kg^{-1})	Fine silt (g kg^{-1})	Clay (g kg^{-1})	Moisture (g kg^{-1})
53.4 ±7.95	32.4 ±14.1	291.4 ±6.4	379.8 ±5.5	243.1 ±18.8	19.6 ±3.2
Chemical properties					
pH in H_2O	**$CaCO_3$ (g kg^{-1})**	**TOC (g kg^{-1})**	**Total N (g kg^{-1})**	**C:N ratio**	**Organic matter (g kg^{-1})**
5.9 ±0.37	<0.1	11.5 ±1.5	1.1 ±0.1	10.9 ±0.4	19.8 ±2.6

*all values (except pH and C/N) are expressed as a percentage on dry matter basis.

The objective of this research was to compare the ability of different miscanthus hybrids grown on the same soil to uptake several radionuclides, using a radioecological characterization of the examined agroecosystem. The characterization was performed by the radiological characterization of the initial source of the radionuclides (soil) and the radiological characterization of the miscanthus hybrids. From there, transfer of radionuclides from soil into crop (miscanthus) can be analysed. One of the important parameters characterizing the transfer of radionuclides from soil into the crops, and the one used here, is the transfer factor. Established transfer factors can later be used in transfer models to predict the concentration of radionuclides in biomass.

2. Materials

In this research, the above-ground biomass of 14 different miscanthus hybrids was used, as well as homogenous soil samples at three soil depths. Hybrids were grown at the University of Zagreb Faculty of Agriculture Experimental field Šašinovec (45°50′59.3″N 16°11′26.2″E) (Fig. 1). The field trial within the H2020 BBI-DEMO project No. 745012 "GRowing Advanced industrial Crops on marginal lands for bioEfineries - GRACE" of 14 seed- or rhizome-based miscanthus hybrids was established in March 2018, in a randomised complete block design with four replicates. GRC1-GRC8 represent novel intraspecific seed-based *M. sinensis x M. sinensis* hybrids, selected from the WUR, Netherlands breeding program; GRC10, GRC 11, GRC13 and GRC14 are novel interspecific seed-based *M. sinensis x M. sacchariflorus* hybrids selected from the Aberystwyth University, UK breeding program, as well as the rhizome-based *M. x giganteus* GRC9 (commercial clonal standard) and GRC15 (TV1) provided by Terravesta (Lincoln, UK). The biomass of each hybrid was sampled from all four replicates. After sampling, the biomass was homogenized, and its dry matter content was determined [30].

For the purpose of this study, the soil and the biomass samples were sampled in March 2020, and March 2021; the latter being the year when the full yield potential was obtained. The biomass was sampled following the dormancy of the plants from the previous year's growing season. This timing allowed the natural leaching of soluble substances due to weathering and a consequent loss of a significant portion of the foliage.

3. Methods

3.1. Soil analyses

Prior to the radionuclides' measurements and determination, the initial soil analysis was performed at INRAE, France (Laboratoire

Table 2
Miscanthus biomass analyses (n = 3).

Parameter		GRC 1	GRC 2	GRC 3	GRC 4	GRC 5	GRC 6	GRC 7
Moisture (%)	2020	6.2 ±0.1**	7.1 ±0.1	6.7 ±0.2	7.6 ±0.1	7.6 ±0.1	8.0 ±0.1	8.5 ±0.2
	2021	20.3 ±3.1	23.2 ±4.6	21.9 ±5.3	19.8 ±5.8	25.0 ±5.4	21.8 ±4.6	20.9 ±5.8
Ash (%db)*	2020	3.6 ±0.22	4.2 ±0.66	3.4 ±0.29	2.8 ±0.17	3.2 ±0.15	3.6 ±0.23	3.7 ±0.06
	2021	1.8 ±0.1	2.9 ±0.1	1.5 ±0.1	2.1 ±0.1	2.2 ±0.1	2.6 ±0.1	3.7 ±0.2
Coke (%db)	2020	14.1 ±0.81	15.0 ±0.1	14.6 ±0.2	12.7 ±0.3	13.6 ±0.3	13.6 ±0.3	12.3 ±1.4
	2021	9.3 ±0.5	10.7 ±0.9	10.3 ±1.3	10.4 ±0.8	10.6 ±0.2	10.6 ±0.3	11.5 ±0.2
Fixed carbon (%db)	2020	10.6 ±0.8	10.7 ±0.1	11.2 ±0.2	9.9 ±0.3	10.4 ±0.3	10.16 ±0.3	8.6 ±1.4
	2021	8.0 ±1.2	7.9 ±1	8.8 ±1.2	8.3 ±0.8	8.4 ±0.2	8.0 ±0.3	7.9 ±0.3
Volatile matter (%db)	2020	78.9 ±0.8	78.0 ±0.1	78.1 ±0.2	79.6 ±0.3	79.4 ±0.3	79.0 ±0.3	80.4 ±1.4
	2021	85.0 ±0.6	83.4 ±0.7	82.2 ±3.6	84.7 ±1.0	85.0 ±1.1	83.8 ±0.2	84.4 ±2.2
HHV (MJ kg^{-1})	2020	17.7	17.1	18.1 ±0.2	18.0 ±0.2	17.9 ±0.2	18.0	18.1 ±0.1
	2021	17.8 ±0.1	17.4	17.6 ±0.1	17.6 ±0.19	17.4 ±0.1	17.6 ±0.1	17.4 ±0.1
Parameter		**GRC 8**	**GRC 9**	**GRC10**	**GRC11**	**GRC13**	**GRC14**	**GRC15**
Moisture (%)	2020	9.1 ±0.2	6.5 ±0.3	6.6 ±0.2	6.7 ±0.1	7.0 ±0.1	6.8 ±0.1	7.0
	2021	24.1 ±5.6	29.9 ±4.0	47.3 ±4.7	42.9 ±3.1	44.4 ±4.9	43.3 ±3.3	33.5 ±5.3
Ash (%db)	2020	3.1 ±0.2	4.9 ±4.3	3.9 ±0.1	7.2 ±0.4	3.7 ±0.1	4.1	4.0 ±0.1
	2021	2.3 ±0.2	1.9 ±0.1	3.4 ±0.1	3.0 ±0.3	3.0 ±0.1	3.1 ±0.1	1.6 ±0.1
Coke (%db)	2020	11.6 ±0.9	13.6 ±0.4	14.8 ±1.0	16.5 ±0.4	14.5 ±0.5	13.9 ±0.4	14.0 ±0.3
	2021	10.6 ±0.6	11.2 ±0.6	11.1 ±0.4	10.3 ±0.4	7.7 ±0.9	11.0 ±0.3	11.9 ±0.4
Fixed carbon (%db)	2020	8.4 ±0.9	8.7 ± 4.4	10.9 ± 1.0	9.3 ± 0.6	10.8 ± 0.6	9.7 ± 0.4	10.0 ± 0.2
	2021	8.3 ±0.71	9.3 ±0.66	7.7 ±0.44	7.3 ±0.42	4.7 ±0.89	7.9 ±0.32	10.3 ±0.5
Volatile matter (%db)	2020	81.4 ±0.9	79.9 ±0.1	78.6 ±1.1	76.8 ±0.4	78.5 ±0.6	79.4 ±0.5	79.1 ±0.3
	2021	83.9 ±0.8	84.0 ±0.7	82.2 ±2.9	83.0 ±2.0	84.7 ±0.9	84.9 ±1.8	84.3 ±2.2
HHV (MJ kg^{-1})	2020	18.1 ±0.0	18.4 ±0.0	18.8	17.8	18.5	18.4	18.3 ±0.2
	2021	17.7 ±0.1	17.8 ±0.1	17.3	17.0 ±0.45	17.3 ±0.1	17.5 ±0.1	17.6 ±0.1

*db = dry basis; GRC1-GRC8 represent novel seed-based *M. sinensis* x *M. sinensis* hybrids, provided by WUR, Netherlands; GRC10, GRC 11, GRC13, GRC14 are novel seed-based *M. sinensis x M. sacchariflorus* hybrids provided by IBERS, UK; rhizome-based *M. x giganteus* GRC9 (commercial) and GRC15 (TV1) were provided by Terravesta.

d'analyse des sols, Arras, Cofrac accredited ISO 17025). For this purpose, in November 2017, eight soil cores (0–30 cm) were extracted by a hand driven soil corer to quantify physical and chemical indicators. Physical analyses comprehended particle size distribution [31] and moisture content [32]. Chemical analyses determined carbonate content [33], total organic carbon [34] and total N [35]. The results of the soil physical and chemical properties, prior to miscanthus plantation establishment are presented in Table 1.

3.2. Biomass analyses

For biomass proximate and ultimate analyses, the following parameters were determined (Table 2): initial moisture content [30], ash content [36], coke, fixed carbon and volatile matter content [37], and the calorific value [38] by using an adiabatic calorimeter.

An average ratio of stem:foliage was found to be 4.0, ranging from 2.1 to 9.7 for *M. sinensis x M. sinensis* and from 2.4 to 7.4 for *M. sinensis x M. sacchariflorus* hybrids.

3.3. Radionuclide analyses

All the radionuclides' measurements were performed in the Laboratory of Radiation Protection Unit of the Institute for Medical Research and Occupational Health. High resolution gamma spectrometry in energy range (40–2000 keV) was used for the purpose of radionuclides' analyses. The radionuclides of interest in the environmental samples were ^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{40}K, and ^{137}Cs [39, 40]. High purity Ge Mirion Technologies (Canberra), Inc. GC5019 photon detector system, 54 % relative efficiency and resolution of 1.9 keV, all at 1.33 MeV, was used to measure miscanthus samples. High-purity Ge photon detector system ORTEC HP GMX, 74 % relative efficiency and resolution of 2.26 keV, all at 1.33 MeV, was used to measure soil samples.

All the selected radionuclides are γ emitters, although ^{238}U and ^{232}Th cannot be detected through their own γ rays. ^{40}K, and ^{137}Cs are also β emitters, and ^{238}U, ^{232}Th, ^{226}Ra, and ^{210}Pb are also α emitters. ^{238}U, ^{232}Th, and ^{226}Ra also have progeny, including short-lived radionuclides, which are α, β, and γ emitters.

α, β, and γ rays can all ionize atoms they collide with. γ rays are the most penetrating, but they ionize relatively few atoms per ray. For γ rays, there is little difference whether they are inside an organism or come from the outside. β rays have much lower penetration than γ rays, but high energy β particles can still penetrate over a cm through a living tissue. Inside an organism, they can ionize many more atoms than γ rays. α rays cannot penetrate outer layer of organism (skin, bark etc.). Inside the organism, however, they can ionize millions of atoms. The actual effect of ionization depends. If affected molecules split or copy, they can transfer the error. If enough molecules in a single area are affected, the function can be impaired.

The soil samples were taken from fifteen different positions within the field, from depths of 0–5 cm, 5–10 cm and 10–15 cm. Before the establishment of the plantation, the soil was deep ploughed (depth >50 cm); hence, soil uniformity, including the uniformity of activity concentrations of radionuclides, was expected in the upper soil layers. The soil samples were collected from the whole field (0.6 ha) using a random sampling method. A single composite sample was obtained from 15 individual subsamples from a given area, and at each depth interval, in order to form a representative sample that reflects the average radionuclide concentration within this micro-location. The approach was based in the understanding that a 1-ha area, as is the case in this research, is perceived as a micro-location in terms of the radionuclide distribution, which is supported by the previous research indicating radionuclide homogeneity in the area of approx. 0.6 ha. The homogenization method effectively reduces the sampling error that might arise from a small-scale variability, which is particularly applicable where large-scale heterogeneity is not expected. Thus, for the scope of this study, a detailed sampling error calculation was deemed unnecessary.

In terms of the origin of the radionuclide pollution in soil, there is no site contamination, except the ^{137}Cs contamination present in Europe as a result of the Chernobyl and Fukushima accidents, as well as the atmospheric nuclear weapon testing.

Samples of ∼2 kg wet weight were placed in PET bags, sealed, and transported to the laboratory. Soil samples were dried out, cleared of impurities (grass, dead leaves and similar), homogenized, and packed into 1-L Marinelli beakers.

Three whole plants of each miscanthus hybrid were taken from each plot within four replicates. Plant samples were dried at 105 °C for 24 h, chopped into small pieces, milled in a laboratory mill, sieved through a 1 mm screen, homogenized and packed into the 200-mL cylindrical containers until the containers were fully filled. The material was packed as tightly as possible by hand and weighted. The uncertainty of mass measurement, no more than 0.1 g, was accounted for in the uncertainty budget of activity concentration. Both geometries are in standard use in the laboratory. Calibrations for both geometries were performed using the calibration standards acquired from the Czech Metrology Institute in the same geometries. Calibrations included correction for coincidence summing.

All the samples were carefully closed and sealed with a tape and left undisturbed long enough to reach the secular equilibrium in Uranium (^{238}U) and Thorium (^{232}Th) decay chains with respect to the Rn gas and its progenies (^{222}Rn in ^{238}U decay chain and ^{220}Rn in ^{232} Th day chain).

3.4. Data processing and analysis

Activity concentrations of ^{210}Pb, ^{40}K, and ^{137}Cs can be directly measured by using gamma ray spectrometry. This method identifies radionuclides and measures their activity in a sample using distinct γ-ray energy emissions. High-purity semi-conductor (Germanium) absorbs γ-ray and emits electric charge proportional to the γ-ray energy. Activity concentrations of ^{238}U, ^{232}Th and ^{226}Ra were determined by measuring the activity concentrations of short-lived radionuclides in ^{238}U and ^{232} Th day chains that are in secular equilibrium with the radionuclides of interest. Those are ^{234}Th for ^{238}U, ^{214}Pb and ^{214}Bi for ^{226}Ra, and ^{228}Ac for ^{232}Th (^{212}Pb, ^{212}Bi, and ^{208}Tl can also be used).

3.5. Transfer factor

The transfer factor is used as an index for the accumulation of a target element in the plant and its concentration in soil [15,16]. Transfer factors were calculated as:

$$\mathrm{Ac(R)_{plant}} \Big/ \mathrm{Ac(R)_{soil}} \tag{1}$$

where Ac is the activity concentration, and R is a radionuclide. In all calculations, soil measurements from the lowest level (10–15 cm) were considered. The activity concentrations are close to uniform (see Table 3), and the lowest layer should be the least susceptible to the surface events (rain, wind) and should be the closest to the activity concentrations of radionuclides up to the depth of ploughing. Both $\mathrm{Ac_{plant}}$ and $\mathrm{Ac_{soil}}$ are calculated for dry weight and are expressed in $\mathrm{Bqkg^{-1}}$.

Table 3
Activity concentrations (expressed in $Bqkg^{-1}$) of the selected radionuclides in soil samples taken from the same field on the same date in 2020 and in 2021. The values presented include 2σ confidence interval.

	Year 2020			Year 2021		
Radionuclide	0–5 cm	5–10 cm	10–15 cm	0–5 cm	5–10 cm	10–15 cm
^{40}K	571 ± 6	546 ± 6	577 ± 6	531 ± 6	552 ± 6	557 ± 6
^{238}U	64 ± 3	64 ± 2	63 ± 3	61 ± 2	63 ± 2	62 ± 2
^{226}Ra	54.2 ± 0.6	53.3 ± 0.6	54.8 ± 0.5	51.7 ± 0.5	51.3 ± 0.6	53.0 ± 0.5
^{210}Pb	80 ± 10	80 ± 20	70 ± 10	70 ± 10	72 ± 9	80 ± 10
^{232}Th	54 ± 2	48.8 ± 0.9	52 ± 1	48 ± 1	49 ± 1	51 ± 1
^{137}Cs	9.3 ± 0.4	9.0 ± 0.4	9.7 ± 0.4	7.6 ± 0.3	8.1 ± 0.4	8.2 ± 0.3

3.6. *Activity concentration index*

The combustion residues, meaning the non-combustible part of biomass, i.e., ash, is a heterogeneous mixture of inorganic and mineral substances suitable for production and/or mixing with the existing building industry products (e.g., cement and concrete). If miscanthus ash is classified as a NORM[1] (Naturally Occurring Radioactive Material) (depending on the local regulations), it has to comply with[2]:

$$I = Ac_{226Ra}/300 + Ac_{232Th}/200 + Ac_{40K}/3000 < 1 \tag{2}$$

where I is the activity concentration index is the Ac_{226Ra} is the activity concentration of ^{226}Ra, Ac_{232Th} is the activity concentration of ^{232}Th and Ac_{40K} is the activity concentration of ^{40}K.

4. Results and discussion

4.1. *Soil activity concentration*

Radionuclides in the soil solution constitute a pool available for the root uptake. This pool of radionuclides is also available for downward migration within the soil profile. The migration depth of radionuclides in contaminated soil is an important factor in the determination of the decrease of external dose rates resulting from the contamination, as well as in the determination of decontamination strategies, like phytoremediation [17]. The soil where the investigated hybrids were grown was deep ploughed in autumn 2017 prior to the establishment of the plantation.

The uptake of radionuclides depends on the various soil and plant factors such as soil types, cation exchange capacity, organic matter content, soil pH, as well as the plant species, root development and root system [14]. In this study, the pH of the soil was found to be slightly acidic (pH around 5.9) which is not in favor of radionuclide sorption process on soil constituents but rather contributes to their mobility. Furthermore, this acidity might have a great effect on the generation of soil colloid, radionuclide hydrolysis and ion exchange reaction, which all affect the adsorption of the radionuclide [41]. The particle size distribution shows that the soil samples from the studied area have a low clay content ranging from 3.6 % to 5.6 % with the soil texture estimated to be silt loam according to the soil textural triangle. It is well established that the mobility of Cs^+ is linked to the sorption to clay minerals, thus the low clay content is more in favor of its mobility. The organic matter content in soil was found to be 19.84 ± 2.60 g/kg. Total N and total organic C in soils ranged from 1.05 ± 0.12 g/kg and 11.50 ± 1.52 g/kg, respectively. The C/N ratio was found to be around 11 which is a classical value for the surface horizon of an agricultural soil. Based on the properties, the soil is characterized as a silt-loam pseudogley, with vertical texture contrast and periodic stagnation of the precipitation water.

The activity of the radionuclide is a number of radioactive decays per second (Bq). When the source is very large, such as in the environmental monitoring (soil, air), the total activity is of very little interest. Hence, the activity concentration, that is, the activity per mass or per volume is a preferred parameter [39,40]. The activity concentrations of radionuclides were measured by using gamma-ray spectrometry method. This method can adequately identify and measure radionuclides of interest – ^{137}Cs and naturally occurring radionuclides. Naturally occurring radionuclides which do not emit γ-rays are in secular equilibrium with their progeny that emits γ-rays, so their activity concentration is determined through their progeny. The results of the soil measurements are presented in Table 3.

Before the investigated miscanthus hybrids were planted in 2018, the specific area where the plantation was established was deep ploughed. The expectation was that it would ensure the soil uniformity, including the uniformity of the activity concentrations of radionuclides. As the results for the year 2020 samples in Table 3 show, the expectation was correct, and the radionuclides are

---

[1] NORM can be used to describe any material which contains natural radioactive elements, but usually NORM is used for the material where human activity increases the likelihood of exposure – for example, uranium ore in the soil would not be referred to as NORM, but once dug up, it would be referred to as NORM.

[2] This formula for activity concentration index is recommended in Ref. [57] and is used in the EU, but the factors may vary. In Croatia, factor 1 is considered conservative, while in some countries conservative factor may be 0.5.

Table 4
Mass concentrations of selected radionuclides in soil and mass concentrations of elements calculated from their significant radioactive isotopes, where applicable.

	Year 2020			Year 2021		
Radionuclide	0–5 cm	5–10 cm	10–15 cm	0–5 cm	5–10 cm	10–15 cm
^{40}K (mg kg^{-1} soil)	2.19	2.10	2.22	2.04	2.12	2.14
	±2	±2	±2	±2	±2	±2
(total potassium)	18.3	17.5	18.5	17.0	17.7	17.8
(g k-1 g soil)	±0.2	±0.2	±0.2	±0.2	±0.2	±0.2
^{238}U (mg kg^{-1} soil)	5.2	5.2	5.2	5.0	5.2	5.1
	±0.2	±0.2	±0.2	±0.2	±0.2	±0.2
(total uranium)	5.3	5.3	5.2	5.0	5.2	5.1
(mg kg-1 soil)	±0.2	±0.2	±0.2	±0.2	±0.2	±0.2
^{226}Ra (ng kg^{-1} soil)	1.5	1.4	1.5	1.4	1.4	1.4
	±0.02	±0.02	±0.01	±0.01	±0.02	±0.01
^{210}Pb (pg kg^{-1} soil)	25	25	22	22	22	25
	±3	±6	±3	±3	±3	±3
^{232}Th (mg kg^{-1} soil) = $^{\text{total thorium}}$	13.3	12.0	12.8	11.8	12.0	12.5
	±0.5	±0.2	±0.2	±0.2	±0.2	±0.2
^{137}Cs (pg kg^{-1} soil)	2.9	2.8	3.0	2.3	2.5	2.52
	±0.1	±0.1	±0.1	±0.1	±0.1	±0.09

uniformly distributed. The activity concentrations of ^{40}K, ^{238}U, ^{232}Th, and ^{210}Pb in soil were found to be higher than the average activity concentrations in soil in Croatia and in the world [39,40,42]. Even though they were slightly elevated, these activity concentrations are not unusual and are not close to the extremes measured in Croatia, let alone in the world [39,40,42]. The activity concentration of ^{226}Ra is similar to the average activity concentration in Croatia [39], and the activity concentration of ^{137}Cs is less than half the average activity concentration in soil in Croatia [40].

^{238}U, ^{232}Th, and ^{40}K are primordial radionuclides; they had existed in the universe from before the Earth was formed [18,39,40]. ^{137}Cs is anthropogenic (man-made) radionuclide, present in the environment in Europe mainly as a result of the Chernobyl accident. Some ^{137}Cs found in the environment in Europe is a result of the atmospheric testing of the nuclear weapons, while small amounts of ^{137}Cs in Europe come from the Fukushima accident. ^{137}Cs is the only anthropogenic gamma-ray emitting radionuclide of interest routinely found in the environment [18,40].

Table 4 presents mass of concentration of analysed radionuclides. From the activities of the primordial radionuclides, total mass of those elements can be calculated – ^{40}K comprises 0.01 % total potassium, ^{238}U comprises 99.3 % of total uranium, and ^{232}Th comprises more than 99.9 % of total thorium.

Comparing the results for the year 2021 to those obtained for the year 2020, the activity concentrations of the radionuclides in the uranium and thorium decay series are similar enough. Moreover, it was determined that the only radionuclides where 2σ uncertainty intervals do not overlap for at least two layers are ^{40}K, ^{226}Ra and ^{137}Cs. It is important to note that the error presented in Table 2 is a measurement error and does not include the sampling error, which may easily explain the ^{226}Ra difference. While ^{40}K difference between the 2020 and 2021 soil samples might also be explained by the sampling error, it should be noted that K is an element routinely taken in by the plants from the soil, and ^{40}K is present in all the natural K (0.012 % of K). Both Cs and K are elements of the first group of the Periodic table of elements. Both elements have one electron in outer shell and therefore their chemical properties are similar [40]. That means that the living organisms can take Cs instead of K, and the other way around. Saturating the soil with K helps in limiting ^{137}Cs intake by plants [43], while ingesting the extra K lowers biological half-life of ^{137}Cs in humans [44].

The results indicate that the soil cultivation ensured that, when miscanthus was planted, a vertical distribution of all the selected radionuclides was uniform or close to uniform. Since ^{137}Cs does not naturally exist in the soil, it usually diffuses from the surface, where it was deposited, into deeper layers. Vertical distribution of ^{137}Cs and ^{210}Pb in various soils is discussed in Ref. [45]. In the surface layers, the similar process exists for ^{210}Pb, where a significant part of ^{210}Pb activity concentration comes not from the decay of its progenies within the soil, but from the decay of the progenies in the air and on the surface, after the ^{222}Ra leched into the atmosphere. Considering solely the mean values in Table 2, there is an indication that ^{210}Pb is leaching into the lower layers (or is being taken up by miscanthus plant from the upper layers); however, large uncertainties mean that it cannot be concluded that the effect is not the result of a measurement uncertainty.

> It is worth noting that the soil remediation is performed on contaminated soil, that is, on soil that contains several orders of magnitude more radionuclides than the average soil. The radionuclide content of soil in this study is close to the world (and Croatian) average.

4.2. Miscanthus activity concentration

After the radionuclides' analyses in soil, the uptake and distribution of the uptake of radionuclides was studied for the miscanthus samples. Table 5 shows the activity concentrations of the selected radionuclides in plant material.

^{40}K comprises 0.012 % of all natural potassium. Of the 14 hybrids measured, only 9 had the ^{40}K activity concentration above the

Table 5

Activity concentrations of the selected radionuclides in miscanthus samples, expressed in Bq kg^{-1}. 14 samples were taken for 14 different hybrids (named GRC 1 through 15, and w/o GRC 12) on the same date in 2020 and in 2021. The values presented include 2σ confidence interval.

Radionuclide		GRC 1	GRC 2	GRC 3	GRC 4	GRC 5	GRC 6	GRC 7
^{40}K	2020	68 ± 2	<11.2	<16.9	<19.4	80 ± 3	<20.5	<20.4
	2021	23.5 ± 0.7	<13.5	24.3 ± 0.6	80 ± 2	37 ± 1	<9.1	<11.8
^{238}U	2020	22 ± 1	<12.3	29 ± 3	<19.3	<15.4	23 ± 2	<18.1
	2021	23 ± 2	<11.5	44 ± 4	37 ± 3	34 ± 2	<8.3	45 ± 3
^{226}Ra	2020	<2.0	<2.4	<2.9	<3.3	<2.7	<3.3	<3.7
	2021	<2.7	<2.4	<1.9	<1.7	3.6 ± 0.6	2.8 ± 0.6	2.7 ± 0.4
^{210}Pb	2020	<80	<88	<133	<147	<127	<167	<182
	2021	140 ± 40	110 ± 20	<73	180 ± 50	120 ± 30	80 ± 20	<90
^{232}Th	2020	5.2 ± 0.6	<4.6	<7.0	<6.9	<8.1	<6.8	<7.3
	2021	<4.4	<4.7	5 ± 1	<3.6	11 ± 2	4.3 ± 0.5	<4.2
^{137}Cs	2020	<1.1	<1.5	<1.8	<1.6	<1.8	<2.1	<2.0
	2021	<1.3	<1.1	<0.7	<1.0	<1.0	<1.0	<1.3

Radionuclide		**GRC 8**	**GRC 9**	**GRC 10**	**GRC 11**	**GRC 13**	**GRC 14**	**GRC 15**
^{40}K	2020	31 ± 1	99 ± 3	223 ± 8	83 ± 3	118 ± 4	110 ± 5	22.4 ± 0.8
	2021	80 ± 2	80 ± 2	168 ± 4	154 ± 4	164 ± 5	128 ± 3	77 ± 2
^{238}U	2020	36 ± 3	2 ± 1	23 ± 3	<13.5	34 ± 4	33 ± 4	<37
	2021	18 ± 1	27 ± 2	13 ± 1	39 ± 3	57 ± 4	23 ± 2	19 ± 1
^{226}Ra	2020	<2.9	<2.3	<3.0	<2.5	<1.9	<3.3	<2.8
	2021	<1.5	<1.4	3.5 ± 0.6	<2	<2.6	<2.1	3.0 ± 0.5
^{210}Pb	2020	<147	29 ± 9	<161	<110	130 ± 40	<135	<129
	2021	130 ± 40	<65	220 ± 60	<70	<104	90 ± 20	<61
^{232}Th	2020	<7.2	<0.4	<7.0	<5.3	6 ± 1	<7.7	<6.3
	2021	<3.0	<2.8	<3.7	<3.4	7 ± 1	7 ± 1	<2.9
^{137}Cs	2020	<1.7	<0.1	<1.8	<1.0	<1.4	<1.5	<1.4
	2021	<0.6	<0.5	<1.0	<0.8	<1.3	<1.0	<0.9

Value < X in table means that the measured value was below the detection limit (DL), the smallest value needed to quantify the result. GRC1-GRC8 represent novel seed-based *M. sinensis* x *M. sinensis* hybrids, provided by WUR, Netherlands; GRC10, GRC 11, GRC13, GRC14 are novel seed-based *M. sinensis x M. sacchariflorus* hybrids provided by IBERS, UK; rhizome-based *M. x giganteus* GRC9 (commercial) and GRC15 (TV1) were provided by Terravesta.

detection limit (DL) in both 2020 and 2021, and in 3 hybrids it was below the DL in both samples. The average measured activity concentration in 2020 samples was 92.7 Bq kg^{-1}, and in 2021 samples it was 92.3 Bq kg^{-1}. The highest measured activity concentration of ^{40}K was 223 (±8) Bq kg^{-1}. The results indicate that these hybrids intake relatively low amount of K.

All measurements of the activity concentration of ^{137}Cs were below the DL. Considering the difference between the ^{137}Cs activity concentrations in 2020 and 2021, it would indicate that it resulted from increased leaching into the deeper layers of soil (possibly as a result of deep ploughing), and not from the transfer into the miscanthus plants.

The activity concentration of ^{232}Th was measured above the DL in both 2020 and 2021 sample in only one hybrid. In 5 other hybrids, the activity concentration of ^{232}Th was measured in one of the samples. Since ^{232}Th levels in soil are close to the world average [42], and not elevated, the results are not unexpected.

Activity concentrations of the long-lived radionuclides – members of the uranium decay chain are not secular equilibrium, which should be expected. ^{238}U and ^{226}Ra enter the miscanthus hybrids by using different chemical and biological processes, whereas ^{210}Pb originates mostly from air [18,40].

In samples from both 2020 and 2021, activity concentration of ^{210}Pb was, on average, the highest, with the average activity concentration of 123 Bq kg^{-1} in 10 samples where it could be measured, and the average DL of 115 Bq kg^{-1} in 18 samples where the activity concentration was below the DL. The latter means that the activity concentration of ^{210}Pb in miscanthus plants was higher than in soil which indicates that the primary source of ^{210}Pb is the atmosphere, from where the ^{210}Pb is deposited either by precipitation or through wind, either in form of ^{222}Rn, ^{210}Pb or one of the short-lived radionuclides in-between.

Activity concentration of ^{226}Ra was above the DL in only 5 samples, all sampled in year 2021. The same as for ^{232}Th, since ^{226}Ra, the levels in soil are close to the world average [42], and not elevated.

Activity concentration of ^{238}U was measured above the DL in both years in half of the hybrids, and only in 1 hybrid it was found to be below the DL in both years. The average measured activity concentration of ^{238}U was 25.2 Bq kg^{-1} in year 2020, and 31.6 Bq kg^{-1} in year 2021. These results are very interesting, since the activity concentration is much higher than for ^{232}Th and ^{226}Ra. The latter result is something to follow, to confirm during the following years whether the observed is a real effect.

4.3. Transfer factors (soil to plant)

The potential of a plant to be used in phytoremediation does not merely depend on the concentration of the element in the plant. It also depends on the transfer and the accumulation ability of the target element [14,46]. One of the significant parameters widely used in the evaluation of internal radiation dose is the soil-to-plant transfer factor (TF). The TF, i.e., the ratio of activity concentration of

Table 6
Transfer factor (TF) ranges in 2020 and 2021.

Radionuclide		GRC 1	GRC 2	GRC 3	GRC 4	GRC 5	GRC 6	GRC 7
^{40}K	2020	0.12	<0.02	<0.03	<0.03	0.14	<0.04	<0.04
	2021	0.04	<0.02	0.04	0.14	0.07	<0.02	<0.02
^{238}U	2020	0.35	<0.20	0.46	<0.30	<0.25	0.37	<0.29
	2021	0.37	<0.19	0.71	0.60	0.55	<0.13	0.73
^{226}Ra	2020	<0.04	<0.04	<0.05	<0.06	<0.05	<0.06	<0.07
	2021	<0.05	<0.05	<0.04	<0.03	0.07	0.05	0.05
^{232}Th	2020	0.10	<0.09	<0.13	<0.13	<0.16	<0.13	<0.14
	2021	<0.09	<0.09	0.10	<0.07	0.22	0.08	<0.08
^{137}Cs	2020	<0.11	<0.15	<0.19	<0.16	<0.19	<0.22	<0.21
	2021	<0.16	<0.13	<0.09	<0.12	<0.12	<0.12	<0.16
Radionuclide		**GRC 8**	**GRC 9**	**GRC 10**	**GRC 11**	**GRC 13**	**GRC 14**	**GRC 15**
^{40}K	2020	0.05	0.17	0.39	0.14	0.20	0.19	0.04
	2021	0.14	0.14	0.30	0.28	0.29	0.23	0.14
^{238}U	2020	0.57	0.03	0.37	<0.21	0.54	0.52	<0.59
	2021	0.29	0.44	0.21	0.63	0.92	0.37	0.31
^{226}Ra	2020	<0.05	<0.04	<0.05	<0.05	<0.03	<0.06	<0.05
	2021	<0.03	<0.03	0.07	<0.04	<0.05	<0.04	0.06
^{232}Th	2020	<0.14	<0.01	<0.13	<0.10	0.12	<0.15	<0.12
	2021	<0.06	<0.05	<0.07	<0.07	0.14	0.14	<0.06
^{137}Cs	2020	<0.18	<0.01	<0.19	<0.10	<0.14	<0.15	<0.14
	2021	<0.07	<0.06	<0.12	<0.10	<0.16	<0.12	<0.11

For the radionuclides where some of the measured activity concentrations were below the DL, the minimum value in the range is expressed as < ("less than") smallest DL divided by the activity concentration in soil. If all the measured activity concentrations were below the DL, instead of a range, only one value is given, expressed as < ("less than") largest DL divided by the activity concentration in soil; GRC1-GRC8 represent novel seed-based *M. sinensis* x *M. sinensis* hybrids, provided by WUR, Netherlands; GRC10, GRC 11, GRC13, GRC14 are novel seed-based *M. sinensis x M. sacchariflorus* hybrids provided by IBERS, UK; rhizome-based *M. x giganteus* GRC9 (commercial) and GRC15 (TV1) were provided by Terravesta.

radionuclide in soil (expressed in Bq kg^{-1}db) and activity concentration of radionuclide in plant (expressed in Bq kg^{-1} db) [16,47] are both affected by soil properties, vegetation type, radionuclide type, and climatic conditions. As they indicate the degree of uptake of radionuclides from soil to plants, the physiological variability of plants (differences between types and species), are likely sources of TF variations [47].

From the data shown in Tables 2 and 3, the transfer factors (TFs) were calculated. TF cannot be calculated for ^{210}Pb. Considering relatively high activity concentration of ^{210}Pb in miscanthus plants compared to the ^{210}Pb present in the soil, and the fact that ^{210}Pb in plants often is deposited from the atmosphere, and not absorbed from the soil (see, for example, discussion on ^{210}Pb in maize in Sarap et al. [16] we cannot reject the assumption that at least a part of the ^{210}Pb present in miscanthus came from the atmosphere, and not from the soil. Table 6 shows the TF ranges for different miscanthus hybrids.

The ranges of the TFs shown in Table 6 are comparable to the ranges given in Refs. [14–16,48–51]. In general, a TF range of 0.001–10 may encompass the most soil-to-plant TF values, considering different plant species and types, soil types, radionuclide origin, and climatic conditions. Furthermore, the researchers found that radioisotope uptake during the middle or late growth stages resulted in higher TF values than those during the early growth stage [16]. Compared to the studies dealing with miscanthus hybrids [14,48], the results in this study were found to be in the lower end of the intervals. However, both latter studies investigated miscanthus growing on uranium tailings, meaning that the soil contained very high concentrations of naturally occurring radionuclides, and further study is needed to assess how TFs depend on the activity concentration of radionuclides in soil.

4.4. *Possible management route of miscanthus contaminated with radionuclides*

Miscanthus is currently being used for either the production of bioenergy or bioproducts. Lately, due to its high potential, it has been considered as a phytoremediation tool, specifically, for phytoextraction. However, since phytoextraction involves contaminant accumulation in the crop biomass during the plant growth, when being used for this purpose, the harvested biomass becomes a highly contaminated biowaste that could be found as a secondary pollution source if mishandled [52–55]. Hence, appropriate management methods should be applied and a serious approach for the disposal and utilization of the biomass contaminated with radionuclides should be taken. Currently, most of the available studies focus on the management of the plants contaminated with heavy metals, whereas very scarce data if any is available on the management of the plants contaminated with radionuclides. The biomass quality after the phytoremediation process and the accumulation of radionuclides from soil is not desirable for its most common subsequent use - energy production exclusively. I.e. after burning the miscanthus biomass to obtain energy, most of the radionuclides still remain in the ash, which may be considered as a radioactive material and its further use and deposition possibilities may be restricted. Such value chains are sensitive to the accumulation rate of radionuclides from soil since both, the functionality and efficacy of the phytoremediation process are based on a high rate of pollutant accumulation, whereas the end-use options are often sensitive to their high concentrations. Therefore, this paper analyses one of the possible routes for its environmentally safe further utilization – the construction sector. The analysis is limited to the radiological suitability and related limitations for further use in the construction sector;

Table 7
Activity concentration index *I* in the ash of miscanthus hybrids.

		GRC 1	GRC 2	GRC 3	GRC 4	GRC 5	GRC 6	GRC 7
I	2020	1.36 ± 0.00	*	*	*	0.83	*	*
	2021	0.43 ± 0.00	*	2.21 ± 0.01	1.28 ± 0.00	3.66 ± 0.01	1.19 ± 0.00	0.25 ± 0.00
		GRC 8	**GRC 9**	**GRC 10**	**GRC 11**	**GRC 13**	**GRC 14**	**GRC 15**
I	2020	0.33 ± 0.00	0.67 ± 0.00	1.89 ± 0.00	0.38 ± 0.00	1.89 ± 0.01	0.89 ± 0.00	0.19 ± 0.00
	2021	1.15 ± 0.00	1.40 ± 0.00	1.98 ± 0.00	1.71 ± 0.00	2.98 ± 0.01	2.51 ± 0.01	2.19 ± 0.00

*refers to the activity concentrations of all the radionuclides relevant for *I* (^{226}Ra, ^{232}Th and ^{40}K) below the DL. GRC1-GRC8 represent novel seed-based *M. sinensis* x *M. sinensis* hybrids, provided by WUR, Netherlands; GRC10, GRC 11, GRC13, GRC14 are novel seed-based *M. sinensis x M. sacchariflorus* hybrids provided by IBERS, UK; rhizome-based *M. x giganteus* GRC9 (commercial) and GRC15 (TV1) were provided by Terravesta.

hence, mineral and chemical suitability was not part of this study. Such utilization route complies with several UN SDGs, including good health and well-being, responsible consumption and production, as well as clean energy. Moreover, further use of energy production by-products is in line with the cascade utilization of biomass and circular bioeconomy EU supporting schemes. On the other side, reducing the energy consumption in buildings is one of the objectives set by the European Union in its roadmap towards a low carbon economy in 2050. The use of waste or biomass to produce biobased materials used in the building sector reduces the greenhouse gas emissions associated with the production of construction materials; due to its low-CO_2 impact, such products are often investigated during the last several years [56]. By using data for the ash content in miscanthus hybrids (Table 2), the activity concentration index *I* for each of the hybrids was determined (Table 7).

Table 7 shows that the activity concentration index *I* for most of the hybrids is higher than 1. That means that the use of ash has certain limitations, similar to the utilization of ash from other sources such as coal or wood. The ash from the investigated miscanthus hybrids could be used in the construction industry, in mixtures with other materials, so that the activity concentration index from the final material (i.e. concrete, tiles) satisfies eq. (2). Depending on the use of the material, it is acceptable to have the activity concentration index higher than 1 (for example, it may be permissible for material used in road construction). The fact that the activity concentrations of all the radionuclides relevant for *I* (^{226}Ra, ^{232}Th and ^{40}K) were below the DL means that such sample is usually considered safe, but strictly conservative approach may require calculation of *I* using the DLs. It needs to be noted that the results shown in Table 7 presumed that all the radionuclides measured in the miscanthus hybrids remain in the ash, which is the presumption used when assessing coal for coal-fired power plants and the potential for further use of coal ash.

5. Conclusion

Fourteen novel miscanthus hybrids were planted on a freshly deep ploughed field, and their radionuclide remediation potential was determined during the two consecutive years. The radioactive content of soil was found to be similar to the global average. In order to have a better insight into the miscanthus phytoremediation potential, transfer factors for radionuclides were calculated.

The transfer factors were found to be very low for ^{226}Ra ($\leq$0.07), but similar transfer factors for ^{226}Ra were measured for a variety of plants. Transfer factors for ^{40}K ($\leq$0.39) and for ^{232}Th ($\leq$0.21) were in the lower limits for these radionuclides, but not unprecedented, as were possible transfer factors for ^{137}Cs ($<$0.22). Transfer factors for ^{238}U were the highest of all the measured ones ($\leq$0.92), but not too high compared to the measurements in other studies. For ^{210}Pb, the transfer factors were not calculated, since the expectation is that a significant part of the measured quantity came from the air, and not through the soil.

Furthermore, from the sustainability and circular bioeconomy approach, utilization of the contaminated biomass was assessed. Radiological content in miscanthus was found to be high enough, whereas the ash content was found to be low enough that it can be expected that miscanthus ash could be considered as a NORM; even when grown on non-contaminated land, and its further use could be obtained under the controlled conditions, similar to the use of ash from the coal-fired power plants in the construction materials.

Data availability statement

Data associated with the study has not been deposited into a publicly available repository. Data will be made available on request.

CRediT authorship contribution statement

Vanja Jurišić: Writing – original draft, Conceptualization. **Davor Rašeta:** Writing – original draft, Investigation. **Mislav Kontek:** Visualization, Investigation. **John Clifton-Brown:** Supervision. **Luisa M. Trindade:** Writing – review & editing. **Isabelle Lamy:** Writing – review & editing, Data curation. **Annie Guerin:** Investigation. **Andreas Kiesel:** Writing – review & editing. **Ana Matin:** Visualization. **Tajana Krička:** Resources. **Branko Petrinec:** Writing – review & editing, Resources, Methodology.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Vanja Jurisic reports financial support was provided by European Regional Development Fund. Vanja Jurisic reports financial support was provided by European Commission and BBI consortium. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The research was financed by the OP "Competitiveness and Cohesion" 2014–2020, project KK.01.1.1.07.0078 „*Sustainable biogas production by substituting corn silage with agricultural energy crops*", and by the European Commission and BBI consortium *via* H2020 BBI-DEMO project No. 745012 „*GRowing Advanced industrial Crops on marginal lands for biorEfineries - GRACE*".Some of the results of this research were obtained using the facilities and equipment funded within the European Regional Development Fund project KK.01.1.1.02.0007 "*Research and Education Centre of Environmental Health and Radiation Protection – Reconstruction and Expansion of the Institute for Medical Research and Occupational Health*".

References

[1] F.O. Ugbede, O.D. Osahon, Soil-to-plant transfer factors of ^{238}U and ^{232}Th in rice from Ezillo paddy fields, Ebonyi State, Nigeria, J. Environ. Radioact. 233 (2021) 106606, https://doi.org/10.1016/j.jenvrad.2021.106606.

[2] E. Meers, S. Van Slycken, K. Adriaensen, A. Ruttens, J. Vangronsveld, G. Laing, N. Du Witters, T. Thewys, F.M.G. Tack, The use of bio-energy crops (Zea mays) for 'phytoattenuation' of heavy metals on moderately contaminated soils: a field experiment, Chemosphere 78 (2010) 35e41, https://doi.org/10.1016/j.chemosphere.2009.08.015.

[3] J. Korzeniowska, E. Stanislawska-Glubiak, Phytoremediation potential of *Miscanthus x giganteus* and *Spartina pectinata* in soil contaminated with heavy metals, Environ. Sci. Pollut. Res. (2015), https://doi.org/10.1007/s11356-015-4439-1.

[4] A. Mahar, P. Wang, A. Ali, M.K. Awasthi, A.H. Lahori, Q. Wang, R. Li, Z. Zhang, Challenges and opportunities in the phytoremediation of heavy metals contaminated soils: a review, Ecotoxicol. Environ. Saf. 126 (2016) 111e121, https://doi.org/10.1016/j.ecoenv.2015.12.023.

[5] W. Nakbanpote, O. Meesungnoen, M.N.V. Prasad, Potential of ornamental plants for phytoremediation of heavy metals and income generation, in: Bioremediation and Bioeconomy, 2016, https://doi.org/10.1016/B978-0-12-802830-8.00009-5.

[6] V. Pidlisnyuk, T. Stefanovska, E.E. Lewis, L.E. Erickson, L.C. Davis, Miscanthus as a productive biofuel crop for phytoremediation, Crit. Rev. Plant Sci. 33 (1) (2014) 1–19, https://doi.org/10.1080/07352689.2014.847616.

[7] B. Barbosa, S. Boléo, S. Sidella, J. Costa, M.P. Duarte, B. Mendes, S.L. Cosentino, A.L. Fernando, Phytoremediation of heavy metal-contaminated soils using the perennial energy crops Miscanthus spp. and *Arundo donax* L, BioEnergy Research 8 (4) (2015) 1500–1511, https://doi.org/10.1007/s12155-015-9688-9.

[8] J. Antonkiewicz, B. Kolodziej, E.J. Bielinska, The use of reed canary grass and giant miscanthus in the phytoremediation of municipal sewage sludge, Environ. Sci. Pollut. Control Ser. 23 (10) (2016) 9505–9517, https://doi.org/10.1007/s11356-016-6175-6.

[9] V. Pidlisnyuk, P. Shapoval, Ž. Zgorelec, T. Stefanovska, O. Zhukov, Multiyear phytoremediation and dynamic of foliar metal(loid)s concentration during application of Miscanthus× giganteus Greef et Deu to polluted soil from Bakar, Croatia, Environ. Sci. Pollut. Control Ser. 27 (25) (2020) 31446–31457, https://doi.org/10.1007/s11356-020-09344-5.

[10] G. Andrejić, J. Šinžar-Sekulić, M. Prica, Ž. Dželetović, T. Rakić, Phytoremediation potential and physiological response of *Miscanthus* × *giganteus* cultivated on fertilized and non-fertilized flotation tailings, Environ. Sci. Pollut. Control Ser. 26 (33) (2019) 34658–34669, https://doi.org/10.1007/s11356-019-06543-7.

[11] A. Zhao, L. Gao, B. Chen, L. Feng, Phytoremediation potential of *Miscanthus sinensis* for mercury-polluted sites and its impacts on soil microbial community, Environ. Sci. Pollut. Control Ser. 26 (34) (2019) 34818–34829, https://doi.org/10.1007/s11356-019-06563-3.

[12] T. Didier, L.G. Philippe, H. Sonia, B. Amar, M.C. Claudia, F. Jairo, Prospects of *Miscanthus x giganteus* for PAH phytoremediation: a microcosm study, Ind. Crop. Prod. 36 (1) (2012) 276–281, https://doi.org/10.1016/j.indcrop.2011.10.030.

[13] A. Mamirova, V. Pidlisnyuk, A. Amirbekov, A. Ševců, A. Nurzhanova, Phytoremediation potential of Miscanthus sinensis And. in organochlorine pesticides contaminated soil amended by Tween 20 and Activated carbon, Environ. Sci. Pollut. Control Ser. 28 (13) (2021) 16092–16106, https://doi.org/10.1007/s11356-020-11609-y.

[14] X. Yan, Uptake of radionuclide thorium by twelve native plants grown in uranium mill tailings soils from south part of China, Nucl. Eng. Des. 305 (2016) 80–83, https://doi.org/10.1016/j.nucengdes.2016.04.019.

[15] H.H. Azeez, H.H. Mansour, S.T. Ahmad, Transfer of natural radioactive nuclides from soil to plant crops, Appl. Radiat. Isot. 147 (2019) 152–158, https://doi.org/10.1016/j.apradiso.2019.03.010.

[16] N.B. Sarap, J.D. Krneta Nikolić, J.Đ. Trifković, M.M. Janković, Assessment of radioactivity contribution and transfer characteristics of natural radionuclides in agroecosystem, J. Radioanal. Nucl. Chem. 323 (2020) 805–815, https://doi.org/10.1007/s10967-019-06986-9.

[17] M.B.S. Singh, D. Singh, N.K. Dhal, Enhanced phytoremediation strategy for sustainable management of heavy metals and radionuclides, Case Studies in Chemical and Environmental Engineering 5 (2022) 100176, https://doi.org/10.1016/j.cscee.2021.100176.

[18] B. Petrinec, M. Šoštarić, D. Babić, The role of physics in radioecology and radiotoxicology, Arh. Hig. Rada. Toksikol. 70 (2019) 3–13, https://doi.org/10.2478/aiht-2019-70-3225.

[19] Food Standards Agency, The removal of post-chernobyl sheep controls, Food Standards Agency Open Board – 20 March 2012, FSA, Great Britain..

[20] L. Yan, Q. Van Le, C. Sonne, Y. Yang, H. Yang, H. Gu, N. Ling Ma, S. Shiung Lam, W. Peng, Phytoremediation of radionuclides in soil, sediments and water, J. Hazard Mater. 407 (2021) 124771, https://doi.org/10.1016/j.jhazmat.2020.124771.

[21] N. Scarlat, M. Banja, Possible impact of 2020 bioenergy targets on European Union land use. A scenario-based assessment from national renewable energy action plans proposals, Renew. Sustain. Energy Rev. 18 (2013) 595–606, https://doi.org/10.1016/j.rser.2012.10.040.

[22] M. Pogrzeba, J. Krzyżak, S. Rusinowski, S. Werle, A. Hebner, A. Milandru, Case study on phytoremediation driven energy crop production using Sida hermaphrodita, Int. J. Phytoremediation 20 (12) (2018) 1194–1204, https://doi.org/10.1080/15226514.2017.1375897.

[23] I. Alkorta, J.M. Becerri, C. Garbisu, Recovery of soil health: the Ultimate goal of soil remediation processes, in: G. Płaza (Ed.), Trends in Bioremediation and Phytoremediation, Research Signpost, Kerala, India, 2010, pp. 1–9.

[24] J. Clifton-Brown, A. Hastings, M. Mos, et al., Progress in upscaling Miscanthus biomass production for the European bio-economy with seed-based genotypes, GCB Bioenergy 9 (1) (2017) 6–17, https://doi.org/10.1111/gcbb.12357.

[25] I. Lewandowski, The role of perennial biomass crops in a growing bioeconomy, in: S. Barth, D. Murphy-Bokern, O. Kalinina, G. Taylor, M. Jones (Eds.), Perennial Biomass Crops for a Resource-Constrained World, Springer, Cham, 2016, https://doi.org/10.1007/978-3-319-44530-4_1.

[26] E. Magenau, A. Kiesel, J. Clifton-Brown, I. Lewandowski, Influence of cutting height on biomass yield and quality of miscanthus genotypes, GCB Bioenergy 13 (10) (2021) 1675–1689, https://doi.org/10.1111/gcbb.12881.

[27] J. Clifton-Brown, K.-U. Schwarz, D. Awty-Carroll, A. Iurato, H. Meyer, J. Greef, J. Gwyn, M. Mos, C. Ashman, C. Hayes, L. Huang, J. Norris, C. Rodgers, D. Scordia, R. Shafiei, M. Squance, T. Swaller, S. Youell, S. Cosentino, R. Flavell, I. Donnison, P. Robson, Breeding strategies to improve Miscanthus as a sustainable source of biomass for bioenergy and biorenewable products, Agronomy 9 (11) (2019) 673, https://doi.org/10.3390/agronomy9110673.

[28] D. Awty-Carroll, E. Magenau, M. Al Hassan, E. Martani, M. Kontek, P. van der Pluijm, C. Ashman, E. de Maupeou, J. McCalmont, G.-J. Petrie, C. Davey, K. van der Cruijsen, V. Jurišić, S. Amaducci, I. Lamy, A. Shepherd, J. Kam, A. Hoogendam, M. Croci, O. Dolstra, A. Ferrarini, I. Lewandowski, L.M. Trindade, A. Kiesel, J.C. Clifton-Brown, Yield performance of 14 novel inter- and intra-species Miscanthus hybrids across Europe, GCB Bioenergy 00 (2023) 1–25, https://doi.org/10.1111/gcbb.13026.

[29] K. van der Cruijsen, M. Al Hassan, G. van Erven, O. Dolstra, L.M. Trindade, Breeding targets to improve biomass quality in miscanthus, Molecules 26 (2) (2021) 254, https://doi.org/10.3390/molecules26020254.

[30] HRN EN ISO 18134-2, Solid Biofuels - Determination of Moisture Content - Oven Dry Method - Part 2: Total Moisture - Simplified Method, 2017, Croatian Standards Institute, Croatia, 2017 (in Croatian).

[31] ISO 11277:2020, Soil Quality — Determination of Particle Size Distribution in Mineral Soil Material — Method by Sieving and Sedimentation, International Organization for Standardization, Switzerland, 2020.

[32] ISO 11465:1993, Soil Quality — Determination of Dry Matter and Water Content on a Mass Basis — Gravimetric Method, International Organization for Standardization, Switzerland, 1993.

[33] ISO 10693:1995, Soil Quality — Determination of Carbonate Content — Volumetric Method, International Organization for Standardization, Switzerland, 1995.

[34] ISO 10694:1995, Soil Quality — Determination of Organic and Total Carbon after Dry Combustion (Elementary Analysis), International Organization for Standardization, Switzerland, 1995.

[35] ISO 13878:1998, Soil Quality — Determination of Total Nitrogen Content by Dry Combustion ("elemental Analysis, International Organization for Standardization, Switzerland, 1998.

[36] HRN EN ISO 18122, Solid Biofuels — Determination of Ash Content, 2015, Croatian Standards Institute, Croatia, 2015 (in Croatian).

[37] HRN EN ISO 15148, Solid Biofuels - Determination of the Content of Volatile Matter, 2010, Croatian Standards Institute, Croatia, 2010. in Croatian.

[38] HRN EN ISO 18125, Solid Biofuels — Determination of Calorific Value, 2017, Croatian Standards Institute, Croatia, 2017. in Croatian.

[39] M. Šoštarić, B. Petrinec, M. Avdić, L.J. Petroci, M. Kovačić, Ž. Zgorelec, B. Skoko, T. Bituh, J. Senčar, G. Branica, Z. Franić, I. Franulović, D. Rašeta, I. Bešlić, D. Babić, Radioactivity of soil in Croatia I: naturally occurring decay chains, Arh. Hig. Rada. Toksikol. 72 (2021) 6–14, https://doi.org/10.2478/aiht-2021-72-3439.

[40] M. Šoštarić, B. Petrinec, M. Avdić, L.J. Petroci, M. Kovačić, Ž. Zgorelec, B. Skoko, T. Bituh, J. Senčar, G. Branica, Z. Franić, I. Franulović, D. Rašeta, I. Bešlić, D. Babić, Radioactivity of soil in Croatia ^{137}Cs, ^{40}K, and absorbed dose rate, Arh. Hig. Rada. Toksikol. 72 (2021) 15–22, https://doi.org/10.2478/aiht-2021-72-3440.

[41] X. Yan, X. Luo, Radionuclides distribution, properties, and microbial diversity of soils in uranium mill tailings from southeastern China, J. Environ. Radioact. 139 (2015) 85–90, https://doi.org/10.1016/j.jenvrad.2014.09.019.

[42] UN, United Nations Scientific Committee on the Effects of Atomic Radiation UNSCEAR 2008 Report to the General Assembly, with Scientific Annexes, Volume I: (Sources) Report to the General Assembly, Scientific Annexes A and B. United Nations, 2010, 978-92-1-142274-0, https://www.unscear.org/docs/publications/2008/UNSCEAR_2008_Report_Vol.I-CORR.pdf.

[43] Ž. Zgorelec, M. Šoštarić, D. Babić, I. Šestak, M. Mesić, A. Perčin, B. Petrinec, Effects of fertilisation on radionuclide uptake by maize from an acidic soil in northwestern Croatia, 8, Soil Tillage Res. 212 (2021) 105030, https://doi.org/10.1016/j.still.2021.105030.

[44] UNSCEAR, Report: "Report of the United Nations Scientific Committee on the Effects of Atomic Radiation", Annex B: Environmental Contamination, 1966 paragraph 104.

[45] R. Fujiyoshi, S. Sawamura, Mesoscale variability of vertical profiles of environmental radionuclides (^{40}K, ^{226}Ra, ^{210}Pb and ^{137}Cs) in temperate forest soils in Germany, Sci. Total Environ. 320 (2–3) (2004) 177–188, https://doi.org/10.1016/j.scitotenv.2003.08.007.

[46] D. Kobayashi, T. Okouchi, M. Yamagami, T. Shinano, Verification of radiocesium decontamination from farmlands by plants in Fukushima, J. Plant Res. 127 (2014) 51–56, https://doi.org/10.1007/s10265-013-0607-x.

[47] M.Y. Mostafa, N.F. Kadhim, H. Ammer, Y. Baqir, The plant transfer factor of natural radionuclides and the soil radiation hazard of some crops, Environ. Monit. Assess. 193 (6) (2021) 1–11, https://doi.org/10.1007/s10661-021-09061-7.

[48] G. Song, Q. Zhu, M. Lu, D. Chen, Y. Chen, Natural radioactivity in miscanthus floridulus plant from the uranium tailing pile at Guangdong, south China, J. Appl. Math. Phys. 2 (2014) 848–854, https://doi.org/10.4236/jamp.2014.29095.

[49] M. Haque, J. Ferdous, Transfer of natural radionuclides from soil to plants in Savar Dhaka, Spanish Journal of Soil Science (SJSS) 7 (2017) 133–145, https://doi.org/10.3232/sjss.2017.v7.n2.05.

[50] F.O. Ugbede, B.C. Aduo, O.N. Ogbonna, O.G. Ekoh, Natural Radionuclides, Heavy Metals and Health Risk Assessment in Surface Water of Nkalagu River Dam with Statistical Analysis, 2020, https://doi.org/10.1016/j.sciaf.2020.e00439. Scientific African. 8:e00439.

[51] M. Tamaoki, T. Yabe, J. Furukawa, M. Watanabe, K. Ikeda, I. Yasutani, T. Nishizawa, Comparison of potentials of higher plants for phytoremediation of radioactive Cesium from contaminated soil, Environ. Control Biol. 54 (1) (2016) 65–69, https://doi.org/10.2525/ecb.54.65.

[52] H.I. Gomes, Phytoremediation for bioenergy: challenges and Opportunities, Environmental Technology Reviews 1 (1) (2012) 59–66, https://doi.org/10.1080/09593330.2012.696715.

[53] R. Attinti, K.R. Barrett, R. Datta, D. Sarkar, Ethylenediaminedisuccinic acid (EDDS) enhances phytoextraction of lead by vetiver grass from contaminated residential soils in a panel study in the field, Environ. Pollut. 225 (2017) 524–533, https://doi.org/10.1016/j.envpol.2017.01.088.

[54] X.M. Gong, D.L. Huang, Y.G. Liu, G.M. Zeng, R.Z. Wang, J.J. Wei, C. Huang, P. Xu, J. Wan, C. Zhang, Pyrolysis and reutilization of plant residues after phytoremediation of heavy metals contaminated sediments: for heavy metals stabilization and dye adsorption, Bioresour. Technol. 253 (2018) 64–71, https://doi.org/10.1016/j.biortech.2018.01.018.

[55] Z. Liu, K.-Q. Tran, A review on disposal and utilization of phytoremediation plants containing heavy metals, Ecotoxicol. Environ. Saf. 226 (2021) 112821, https://doi.org/10.1016/j.ecoenv.2021.112821.

[56] A.V. Díaz, A.F. López, P.M. Bello Bugallo, Analysis of biowaste-based materials in the construction sector: evaluation of Thermal Behaviour and life Cycle assessment (LCA), Waste and Biomass Valorization 13 (2022) 4983–5004, https://doi.org/10.1007/s12649-022-01820-y.

[57] EC, Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, 2000. https://op.europa.eu/en/publication-detail/-/publication/988f3243-5259-43a5-b621-fbff662deeb0/language-en. European Commission, Bruxelles.